Dimensional crossover in spin diffusion: A manifestation of the quantum Zeno effect

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The quantum Zeno effect (QZE) implies that a too frequent ($\omega_\phi \to \infty$) observation of a quantum system would trap it in its initial state, even though it would be able to evolve to some other state if not observed. In our scheme, interacting spins in a three-dimensional cubic lattice, ''observe'' each other with a frequency $\omega_\phi \propto \sqrt{J_x^2+J_y^2+J_z^2}/\hbar$, where the J's are the coupling constants. This leads to a ''diffusive'' spread of a local excitation characterized by the constants $D_\mu \propto J_\mu^2/\omega_\phi$. Thus, a strongly asymmetric interaction (e.g., $J_y/J_{x(z)} \gg 1$), would hinder diffusion in the perpendicular directions ($D_{x(z)} \to 0$) manifesting the QZE. We show that this effect is present in numerical solutions of simple two-dimensional systems. This reduction in the diffusion kinetics was experimentally observed in paramagnetic compounds where the asymmetry of the interaction network manifests through an exchange narrowed linewidth. Experimental designs are proposed. [S0163-1829(98)04409-9]

Quantum dynamics of magnetic excitations in a system of interacting spins at high temperature is an active field of research.[1] From a macroscopic scope, i.e., for long times and large wavelengths, one expects that excitations should evolve irreversibly. The resulting hydrodynamic equations[2] describe the ''spin diffusion.'' In this regime a local excitation decays as $(Dt)^{-d/2}$, where d is the dimension of the space. From the microscopic point of view, however, dynamics is governed by reversible quantum mechanics as long as quantum coherence is maintained. Although this dynamics *seems* to be ''diffusive'' already at intermediate times, a careful study[3] of low-dimensional systems can recognize quantum interferences. Its experimental observation[4,5] constitutes a fingerprint of the bounded regions where the dynamics occurs. In addition an experimental realization of a ''Loschmidt daemon,''[6] which allows the evolution backwards in time, can be achieved by inverting the sign of the effective Hamiltonian. In this case, ''irreversible'' interactions are simply those we do not control. Hence, reversibility is not total.[7] This effect is stronger than what can be inferred from the magnitude of the noninverted terms. In fact, the apparently ''diffusive'' dynamics of the many-body interaction seems to transform small residual interactions into efficient mechanisms to stabilize an irreversible diffusion.[8] Irreversible effect is often amplified even by those interactions we *can* control. This is analogous to the Drude approximation for the electrical resistance of an impure metal at low temperatures. There, the *reversible* elastic scattering with impurities with rate $(1/\tau_{\rm imp})$ facilitates the interaction with the thermal bath. This bath acts through the uncontrollable electron-phonon interactions or other dephasing collisions with rate $1/\tau_\phi$. The remarkable consequence is that in first approximation the diffusion constant does not depend on τ_ϕ but it is $D \propto v^2 \tau_{\rm imp}$, where v is a typical velocity for ballistic propagation of the excitation. Its lesson is that a reversible interaction can provide an evolution close enough to diffusion. Then, when actual irreversible processes occur, they stabilize[9] the evolution into an irreversible diffusion. The observed diffusion constant remains unchanged. This gives the ultimate justification for the *stosszahlansatz* or assumption of randomness after successive collisions in which no memory of the previous quantum state is retained. This is equivalent to considering a collision with an impurity as a classical measurement or wave function collapse. This, of course, is an approximation which breaks down close to the localized regime where interferences play a fundamental role.

The goal of the many-body techniques is to provide the match between the quantum and hydrodynamic regimes providing the machinery for the calculation of the diffusion constant and other macroscopic observables. However, because of the implicit approximations, this is sometimes done at the cost of the physical insights into the nature of irreversibility. To understand the dynamics of a local excitation, we notice that one up spin, in a lattice with all down spins, propagates with a typical ''ballistic'' velocity proportional to the exchange constant J. At high temperature, there are as many up spins as down spins. Hence, this propagation in the sublattice of down spins is interrupted by the modification of this lattice. This produces a ''collision'' rate $(1/\tau_{\rm mb})$ also proportional to J. If we neglect quantum interferences (*stosszahlansatz*) produced by multiple collisions, a diffusion constant $D \propto v^2 \tau_{\rm mb} \propto J$ is obtained. Again the point is that a diffusive behavior is a good approximation for the dynamics. It will be stabilized by later irreversible interactions. Keeping this in mind, we may *consider* each ''*collision*'' *as a measurement* process.

