

Philip C. E. Stamp

Statement

and

Readings

MECHANISMS of DECOHERENCE

Philip Stamp

The topic of decoherence has a long history, beginning in the 1950s by Ludwig, Green and others; by the 1970s simple analyses of real systems had begun (Zeh, Simonius), and the idea that the environment could impose ‘selection rules’ (what later was called by Zurek the ‘pointer basis’) had been analysed (see eg., Simonius). A fundamental change in attitude began with the idea of Leggett et al. in the 1980’s, that one could give detailed theoretical predictions for the quantum mechanics of large systems, and that SQUIDs could show macroscopic coherence properties (this was finally seen by Chiorescu et al. in 2003). This forced the well-known ‘measurement problem’ to become a serious topic of investigation in mainstream condensed matter physics; it also stimulated serious experimental tests of the validity of QM on the mesoscopic scale. Perhaps an even more fundamental change in focus was engendered in the discussion, mainly in the last 15 years, of large-scale entanglement, which is required for most kinds of quantum information processing. The quest for quantum communication devices, and for a workable quantum computer, has led to an avalanche of experimental work, in solid-state systems and in quantum-optical systems. As a result of these developments, it has become clear that we need a proper theory of decoherence, which explains not only general features like the connection to quantum measurements, the relationship to dissipative processes, and the possibility that there may be ‘intrinsic decoherence’ processes in Nature, but which also elucidates the detailed mechanisms involved in decoherence, and which can make quantitative predictions for the dynamics of decoherence in real systems.

This talk will begin by reviewing some of the history, and standard questions that arise, such as the relationship between decoherence and the ‘classical’ limit of QM, as well as to quantum measurements, dissipation, and so on. I will briefly discuss some recent ideas such as ‘intrinsic decoherence’ mechanisms, and ‘3rd party decoherence’. However the main focus of this talk will be on the mechanisms of decoherence arising in Nature, and the ways in which one can try to control or suppress them in the lab. I will emphasize decoherence in condensed matter systems, discussing how one can reduce the description of environmental decoherence to one of two models, in which the environment is described as either an oscillator bath or a spin bath. The implications for important contemporary problems in physics are discussed, with emphasis on solid-state qubits, and on ‘quantum critical phenomena’.

Decoherence is often assumed to rule out coherence phenomena at high temperatures. Some of the most interesting examples of large-scale low-temperature quantum coherence will be referred to (involving SQUIDs and magnetic systems) ^[1]. However one can actually get remarkable examples of room-temperature coherence, even in condensed matter systems: I will make some brief remarks on decoherence in some biological systems, with specific reference to the light-harvesting molecules ^[2].

[1] *Experimental work on decoherence in solid-state systems is being reviewed in B Barbara’s talk.*

[2] *Decoherence in biological systems will be discussed in detail in H Briegel’s talk.*

READINGS:

1. PCE Stamp, “The decoherence puzzle”, *Stud. Hist. Phil. Mod Phys.* **37**, 467 (2007) [included in reader]
2. M Simonius, “Spontaneous symmetry-breaking and blocking of metastable states”, *Phys Rev Lett* **40**, 980 (1978)
3. HM Ronnow et al., “Quantum Phase transition of a magnet in a spin bath” *Science* **308**, 389 (2005) [included in reader—see Bernard Barbara’s section]
4. E Collini et al., “Coherently wired light-harvesting in photosynthetic marine algae at ambient temperature” *Nature* **463**, 644 (2010).

The decoherence puzzle

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Abstract

The understanding of decoherence is critical to philosophical debates on several different topics, including measurements, the ‘emergence’ of classical mechanics from quantum mechanics, and the arrows of time. This paper first reviews the basic mechanisms of decoherence in Nature, stressing recent discoveries and the crucial importance of ‘low-energy’ physics. The way in which the interpretation of some recent experiments relates to the problem is also delineated. Finally, some of the more common questions posed by philosophers about decoherence are reformulated, and partial answers are given to these. Throughout the article, the incomplete nature of our understanding is stressed, and the way it depends on several different unresolved questions in both low- and high-energy physics.

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Keywords: Decoherence; Quantum environment; Quantum measurement

1. Introduction

‘Decoherence’ means different things to different people. To most physicists, phase decoherence is a fact of life, important throughout physics (and large parts of chemistry). For those interested in the foundations of quantum mechanics, and for historians and philosophers of physics, decoherence is interesting because of its connection to three main problems, viz., (i) the ‘quantum measurement’ problem; (ii) the ‘emergence’ of classical

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from quantum mechanics (and the hinterland between the two); and (iii) the arrows of time. The philosophical literature on decoherence over the last two decades has mostly focussed on what is sometimes called the ‘decoherence programme’,¹ viz., the effort to explain away problems like the three just mentioned as decoherence phenomena.

Curiously most physicists are not interested in either the decoherence programme or philosophical discussions of it—this in spite of their strong professional interest in decoherence phenomena. One reason for this is probably the empirical bent of most physicists, who quickly lose interest in grand ‘scenarios’ or ‘programmes’ when details are not forthcoming,² or when the scenario is not experimentally testable (necessarily the case for many discussions of the arrows of time (Halliwell et al., 1994; Savitt, 1995; Schulman, 1997; Zeh, 1989)). However there is also somewhat of a schism, between (a) physicists who feel that decoherence is a fairly trivial process, ubiquitous in physics, about which no interesting general statements can be made, and (b) those who feel it is highly non-trivial, but that meaningful discussion requires models that are both realistic and of some generality. The problem here is that most discussions of decoherence in the context of foundational problems (e.g., quantum measurements) have been based on simple idealised models. There is an obvious need for realistic models of complex macroscopic systems, if we are to address any of the three big questions mentioned above. This problem has occasionally been acknowledged by proponents of the decoherence programme. For example, Omnés (1994, Chapter 7), in his book does recognise some of the limitations of simplified models of decoherence—although this does not stop him from claiming some rather general results for macroscopic systems! For remarks on the validity of such results, see Section 4.

Ironically, in the last 20 years a quiet revolution has taken place in our understanding of the quantum mechanics of large systems, and of decoherence phenomena. The revolution is by no means complete, and we will see that several crucial problems remain to be solved. However we now have at hand many of the details missing from earlier discussions. As often happens, many early general assertions made on the basis of the idealised models can now be seen to be misleading, or just plain wrong. Despite this, the newer advances have had little impact on the philosophical literature. This is surprising and unfortunate, since the results do radically change our perspective on at least the first two questions mentioned above, and possibly also the third.

¹For an introduction to the philosophical literature on decoherence, see Bacciagaluppi (2005). What is called the ‘decoherence programme’ by, e.g., Joos et al. (2003) and Zeh (2002), can actually be separated into various strands, depending on whether one is dealing with non-relativistic physics or quantum gravity, and on which question one is interested in (quantum measurements, the interpretation of quantum mechanics, large-scale quantum phenomena, cosmology, etc.). For extensive reviews, see Joos et al. (2003), Omnés (1992, 1994), Zeh (2002) and Zurek (2003), and for reviews of the ‘decoherence histories’ approach, see Griffiths (1984, 1986), Hartle (1991) and Gell-Mann & Hartle (1993). For discussions of the arrows of time, which touch upon the connections to decoherence, the quantum arrow, etc., see Schulman (1997), Zeh (1989), Halliwell, Pérez-Mercader, & Zurek (1994) and Savitt (1995).

²Typically what experimentalists are looking for is testable predictions—or at least something sufficiently precise and realistic that it can be related to some present or future class of experiments. Theorists are also looking for something quantitatively precise, which acquires much greater interest if it is both realistic (i.e., not oversimplified) and of some broad generality. Note that ‘theoretical programmes’ sometimes have a bad name in physics—an attitude summed up in Pauli’s famous letter to Gamow in 1954 (referring to Heisenberg’s ‘programme’ for a unified field theory). Writing “This is to show I can paint like Titian”, he drew a simple rectangle, and then wrote “Only technical details are missing”.

The purpose of this paper is to

- (a) review quickly, for non-specialists,³ what we now know about the physical mechanisms of decoherence, stressing the recent developments and their broader implications (Section 2). Then, in Section 3, I discuss what this means in the lab—how do experiments bear on the fundamental questions mentioned above? The main interest here for philosophers is to see just how much the interpretation of experiments depends on how one feels about the ultimate validity of quantum mechanics. For those wishing to follow up any of the themes mentioned in these two sections, I have given extensive references;
- (b) in the light of the recent developments, to reconsider some of the more general questions mentioned above—concentrating on whether there exist ‘intrinsic’ sources of decoherence in Nature, how decoherence relates to the emergence of classical physics, to irreversibility and dissipation, to quantum measurements, and to the arrows of time (all in Section 4). The conclusion (Section 5) summarises where we are now.

2. Decoherence and quantum relaxation: models, mechanisms, dynamics

Discussions of decoherence usually begin with the interaction of a physical system \mathcal{S} with an environment \mathcal{E} . One imagines that \mathcal{S} starts off in some simple superposition of states, say $\Psi = \sum_j c_j \psi_j$, which upon interaction with \mathcal{E} , becomes entangled with it, so that the final state cannot be decomposed into a product state. Averaging over the environmental variables then produces a full or partial mixture, rather than a superposition of states, for \mathcal{S} . How this all happens in the real world is part of the ‘decoherence problem’, and it is interesting to see how views on this have evolved over the years.

Even before decoherence was discussed as such in the literature, mechanisms for it were being discussed in the context of the measurement problem, in the wake of the analyses of the 1930s of quantum measurements (London & Bauer, 1939/1983; Neumann, 1932/1955; Pauli, 1980). Early discussions of decoherence processes emphasised the role of randomisation of phases, and analysed this in terms of simple models of system/environment interactions, leading to irretrievable loss of phase correlations in the environment.⁴ Very interesting ideas emerged from these discussions, including the possible role of amplification and relaxation, at least in measurements (Daneri et al., 1962, 1966), and the idea that the structure of interactions in the world might inevitably lead to decoherence in certain ‘preferred bases’.⁵ Simple models of decoherence were analysed in some of these papers, including Geiger counters, cloud chambers (an analysis going back

³It is assumed that the reader is familiar with elementary quantum mechanics. An intuitive understanding like that provided by Feynman (1965) is also useful.

⁴The idea of decoherence goes back at least to Ludwig (Born & Ludwig, 1958; Ludwig, 1953, 1958). Another early paper, concentrating on the role of the environment in the measurement problem, is Green (1958). These papers all argued that environmental dephasing (what we now call decoherence) would destroy large-scale quantum behaviour. This idea was picked up and further developed in Daneri, Loinger, & Prosperi (1962, 1966), Zeh (1970, 1973), Joos & Zeh (1985) and Simonius (1978), amongst others.

⁵The idea of preferred bases and preferred states, selected by decoherence, is described in, e.g., Simonius (1978), where these states are called ‘inert states’; and in Zurek (1981, 1982), where they are called ‘pointer states’. See also (Zurek, 2003).

to Mott), Stern–Gerlach experiments, sugar molecules, damped oscillators, etc. However, insofar as any macroscopic features of these examples were discussed, this was done in a very crude way, the aim being to demonstrate that decoherence would *always* suppress quantum interference effects except at the atomic or molecular scale (an assertion repeated (Van Kampen, 1988) as late as 1988).

This old orthodoxy was severely upset at the beginning of the 1980s by the now well-known work of Leggett et al. (Caldeira & Leggett, 1983; Leggett, 1984; Leggett et al., 1987), who pointed out that in fact one could expect superconducting SQUIDs to show quantum tunneling and interference properties at the macroscopic scale—and that moreover, one could test quantum theory at the macroscopic scale in this way. Initial scepticism has yielded in most quarters to the weight of experimental evidence—both macroscopic tunneling and coherence have now been seen in superconductors (see Section 3). In related developments, experimentalists have succeeded in the multi-particle entanglement of atoms in traps (Häffner et al., 2005; Leibfried et al., 2005), as well as superpositions of photon states in cavities (Zhao et al., 2004, and refs. therein); and new schemes, involving ideas like ‘quantum non-demolition’ measurements,⁶ have been employed to reduce decoherence and dissipation effects in optical systems and in large Al bars (for gravity wave experiments).

Leggett et al. used an “oscillator bath” representation of the environment—a ploy first described by Feynman and Vernon (1963) and developed much further by Leggett et al. (Caldeira & Leggett, 1983; Leggett, 1984; Leggett et al., 1987). These models clearly lend themselves to problems in particle and string physics, quantum optics, and cosmology,⁷ and they are also often used in condensed matter systems at low temperatures (Weiss, 1999). In contrast to the qualitative pre-1980 discussions of decoherence, we have a real theory, quantitatively testable on a large variety of systems. This completely changes the nature of both the scientific and the philosophical debates, as we shall see in the rest of this article.

However, in spite of this remarkable success, there is an important quantitative problem, particularly in solid-state systems—when one comes to compare the decoherence rates predicted by Caldeira–Leggett theory with the measured rates, the experimental rates are typically several orders of magnitude larger than theory predicts (see Section 3). This discrepancy indicates that most of the decoherence is coming from somewhere else, in ways not described by oscillator bath models. Whether this constitutes in some way a real problem of principle, particularly for tests of quantum mechanics at the macroscopic scale, is one of the topics addressed herein.

In Section 2.1 the main features of environmentally induced decoherence are explained, with an emphasis on the physical mechanisms responsible. Since there is a widespread belief that *all* decoherence is caused by direct interaction with an environment, in Section 2.2 I briefly outline another way decoherence can happen. The material of Section 2 is essential if one wishes to address the more philosophical questions associated with decoherence.

⁶For discussions of some novel measurement schemes, including quantum non-demolition schemes, see Braginsky & Khalili (1992) and Caves, Thorne, Drever, Sandberg, & Zimmermann (1980).

⁷For some examples of the use of oscillator bath models in cosmology and string theory see Cornwall & Bruinsma (1988), Callan & Freed (1992) and Callan, Felce, & Freed (1993, and refs. therein).

2.1. Quantum environments

2.1.1. Extended environmental modes; oscillator baths

Research on the dynamics of polarons and related problems led Feynman in the early 1960s to a general discussion of the interaction of a quantum system with its background environment. Feynman and Vernon (1963) considered the case where each environmental mode coupled only weakly to the central system. Arguing that for this weak-coupling case, the effect of *any* environment could be mapped to that of a set of oscillators, they treated a model Hamiltonian in which a central system \mathcal{S} , with generalised coordinates P, Q and Hamiltonian $H_0(P, Q)$, interacted with an environment \mathcal{E} of oscillators with generalised coordinates $\{p_q, x_q\}$ and Hamiltonian $H_{\text{env}}^{\text{osc}}(\{p_q, x_q\})$, via a simple bilinear coupling:

$$H_{\text{eff}}^{\text{osc}} = H_0 + H_{\text{int}} + H_{\text{env}}^{\text{osc}}, \quad H_{\text{int}}(Q, \{x_q\}) = \sum_{q=1}^N c_q x_q Q. \quad (1)$$

We assume that the entire Hamiltonian $H_{\text{eff}}(\Omega_0)$ is defined with an ultraviolet cutoff energy Ω_0 . The important points to bear in mind here are:

- (i) the oscillators have bosonic statistics, and typically represent delocalised modes, extending over the whole region of the environment. Typical examples are phonons, magnons, electron–hole pairs, or photons, which are wave-like oscillations of some background field. These are the low-energy modes of the environment—at higher energies the model usually breaks down;
- (ii) the couplings $\{c_q\}$ are weak—in fact $c_q \sim O(N^{-1/2})$, where N is the number of low-energy environmental modes (N is thus proportional to the size of the environmental domain). This typically follows because we must normalise the oscillator wave functions (so they are $\sim O(N^{-1/2})$). Typically N is very big, so that mathematical treatments often just adopt the ‘thermodynamic limit’ $N \rightarrow \infty$. Since the effect of each oscillator to second order is $\sim |c_q|^2 \sim O(1/N)$, their total effect is then independent of N , as it should be in this limit. Thus each oscillator is only very weakly affected by the system, but the system may be quite strongly affected by the oscillators.

Curiously, the work of Feynman and Vernon had no impact whatsoever on the discussion of quantum measurements or decoherence for two decades—possibly because it was phrased in the then unfamiliar language of path integrals, and because the community working on the foundations of quantum mechanics was less interested at that time in detailed models.