Quantum dynamics is strongly modified by recurrent measurements. This phenomenon, known as the quantum Zeno effect (QZE),[10] has been applied with particular success in quantum optics.[11] In simple words, it affirms that if the evolution of a quantum system is observed too fre-

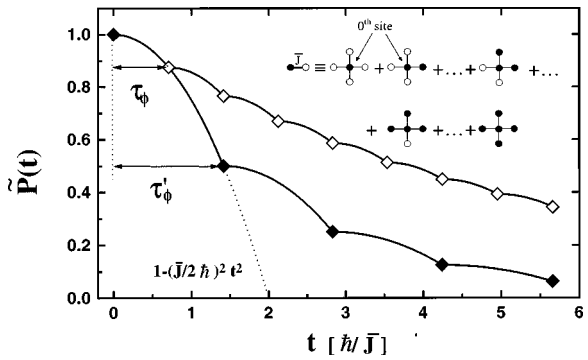

FIG. 1. Probability of stay for a particle whose quantum evolution is interrupted by a measurement process with a period τ_ϕ and $\tau'_\phi = 2\tau_\phi$. The coherent evolution is approximated up to the quadratic term and the return probability is neglected. The increase of the probability of stay is a manifestation of the QZE. The evolution shown is equivalent to the average evolution of the ensemble in the inset. Only 5 of the 16 configurations corresponding to a local excitation in the high-temperature limit are sketched.

quently, there will be no evolution to be seen. While this might sound paradoxical from a classical point of view, within quantum logic this is not a paradox at all, since a measurement involves a collapse of the wave function and the start of a new quantum evolution. For short times, the probability of staying in the initial state is $P_{i,i}(t)=1-(\overline{J}t/2\hbar)^2+\cdots$, with $\overline{J}$ being an average exchange energy. The lack of a linear term on this expansion has very important consequences. If the evolution in a time t_s is interrupted by N observations, the final probability of stay is $\tilde{P}_{i,i}(t=t_s)=[P_{i,i}(t_s/N)]^N \to 1$ when $N\to\infty$. Here, $t_s/N=\tau_\phi$ defines the decoherence time. Hence one sees why a frequent collapse (i.e., $\omega_\phi=1/\tau_\phi \gg 1$) hinders evolution.[12] The result of the successively interrupted evolution is shown schematically in Fig. 1.

This article addresses a paradoxical aspect of the dimensional dependence in the dynamics of magnetic excitations in light of the QZE. In order to make it more obvious, let us first discuss the particular limit of a cubic lattice where the interaction along direction y grows ($J_y/J_{x(z)}\to\infty$). The diffusion coefficient in that direction will also grow, $D_y \propto J_y$. However, the spreading rate in the other directions will decrease, $D_{x,(z)} \propto 1/J_y$. The essence of the argument that we are going to develop is that spins spreading in each xz plane register the evolution in the other parallel planes through the many-body coupling J_y (i.e., they ''observe'' each other with a frequency $\omega_\phi \to J_y/\hbar$), producing the reduction of the dynamics within the planes. This is a general behavior also valid for the nonperturbative limit of $J_y \approx J_{x(z)}$. To our knowledge this has not been noticed previously.

The calculation of the dynamics of a local spin polarization in the high-temperature regime can be mapped to a system of Fermi particles on a lattice. Up spins are identified with particles and down spins with holes. A particle is localized at the excited site with the rest of the sites being occupied with probability $\frac{1}{2}$. The initial state is an incoherent superposition of all the possible initial states a few of which are shown in the inset of Fig. 1. The particle initially at zeroth site starts to evolve having a finite probability amplitude to jump into empty neighboring sites. Meanwhile, other particles also can move in and out of neighboring sites creating a fluctuating effective potential. If this is approximated by a stochastic potential, one obtains an irreversible equation satisfying the hydrodynamic limits. The essential point is that the correlation times of the potential are the same as those that characterize the dynamics of a particle in its fluctuating environment. This leads to a self-consistent equation which is the core of classic many-body calculations of spin diffusion such as that of Blume and Hubbard[13] for symmetric lattices. However, those calculations do not show the mind-teasing behavior of the asymmetric lattices which are the purpose of this work. Let us do a simplified calculation valid for both symmetric and asymmetric systems, which will clarify the effect of dimensional crossover in the dynamics. Consider a d-dimensional (hyper)cubic lattice of spins 1/2 interacting through the Hamiltonian