At the beginning of the 1980s Caldeira and Leggett (1983) introduced a somewhat generalised Feynman–Vernon model, in which the coupling $\sum_q c_q x_q Q$ was replaced by

$$H_{\text{int}}^{\text{osc}} = \sum_{q=1}^N [F_q(Q)x_q + G_q(P)p_q]. \quad (2)$$

The Hamiltonians (1) and (2) are effective ones, which means amongst other things that the couplings c_q, F_q , and G_q , the oscillator frequencies ω_q , and even the system Hamiltonian H_0 depend not only on the UV cutoff Ω_0 but also on the bath temperature T . This may seem strange to some (particularly readers more at home with the models used in particle physics). Recall however that *all* Hamiltonians in physics are effective ones,

written in a quantum system in terms of operators defined over some restricted Hilbert space, depending implicitly or explicitly on energy cutoffs, temperature, and possibly other boundary conditions.⁸ It is only when dealing with a very rarified medium that one can ignore these complexities.

Caldeira and Leggett gave arguments for the very general applicability of such effective Hamiltonians to systems at low energy (along with specific application to superconducting SQUIDS). Consider some arbitrary environment, with eigenstates $\phi_\alpha(\mathbf{X})$ and eigenenergies ε_α defined over the environment's full multi-dimensional coordinate space \mathbf{X} . Assume the system interacts with this environment via some interaction $V(Q, \mathbf{X})$. Then the arguments go as follows:

- (a) Certainly we can recover an oscillator bath model if the coupling between different eigenstates induced by the interaction $V(Q, \mathbf{X})$ is weak, i.e., under the Feynman–Vernon condition that

$$|V_{\alpha\beta}| \ll |(\varepsilon_\alpha - \varepsilon_\beta)| \quad (3)$$

for all relevant environmental states, where $V_{\alpha\beta} = \int d\mathbf{X} \phi_\alpha^*(\mathbf{X}) V(Q, \mathbf{X}) \phi_\beta(\mathbf{X})$. The oscillator modes then correspond to the transitions between these states, and $\omega_q \equiv (\varepsilon_\alpha - \varepsilon_\beta)$.

- (b) However, even if the weak-coupling condition is not obeyed, we can use a Born–Oppenheimer argument to derive a similar criterion. We first define adiabatic environmental eigenstates $\tilde{\phi}_\alpha(\mathbf{X}, Q)$ and eigenenergies $\tilde{\varepsilon}_\alpha(Q)$, which depend on the instantaneous system coordinate Q . Now suppose that these states have a *fast* dynamics compared to the slower dynamics of the system coordinate Q (formally, that if Q moves on a frequency scale E_0 , then $E_0 \ll \tilde{\varepsilon}_\alpha$). One then defines a fake ‘gauge potential’ $A_{\alpha\beta}$, describing the effect of the slowly changing Q on the bath modes, given by $iA_{\alpha\beta} = \int d\mathbf{X} \tilde{\phi}_\alpha^*(\mathbf{X}) \partial/\partial Q \tilde{\phi}_\beta(\mathbf{X})$; there is no reference to the original interaction between Q and the bath modes, because this has already been incorporated into the renormalised $\tilde{\varepsilon}_\alpha$. Standard manoeuvres then show that we can make a mapping to an oscillator bath provided

$$|A_{\alpha\beta}| \ll |(\tilde{\varepsilon}_\alpha - \tilde{\varepsilon}_\beta)| \quad (4)$$

for all the relevant modes. If (4) is satisfied, then the oscillators now describe transitions between the new adiabatic bath modes, with frequencies $\omega_q \equiv (\tilde{\varepsilon}_\alpha - \tilde{\varepsilon}_\beta)$; and one can also derive the couplings F_q, G_q in terms of the gauge coupling in (4).

- (c) Leggett et al. then argued that the low- T , low-energy quantum dynamics of such a system could be related to its higher T dissipative classical dynamics (cf. Fig. 1). From the classical dissipative dynamics one *infers* a low-energy effective Hamiltonian (having the form (1), with the generalised interaction in (2)); in particular, one finds the form of the couplings in (2). This is crucially important—instead of trying to derive the form of H_{eff} from some theory (a move which is always open to criticism given the

⁸The idea of the ‘effective Hamiltonian’ (or the effective Lagrangian) is rather subtle, and bound up in the recent history of physics with the idea of the renormalisation group (although discussions go back at least to the 19th century). See, e.g., Anderson (1984). For a recent discussion of the effective Hamiltonian, directed to a philosophical readership, see Stamp (to be published).

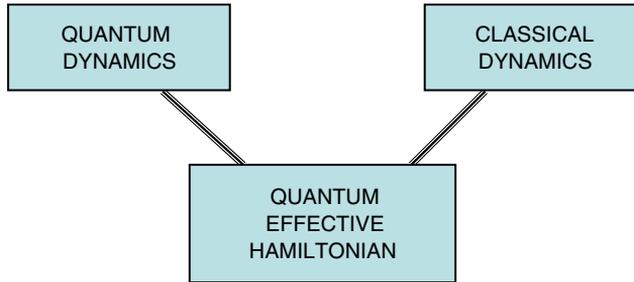


Fig. 1. The epistemological connection between the observable classical (usually high-temperature) dynamics and the low-temperature quantum dynamics (often not so easy to observe), for a system with many degrees of freedom. Both can be derived from the correct quantum effective Hamiltonian. Often (as in the approach of Caldeira and Leggett) one infers the quantum Hamiltonian from experiments on the classical behaviour.

huge complexity of large systems), one instead infers it directly from experiment.⁹ One then *derives* the quantum dynamics of the system from this effective Hamiltonian.

At first glance the assumptions behind the oscillator bath model seem restrictive—small oscillations and weak coupling to each mode, use of a Born–Oppenheimer approximation, etc. However appearances are deceptive—oscillator bath models are quite robust in the real world. A large class of effective Hamiltonians (sometimes called a *universality class*), which will describe many physical systems, can be mapped to models of the oscillator bath type (Dubé & Stamp, 2001). Examples include: (i) itinerant fermion baths (e.g., a bath of interacting conduction electrons), in three, two or one dimensions; (ii) systems having weak higher-order ‘anharmonic’ couplings to extended bath modes—these can be absorbed into modified couplings to a new set of oscillators (the couplings and oscillator frequencies now being very strongly T -dependent); and (iii) systems where bath modes are strongly coupled to the system, provided the condition (4) is not violated (i.e., provided the effective coupling between two environmental states goes to zero fast enough as one reduces the energy difference between them). It is worth remarking here on a point which is crucially important for decoherence. The reduction in the strength of coupling to oscillator bath modes at low energies is a general feature of extended environmental states, whose density of states always goes down with energy, because of decreasing available phase space volume. This means that at as one lowers energies and temperatures towards zero, we can naively expect the decoherence from oscillator baths to also decrease to zero.

We have seen that oscillator bath models of quantum environments are thus much more general than is often assumed in the literature. However they certainly cannot always work, and they clearly fail in many solid-state systems at low temperatures. In order to understand why, we make a little diversion into the real world of low-energy physics.

⁹In Caldeira–Leggett theory, the interaction between system and environment is summarised in a ‘spectral function’ $J(\omega, T)$, a function of frequency and temperature. If the Caldeira–Leggett effective Hamiltonian applies to some physical system, and if one knows $J(\omega, T)$, then the behaviour can be derived theoretically in both classical and quantum regimes. More typically, one *infers* $J(\omega, T)$ from the classical and/or quantum behaviour in experiments.

2.1.2. *Interlude: real condensed matter*

Condensed matter is all around us—we are directly aware of little else. All measuring systems are made from condensed matter. It is clearly messy, and complex structures and order are evident everywhere (not least in living things). As a result, except for the He liquids (which go superfluid at low T and which can be made in essentially completely pure form) and rarified gases and plasmas, the low-energy effective Hamiltonians of *real* condensed matter systems are extraordinarily subtle (and very far from the descriptions usually given in student textbooks). There is a common misunderstanding that these subtleties have to do with ‘dirt’ effects (the ‘squalid state’, in Pauli’s famous phrase). In fact they are mostly intrinsic, for the following reasons (footnote 8):

- (i) *Topology*. Many-particle wave-functions have topological properties which restrict and sometimes control the dynamics. This often leads to new branches of low-energy ‘topological excitations’, with their counterpart in the effective Hamiltonian (Thouless, 1998).
- (ii) *Lattices + interactions*. In solids, electrons are constrained to move between different atomic orbitals. Strong repulsive interactions between electrons can prevent more than one particle per orbital, imposing a highly non-trivial structure on the Hilbert space of the effective Hamiltonian and even causing the low-energy states to localise.
- (iii) *Boundaries or edges*. All systems have boundaries. In conjunction with long-range forces and/or the topological properties of wave-functions, the boundaries and the states localised near them can control the low-energy properties of the whole system.
- (iv) *Frustration*. Interactions between two different pairs of particles or spins are often ‘incompatible’ (i.e., lead to contradictory effects on any one of the particles). The result is typically a large number of almost degenerate low-energy states which hardly communicate.¹⁰ The system can never reach its putative ground state (which then becomes a mere mathematical chimera). Because of frustration, most pure solids, without impurities, are intrinsically disordered. States pile up at low energies—many of these low-energy states are localised (footnote 10).

Clearly none of these effects come from ‘junk’ or ‘dirt’; moreover, because they arise from very general mechanisms, they lead to effects that are ubiquitous in low-temperature experiments. These include peculiar structure in the low-energy density of states, complex and often non-linear long-time relaxation phenomena, including ‘glassy’ behaviour (the freezing out of dynamics caused by frustration), increasingly subtle kinds of quantum ordering as one lowers the temperature, etc. Over the last four decades a phenomenological description has emerged for these low-energy phenomena, in terms of a set of low-energy discrete modes (i.e., each having a discrete finite set of states, often only two, in the energy range of interest), appropriate to localised states (Anderson, 1994; Binder & Young, 1986; Esquinazi, 1998; Mézard et al., 1987). These states interact both amongst themselves, and with the extended ‘oscillator modes’. Thus one ends up with a low energy description in terms of a set of interacting ‘two-level systems’; usually the interactions are fairly weak,

¹⁰The only elementary review of some of the low-energy complexities in real solids seems to be the five short articles by Anderson on ‘spin glasses’ (Anderson, 1994). More sophisticated reviews are by Binder & Young (1986) and Mézard, Parisi, & Virasoro (1987); this latter book also makes the connection with work in computation and biology.

although they can have important effects. There is certainly no universal agreement about this picture (Yu & Leggett, 1988), but in many cases there is extensive evidence that it gives a good description of the low-energy physics (Anderson, 1994; Binder & Young, 1986; Esquinazi, 1998; Mézard et al., 1987). I emphasise again that these effects are pretty much universal in solids, although their effects are sometimes not obvious until very low temperatures. Their effects on ordinary transport and other dissipative properties can be very small (making them almost invisible at higher temperatures), but we shall see that their contribution to decoherence can be very large.

One is often met by surprise at this situation. How, it is asked, can a simple solid show such ‘pathological behaviour’, when after all it is made up electrons, protons, etc., which can be described by a simple continuum theory having none of these complexities? The fallacy in this argument is the assumption that the effective Hamiltonian of a composite system will somehow be analysable into that of its constituents.¹¹ This is not true—the effective theory of the constituents is still an effective theory, applicable only in a certain energy range and assuming a restricted Hilbert space. For this reason neither the vacuum nor the low-energy eigenstates of the high-energy Hamiltonians used in particle physics look anything like a condensed matter system (even though this is physically what a high-energy system becomes if it is cooled!). In many real solids, an infinite hierarchy of effective Hamiltonians, ever more complex, is expected as one lowers the energy scale (footnote 8), and we only have a dim understanding of what their structure might be. In other words, we do not really understand the basic structure of the lowest energy states or Hilbert spaces of most many-body systems. An understanding of this low-energy structure is one of the holy grails of condensed matter physics—in many ways it seems more elusive now than it did 30–40 (or even 100) years ago. One hundred years ago, with the vindication of the atomic hypothesis, but before quantum mechanics, a simple reductionist view of condensed matter looked very reasonable. Thirty to forty years ago, a unification of methods between quantum field theory and condensed matter physics looked imminent—the Ginzburg–Landau–Wilson theory of phase transitions, and the BCS theory of superconductivity, were shaping much of modern particle theory. This unification has happened, but only in the study of ‘simple’ systems. For a more realistic perspective see Anderson (1994), Binder and Young (1986), Esquinazi (1998) and Mézard et al. (1987).

If some day we ever have a “complete theory of everything”, with a ‘universal Hamiltonian’ whose eigenstates (including the ground state) represent the real states of the universe, over all energy scales, then we would presumably find that the low-energy states of this Hamiltonian contain the full complexity of real condensed matter. Right now we have little idea if such a theory would even be meaningful (it is perhaps more likely that the whole Hamiltonian structure will be replaced by something more fundamental). We certainly have not the slightest idea whatsoever what it would look like. Current efforts towards progress range from theory at supra-Planck scale energies, to the exploration of coherence phenomena at temperatures below 10^{-9} K.

¹¹It is commonly argued that the ‘complexity’ of low-energy physics comes only from the large number of constituents (this is certainly the point of view of ‘reductionists’). This argument is refuted in a well-known paper by Anderson (1972), which inspired a very large subsequent literature.

2.1.3. Localised modes: spin baths

We return now to the question at hand, which is to understand the sources of decoherence at low energies in real condensed matter systems. The importance of the previous discussion is that we now see we must deal with the large number of low-energy localised states existing in solids, or more generally, low-energy modes having a finite Hilbert space, with discrete excitations. The general nature of these was described above; they include the eigenstates of nuclear spins, of topological defects, and of various more subtle modes associated with frustration, boundaries, and intrinsic disorder. In any real system there will also be ‘junk’ effects, coming from paramagnetic impurities, ‘charge trap’ excitations, etc. In many systems we may not know exactly what these discrete modes are, but as noted above, their presence is often very obvious in experiments (Anderson, 1994; Binder & Young, 1986; Esquinazi, 1998; Mézard et al., 1987).

Now one can always map a system having a set of M discrete states to a spin system, with spin σ , such that $2\sigma + 1 = M$. Thus we can in all cases describe an environment of these states as a ‘spin bath’ (Prokofev & Stamp, 2000). Spin baths have the following general characteristics:

- (i) The generic model for a quantum system interacting with a spin bath (corresponding to the generic oscillator bath model defined by Eqs. (1) and (2)) has the effective Hamiltonian:

$$H_{\text{eff}}^{\text{sp}}(\Omega_0) = H_0 + H_{\text{int}}^{\text{sp}} + H_{\text{env}}^{\text{sp}}, \quad (5)$$

where $H_0(P, Q)$ describes the system as before; but now the interaction term is a vector coupling to a set of ‘spins’ $\{\sigma_k\}$ (which for simplicity we take here to be two-level systems, i.e., spin- $\frac{1}{2}$ systems):

$$H_{\text{int}}^{\text{sp}} = \sum_k^{N_s} \mathbf{F}_k(P, Q) \cdot \sigma_k, \quad (6)$$

and the spin bath Hamiltonian itself has the form:

$$H_{\text{env}}^{\text{sp}} = \sum_k^{N_s} \mathbf{h}_k \cdot \sigma_k + \sum_{k,k'}^{N_s} V_{kk'}^{\alpha\beta} \sigma_k^\alpha \sigma_{k'}^\beta, \quad (7)$$

with a set of external fields $\{\mathbf{h}_k\}$, and interspin interactions $V_{kk'}$. The generalisation of this model to bath modes having $M > 2$ discrete states is straightforward.

- (ii) Each bath ‘spin’ interacts only weakly with its compatriots—formally we require that $\{|\mathbf{F}_k|\} \gg |V_{kk'}|$. If the $\{\sigma_k\}$ describe localised modes, this is quite typical. The different bath excitation wave-functions do not overlap and can only communicate via weak long-range interactions $V_{kk'}$, whereas there is nothing limiting the size of the $\{|\mathbf{F}_k|\}$ (which are no longer $\sim O(1/N^{1/2})$). The bath dynamics is then under the direct control of the central system (note that inequality (4) is now violated), with its own ‘intrinsic dynamics’ playing second fiddle. Recall that this is exactly opposite to the oscillator bath system, where the intrinsic dynamics of the oscillator bath is only weakly perturbed by the central system, because the oscillator frequencies $\{\omega_q\}$ are much larger than either the $\{c_q\}$ or the F_q, G_q in (2). This situation is illustrated in Fig. 2.