$$\mathcal{H}_{II}=\sum_{k,j>k} J_{jk}\left[\alpha 2S_j^z S_k^z-\frac{1}{2}(S_j^+ S_k^- + S_j^- S_k^+)\right], \qquad (1)$$

where S are the usual spin operators with subscripts indicating spin sites and $J_{jk}=J_\mu$ are nearest-neighbor interaction parameters depending only upon the direction $\hat{\mu}$ along sites j and k at distance $a_\mu \equiv a=1$. For $\alpha=-\frac{1}{2}$, it describes the Heisenberg model (isotropic exchange), $\alpha=0$ defines the XY model, while $\alpha=1$ is a truncated dipolar Hamiltonian.

For each initial state $|i\rangle$, with the zeroth site polarized (i.e., one of the states shown in Fig. 1), the probability of finding the same site polarized in the state $\langle f|$ after a time t, is

$$P_{f,i}(t)=|\langle f|\exp[-(i/\hbar)\mathcal{H}_{II}t]|i\rangle|^2. \qquad (2)$$

A total ensemble averaged probability that a spin initially up at the zeroth position is still up at time t can be calculated summing over all the N_i and N_f possible initial and final states:

$$\langle P(t)\rangle=\sum_f^{N_f}\sum_i^{N_i}\frac{1}{N_i}P_{f,i}(t)$$
$$=1-\frac{1}{4}\sum_\mu^d \langle Z_\mu\rangle J_\mu^2 t^2/\hbar^2+\mathcal{O}(t^4)+\cdots. \qquad (3)$$

$\langle Z_\mu\rangle=1$ is the average number of neighbors along direction μ with down spin. In the second order term only the flip-flop terms produce the exchange of the originally polarized zeroth spin with its neighbors. This corresponds to the one-body dynamics of one up spin in a lattice in which the other up spins remain frozen. Higher order terms contain the dynamics of those other spins and the many-body interactions.

A normalized magnetization can be calculated from the spin autocorrelation function as

$$M(t)=\langle S_0^z(t)S_0^z\rangle/\langle S_0^z S_0^z\rangle=2(\langle P(t)\rangle-\tfrac{1}{2}). \qquad (4)$$

This magnitude is experimentally accessible. For short times, the mean square displacement of the magnetization in terms of the nearest-neighbor spin correlation functions $\langle r_\mu^2\rangle \propto a_\mu^2\langle S^z_{0\pm a_\mu}(t)S_0^z\rangle$. The truncated quantum dynamics given by Eq. (3) gives

$$\langle r_\mu^2\rangle_{\text{quant}} = \tfrac{1}{2}\, a_\mu^2 J_\mu^2 t^2/\hbar^2. \tag{5}$$

However, the perturbative expansion of Eq. (3) is not a practical way to obtain the long time dynamics but for few simple one-dimensional systems.[1] Therefore, we need an entirely different approach. First, we notice that for infinite lattices the long time evolution is very complex and it presents the *apparently* diffusive behavior (eventually stabilized by interactions with the thermal bath) that we want to evaluate. While evolution with Eq. (1) is not an irreversible process, it is close enough to a diffusive evolution so that a self-consistent condition is already achieved for intermediate times.

When $\langle P(t)\rangle$ has decayed substantially, let us say when the second term in Eq. (3) is half of the first, the environment has changed completely and no phase coherence with the initial one-body state is retained. This defines the dephasing time

$$\tau_\phi = \hbar 2/\sqrt{2\sum_\mu^d J_\mu^2} = 1/\omega_\phi. \tag{6}$$

It is important to note that this functional dependence on J_μ does not depend on the amount of decay chosen to determine τ_ϕ. The change in the environment, following a quantum dynamics according to Eq. (3), is slower than linear at early times, becoming important only at about τ_ϕ. Then, it can be described by a discrete time Markovian process leading to a classical random walk. At the dephasing time the mean square displacement of the magnetization is