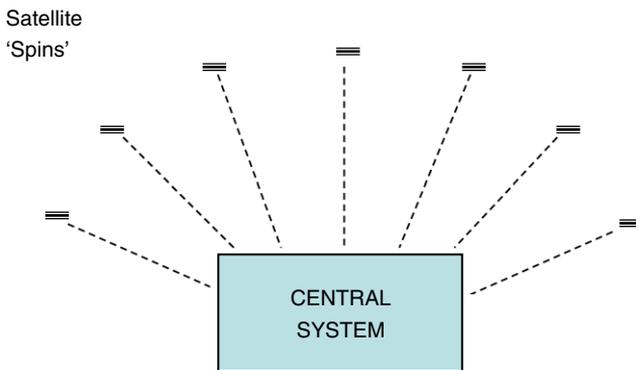


Fig. 2. The ‘spin bath’ environment—a set of satellite spins couples to the central quantum system of interest. The spins typically represent localised modes (not necessarily spins!) in the environment, each with a finite Hilbert space (often two-dimensional). The coupling between spins is weak compared to the coupling of each to the central system.

Clearly under some circumstances we can map the spin bath onto an oscillator bath. For example, if the interactions $V_{kk'}$ are strong (i.e., if $\{|\mathbf{F}_k|\} < |V_{kk'}|$ and if $|\mathbf{h}_k| < |V_{kk'}|$), then the bath spins can couple together to form extended ‘spin waves’, and $H_{\text{eff}}^{\text{sp}}(\Omega_0)$ then maps back to a Caldeira–Leggett model. If the central system dynamical energy scale $E_0 \gg |\mathbf{F}_k|$, then one goes to an anti-adiabatic (or ‘anti-Born–Oppenheimer’) limit, in which the system–bath couplings can be treated perturbatively. One can give more complete criteria for the mapping of spin baths to oscillator baths (Prokofev & Stamp, 2000), which we see must also involve the static fields $\{\mathbf{h}_k\}$.

In real physical systems the coupling energies $|\mathbf{F}_k|$ and static field strengths $\{\mathbf{h}_k\}$ are often spread over a very wide range, particularly in systems with frustration, disorder or impurities (note that ‘impurities’ include nuclear spins, which are almost everywhere; they live in some finite fraction of the nuclei of almost all the elements in solids). We cannot then use either or a Born–Oppenheimer or an anti-Born–Oppenheimer approximation, there are many environmental modes which must be treated directly as localised modes. Because these modes then have characteristic frequencies similar to those of the central system we are interested in, they cause a lot of decoherence.

2.2. Bath-induced decoherence and relaxation

Although the detailed calculation of the dynamics of decoherence is a complicated business, many of the main points can be understood by simple (although qualitative) arguments.

As noted earlier, in the early development of this subject, the idea of decoherence was very much bound up with quantum measurements. Decoherence was, in effect, viewed as a process in which the environment \mathcal{E} ‘measured’ the state of the system \mathcal{S} being decohered, via a transition

$$\sum_j c_j \psi_j \Phi_0 \rightarrow \sum_j c_j \lambda_j \Phi_j, \tag{8}$$

for the combined $\mathcal{S} \otimes \mathcal{E}$, with the final states $\{\Phi_j\}$ of \mathcal{E} uniquely correlated to the original system states $\{\psi_j\}$. The superposition still exists in the combined state of $\mathcal{S} \otimes \mathcal{E}$, but tracing out the environment gives a reduced density matrix $\rho_{ij}^S = |c_j|^2 \delta_{ij}$ for \mathcal{S} , in which all correlations between the ψ_j (i.e., all off-diagonal matrix elements in ρ_{ij}^S , in this particular Hilbert space basis) have disappeared. Recall that the infamous ‘measurement problem’ (d’Espagnat, 1976; Wheeler & Zurek, 1983) centres around states like (8).

Any real environment would not align its states so exactly with those of \mathcal{S} ; such precise correlations can only be engineered by an experimentalist, by deliberate ‘state preparation’ (Margenau, 1973a,b). Nevertheless over the last 50 years there has been great interest in how the environment might cause ρ_{ij}^S to diagonalise in certain preferred bases (in the context of measurement theory these states are called the ‘inert’ or ‘pointer’ states (footnote 5)). The most popular is the basis set $\{Q\}$ of position eigenstates $\{Q\}$, and one assumes that $\rho_{Q,Q}^S$ tends over some timescale to a function which is narrowly focussed around $\delta(Q - Q')$ (e.g., a Gaussian function $\rho_{Q,Q}^S = (1/2\pi\sigma^2)^{1/2} \exp[-(Q - Q')^2/2\sigma]$, with small variance σ^2). This certainly can happen in simple models. The easiest way is to couple some bath to \mathcal{S} with a coupling linear in the system coordinate Q , and the model of a central oscillator coupled bilinearly to a bath of oscillators has been the object of many papers (which usually assume an Ohmic coupling) like Grabert, Schramm, and Ingold (1988).

However these models and their behaviour lack generality, as we will see. Before continuing, it is useful to give some intuition for the dynamics of decoherence, i.e., the time evolution of ρ^S . This is described by a propagator K , which relates the density matrix at some time t_2 to its state at an earlier time t_1 . Now let us go to a particular basis, the position basis, which allows us to look at how K evolves in real space, using the highly intuitive path integral formulation of quantum mechanics (Feynman & Hibbs, 1965; Feynman, Leighton, & Sands, 1965). As discussed by Feynman, one can usefully write (see also Feynman & Vernon, 1963):

$$K(Q_2, Q'_2; Q_1, Q'_1; t, t') = \int_{Q_1}^{Q_2} \mathcal{D}q \int_{Q'_1}^{Q'_2} \mathcal{D}q' e^{-i/\hbar(S_0[q] - S_0[q'])} \mathcal{F}[q, q'], \quad (9)$$

where $\mathcal{F}[q, q']$ embodies all the effects of the bath on the dynamics of \mathcal{S} , after we have averaged over the bath. To interpret (9), suppose first that $\mathcal{F}[q, q'] = 1$, i.e., that the system \mathcal{S} is completely decoupled from the bath, and propagates freely. Then K propagates along two paths $q(\tau)$ and $q'(\tau)$ between the limiting arguments, and in the usual quantum way, one sums over all possible pairs of paths. Thus $\mathcal{F}[q, q']$ is just a *weighting factor*, defined over these two paths, and it couples them. Moreover, $\mathcal{F}[q, q']$ has a simple form; one can always write $\mathcal{F}[q, q'] = \exp[-i\Phi - \Gamma]$, where the phase $\Phi[q, q']$ and ‘damping’ $\Gamma[q, q']$ are real.

Now suppose, for example, that $\mathcal{F}[q, q']$ falls off rapidly when the paths q and q' move apart from each other. Then the density matrix will be forced towards an approximate ‘pointer basis’ in Q -space. Many other behaviours are also possible. The advantage of dealing with $\mathcal{F}[q, q']$ is that it can also be connected in a transparent way with (and calculated from) the effective Hamiltonian, and the behaviour of $\mathcal{F}[q, q']$ is easily visualised.

Decoherence and relaxation in oscillator bath models. The essential properties of $\mathcal{F}[q, q']$ for a system in contact with an oscillator bath were defined by Feynman (Feynman &

Hibbs, 1965; Feynman et al., 1965; Feynman & Vernon, 1963), and as noted above, Caldeira and Leggett were able to relate the quantum and classical dynamics of \mathcal{S} for such environments. This crucial step allows us to understand decoherence for such models in the same terms as we understand ordinary dissipation.

In this way one arrives at the following intuitive picture. The ‘fast’ environmental modes cause little decoherence or dissipation. They simply adapt their dynamics to the much slower system dynamics, and their main effect is simply to renormalise this slow dynamics (i.e., change its frequency scale somewhat). Dissipation and decoherence both arise from energy exchange between system and bath, either because of thermal or quantum fluctuations in the bath (one can also think of this as a combination of stimulated and spontaneous emission/absorption processes). The connections established for oscillator baths between decoherence and dissipation and between the classical and quantum dynamics, are amongst the most remarkable results derived by Caldeira and Leggett.

The crux of the connection between decoherence and relaxation in oscillator bath models lies in the weak-coupling assumption (i.e., the assumption that each bath mode is only weakly perturbed by the central system; note again that the system itself may be very strongly affected by the combined effect of all the bath modes). Decoherence and dissipation are arising then from the same second-order processes, in which a single bath mode intervenes to exchange energy with the system (footnote 9). By summing over all the bath modes, one rapidly introduces standard results for a situation like this (Leggett, 1984; Caldeira & Leggett, 1983). The ‘fluctuation–dissipation’ theorem, connecting the fluctuation spectrum of the bath with the dissipation it causes on the system, is an immediate consequence of the weak-coupling assumption (the derivation uses ‘linear response’ theory). We can thereby connect the decoherence and dissipation in the system directly to the noise spectrum of the environment.

To give a quick intuitive picture of all this, let us pick a really simple central system \mathcal{S} , viz., a ‘qubit’, the elementary component of a quantum computer, which we then couple to an oscillator bath. This model (known as the ‘spin–boson’ model) has been studied rather exhaustively (Leggett et al., 1987; Weiss, 1999). The qubit itself can be described by a Pauli spin- $\frac{1}{2}$ vector τ , and we choose a Hamiltonian $H_0(\tau) = A_0\tau_x + \varepsilon\tau_z$. Working in the basis of the eigenstates $|\uparrow\rangle$ and $|\downarrow\rangle$ of τ_z , the propagation of the density matrix in time can then be visualised very easily in path integral language (see Fig. 3). The qubit simply ‘flips’ back and forth between $|\uparrow\rangle$ and $|\downarrow\rangle$. This happens on a very short timescale $\sim 1/\Omega_0$, governed by the high-energy physics of the qubit at energy $\sim \Omega_0$. If we now add the coupling $\sum_q c_q \tau_z x_q$ to the oscillators (the analogue of the Feynman–Vernon coupling $\sum_q c_q Q x_q$ to Q), the effect of the bath is to allow ‘second-order’ interaction processes between the bath and qubit,¹² with the oscillators shown as wavy lines.

The qubit–bath coupling distinguishes the states $|\uparrow\rangle$ and $|\downarrow\rangle$, so that the bath is in effect ‘watching’ the qubit. It is not surprising that the general effect of this coupling is to slow down the qubit dynamics and to degrade coherence (i.e., superpositions) between $|\uparrow\rangle$ and $|\downarrow\rangle$. In path integral language, attractive interactions are generated between the jumps, both on the same path and between paths; this causes them to bind together and thereby disappear (thereby making jumps less frequent and also suppressing ‘off-diagonal’ states

¹²Each wavy line in the figure represents the emission and absorption of a bath excitation, and is thus second order in the interaction. Multiple interaction lines appear in the figure because the influence functional in the path integral (Eq. (9)) is an exponential function of the interaction.

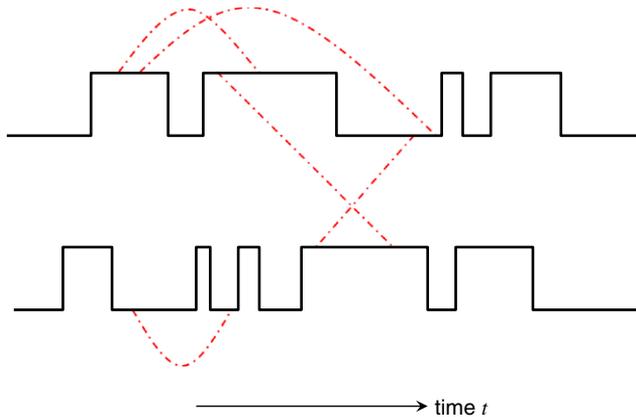


Fig. 3. The behaviour in time t of the density matrix of a simple two-level system (a ‘qubit’) which is interacting with a bath. The density matrix always involves two paths (the ‘forward’ path 1 and ‘return’ path 2); each switches between the two available qubit states as time goes on. The qubit–bath coupling mediates interactions between paths (hatched lines), as well as ‘self-energy’ interactions between states of the same path at different times.

corresponding to interference). If the qubit-bath interaction is strong enough, this can even cause all transitions to disappear, and the qubit is then frozen by the bath. The dissipative slowing down can be related directly to the decoherence rate, and both can be related to the quantum fluctuations of the bath, as we expect from the remarks made above.

All this is very much what one might expect from the bath. Study of other models, such as a moving particle coupled linearly to the bath according to (1) or (2), and either in a homogeneous medium or in some potential well (or tunneling from it), give similar results.

Finally, we re-emphasise the point already made above that as one lowers the bath temperature and the operating energy scale Δ_0 of the qubit, the decreasing available phase space for transitions in the bath states means that decoherence also falls rapidly (particularly rapidly in insulators) and eventually goes to zero, as T and Δ_0 go to zero. High-energy bath modes cause little decoherence (one has to be careful to distinguish simple renormalisation effects caused by these modes from genuine decoherence (Unruh, 1999)). Under certain circumstances, one can then *engineer* the oscillator bath environment to have very few low-energy states (for example, in a superconductor one has a gap in the low-energy spectrum). In this case we expect very little decoherence from the oscillator bath.

Decoherence from spin baths. Decoherence works in a very different way for a spin bath, and the differences with oscillator bath decoherence are very illuminating (Dubé & Stamp, 2001; Prokofev & Stamp, 2000). Consider a particular bath spin σ_k . Its dynamics is controlled by (i) a *static* field \mathbf{h}_k , and (ii) a dynamic field $\mathbf{F}_k(P, Q)$ caused by the central system, whose state is evolving in time. The interaction $V_{kk'}$ with other spins is a small perturbation on this.

How σ_k actually evolves in time depends on a third energy/frequency scale—the rate $\dot{\mathbf{F}}_k(P, Q)$ at which $\mathbf{F}_k(P, Q)$ is changing. This rate is controlled by the dynamics of the central system \mathcal{S} . If the characteristic frequency scale for changes of $\mathbf{F}_k(P, Q)$ is Ω_0 , so that $|\dot{\mathbf{F}}_k/\mathbf{F}_k| \sim \Omega_0$, then we have two limits, viz., (a) if $u_k \ll 1$, where $u_k = |\mathbf{F}_k|/\Omega_0 \sim |\mathbf{F}_k^2/\dot{\mathbf{F}}_k|$, the

system is moving too fast for σ_k to follow the field F_k (the ‘fast’ or ‘sudden’ limit); whereas (b) if $u_k \gg 1$, the system dynamics is slow, and σ_k can track F_k fairly accurately (the adiabatic limit).

To get an idea of how decoherence works here, imagine that \mathcal{S} is a qubit τ , so that its typical paths are those just discussed for the spin–boson model. The coupling between σ_k and τ can be assumed quite generally to take the form $H_{\text{int}} = \hat{\tau}_z \omega_k \cdot \sigma_k$, so that the field on σ_k from the central qubit, when it is in state $|\uparrow\rangle$, is ω_k , and this field reverses its direction when the qubit flips. Note that there is also another *static* field \mathbf{h}_k acting on σ_k (cf. Eq. (7)); the total field is the sum of the two (we ignore the interaction $\{V_{kk'}\}$ for the moment).

How this then affects the dynamics of our bath spin σ_k is shown schematically in Fig. 4, which shows a typical path for the bath spin (*not* for the qubit!). Each time τ flips, the total field jerks suddenly (on a time scale Ω_0^{-1}) between two orientations. Suppose the bath spin begins in a state oriented along one of these fields. A qubit flip then starts it precessing about the new field; in general when τ flips back the bath spin will be oriented in some other direction, and it will begin to precess anew around this field. We see immediately that: (i) the bath spin’s dynamics is now *entangled* with that of the qubit. In particular it accumulates a ‘precessional phase’ that depends on the qubit path; and thence (ii) averaging over the bath spins now gives decoherence in the dynamics of the central system τ , this is called ‘precessional decoherence’.

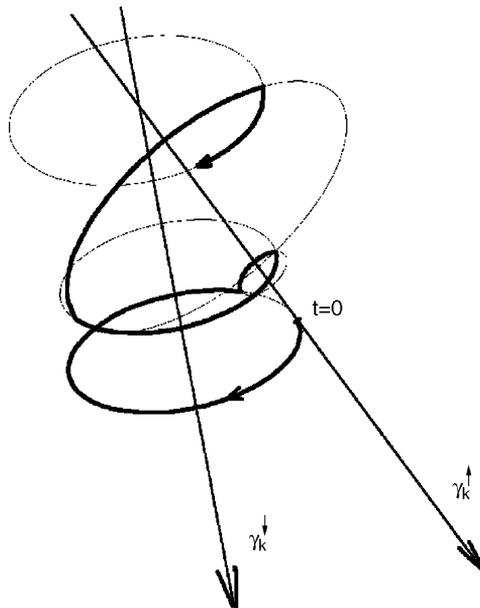


Fig. 4. The dynamics of a bath spin (the ‘kth’ bath spin) under the influence of a qubit. The qubit exerts fields γ_k^\uparrow or γ_k^\downarrow , depending on whether the qubit is in state $|\uparrow\rangle$ or $|\downarrow\rangle$. Each time the qubit flips, the bath spin must begin to precess in the new field, causing the bath spin trajectory shown. The dependence of this trajectory on the qubit trajectory means that their quantum dynamics are strongly entangled, even though no energy is exchanged.

Precessional decoherence is almost always the most important mechanism of decoherence coming from a spin bath. However it has very different characteristics from that coming from oscillator baths. In particular

- (i) Notice that *no energy transfer* between system and bath is involved in precessional decoherence; no transitions in the bath spin state occur.¹³ Nevertheless very strong decoherence can occur—if the total phase accumulated by all the bath spins over a time τ is $\gtrsim 2\pi$, then averaging over this phase will give very strong decoherence in the qubit dynamics over this time.
- (ii) The bath spin dynamics is being driven by the qubit. If the qubit dynamics is switched off, only the very weak interspin interactions $\{V_{kk'}\}$ can drive the bath spins. Thus the bath spin dynamics is largely *slaved* to the qubit dynamics and will slow down drastically if the bath spin dynamics is frozen. Again, this is totally different from an oscillator bath, whose internal dynamics is only weakly affected by coupling to the central system. Incidentally this means that the intrinsic noise coming from the spin bath has little connection to the decoherence.

These results underline the fact that decoherence caused by a quantum environment is really about phase exchange between system and environment, and has no necessary connection with either environmental noise or dissipation at all. It can proceed in the complete absence of either. Some physicists, used to the framework of linear response and fluctuation dissipation theorems, are quite surprised by this. It is important to remember the limitations of the fluctuation–dissipation framework—it only works if the bath is weakly perturbed by the central system (or by some probe). In the present case the spin bath will only obey linear response and the fluctuation–dissipation theorem if it is weakly perturbed, which is precisely the point at issue here. Indeed, it should be remarked that all the standard ideas about linear response are more and more difficult to apply as one lowers the temperature, since ever smaller perturbations will take the bath outside the linear response regime.

This naturally leads one to ask how decoherence from spin baths behaves as one goes to the low temperature limit. In contrast to oscillator baths, we can no longer assume that the oscillators will go away in the low-energy limit. Indeed, as remarked above, localised states tend to *pile up* at low energies, in many solids. Thus one can expect very large contributions to decoherence from these states. Thus on purely theoretical grounds one can expect that spin bath decoherence will dominate over oscillator bath decoherence at low temperature. There is no reason to expect it to go to zero, even as $T \rightarrow 0$. Thus we see that the intuitive connection between decoherence, dissipation, and environmental noise, all gained from the oscillator bath models, is in no way generic to decoherence.

Nevertheless very surprising features can emerge. For example, a particle hopping quantum mechanically around a lattice of some topology will, if coupled to an oscillator bath, *always* tend at long times to show diffusive dynamics, and this feature is often cited as an example of the inevitable crossover of quantum behaviour to classical stochastic

¹³We omit here discussion of ‘topological decoherence’ from the spin bath, which is usually much weaker than precessional decoherence (and also causes no dissipation). See, e.g., Prokofev & Stamp (1993, 2000). Note that the analogue of this can exist in special oscillator bath models where the longitudinal coupling $c_q x_q \tau_z$ is absent, but a transverse coupling like $c_q^\perp x_q \tau_x$ is present—again there will be no dissipation.

behaviour in the presence of decoherence. But one can show (Prokofev & Stamp, 2006) that if the particle is instead coupled to a spin bath, this is not so. In fact, in the long time limit, there will always be some part of the particle density matrix which still shows quantum interference behaviour. Since such lattices can be used as very general models for the propagation of quantum information (Kempe, 2003; Kendon, 2003), this result is of some importance.

Finally, I emphasise that none of these results are mere remarks about abstract models. As we will note in Section 3, they are crucial for recent experiments.

2.3. *Third-party decoherence*

The literature on decoherence deals entirely, as far as I know, with environmentally induced decoherence, in which phase correlations (and possibly also energy, etc.) are transferred from system to environment by some physical coupling between them. The purpose of this section is cautionary, to emphasise that entanglement between system and environment can be set up without such a direct coupling, or even an indirect one. The basic idea discussed briefly here (more details appear elsewhere¹⁴) is that of ‘third-party decoherence’, in which decoherence emerges eventually in the dynamics of some system \mathcal{S} , not via any direct coupling to the environment \mathcal{E} , but through the influence of a third party.

Clearly there is a trivial way in which phase correlations can be set up between a system \mathcal{S} and an environment \mathcal{E} , even when they are not directly coupled. One couples \mathcal{S} to a ‘third party’ \mathcal{P}_3 which is in contact with (or is later brought into contact with) \mathcal{E} . Thus phase correlations, entanglement, etc., pass through the chain $\mathcal{S} \rightarrow \mathcal{P}_3 \rightarrow \mathcal{E}$. However this is clearly not a fundamentally new situation. Theoretically, we can simply expand our original environment to a new environment $\mathcal{E}' = \mathcal{E} + \mathcal{P}_3$. The details may be non-trivial and important for experiment, and interesting things may happen, since the entanglement held between \mathcal{S} and \mathcal{P}_3 may take some time to reach \mathcal{E} (particularly if \mathcal{P}_3 is only later allowed to interact with \mathcal{E}). One can also extend this chain to include fourth, fifth, etc., parties.

There are however more interesting kinds of third-party decoherence. Consider as an example the famous two-slit experiment, in which particles pass through two slits and an interference pattern is produced on a screen. There will be simple decoherence mechanisms here, in which the particle interacts with a photon bath (the dipolar EM interaction allows photons to track the particle path $\mathbf{Q}(t)$) or even phonons emitted by the particle if it collides inelastically with the slit system on its way through. However a more subtle effect can arise if the particle itself possesses internal degrees of freedom $\{x_I\}$, which themselves do not interact with the particle centre of mass coordinate $\mathbf{Q}(t)$, but which do interact with the slit system. In this way it is possible to entangle the environmental wave-function $\Phi(\{x_I\})$ with the system wave-function $\Psi(\mathbf{Q})$, not through any interaction mediated by the slit, but simply because they interact in similar ways with the slit system.

Without going into details (footnote 14), we can easily see how this works in a ‘toy’ calculation. If we ignore the internal modes of the particle, we have the usual situation depicted in Fig. 5. Assuming slit states $\psi_j \sim |\psi_j| e^{i\phi_j}$, where $j = A, B$, we then find the

¹⁴The discussion of third-party decoherence here is simplified and does not include energy relaxation and equilibration, or any dynamics. For a proper analysis, see Stamp (to be published).

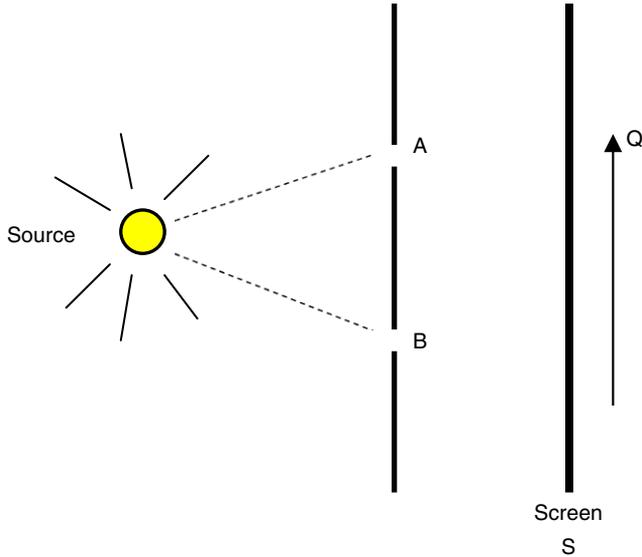


Fig. 5. The two-slit experiment—a particle can pass via either slit A or slit B to reach a point with coordinate Q on the screen S . The probability $P(Q)$ of arrival at Q then shows the standard interference pattern.

probability of arrival of particles at coordinate Q on the screen is given by

$$P(Q) = P_A(Q) + P_B(Q) + 2[P_A(Q)P_B(Q)]^{1/2} \cos \phi_{AB}(Q), \tag{10}$$

where the phase $\phi_{AB}(Q) = \phi_A(Q) - \phi_B(Q)$ in the interference term comes from the difference in phase accumulated by particles traveling through the A or B slits.

Now suppose that when the particle goes through the slit system, internal modes are excited, and these are excited *differently* depending on which slit the particle goes through. In a real experiment on, e.g., buckyballs, which could involve the excitation of phonon modes via the deformation of the buckyball, the deformation will certainly depend on which slit the particle goes through. In this case the internal vibration modes will be excited rather differently. After a passage through slit A, the l th mode will be in some state $\phi_l^A = \sum_{n_l} c_{n_l}^A \chi_{n_l}$, with amplitude $c_{n_l}^A$ to excite this mode into its n th excited state; however passage through the other slit B will give different amplitudes $c_{n_l}^B$, so that the overlap

$$f_l = |\langle \phi_l^A | \phi_l^B \rangle| = \sum_{n_l} |(c_{n_l}^A)^* c_{n_l}^B| < 1. \tag{11}$$

We see that the wave-functions of the internal modes are now entangled with that of the centre of mass motion, even though they have never interacted with them. It is simple to now show that after tracing over these internal modes, we get a suppression of the interference term above by a factor $D = \prod_l f_l$.

The crucial point in the above discussion is that at no point ever do the internal modes of the particle interact with its centre of mass coordinate, either directly or indirectly—instead, they both happen to interact in a similar way with the slit system. In other words, because of the symmetry of the system, a kind of underlying constraint, the two different

systems (centre of mass coordinate, and internal modes) are forced to separately interact with the slit system in such a way that afterwards their states are entangled. One can extend this discussion to other examples, for which there is no space here.

Considering the problem from a more general standpoint, we note that third-party decoherence can affect any system whose behaviour is conditioned by some agency which also happens to condition the behaviour of an environment which we trace over. This means that it can be quite discreet, and not so easy to eliminate from an experiment. As with the spin bath, there is no dissipation in the motion of the central system coordinate Q , yet it still experiences decoherence. The results also mean that the usual discussions of decoherence in terms of interacting system–bath models, described by some effective Hamiltonian, are incomplete. I emphasise that none of the above results are beyond the reach of standard formalism (one can describe them equally well with reduced density matrices or with the decoherence functional formalism, by suitably generalising the averages). The novelty is the necessity for inclusion of the apparently innocuous third party.

3. Decoherence in the lab

The 21st-century lab is the battlefield upon which our ideas on decoherence, confronted by experiment, are going to live or die. Two points are worth emphasising:

- (i) The stakes are very high. Questions at issue include: do we really understand what causes decoherence and are there ineluctable or even intrinsic decoherence sources in Nature? Is quantum mechanics valid at large scales? If so, can we use highly entangled multi-particle states (in spite of decoherence)? The experimental answers to these questions will play a major role in the future evolution of physics.
- (ii) The relationship between experiment and theory is very complex here. On some fronts, experiment is loath to challenge theory, even where there is striking disagreement. In many cases, the interpretation of the experiments often depends on what theoretical question the experimentalists decide they are probing. Any experiment can be examined through different theoretical lenses.

In the last four decades some landmark experimental tests of quantum mechanics have been formulated and enacted, particularly associated with Bell's theorem and entanglement (Aspect, Dalibard, & Roger, 1982; Clauser & Shimony, 1978). In some of this work, and in offshoots of it, quantum entanglement and superposition have been tested over length scales of many km (Marcikic et al., 2004). However none of these tests has involved a large number of particles; rather, they have involved small molecules or a few entangled photons or ions. A number of experimental tests of quantum mechanics at the macroscopic scale, involving very large numbers of particles, were suggested by Leggett et al. (Caldeira & Leggett, 1983; Leggett, 1984, 2002; Leggett & Garg, 1985). These tests all involved the use of superconductors. One set of tests looked at 'macroscopic quantum tunneling' of superconductors—the quantitative theoretical predictions of tunneling rates vs. temperature and applied field (Caldeira & Leggett, 1983; Leggett, 1984) included the dissipative effect on tunneling of the environment (Caldeira–Leggett theory). The later experiments (Clarke, Cleland, Devoret, Esteve, & Martinis, 1988) agreed with this theory over the

whole range of experimental parameters, to within experimental error—a remarkable result. None of these experiments probed decoherence.

Tests of our understanding of decoherence have come both from quantum optics and from solid-state physics. In the former, decoherence in the dynamics of entangled ions is expected to come from interaction with photons (Myatt et al., 2000). Experiment indicates that the mechanisms are understood; there seem to be no hidden sources of decoherence. Tests in the solid state have looked at (i) coherent electron dynamics in mesoscopic conductors, and (ii) coherent tunneling in superconductors and magnetic systems. The main results here are as follows:

(a) Many experiments on mesoscopic conductors measure the time it takes for phase coherence to be lost in the dynamics of the electrons. Some of these experiments indicate that strong decoherence persists down to very low temperature. Since this result conflicts with the standard theory (in which decoherence comes from interactions of electrons between themselves and with phonons and impurities), it has caused much controversy (Aleiner, Altshuler, & Gershenson, 1999; Mohanty, Jariwala, & Webb, 1997). Some more recent experiments (Pierre et al., 2003) indicate that interaction of the electrons with spin impurities may be responsible (i.e., a ‘junk’ effect, with the junk being the bath of spin impurities).

(b) In a large number of different molecules, the electronic spins lock strongly together to give a ‘giant spin’, which at low temperatures can quantum tunnel through the energy barrier between two different spin orientations. Many experiments have examined this tunneling (Wernsdorfer, 2001; Tupitsyn & Barbara, 2001), as well as related phenomena in rare earth magnets (Ronnow et al., 2005, and refs. therein). It is now clear what controls the tunneling dynamics of these giant spins. At low temperatures the nuclear spins in the system (coupled strongly to the central giant spin via hyperfine interactions) disrupt the coherent dynamics of the central spin, so that the tunneling is completely incoherent. Present efforts to make spin qubits (for quantum computation) concentrate on suppressing this nuclear spin-mediated decoherence by making the qubit dynamics much faster than the nuclear spin dynamics, bringing in the risk of significant decoherence from phonons (Stamp & Tupitsyn, 2004) (an oscillator bath effect). One can also try to eliminate the nuclear spins by isotopic purification, but this will not be easy.

(c) Tests of the coherent dynamics of a superconducting SQUID between two potential wells are a solid-state realisation of a ‘Schrödinger’s Cat’, in which a macroscopic number of electrons are in a coherent superposition of two different current states (Leggett et al., 1987). In the last few years several experiments have given very strong evidence for Cat states in superconductors (Chiorescu, Nakamura, Harmans, & Mooij, 2003; Nakamura, Pashkin, & Tsai, 1999; Pashkin et al., 2003; Vion et al., 2002). Leggett and Garg (1985) also formulated a criterion of ‘macroscopic realism’ which can be tested on systems of this kind. The criterion of macrorealism has a clear physical meaning—it distinguishes those properties of a macroscopic system which can be treated as objectively real, in a similar spirit to that discussed by EPR and Bell for microscopic systems. The formal criterion for testing macrorealism involves a set of inequalities pertaining to measurements at different times on a macroscopic variable. These inequalities test quantum mechanics explicitly on the macroscopic scale, but experiments on them have yet to be done. Some of the existing experiments have explicitly measured decoherence rates (Chiorescu et al., 2003; Nakamura et al., 1999; Pashkin et al., 2003; Vion et al., 2002). The experimental decoherence rates in superconductors are always found to be much larger (by up to six orders of magnitude)

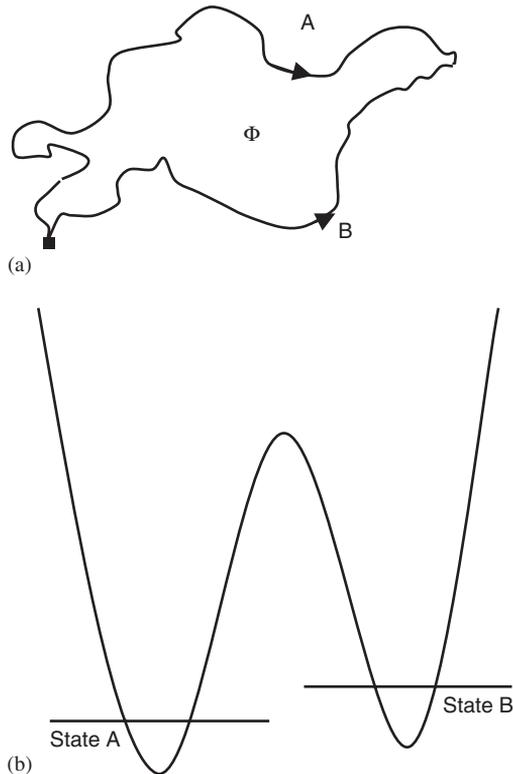


Fig. 6. Tests of large-scale quantum phenomena in condensed matter systems usually involve either interference between propagation along two different paths, which may have some flux Φ enclosed between them (a); or they involve interference between two states quasi-localised in two different potential wells, which communicate weakly by tunneling (b). Experiments on magnetic molecules combine both features (see text).

than those predicted by Caldeira–Leggett theory. Note that this is the same theory which works so well for dissipative tunneling experiments in the same superconductors!