$$\langle r_\mu^2\rangle_{\text{class}} = 2D_\mu \tau_\phi. \tag{7}$$

At $t=\tau_\phi$, both quantum and classical diffusive regimes must coincide. This statement was rigorously proved in Ref. 9, where we used the Keldysh formalism[14] to achieve a nonperturbative description of the crossover from the quantum to the diffusive regime for a particle interacting with a dephasing field. In our model this field acts at a typical time with an interaction probability $p(t)dt=\delta(t-\tau_\phi)dt$. Then, the coherence with the initial state has a survival probability $\theta(\tau_\phi - t)$. This step function is more appropriate to describe the quantum dynamics of the dephasing field than the usual $\exp[-t/\tau_\phi]$. According to Ref. 9, within this approximation the self-consistent propagation of density excitations (satisfying the integral Keldysh equation) requires that both quantum and Markovian descriptions[12] give the same probability distribution at $t=\tau_\phi$. This condition is equivalent to equate Eqs. (5) and (7), from which we obtain the diffusion constant for each direction:

$$D_\mu = \frac{a_\mu}{2\hbar}\times J_\mu^2/\sqrt{2\sum_\mu^d J_\mu^2} = \frac{a_\mu^2}{4\hbar^2} J_\mu^2/\omega_\phi. \tag{8}$$

This important result contains the paradoxical aspects of spin dynamics we discussed in the introductory paragraphs. While our procedure has been mainly qualitative, we believe it catches the fundamental phenomena, and therefore the correct functional dependence. For the symmetric three-dimensional lattice this gives $D=Ja^2/(\hbar 2\sqrt{6})$, in fair agreement with the values calculated by a number of previous authors[13,15] and consistent with simulations[13] in a system of classical spins.

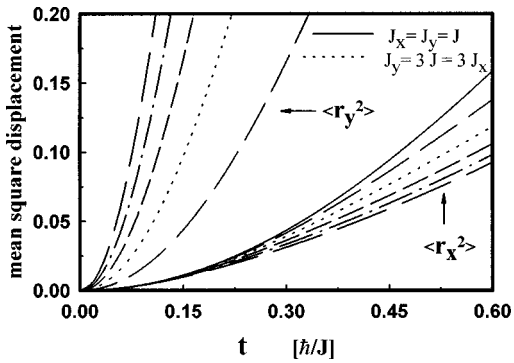

FIG. 2. Mean square displacement of magnetization as function of time for increasing asymmetrization of the 2D lattice with Heisenberg interaction. The full line is $J_y=J_x=J$, the other lines are the sequences $J_y/J=2$, 3, 4, 5, and 6. J_x is kept constant. Upper curves are displacements along y, while lower ones are along x. The QZE is manifested in the slowdown of the spreading along x.

While the numerical solution of the quantum dynamics of systems with a large number of spins is a formidable task, we can attempt to see signatures of the discussed phenomena for small two-dimensional systems. We consider a nine-spin system with periodic boundary conditions and evaluate the eigenstates of all the spin configurations to study the dynamics according to Eqs. (1) and (2) with $\alpha=-1/2$. In Fig. 2 we show the numerical evaluation of $\langle r_x^2\rangle$ and $\langle r_y^2\rangle$ as a function of time t for different values of J_y while keeping $J_x=1$. While the diffusive regime ($\langle r_y^2\rangle \sim 2D_y t$) cannot be reached in a small system, the plot shows that an increase in J_y is correlated with an increase in the spreading dynamics. The paradox that is manifested in the QZE is that the growth of $\langle r_x^2\rangle$, showing the dynamics in the perpendicular direction, is slowed down by the increase of J_y. While eventually the hydrodynamic limit could be described by a diffusion equation in which variables can be separated, in the many-body Schrödinger equation variables appear intimately entangled, leading to the interdependence of the diffusion constants of Eq. (8).

We want to show that even when an asymmetry of the lattice could lead to a faster quantum decay of the polarization for short times, it produces a reduction of the effective dimensionality of the lattice where diffusion occurs which slows down the spreading. For this purpose we study the time decay of the local polarization in a square lattice. The inset in Fig. 3 shows the magnetization $M(t)$ in the symmetric lattice up to intermediate times [for long times weak mesoscopic beats[4] resembling those of one-dimensional (1D) rings would appear]. The symmetry of the interaction network can be broken by slightly increasing the coupling along direction y, $J_y=J+\delta J$, while in the other direction it is decreased by the same amount $J_x=J-\delta J$. The thick line in the main plot shows the difference between the local magnetizations calculated for the asymmetric and the symmetric networks $\delta C=M^A(t)-M^S(t)$, with $\delta J/J=0.1$. For very short times the spreading in the asymmetric lattice is faster than in the symmetric one and δC follows the parabolic approximation $\delta C \approx -(\delta J)^2 t^2/\hbar^2$ shown by the thin line. QZE manifests for intermediate times, slowing down the diffusion in the asymmetric case. This compensates the fast decay of short times at around $t=1.8\hbar/J$, when $\delta C=0$. This crossover from faster decay of M^A as compared with M^S to a