All of the above experiments are of course described using certain theoretical models, and I have summarised the most important ones in Fig. 6. The experiments on decoherence in mesoscopic conductors rely on interference between single electrons following two different paths (Fig. 6(a)); if there is flux enclosed between these paths, we can use the Aharonov–Bohm effect (actually, its suppression) to detect decoherence. In experiments on magnetic molecules, or superconductors (Fig. 6(b)), the relevant mesoscopic or macroscopic coordinate (magnetisation for the molecules, flux for the superconductors) is confined to tunnel between two potential wells. If it moves coherently, one can use it as a qubit, whereas decoherence gradually converts its motion to incoherent tunneling.

We may now summarise the results of the experiments insofar as they concern decoherence. We apparently do know what is causing decoherence in some of the experiments (ions in cavities, possibly mesoscopic conductors, possibly magnetic molecules), but so far theory has not described the decoherence in superconductors. The claims made for intrinsic zero-temperature decoherence in some of these experiments have yet to be properly tested.

Let us now come to a more general discussion of these results, which from a philosophical standpoint is of some interest. The main point I wish to make is that how we interpret the experiments depends mainly on what question we think they are asking. The following questions (amongst others) are thought to be important:

- (i) Do we understand the decoherence in these experiments (where it is coming from, how it works)? Are there hidden sources of decoherence? Are there even *intrinsic* mechanisms of decoherence in Nature?
- (ii) Is the whole idea of quantum information processing with massively entangled states possible in practice, or even in principle? Can we get rid of decoherence?
- (iii) Has macrorealism been tested (and what is the verdict)?

Any response to these questions depends on how one feels about the gap between theory and experiment—in particular, on whether it is felt that standard quantum mechanics, using one mechanism or another, can eventually explain all the decoherence in *all* of the experiments. If so, then one can adopt the view that even very large existing discrepancies are basically just a question of detail. With a lot of work theory and experiment will eventually be brought to agree. If not, then these tests of decoherence mechanisms and rates become of supreme importance in our quest to understand quantum mechanics properly and possibly even to go beyond it. Disagreement between theory and experiment is then very far from being a mere detail.

These are of course two extreme points of view, and there are others lying between them. Nevertheless the point is clear: how the experiments are interpreted depends less on the experiments themselves than on a faith about the validity of the existing theoretical framework.

This point is rather obvious as far as the first two questions are concerned, so I will not belabour it. The question about macrorealism brings the relation between experiment and theory into acute relief. Many (indeed most) physicists, faced with the observations in superconductors of macroscopic coherence, simply remark that the verification of quantum mechanics at the macroscopic scale is not surprising and are then less interested in hearing about tests of macrorealism. The expectation is that quantum mechanics will always prevail. The existence of large amounts of decoherence is then again regarded as a detail, a problem to be solved within the framework of quantum mechanics.

On the other hand there are those who think such tests important, that quantum mechanics does need to be tested at the macroscopic scale and may be found wanting. Apart from the Leggett school of thought (Leggett, 2002; Leggett & Garg, 1985), many papers have discussed non-linear extensions of quantum mechanics, where the non-linearity appears for sufficiently large systems and would be hard to distinguish from decoherence in experiments (Ghirardi, Pearle, & Rimini, 1990; Ghirardi, Rimini, & Weber, 1986; Pearle, 1976, 1989). There are also more exotic ideas, involving intrinsic decoherence sources, coming either from spacetime curvature (intrinsic gravitational decoherence)¹⁵ or from ultra-Planck scale physics ('t Hooft, 1999, 2001), the latter idea having an interesting

¹⁵For gravitationally induced wave-function collapse, see Diósi (1989), Ghirardi, Grassi, & Rimini (1990) and Penrose (1994, Sections 6.10–6.12). A possible experimental test was suggested by Marshall, Simon, Penrose, & Bouwmeester (2003).

history.¹⁶ All of these ideas attempt to go beyond existing theory and try to remove some of the paradoxical features of quantum mechanics. Although none of these programmes has actually constructed a comprehensive theory, they do provide possible experimental tests. These test macrorealism in the case of experiments on superconductors, and in the other cases predictions are made for what looks like an intrinsic decoherence rate in Nature, in a way which violates conventional quantum theory. This intrinsic decoherence would show up, in all these cases, in experiments on large-scale quantum phenomena. As noted by 't Hooft (1999, 2001), any such intrinsic decoherence mechanism would put severe limits on quantum information processing (in the holographic approach of 't Hooft and Susskind, it would be impossible for a quantum computer or quantum memory to involve more than roughly 400 entangled qubits).

An important point I wish to make here is that no experiment purporting to test quantum mechanics, according to any of these scenarios, can afford to ignore disagreements between experimental and theoretical decoherence rates; these are no longer a question of detail. One certainly cannot treat any disagreement as a 'dirt' or some other uncontrolled extrinsic effect. This would automatically dismiss any real breakdown of quantum mechanics as a dirt effect and make tests of large-scale quantum mechanics impossible in principle.

The aim of this section has been to give readers a feel for how current experiments bear on some of the really fundamental questions associated with decoherence and possibly on even more fundamental questions about quantum mechanics itself. Perhaps not surprisingly, we see that how the experiments are interpreted depends very much on prevailing views and prejudices, about the expected answers to these questions.¹⁷

4. Six questions about decoherence and quantum relaxation

With the material in the two previous sections in hand, we may now address directly some of the larger problems mentioned in the introduction. Rather than a lengthy analysis of these, it is simpler to frame the discussion in terms of a set of six questions. Some of these have frequently been posed before, others less so. However in all cases the answers depend in one way or another on what we have been discussing, i.e., on what are the mechanisms of decoherence.

Question 1: What causes decoherence in Nature? Is there a 'generic model' of decoherence (and if so what is it)?

Answer: We have certainly now elucidated some of the decoherence mechanisms operating in Nature, and there is a large variety of them. While the three models discussed in Section 2 (spin bath, oscillator bath, and third-party decoherence) themselves cover

¹⁶The idea that quantum fluctuations of spacetime at very high energies, up to the Planck scale, might cause decoherence at low energies has been discussed in various contexts. See, e.g., Hawking (1982), Hawking & Laflamme (1988), Coleman (1988) and Ellis, Mohanty, & Nanopoulos (1989).

¹⁷I emphasise that we are interested here in the theoretical context in which genuine experimental challenges to an established theory (here, quantum mechanics) are mounted and what criteria are used to decide how successful is the challenge. There are currently several controversies raging about the relation between theory and experiment in science, notably over the misuse of experimental data (in, e.g., the debate over evolution vs. 'intelligent design', or in the Schön–Batlogg debacle, where some 20 papers based on fabricated data were published by Nature and Science). This is of course a very different issue, and should not be confused in any way with the present discussion.

many different physical systems, there is no reason to suppose we have found *all* possible mechanisms of decoherence!

At this point one has to insist that the real verdict must come from experiment. Without a quantitative explanation of experimental decoherence rates in terms of known theoretical models, one can always posit undiscovered sources of decoherence ‘out there’. As emphasised above, many experimental systems at present show anomalously large experimental decoherence rates (although some discrepancies can probably be explained by spin bath effects). To have a generic model for decoherence would suppose a much better understanding than we presently have of most condensed matter systems. One should beware of general theorems on decoherence rates for large systems, since they usually make very restrictive (and unrealistic) assumptions about the structure of the many-body states.

Thus we are not yet in a position to be talking about a generic model for decoherence.

Question 2: Is decoherence necessarily related to irreversibility and dissipation/relaxation? If so, does decoherence then go to zero with temperature, and can it be eliminated in the real world?

Answer: All these questions have been controversial and are also of fundamental interest. If decoherence were tied to dissipation, then at low energy, with a cold environment, decoherence rates would be very low, going to zero with temperature; and moreover vacuum fluctuations would not cause decoherence at all. Such a conclusion would be of great importance, if true.

As we saw in Section 2, dissipation and decoherence *are* tied together in the oscillator bath models of the environment. However, as we also saw, this result is *not* true for spin baths, where one can have decoherence with no dissipation, even at $T = 0$; and in the case of third-party decoherence there cannot possibly be any environmental dissipation, at any T , since there is no direct coupling to the environment. Thus there is no necessary connection between decoherence and dissipation in the real world, and no necessary reason for it to go to zero at $T = 0$.

This is a problem of real practical interest right now, both for the construction of quantum information processing systems and for the standard physics of solids. There is thus a massive worldwide quest going on for ways to eliminate environmental decoherence. If the three sources of decoherence just mentioned are in fact the only kinds that exist, then one might still entertain hopes of eliminating them; indeed, some very interesting idea for doing this are under present investigation. However, what if there are other decoherence sources? This suggests the next question:

Question 3: Are there ‘intrinsic’ sources of decoherence in Nature, impossible to eradicate?

Answer: By ‘intrinsic’ sources, is meant sources which are inevitable in the world as it is, not arising from dissipative processes and perhaps even arising as part of the basic structure of the universe. Such intrinsic sources of decoherence in Nature, operating even at $T = 0$, would not only provide a way of explaining the ‘emergence of classical physics’ in fields ranging from quantum cosmology to condensed matter physics; they would also place a fundamental limit on the observability of quantum phenomena. This would limit the possibility of seeing macroscopic quantum phenomena, and also place fundamental limits on the superpositions required for quantum computing.

Possibilities for intrinsic decoherence mechanisms have already emerged from both low- and high-energy physics. From low-energy physics there has been a suggestion that zero

point modes of continuous quantum fields (in particular, the photon field) could cause $T = 0$ decoherence. This has, for example, been suggested as an explanation of the decoherence saturation at low T in mesoscopic conductors (Mohanty et al., 1997). This suggestion is controversial and was discussed in Section 3; many feel that the explanation lies instead with magnetic impurities (a spin bath effect). There is also the suggestion of non-linear terms in the dynamics of macroscopic quantum systems (Ghirardi, Pearle et al., 1990; Ghirardi et al., 1986; Pearle, 1976, 1989); this has hardly been tested yet.

On the high-energy side a wide range of possibilities has been canvassed and already noted in Section 3. These include, again, low-energy decoherence from zero point modes—this time from gravitons, or string fields, or from vacuum fluctuations of the spacetime metric (footnote 16) (including the so-called ‘baby universe’ fluctuations). So far these suggestions have not been met enthusiastically for they fly in the face of conventional ideas about renormalisation, according to which neither very high energy modes nor vacuum fluctuations can enter into any dynamic processes in a low-energy effective Hamiltonian. There are also more exotic ideas, involving modifications of quantum theory. Two recent proposals are an intrinsic decoherence arising from spacetime curvature (intrinsic gravitational decoherence) (footnote 15), and a source arising from ultra-Planck scale physics, suggested by ’t Hooft (1999, 2001). Although neither of these programmes has actually constructed a comprehensive theory, they do provide possible experimental tests, in both cases involving an intrinsic decoherence rate, which violates conventional quantum theory. This intrinsic decoherence would show up in both cases in experiments on large-scale quantum phenomena. As noted by ’t Hooft, any such intrinsic decoherence mechanism would put severe limits on quantum information processing (in the holographic approach of ’t Hooft and Susskind, it would be impossible for a quantum computer or quantum memory to involve more than roughly 400 entangled qubits).

Clearly some pretty crucial experiments are required here. This is one of the very interesting frontiers of physics right now. Such experiments will have to be done with great care, to eliminate, for example, the influence of third-party decoherence processes, not reflected in the effective Hamiltonian of the experimental system but in its previous history. Indeed it is not obvious to the present author how one can eliminate third-party decoherence with certainty.

Question 4: Does decoherence give rise to the ‘emergence’ of classical physics? If so, then what kind of a theory is quantum mechanics (often held to depend on classical mechanics for its definition in the first place)?

Answer: One interpretation of this question focuses on the more physical question of how classical quasi-deterministic behaviour emerges for large systems, and/or how quasi-classical stochastic behaviour emerges, even for small systems. It should now be completely evident, from Sections 2 and 3, that a proper answer to this question requires understanding the real decoherence mechanisms operating in Nature and that these are not so simple, or necessarily completely understood. Thus we do not yet have a theory which derives classical physics from quantum physics solely using ideas from decoherence, even though we do have some derivations of classical behaviour within certain models. It is important to note that in some other models one can actually find non-classical behaviour emerging in the large-scale dynamics, because of decoherence (this happens, for example, when one is dealing with a spin bath environment (Prokofev & Stamp, 2006)). Thus there is nothing inevitable about classical behaviour!

In a second interpretation, it is suggested that decoherence might not only derive classical physics as a limiting case of quantum mechanics but also show how strictly classical concepts such as momentum and position are inevitable in the very formulation of quantum mechanics. The basic argument here is that the structure of interactions in Nature inevitably leads to a preferred ‘inert’ or ‘pointer’ basis for the states of macroscopic objects (Simonius, 1978; Zurek, 1981, 1982, 2003). Again, however, this argument has relied on simple models, and a general demonstration would require the use of more general models. It is extremely interesting to ask whether more general models could yield instead ‘non-classical’ pointer bases. Just as interesting is to ask what physicists will do if, as seems very possible, experiments find that ‘macrorealism’ (in the Garg–Leggett sense) fails. How then will we formulate quantum mechanics?

Question 5: With the understanding gained into the mechanisms of decoherence, can we now say how quantum measurements work? And does decoherence ‘solve’ the measurement problem?

Answer: One of the remarkable paradoxes of quantum physics is how difficult to give a theoretical description of most measurement schemes, even though they are being used all the time to do experiments! Detailed accounts, including all steps from the measured degrees of freedom up to the final ‘classical’ state of the measuring apparatus, are mostly confined to experiments designed for tests of quantum phenomena (often in quantum optics labs) or to sensitive experiments designed to search for very weak effects (e.g., gravity waves (Braginsky & Khalili, 1992; Caves et al., 1980)). Usually in these descriptions assumptions are made about how irreversible amplification processes, accompanied by strong decoherence, lead to definite results, FAPP (For All Practical Purposes). There is no question that if serious tests of quantum mechanics are to be made at the macroscopic scale (e.g., of macrorealism), a more complete analysis will need to be done, carrying the full quantum description right up to the macroscopic scale and including all sources of decoherence at each stage. It hardly matters which verdict the experiments give here. In either case a convincing experimental result will only be attained if all sources of decoherence are understood (including third-party decoherence).

Whether such analyses will ‘solve’ the measurement problem depends on what the problem is supposed to be. As with question 4, we remark that there is nothing inevitable about a classical behaviour for the measuring system (unless one defines measuring systems so that they must be classical!). On the other hand if tests of quantum mechanics at the macroscopic scale do actually vindicate it, so that macrorealism is falsified, then the measurement problem will surely undergo a radical transformation to a new problem, viz.: how far can we push the ‘FAPP barrier’ (between the quantum and classical worlds) into what is now considered the classical world? Certainly a new vocabulary will be required by physicists to deal with genuinely macroscopic quantum states.

Question 6: Is decoherence connected to the ‘Arrow of Time’? If so, how?

Answer: It is commonly assumed that all arrows, including the thermodynamic arrow, derive from the cosmological arrow. In this view, irreversibility is caused ultimately by the cosmological arrow. If one assumes that decoherence is connected with irreversibility, then the ‘quantum arrow’ results from the thermodynamic arrow (a commonly adopted point of view) and is also then subservient to the cosmological arrow. In this picture, everything in the universe, even something as basic as classical spacetime, has resulted from special initial conditions.

However, this point of view is by no means universally accepted, and it is possible to write quantum mechanics, including measurements, in a time-symmetric form (Aharonov, Bergmann, & Lebowitz, 1964). We have also seen above that some kinds of decoherence are not at all connected with dissipation or irreversibility and that decoherence does not even necessarily have to lead to classical behaviour. Thus we are driven back to the familiar question about mechanisms—whether or not we should associate decoherence with either the thermodynamic or the cosmological arrow of time depends on what mechanisms are responsible for decoherence. Certainly the results about the mechanisms of decoherence, discussed in Sections 2 and 3, make it clear that there is no necessary or logical connection between decoherence and the thermodynamic arrow. If decoherence is logically independent of the thermodynamic arrow, it is much less obvious that it is connected with the cosmological arrow.

A real handicap in analyses of this question is that many of the current discussions of the arrow of time are framed in terms of theories about the beginning of the universe, or of vague ideas like the ‘anthropic principle’, which have not been really tested and which change fairly rapidly with time. The most prudent course of action here may be to suspend judgement on any possible connection between the cosmological arrow of time and decoherence until both are understood a little better.

5. Conclusions

In this article I have discussed how general questions about the nature of solids and about the low-energy physics of macroscopic systems have consequences for old questions about quantum measurements, about the relation between classical and quantum mechanics, and about the validity of quantum mechanics itself. If there is a central point here, it is that facts about the physical mechanisms of decoherence are crucial to answering these questions. We now know something about these mechanisms, and what we have found out has radically changed our perspective. Far from asking “how do decoherence and/or dissipation produce classical mechanics at the macroscopic scale?”, we are now asking “how can we evade decoherence at the macroscopic scale?”. The preparation and use of states with high-level entanglement (i.e., N -entangled states with $N \gg 1$), instead of being treated as a theoretical impossibility, is now a target in many experimental research programmes. Most radical of all, the idea that the investigation of such states could lead to a failure of quantum mechanics itself is being taken seriously by both high- and low-energy theorists, with experiments to test this idea in preparation.

If there is a thread running through all of this, it is that to make progress we need a firm understanding of the physical mechanisms governing decoherence. Decoherence, according to the older ideas, is supposed to explain away the quantum measurement problem and to explain how classical mechanics emerges from quantum mechanics. And yet in the last few years experiments have been gradually bringing decoherence under control, inexorably pushing quantum mechanics to scales that were formerly the preserve of classical physics. Along the way a new picture, a picture of how decoherence operates, has begun to emerge. Far from being associated with ordinary relaxation, the decoherence in most experiments (certainly those in solid-state systems) appears to come from ‘sleepers’ modes, modes nearly invisible in most experiments because they cause almost no dissipation. Thus decoherence is more subtle, and perhaps more pervasive, than previously thought. There are many

things we still do not understand about decoherence and what causes it, and it should now be clear that this is a very pressing problem.

We have seen that how one views all this depends much on pre-existing prejudices, both about our present understanding of solids and about the validity of quantum mechanics itself. The point of view I have taken is that there are still many things we do not understand about solids, particularly at very low energies, and that the failure of quantum mechanics is a possibility which is certainly worth considering and testing experimentally. It then follows that we cannot dismiss disagreement between theoretical and experimental decoherence rates, which may conceal the very failure we are looking for, whether it comes from the ultra-Planck scale or from very low energies.

It is always remarkable when a combination of theory and experiment has larger philosophical consequences. Perhaps the most dramatic example in recent times has been the impact of Bell's inequalities, where a set of experiments in atomic physics was able to rule out a whole class of possible theories about Nature, and in doing so, consign a widely accepted philosophical view about 'reality' to the dustbin. The fascinating prospect is that future experiments at low temperatures in condensed matter systems looking for 'gravitational decoherence', or non-linear terms in a future quantum mechanics, or something else, may have a similar impact. But this remains to be seen.

Acknowledgements

I was stimulated to write this paper by a PITP workshop which brought together philosophers, historians, and physicists, in December 2004. The paper has benefited from conversations with my present and former colleagues A. Morello, I. S. Tupitsyn, W. G. Unruh, and G. 't Hooft, and also from discussions with M. Devoret, D. Howard, A. J. Leggett, N. V. Prokof'ev, Y. Nakamura, and R. Wald. However none of them are responsible for what is said herein! The research was supported by the National Science and Engineering Council, by the Canadian Institute for Advanced Research, and by the Pacific Institute of Theoretical Physics (PITP).

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Spontaneous Symmetry Breaking and Blocking of Metastable States

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(Received 27 October 1977)

A mechanism for spontaneous symmetry breaking and related phenomena and the corresponding blocking of metastable states is discussed. It is based on the interaction of an object with a background of "probes" like photons or particles, etc., in its natural surrounding. Applications include quasilocalization of macroscopic bodies, spontaneous parity nonconservation of sugar crystals, localization of atoms in molecules (Born-Oppenheimer approximation), stability of metastable compounds, and perhaps also intrinsic symmetries of elementary particles.

The object of this Letter is to discuss a quantum mechanical mechanism of spontaneous symmetry breaking and related phenomena and the corresponding blocking of states which are not eigenstates of the Hamiltonian of the object in question. This mechanism differs from, and is much more powerful than, the one usually discussed in the current literature based on nonsymmetric solutions to symmetric equations.

Consider an object O and a probe P described in Hilbert spaces H^O and H^P , respectively. For simplicity H^O is assumed to be two dimensional; generalizations will be mentioned at the end. Consider further two orthogonal states φ_1^O and φ_2^O of the object and an interaction between object and probe leading to the following transitions of the combined system:

$$\varphi_i^O \otimes \varphi_0^P \rightarrow \varphi_i^O \otimes \varphi_i^P, \quad i=1, 2, \quad (1)$$

where φ_0^P is the assumed initial state of the probe. By linearity this gives the transition for arbitrary initial state $a\varphi_1^O + b\varphi_2^O$. If no further observation is performed on the probe, and the object alone is considered after separation of the two, the object has to be described by a density matrix ρ^O on H^O and the trace has to be taken over H^P .

Taking φ_1^O, φ_2^O as basis in H^O and assuming $(\varphi_1^P, \varphi_2^P) = 0$, the transition (1) for an initial su-

perposition $a\varphi_1^O + b\varphi_2^O$ with $|a|^2 + |b|^2 = 1$ leads then for the density matrix ρ^O of the object to

$$\begin{pmatrix} |a|^2 & ab^* \\ a^*b & |b|^2 \end{pmatrix} \rightarrow \begin{pmatrix} |a|^2 & ab^*(\varphi_2^P, \varphi_1^P) \\ a^*b(\varphi_1^P, \varphi_2^P) & |b|^2 \end{pmatrix} \\ = \begin{pmatrix} |a|^2 & 0 \\ 0 & |b|^2 \end{pmatrix}. \quad (2)$$

This constitutes a "reduction of the state vector" of a pure initial to a mixed final state of the object.

There are two important aspects in this connection. One is the compatibility of this reduction with the linearity of the law of motion. This is the case by construction. The second is the assessment of the relevance and frequency of occurrence of this phenomenon. This may be judged from its connection to the process of measurement. Indeed Eq. (1) is a (simplified) model of a measurement, in which information is transferred from the object to the probe in such a way that subsequent observation of the probe alone could discriminate exactly between the two cases where the object is initially in the state φ_1^O or φ_2^O . The possibility of this discrimination requires φ_1^P and φ_2^P in Eq. (1) to be orthogonal and thus leads to the exact depletion of the off-diagonal elements of ρ^O in Eq. (2) if the object alone is considered, i.e., even, and in particular, in

the absence of any actual observation of the probe.¹ The following two examples illustrate some important application. (I omit the super-script O for the object where this does not lead to confusion.)

Example 1.—The object is a macroscopic body. φ_1 and φ_2 are two localized spacially well-separated wave functions. The probe consists of one or several photons in the region where φ_1 or φ_2 is localized. By observation of the photons one could discriminate between the case where the object is in the state φ_1 or φ_2 . It follows that the passing, emission, or absorption of such photons destroys any coherence which might have prevailed before. Thus under the usual conditions of the macroscopic world where one can see the objects, i.e., discriminate between different locations by observing photons, it is not possible to preserve the coherence between states with macroscopically different localization.² There is no particular (e.g., cosmological) initial condition needed for this. This is responsible also, for instance, for the appearance of bubbles, localized in space, in an overheated liquid.

Example 2.—Consider sugar. It can be in a "right" (φ_1) and "left" (φ_2) state. If the sugar is crystallized, the two may be discriminated by eye (or microscope) by observing the light scattered from it. Thus, again, any coherence between the two states φ_1 and φ_2 is immediately destroyed by the interaction with this light. Now if parity is conserved and the ground state is non-degenerate it is of the form $\varphi = \alpha\varphi_1 + \beta\varphi_2$ with $|\alpha|^2 = |\beta|^2 = \frac{1}{2}$. Symmetry breaking cannot be due to lack of symmetry of the ground state of a symmetric Hamiltonian unless the ground state is degenerate, which is not expected to be the case in general. In both examples the probes could as well be electrons or molecules from the surrounding (in particular for sugar dissolved in a liquid).

An important question which remains to be answered is, what singles out the particular states φ_1 and φ_2 for the reduction (2), i.e., why does the reduction in the examples above not sometimes produce an incoherent mixture between $\varphi_+ = (\sqrt{2})^{-1}(\varphi_1 + \varphi_2)$ and $\varphi_- = (\sqrt{2})^{-1}(\varphi_1 - \varphi_2)$, say, instead of φ_1 and φ_2 . But this would imply that if the object is originally in the state φ_1 , say, it could be found afterwards with 50% probability in the state φ_2 . This means that the interaction with the probe, for which I took photons in my examples, could cause with 50% probability a transition between φ_1 and φ_2 which for large enough dis-

tance between the two cannot be the case for a massive body. Thus the inertia of massive objects singles out quasilocalized states for a reduction by interactions with light probes.

I now turn to an important consequence of reduction (2) in case it occurs frequently (in a sense to be discussed), namely the *blocking* of the states φ_1 and φ_2 by stochastic (repeated) reduction.

Consider for concreteness example 2 with a parity-conserving Hamiltonian H . For proper choice of the phases of φ_1 and φ_2 the two eigenstates of H are $\varphi_{\pm} = (\varphi_1 \pm \varphi_2)/\sqrt{2}$. Their energy difference is $\hbar\omega$. Then, after a reduction the density matrix on the right-hand side of (2) will evolve in a time interval τ according to $\rho \rightarrow \exp\{-i\tau H/\hbar\}\rho \exp\{i\tau H/\hbar\}$ to

$$\frac{1}{2} \begin{pmatrix} 1 + \delta_0 \cos \omega \tau & i \delta_0 \sin \omega \tau \\ -i \delta_0 \sin \omega \tau & 1 - \delta_0 \cos \omega \tau \end{pmatrix}, \quad (3)$$

where $\delta_0 = |a|^2 - |b|^2$. For $\delta_0 \neq 0$, in particular for $\delta_0 = \pm 1$, i.e., pure initial state φ_1 or φ_2 , this exhibits the expected oscillations with circular frequency ω . Thus the original property (right-handedness of some sugar crystals) disappears after a time $T \sim \omega^{-1}$. The fact that this time is long is usually assumed to be the reason for stability. It will be seen that this is only part of the truth.

If after a time interval τ the object is again hit by a probe leading to a reduction of the density matrix to diagonal form, then

$$\rho \rightarrow \frac{1}{2} \begin{pmatrix} 1 + \delta & 0 \\ 0 & 1 - \delta \end{pmatrix}, \quad \text{with } \delta = \delta_0 \cos \omega t. \quad (4)$$

If this is repeated n times, say, then, after a time $t = \sum_{\nu} \tau_{\nu} \approx n\bar{\tau}$, where τ_{ν} are the time intervals between subsequent reductions and $\bar{\tau}$ their mean value, one obtains $\delta = \delta_0 \prod_{\nu} \cos \omega \tau_{\nu}$ in Eq. (4).

I now consider the important cases where³

$$\omega \tau_{\nu} \ll 1, \quad \text{i.e., } \tau_{\nu} \ll T = 1/\omega. \quad (5)$$

Then

$$\prod_{\nu=1}^n \cos \omega \tau_{\nu} \approx \prod_{\nu=1}^n \left[1 - \frac{1}{2} (\omega \tau_{\nu})^2 \right] \approx \exp \left(-\frac{1}{2} \omega^2 \sum_{\nu=1}^n \tau_{\nu}^2 \right).$$

If all τ_{ν} were equal this would lead to $\exp\{-n\omega^2\bar{\tau}^2/2\}$. Assuming more realistically a stochastic distribution with distribution function $e^{-\tau/\bar{\tau}}$ corresponding to a Poisson distribution for the counting rate within a given time interval, then $\sum_{\nu} \tau_{\nu}^2 = 2n\bar{\tau}^2 = 2\bar{\tau}t$ for large n and one obtains

$$\delta_t = \delta_0 e^{-\lambda t}, \quad \text{with } \lambda = \omega \times \omega \bar{\tau} = \omega \times \bar{\tau} / T. \quad (6)$$

Thus the relaxation time $1/\lambda = T \times T/\bar{\tau} \gg T$ is much larger than $T = 1/\omega$ if (5) holds. This blocking of the states φ_1 and φ_2 by stochastic "reduction of the state vector" prevents oscillation between φ_1 and φ_2 and is responsible for enhanced stability.

The same analysis may be performed also if the Hamiltonian has not the high symmetry (parity conservation) with respect to φ_1 and φ_2 as assumed above. If its eigenstates are $\varphi = \alpha\varphi_1 + \beta\varphi_2$ and $\varphi' = -\beta^*\varphi_1 + \alpha^*\varphi_2$ with $|\alpha|^2 + |\beta|^2 = 1$, one obtains $\delta_t = \delta_0 \prod_{\nu} (1 - 3|\alpha\beta|^2 \sin^2 \frac{1}{2} \omega \tau_{\nu})$. With (5) this leads to the replacement of λ in (6) by $\lambda = 4|\alpha\beta|^2 \times \omega\bar{\tau}/T \leq \omega\bar{\tau}/T$ where one notes that $|\alpha|^2 + |\beta|^2 = 1$ implies $4|\alpha\beta|^2 \leq 1$ with equality holding in the symmetric case $|\alpha|^2 = |\beta|^2 = \frac{1}{2}$.