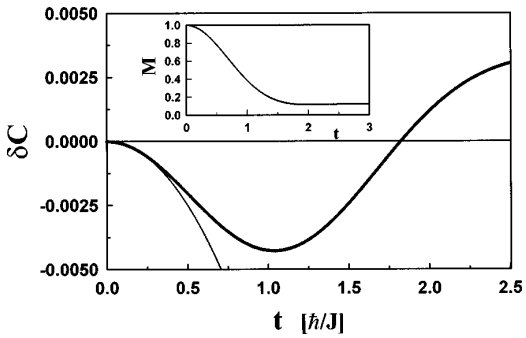

FIG. 3. The inset shows the spin autocorrelation function in a symmetric 2D lattice with Heisenberg interaction as function of time. Thick curve in the main frame shows the difference δC between the autocorrelation function of the symmetric ($J_y = J_x = J$) and asymmetric ($J_y = J + \delta J$ and $J_x = J - \delta J$, with $\delta J/J = 0.1$) lattices. The thin line is a parabolic approximation.

slower one is a very remarkable result of our simple theory consistent with the numerical solutions.

A simple experimental test of the spin dynamics is the linewidth of a magnetic resonance spectrum. Different local environments in a system of noninteracting spins produce an inhomogeneously broadened absorption line, where each frequency corresponds to spins seeing a different local field. However, through the flip-flop mechanism, each spin excitation explores different lattice sites producing an averaging of the field. Thus, the narrowed linewidth[16] is $\Delta\nu \propto \int_0^\infty \langle S_0^z(t) S_0^z \rangle dt$ (eventually the integral extends up to a cutoff time). Then, the slower the dynamics the wider the line.

It is not usually possible to control the magnitude of the exchange interaction for electronic spins. However, this is just the situation observed[17] for a family of Cu$(aa)_2$ single crystals where aa stands for amino acid. If the aa is present in a mixture 50% dextrogyre and 50% levogyre (D,L), the Cu^{2+} paramagnetic centers occupy sites with inversion symmetry in a two-dimensional coupling network. The Cu atoms are connected by OCO bridges, with the different aa residues playing the role of separators between layers. In Cu$(L\text{-}aa)_2$ crystals (100% L-aa), however, a breakdown of this symmetry occurs. The amount of asymmetrization of the lattice depends on the aa. The very important increase of the electron paramagnetic resonance linewidth that is observed when going from D,L to L crystals can be interpreted as a signature of the discussed QZE.

Systems with interacting nuclear spins seem quite promising for the study of dynamics in asymmetric lattices. Macromolecules could be engineered to present a sequence of through bond isotropic couplings when studied in solution. In crystals, where the dipolar interaction is the dominant one, its dependence on the angle θ between the vector connecting dipoles and dipole orientation (fixed by an external field) can be used to change its magnitude and sign. For example, in a cubic lattice of spins, by varying the magnetic field in the plane [001] from the [110] direction toward one at a magic angle ($\theta_m = \arccos[1/\sqrt{3}]$) with the [010] axis, one could see how the dynamics changes from a three-dimensional behavior ($J_x = -d/2$, $J_y = -d/2$, $J_z = d$), with a symmetric diffusion in the xy plane, that of a two-dimensional system ($J_x = -d$, $J_y = 0$, $J_z = d$). The change from the initial orientation would allow a study of a crossover in δC similar to that discussed above. The application of these concepts would be even more direct for magnetically two-dimensional systems ($J_z \approx 0$), where J_x and J_y can be controlled independently. In both cases multiple quantum coherence[18] experiments and spin diffusion pulse sequences[7] could be used to obtain complementary information[19] about the asymmetric spreading of magnetic polarization. A more conventional experiment is to apply a magnetic field gradient along one crystal direction (say y) which, by detuning the resonance frequencies of nuclei at different xz planes, is equivalent to a decrease of the interplane coupling. Results obtained using this technique[20] are also consistent with our predictions. In summary, we have put the problem of spin diffusion under a perspective that could stimulate a series of experiments and calculations to understand spin dynamics.

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