The model considered so far is rather schematic in several respects. First of all it is clear that, in particular for microscopic systems, the interaction with a probe like a photon does not necessarily lead to orthogonal states φ_1^P and φ_2^P for given incoming state φ_0^P . If it does not, then one obtains only a *partial reduction* of the density matrix ρ :

$$\begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} \rightarrow \begin{pmatrix} \rho_{11} & \xi\rho_{12} \\ \xi^*\rho_{21} & \rho_{22} \end{pmatrix}, \quad (7)$$

where $\xi = (\varphi_2^P, \varphi_1^P)$. Since the states are normalized to unity, $|\xi| < 1$ unless $\varphi_1 = \alpha\varphi_2$. After m reduction with ξ_{μ} the off-diagonal elements are mul-

tiplied by

$$\xi^{(m)} = \prod_{\mu=1}^m \xi_{\mu},$$

which approaches zero for large m if $|\xi_{\mu}| < 1$. This is in line with the previously discussed relation to the process of measurement since even if the object may not be localized with one photon, in example 1 for instance, this may still be done with many of them, which then may be considered as one probe¹ leading to a (almost) complete reduction, or as many individual probes each leading to a partial reduction. From the physics involved one thus expects the blocking effect to be working similarly where, of course, one has to require now that m partial reductions with $|\xi^{(m)}| \ll 1$ should take place in a time interval which is short compared to $T = 1/\omega$. This is borne out also by the formal analysis whose result is given here for symmetric (parity-conserving) Hamiltonian as in Eqs. (3)–(6). Under the condition that subsequent reductions are statistically independent one can—for the analysis of the average relaxation—replace τ_{ν} by $\bar{\tau}$, τ_{ν}^2 by $2\bar{\tau}$ (see above), and the ξ_{ν} by their *average* ξ with $|\xi| < 1$. The relevant condition replacing (5) turns out to be

$$\omega\bar{\tau}/(1 - |\xi|) \equiv \omega\tau_0 \ll 1. \quad (8)$$

After a sequence of n evolutions under H as in (3) and subsequent reductions according to (7), an initial density matrix $\rho(0)$ turns into $\rho(t)$, where $t = \sum_{\nu} \tau_{\nu} = n\bar{\tau}$,

with⁴

$$\begin{aligned} \delta_t &= e^{-\lambda' t} \left\{ \delta_0 - 2 \operatorname{Im} \left[\frac{\omega\bar{\tau}}{1 - \xi} (1 - \xi^n) \rho_{12}(0) \right] + O_1(\omega^2 \tau_0^2) \right\}, \\ \rho_{12}(t) &= e^{-\lambda' t} \left\{ \xi^n \rho_{12}(0) + \frac{i}{2} \frac{\omega\bar{\tau}\xi}{1 - \xi} (1 - \xi^n) \delta_0 + O_2(\omega^2 \tau_0^2) \right\}, \quad \lambda' = \omega^2 \bar{\tau} \operatorname{Re} \frac{1}{1 - \xi} \equiv \omega^2 \tau_{\text{eff}}, \end{aligned} \quad (9)$$

where $\delta_t = \rho_{11}(t) - \rho_{22}(t)$ as in Eq. (4). Now $|\xi|^n = \exp(t \ln |\xi|/\bar{\tau}) < \exp(-t/\tau_0)$, implying that the off-diagonal element ρ_{12} is effectively reduced to zero after a run-in time of a few times τ_0 . The condition (8) implies $\tau_0 \ll \omega^{-1} \ll \lambda^{-1}$, and $|\omega\bar{\tau}/(1 - \xi)| \ll 1$ in (9) so that spontaneous symmetry breaking and blocking is recovered with modified relaxation time $1/\lambda'$ as expected. This may, of course, be generalized to nonsymmetric evolution as discussed for complete reduction.

In conclusion it is seen that even if single reductions are very weak, i.e., if $(1 - |\xi|) \ll 1$, spontaneous symmetry breaking and blocking persist as long as (8) holds. This ensures a wide

applicability of this phenomenon.

The second oversimplification of the model analyzed is that H^0 is two dimensional and that φ_1^0 and φ_2^0 are reproduced exactly. Of course, the whole discussion can be generalized to l different mutually orthogonal states $\varphi_1^0 \dots \varphi_l^0$ without any principal change. Of some interest, however, is the generalization to groups of states or subspaces H_i^0 of H^0 such that starting from given initial states $\varphi_i^0 \in H_i^0$ subsequent interactions with probes not necessarily reproduce φ_i^0 but still lead to (not necessarily pure) states which are again in H_i^0 for all i . This interaction

then leads to the depletion of matrix elements of the density matrix between states from different H_i^0 in the same way and under the same circumstances as for the case of one-dimensional H_i^0 analyzed explicitly above. The general features of the blocking effect are expected to prevail.

In conclusion I restate the main ingredients of the mechanism presented here. It is based on the interaction of the object under consideration with a background of probes in its natural surrounding. The relevant background may consist for instance of electromagnetic or corpuscular radiation and perhaps even neutrinos, or molecules within a gas or fluid, and eventually also phonons, etc., in a solid, depending on the kind of object studied. The characteristic behavior of the object under the influence of this background is governed by two criteria:

(i) *The inertness criterion* (a) singles out the *inert states* to which reduction takes place such that (b) the interaction with the background (probes) does not induce transitions between these states. It thus defines the axes of spontaneous symmetry breaking. For macroscopic bodies it obviously singles out macroscopically localized states. If some interactions violate condition (b) independent of the choice of inert states, the state relaxes to unit density matrix, i.e., "total chaos." Of course, small violation of condition (b) can be tolerated as long as the relaxation time obtained from these transitions is long as compared to the other characteristic times of the problem at hand.

(ii) *The frequency criterion* for the blocking effect defines the regime in which the latter dominates over the free evolution under the Hamiltonian describing the object. In the cases calculated here it is given by Eqs. (5) and (8).

From the thermodynamical point of view the frequency criterion implies that the object is in a temperature bath with $kT \gg \hbar\omega$ (T = absolute temperature, k = Boltzmann constant) implying unit equilibrium density matrix. However, under the conditions of the inertness criterion, *the shorter $\bar{\tau}$ and the smaller ξ , i.e., the more intense the coupling, the longer is the relaxation time $1/\lambda$ or $1/\lambda'$.*

The mechanism of spontaneous symmetry breaking and blocking of metastable states so obtained applies to many macroscopic systems and compounds which would not be stable otherwise and thus seems to play a crucial role for the stability and classical property of the macroscopic world.⁵ It eliminates the need for special (cosmological)

initial conditions in order to obtain macroscopically localized states.⁶ It is expected to be important also in molecular physics for the localization of atoms in molecules (and thus the applicability in the widely used Born-Oppenheimer approximation), guaranteeing in particular the stability of stereoisomers, chiral molecules (such as sugar), etc. To what extent it operates also for intrinsic degrees of freedom (including P and/or CP) of elementary particles is an open question, which seems, however, certainly worthwhile studying.

This work was supported in part by the Swiss National Science Foundation.

¹The connection with the problem of measurement and the interpretation of quantum mechanics will be discussed in more detail elsewhere. Here I only remark that starting from a purely statistical (ensemble) interpretation of quantum mechanics one obtains in this way under macroscopic conditions (see example 1 below) a description of individual systems.

²In principle one could recover coherence by including in the observation the final states of *all* such photons.

³Of course I implicitly assume also that the collision time between object and probe is short compared to τ_v . This is, however, relevant only for the exact calculation, not for the general feature of the effect discussed. (See the partial reduction discussed below.)

⁴ $|O_i(x^2)| \leq C_i x^2$ for $x \ll 1$ with C_i independent of n .

⁵For instance, $T = 1/\omega \approx 1$ yr and $\tau \approx 10^{-5}$ sec imply $1/\lambda \approx 3 \times 10^{12}$ yr according to (6). On the other hand, for molecules with $T = \omega^{-1} \approx 10^{-2}$ sec it may be possible to study the blocking effect experimentally, though perhaps not in the extreme limit (8), for instance in a molecular beam crossing laser light or going through a gas of appropriate pressure.

⁶For a recent synopsis (with references) of other approaches to establish quantum mechanics as a general theory whose applicability includes the classical domain see J.-M. Lévy-Leblond, in *Quantum Mechanics, A Half Century Later*, edited by J. Leite Lopes and M. Paty (Reidel, Dordrecht, 1977), pp. 187-206. Note, however, that no attempt is made here to prove a reduction of the form (2) for the combined system described on $H^O \otimes H^P$. Unlike other approaches the probe can therefore be a simple microscopic system, which is obviously an important feature. On the other hand, no coherence can prevail between different "pointer" positions (or living and dead cats, etc., so to speak) in the case of a *macroscopic* apparatus under usual conditions, i.e., in the presence of radiation, air, etc. (necessary for living cats).

LETTERS

Coherently wired light-harvesting in photosynthetic marine algae at ambient temperature

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Photosynthesis makes use of sunlight to convert carbon dioxide into useful biomass and is vital for life on Earth. Crucial components for the photosynthetic process are antenna proteins, which absorb light and transmit the resultant excitation energy between molecules to a reaction centre. The efficiency of these electronic energy transfers has inspired much work on antenna proteins isolated from photosynthetic organisms to uncover the basic mechanisms at play^{1–5}. Intriguingly, recent work has documented^{6–8} that light-absorbing molecules in some photosynthetic proteins capture and transfer energy according to quantum-mechanical probability laws instead of classical laws⁹ at temperatures up to 180 K. This contrasts with the long-held view that long-range quantum coherence between molecules cannot be sustained in complex biological systems, even at low temperatures. Here we present two-dimensional photon echo spectroscopy^{10–13} measurements on two evolutionarily related light-harvesting proteins isolated from marine cryptophyte algae, which reveal exceptionally long-lasting excitation oscillations with distinct correlations and anti-correlations even at ambient temperature. These observations provide compelling evidence for quantum-coherent sharing of electronic excitation across the 5-nm-wide proteins under biologically relevant conditions, suggesting that distant molecules within the photosynthetic proteins are ‘wired’ together by quantum coherence for more efficient light-harvesting in cryptophyte marine algae.

Cryptophytes are eukaryotic algae that live in marine and freshwater environments. They are members of an evolutionary group notable because their photosynthetic apparatus was acquired from red algae by a sequence of endosymbiotic events. As a result, cryptophyte photosynthetic antenna proteins (phycobiliproteins) exhibit exceptional

spectral variation between species because they use mainly tunable linear tetrapyrroles (bilins) for light-harvesting. Another remarkable feature of cryptophytes is that they can photosynthesize in low-light conditions, which suggests that the absorption of incident sunlight by phycobiliprotein antennae in the intrathylakoid space¹⁴ and the subsequent transfer of that energy among these proteins and eventually to the membrane-bound photosystems is particularly effective¹⁵. Theory indicates that fast energy transfer is facilitated by small interchromophore separations², yet the average nearest-neighbour centre-to-centre separation of chromophores within cryptophyte light-harvesting antenna proteins (Fig. 1) is ~ 20 Å (ref. 16)—about double that for the major light-harvesting protein in plants. To explore how a light-harvesting antenna can function efficiently with such a counter-intuitive design, we study the antennae of two marine cryptophytes, phycoerythrin PE545 from *Rhodomonas* CS24 and phycocyanin PC645 from *Chroomonas* CCMP270 at ambient temperature (294 K) using two-dimensional photon echo (2DPE) spectroscopy^{10–13}.

PC645 contains eight light-absorbing bilin molecules covalently bound to a four-subunit protein scaffold¹⁷. Its structure, determined to 1.4-Å resolution by X-ray crystallography¹⁸ and shown in Fig. 1a, exhibits approximate twofold symmetry. A dihydrobiliverdin (DBV) dimer (green) located in the centre of the protein and two mesobiliverdin (MBV) molecules (blue) located near the protein periphery give rise to the upper half of the complex’s absorption spectrum (Fig. 1c), spanned by our laser pulse spectrum. The electronic coupling of ~ 320 cm⁻¹ between the DBV molecules C and D (labelled according to the protein subunit that binds them) leads to delocalization of the excitation and yields the dimer electronic excited states, or so-called molecular excitonic states¹⁹, labelled DBV₊ and DBV₋. Excitation

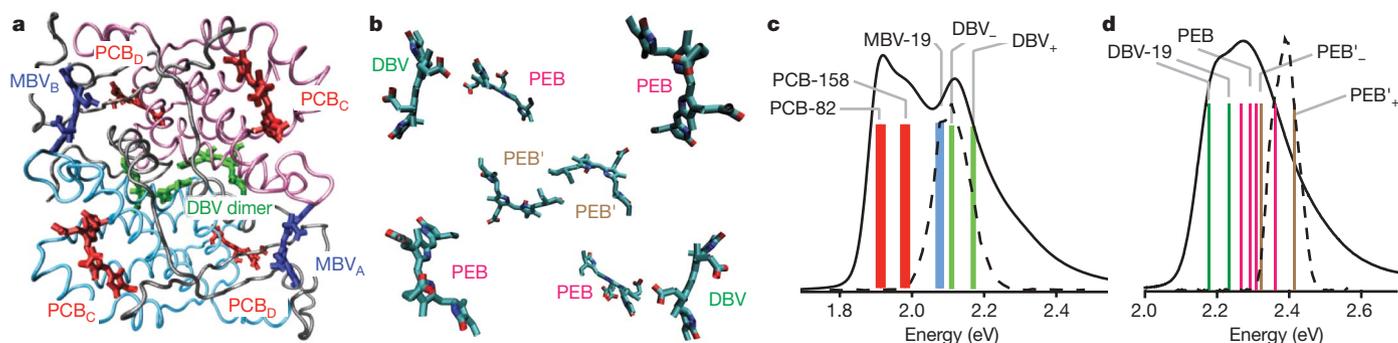


Figure 1 | Structure and spectroscopy of cryptophyte antenna proteins. **a**, Structural model of PC645. The eight light-harvesting bilin molecules are coloured red (PCB), blue (MBV) and green (DBV). **b**, Chromophores from the structural model for PE545 showing the different chromophore incorporation. **c**, Electronic absorption spectrum of isolated PC645 protein

in aqueous buffer (294 K). The approximate absorption energies of the bilin molecules are indicated as coloured bars. **d**, Electronic absorption spectrum of isolated PE545 protein in aqueous buffer (294 K) with approximate absorption band positions indicated by the coloured bars. The spectrum of the ultrafast laser pulse is plotted as a dashed line in **c** and **d**.

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energy absorbed by the dimer flows to the MBV molecules, which are each 23 Å from the closest DBV, and ultimately to four phycocyanobilins (PCB, coloured red) that absorb in the lower-energy half of the absorption spectrum.

The structure of PE545 (Fig. 1b) is closely related to that of PC645 except that the bilin types differ^{16,20}. The lowest-energy chromophores are DBV bilins. The dimer consists of phycoerythrobilin chromophores PEB', with the prime indicating they are doubly covalently bound to the protein. The remaining chromophores are singly bound PEBs. The electronic couplings between the chromophores are reported elsewhere²¹. The approximate absorption spectrum and band positions are shown in Fig. 1d.

For the experiments the proteins were isolated from the algae and suspended at low concentration in aqueous buffer at ambient temperature (294 K). The femtosecond laser pulse (25-fs duration) excites a coherent superposition of the antenna protein's electronic-vibrational eigenstates (absorption bands). The initial state of the system is thus prepared in a non-stationary state²², where electronic excitation is localized to a greater or lesser degree compared to the eigenstates. The time-dependent solution to quantum dynamics for electronically coupled molecules with this initial condition predicts that excitation subsequently oscillates among the molecules under the influence of the system Hamiltonian until the natural eigenstates are restored owing to interactions with the environment. 2DPE provides a means of observing this experimentally, enabling us to explore the significance of quantum coherence.

Representative 2DPE data for PC645 are shown in Fig. 2 with positions on the diagonal assigned to absorption bands. Rich features such as cross-peaks and excited state absorptions are evident. In the 2DPE experiment the two-pulse excitation sequence (sweeping $\tau > 0$) can prepare population density, for example $|\text{DBV}_-\rangle\langle\text{DBV}_-|$, that evolves during the delay time T and can be probed as a bleach signal on the diagonal part of a rephasing 2DPE spectrum. Alternatively, off-diagonal contributions like $|\text{DBV}_+\rangle\langle\text{DBV}_-|$ can be excited when the pump pulse sequence interacts coherently with both absorption bands. The resulting signal will be probed as a cross-peak above the diagonal in rephasing spectra that oscillate as a function of T with frequency $\phi = 2\pi(E_{\text{DBV}_+} - E_{\text{DBV}_-})/h$ because it carries a phase

$\exp(-i\phi T)$. Similarly the complementary coherence $|\text{DBV}_-\rangle\langle\text{DBV}_+|$ will contribute a cross-peak below the diagonal in rephasing spectra that will carry an opposite phase, $\exp(+i\phi T)$.

These predicted coherent oscillations can be reproducibly seen in our 2DPE spectra by plotting the intensity of rephasing spectra at lower and upper cross-peaks as a function of waiting time T (Fig. 2b and c). The red line indicates the cross-peak above the diagonal, the black line is that below. As mentioned above, the upper and lower cross-peak oscillations should differ by a phase factor determined by the sign of the energy difference between the states in superposition, leading to anti-correlated upper and lower cross-peak beats with a dominant frequency component equal to the eigenvalue energy difference. Such behaviour is indeed clearly seen in the experimental data, with the anti-correlated oscillations providing striking evidence that both DBV dimer and DBV–MBV electronic superposition states persist for more than 400 fs after photo-excitation. It is remarkable that electronic coherence spans from the DBV dimer to the peripheral MBV molecules, over a distance of 25 Å.

The PC645 cross-peak beating is complex; multiple frequencies arise for the same reason they do in a simple mechanical system comprised of a mass connected by a weak spring to a pair of masses coupled by a strong spring. And because these data were recorded at room temperature, line broadening is significant, so that overlapping bands partly obscure oscillating features. Nevertheless, Fourier transforms of these data (Supplementary Fig. 1) suggest the presence of frequencies in these beating patterns that can be related to the frequency differences between absorption bands. A careful global analysis of the data (see Supplementary Information) provides evidence that the oscillations in the 2DPE data can be decomposed into components corresponding to frequency differences between absorption bands and that—most importantly—the cross-peak beats at each frequency are anti-correlated.

For a comparison with the PC645 results, we also undertook experiments on the PE545 antenna protein by exciting the blue side of the absorption. A typical rephasing 2DPE spectrum (that is, scanned so that $\tau > 0$) is shown in Fig. 3a. To show beats across the entire anti-diagonal slice through the PEB/PEB' cross-peaks we plot the intensity of the 2DPE rephasing spectrum along the anti-diagonal line drawn in Fig. 3a as a function of population time T (Fig. 3b).

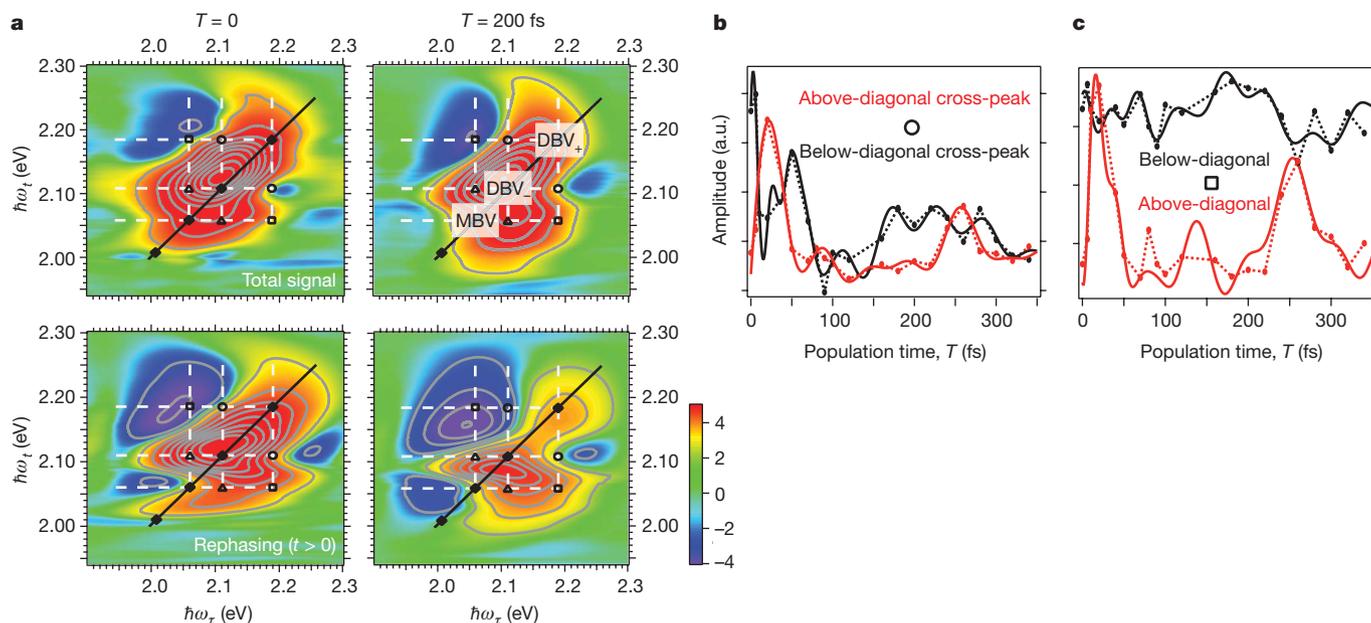


Figure 2 | Two-dimensional photon echo data for PC645. **a**, The left column shows the total real 2DPE spectrum recorded for PC645 at zero waiting time ($T = 0$), together with the rephasing contribution to this signal. The right column shows the data for $T = 200$ fs. The 2DPE spectra show the signal intensity on an arcsinh scale (colour scale, arbitrary units) plotted as a function

of coherence frequency ω_r and emission frequency ω_t . **b**, Intensity of the DBV dimer cross-peaks (open circle) as a function of time T . **c**, Intensity of the MBV–DBV₊ cross-peaks (open square) as a function of time T . The dashed lines interpolate the data points (solid circles). The solid line is a fit to a sum of damped sine functions (Supplementary Information). a.u., arbitrary units.

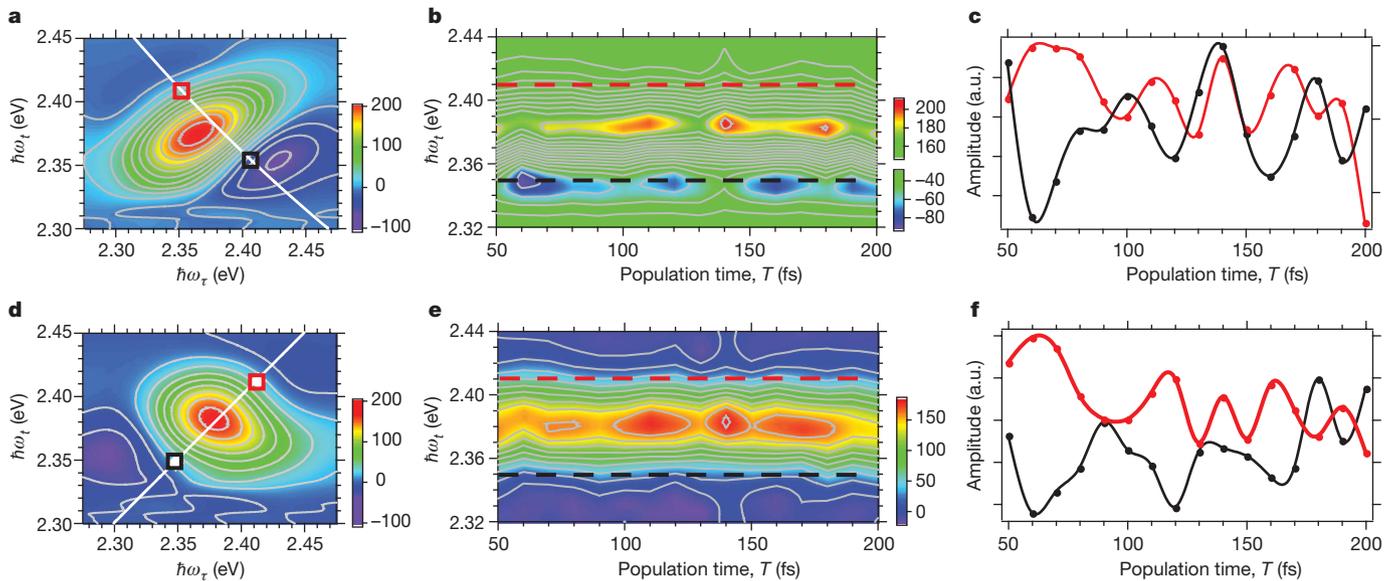


Figure 3 | Two-dimensional photon echo data for PE545. **a**, 2DPE spectrum (rephasing real signal) for PE545 recorded at $T = 100$ fs. **b**, The intensity of the 2DPE rephasing spectrum along an anti-diagonal slice through the cross-peaks versus population time T . Upper and lower cross-peaks are indicated by red and black dashed lines respectively. **c**, Intensity

Oscillations of the main bleach and excited-state absorption peaks are clearly evident. The beats in the centre of this plot are cross-peaks excited because the PEB and PEB' absorption bands overlap owing to spectral line broadening. The cross-peaks, indicated by dashed horizontal lines in Fig. 3b (red is the cross-peak above the diagonal, the black line is that below), are more clearly seen in Fig. 3c, in which the beats are well resolved and markedly anti-correlated—a signature of quantum coherence. These oscillations are directly analogous to those observed for PC645 (compare Fig. 2b).

Figure 3d–f shows plots similar to those in Fig. 3a–c, but for non-rephasing spectra ($\tau < 0$) of PE545. The same electronic coherences giving rise to oscillating cross-peaks in rephasing spectra are predicted to cause oscillations at the diagonal positions of non-rephasing 2DPE spectra²³. Indeed, we observe a clear phase relationship between beats within the rephasing and non-rephasing spectra and also between the data sets, which is compelling evidence for the presence of long-lived quantum coherence. The first ~ 130 fs of these spectra reproducibly show clear oscillations with a period of ~ 60 fs ($\nu \approx 500$ cm^{-1}). After this time the oscillation pattern becomes more complicated, suggesting that the initial coherence may evolve owing to coupling with other molecules in the protein.

In our experiments the light-harvesting process in both PC645 and PE545 antenna proteins involves quantum coherence at ambient temperature, suggesting that coherence may more generally be used by cryptophyte algae. Quantum coherence occurs in an intermediate regime of energy transfer where there is a complex balance between quantum interference among electronic resonances and coupling to the environment causing decoherence²⁴. There still remains the question of precisely how quantum coherence can persist for hundreds of femtoseconds in these biological assemblies. In an isolated molecule, electronic decoherence arises from the decay of the overlap $S(t) = \langle v_2(t) | v_1(t) \rangle$ between the unobserved vibrational wavepackets $|v_1(t)\rangle$ and $|v_2(t)\rangle$ associated with the lower and upper electronic states respectively^{25,26}. In 2DPE experiments, the observable includes both vibrational and electronic components and, as such, decoherence due to decay of $S(t)$ is not manifest in the data. Rather, the slow decay of electronic coherence reflects the interaction of vibronic superposition states with the external environment.

Recent studies have attributed the slow dephasing of electronic coherence to the presence of shared or correlated motions in the

oscillations in the cross-peaks (red and black squares in **a**). **d**, 2DPE spectrum (non-rephasing real signal) for PE545 recorded at $T = 100$ fs. **e**, **f**, As for **b** and **c** but for the 2DPE non-rephasing spectrum along the diagonal slice. The 2DPE spectra are plotted on a linear intensity scale.

surrounding environment^{6,7,27,28}. In this context, we note that, unlike most photosynthetic pigments that are non-covalently complexed to their protein environment (chlorophyll via histidine residues, for example), the bilins in PC645 are covalently bound to the protein backbone. Covalent attachment of the chromophores to their protein environment may support or strengthen correlated motions between chromophores and protein and thus be an important factor in slowing down decoherence in cryptophyte antenna proteins at ambient temperature, thereby differentiating them from many other photosynthetic light-harvesting antennae. We also note that the precise manifestation of long-lived quantum-coherence depends on the photo-excitation conditions^{22,29,30}, and cryptophyte algae are obviously using sunlight that does not arrive in the form of laser pulses as used in our experiments. Nevertheless, the couplings giving rise to the long-lived quantum coherence that we clearly observe at ambient temperature will still be present and strongly suggest that quantum effects facilitate the efficient light-harvesting by cryptophyte algae. That is, long-lived quantum coherence can facilitate energy transfer by 'wiring' together the final energy acceptors (PCB in the case of PC645 and DBV for PE545) across a single protein unit, and thereby help to compensate for the exceptionally large average interchromophore separations in these antenna proteins.

METHODS SUMMARY

Cryptophyte *Chroomonas* sp. (CCMP270 strain, National Culture Collection of Marine Phytoplankton, Bigelow Laboratory for Ocean Sciences, USA) and *Rhodomonas* sp. (CS24) were cultivated and harvested, and the phycobiliproteins were isolated by usual procedures²⁰. 2DPE experiments were performed as described in refs 11 and 27. The laser pulse duration and chirp were measured using transient grating frequency resolved optical gating (TG-FROG) experiments on a solvent (typically ethanol). The time-bandwidth product was estimated to be about 0.53, close to the ideal transform-limited condition for a Gaussian pulse. During data collection for PC645, for any given population time T , τ was scanned from -200 to 200 fs with 0.25-fs steps. Each 2D map at a fixed T is the average of at least three separate scans, and each series of 2D scans at different T was further repeated on different days for comparison. For PE545, τ was scanned from -60 to 60 fs with 0.15-fs steps, and each 2D map is the average of two separate scans. Additionally, PE545 was measured at this wavelength numerous times with different T steps, on different days. The samples were moved after each scan and absorption spectra taken before and after each series of scans confirmed that the sample did not degrade during the measurements.

Full Methods and any associated references are available in the online version of the paper at www.nature.com/nature.

Received 14 July; accepted 17 December 2009.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

Acknowledgements This work was supported by the Natural Sciences and Engineering Research Council of Canada and the Australian Research Council. G.D.S. acknowledges the support of an EWR Steacie Memorial Fellowship.

Author Contributions E.C. performed the experiments on PC645 and analysed those data. C.Y.W. performed the experiments on PE545 and analysed those data. K.E.W. prepared the samples. P.M.G.C. and G.D.S. designed the research. P.B. and G.D.S. examined the interpretation of the results. G.D.S. wrote the paper. All authors discussed the results and commented on the manuscript.

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METHODS

Cryptophyte *Chroomonas* sp. (CCMP270 strain, National Culture Collection of Marine Phytoplankton, Bigelow Laboratory for Ocean Sciences, USA) was cultured at 20 °C under constant low light illumination (12-V white fluorescent tubes, 300 lx at 0.3 m) in a modified 'Fe' medium. The algal cells were harvested and passed twice through a French press cell at a pressure of 1,000 psi. The resultant solution was centrifuged, yielding a supernatant solution containing PC645. PC645 was isolated using gradual ammonium sulphate precipitation from 0 to 80%. The pellets were re-suspended in a minimal volume of 25 mM phosphate buffer at pH 7.1. Further purification was continued using a combination of ion-exchange and size-exclusion chromatography. About 20 ml of pure PC645 was concentrated to approximately 200 μ l using a 10-kDa Amicon Centriprep and then frozen using liquid nitrogen before being stored at -80 °C. A buffer solution, prepared from a 25 mM solution of HEPES (4-(2-hydroxyethyl)-1-piperazineethanesulphonic acid) in deionized water, adjusted to pH = 7.5 by the addition of concentrated NaOH solution, was used to prepare dilute PC645 samples for the experiments.

Rhodomonas sp. (CS24) was cultivated and harvested as previously reported²⁰. Cell pellets were re-suspended in buffer A (0.05 M Mes (pH 6.5) with 1 mM NaN₃) and homogenized in a Teflon glass homogenizer followed by passage through a French press at a pressure of 1,000 psi. The resultant solution was centrifuged for 30 min at 17,000g, producing a pellet of cell debris containing thylakoid membranes and a supernatant containing phycoerythrin. PE545 was purified from the supernatant as described elsewhere²⁰.

2DPE experiments were performed as described in refs 11 and 27. A Ti:sapphire regeneratively amplified laser system was used to pump a Noncollinear Optical Parametric Amplifier (NOPA) to produce 25-fs duration pulses centred at 590 nm for PC645 experiments or at 520 nm for PE545 for the results reported here, with a

spectral bandwidth of about 25 nm and repetition rate of 1 kHz. The laser pulse duration and chirp were measured using TG-FROG experiments on a solvent (typically ethanol). The time-bandwidth product was estimated to be about 0.53, close to the ideal transform-limited condition for a Gaussian pulse. The pulse from the NOPA was split by a 50% beam splitter and the two resulting beams were overlapped in a diffractive optic, producing two pairs of phase-locked beams in a boxcars phase-matched geometry. The delay time T was controlled by a motorized translation stage inserted in one beam path before the diffractive optic, whereas the delay time τ was introduced by means of movable glass wedge pairs, calibrated by spectral interferometry¹¹.

During data collection for PC645, for any given population time T , τ was scanned from -200 to 200 fs with 0.25-fs steps. Each 2D map at a fixed T is the average of at least three separate scans, and each series of 2D scans at different T was further repeated on different days for comparison. For PE545, τ was scanned from -60 to 60 fs with 0.15-fs steps, and each 2D map is the average of two separate scans. Additionally, PE545 was measured at this wavelength numerous times with different T steps, on different days. The samples were moved after each scan and absorption spectra taken before and after each series of scans confirmed that the sample did not degrade during the measurements.

To ensure that the local oscillator did not influence the response of the system, its intensity was attenuated by about three orders of magnitude relative to the other beams and the time ordering was set so that the local oscillator always preceded the probe by \sim 500 fs. The resulting local oscillator-signal interference intensity was focused into a 0.63-m spectrograph (25- μ m slit) and recorded using a 16-bit, 400 \times 1,600 pixel, thermo-electrically cooled charge-coupled device (CCD) detector. Subtraction of unwanted scatter contributions, Fourier windowing, transformation and phase retrieval were performed as reported previously¹¹.