Spin relaxation and decoherence of two-level systems

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(Dated: February 2, 2008)

We revisit the concepts of spin relaxation and spin decoherence of two level (spin-1/2) systems. From two toy-models, we clarify two issues related to the spin relaxation and decoherence: 1) For an ensemble of two-level particles each subjected to a different environmental field, there exists an ensemble relaxation time $T_1^*$ which is fundamentally different from $T_1$. When the off-diagonal coupling of each particle is in a single mode with the same frequency but a random coupling strength, we show that $T_1^*$ is finite while the spin relaxation time of a single spin $T_1$ and the usual ensemble decoherence time $T_2^*$ are infinite. 2) For a two-level particle under only a random diagonal coupling, its relaxation time $T_1$ shall be infinite but its decoherence time $T_2$ is finite.

PACS numbers: 73.23.-b, 72.70.+m, 71.70.Ej

Spin relaxation and decoherence is of central importance in quantum computation and spintronics\cite{1, 2, 3}. Two phenomenological quantities, $T_1$ and $T_2$ known as the spin relaxation time and spin decoherence time for a two-level (spin-1/2) system, were introduced to describe the spin dynamics under the influence of an environment or external field in early theory\cite{4, 5}. As illustrated in Fig. 1(a), a spin in its upper level, denoted by $|+\rangle$, may jump to its lower energy level, denoted by $|-\rangle$, due to spin-environment coupling. $T_1$ is defined as the average transition time. A quantum spin can also be in a superposition state, say $(|+\rangle + |-\rangle)/\sqrt{2}$ initially. Without the environmental interaction, the system undergoes a constant precession with a frequency $\omega = E/\hbar$, where $E$ is the energy difference between $|+\rangle$ and $|-\rangle$. Under the environmental influence, the phases of the expansion coefficients in the superposition state will become random, and their relative phases, which describe the precession angle $\alpha$ in Fig. 1(b), will be completely undetermined (decoherence). The time for the superposition state to lose its coherence is called $T_2$. $T_1 = T_2$ is believed to be true when the spin is isotropic while $T_2 \leq 2T_1$ holds in general\cite{6, 7, 8}.

In recent years, it has been realized that another quantity, $T_2^*$, is important for a large number of experiments that measure the decoherence of an ensemble of spins rather than a single spin $T_2^*$. Illustrated in Fig. 1(c), in addition to the uncertainty in the precession angle of a single spin at a given time, there will be an uncertainty in the relative precession angles among different spins due to the spatial inhomogeneity. This additional decoherence source leads normally to $T_2^* \leq T_2$, within which $T_2^*$ may deviate notably from $T_2$. For example, $T_2^*$ of electron spins in quantum dots was shown to be several orders smaller than $T_2$\cite{9}.

![FIG. 1: (Color) An illustration of $T_1$, $T_2$ and $T_2^*$. (a) $T_1$ is the typical time for a spin jumps from the upper level (up spin) to the lower level (down spin). Inset: The polar angle $\beta$ and the azimuthal angle $\alpha$ of a spin along unit direction $\hat{n}$. (b) The precession of spin in xy-plane. At $t = 0$, the spin points to x-direction. Due to its irregular precession, the spin direction in a later time may be in a range of directions denoted by grey (blue) arrows. The spin loses its direction completely at $T_2$, called the decoherence time. (c) For an ensemble of spins, all spins point to the x-direction at the beginning. As time goes on, different spins will precess to different directions due to the fluctuating field is different at different places. After $T_2^*$, spins point to all directions all the time.](attachment:fig1.png)

Our current understanding of $T_1$, $T_2$ and $T_2^*$ is more accurate and much deeper than that of the early years\cite{10}. Its applications to various systems, especially those of nanostructures are of current focus. However, there are still many vague conceptions or even misconceptions about the description, origins and mechanisms...
of these quantities. In this letter, we revisit the concepts of the spin relaxation and decoherence of two-level systems. We shall argue that, besides an ensemble decoherence time $T_2^*$, an ensemble relaxation time $T_1^*$ is also necessary to interpret experimental results. We show, through a toy-model calculation, that $T_1^*$ distinguishes itself from $T_1$. In the case of our study, the total polarization of the system decreases with time while each spin undergoes a well-defined periodic oscillation. Thus, there is a finite pseudorelaxation time $T_1^*$ for the whole system, but an infinite $T_1$. We investigate also the effects of a random diagonal coupling on spin relaxation and decoherence. We show that both slow and fast fluctuation are harmful to $T_2$ while its effect on $T_1$ must go through the off-diagonal coupling.

**Formulation of the dynamics of a two-level system.** To mimic the motion of a two-level (spin-1/2) system coupled to an environment, we assume that the system may jump from one state into the other through the coupling, or the system in a superposition state may obtain random phases to its expansion coefficients. The Hamiltonian of the system can be properly described by a $2 \times 2$ matrix,

$$
H = \frac{E}{2} \sigma_z + K(t) \cdot \sigma = \begin{bmatrix} \frac{E}{2} + K_z & K_x - iK_y \\ K_x + iK_y & -\frac{E}{2} - K_z \end{bmatrix}.
$$

(1)

$$
\sigma = \sigma_x \hat{x} + \sigma_y \hat{y} + \sigma_z \hat{z}
$$

is the Pauli operator. The environment-system interaction is approximated by a randomly fluctuating field $K(t) = K_x \hat{x} + K_y \hat{y} + K_z \hat{z}$, which could originate from the interactions of the spin with phonons, photons or with other surrounding spins. In general, the field is correlated both in time and in space. Its covariance measures the strength of the coupling which should also depend on the temperature besides of other factors. How to obtain the field from a microscopic Hamiltonian of both system and the environment is an interesting problem in both physics and chemistry. For example, it has been shown that electron-phonon, spin-orbit, and/or hyperfine interactions for electron spins in a quantum dot are equivalent to off-diagonal couplings. We will see that the diagonal coupling $K_z(t)$ and the off-diagonal coupling $K_x(\gamma)(t)$ play different roles in determining $T_1$ and $T_2$ respectively. Without losing the generality, the energies of the upper and the lower levels (|±⟩) are assumed to be ±$E/2$, respectively, in the absence of the fluctuating field. The energy may be due to the Zeeman interaction for a spin in a constant magnetic field.

Express the general wave function of our two-level system as $\psi(t) = a(t)|+⟩ + b(t)|-⟩$, the equations for the expansion coefficients $a(t)$ and $b(t)$, derived from the time-dependent Schrödinger equation, are (set $\hbar = 1$)

$$
\begin{align*}
\frac{da}{dt} &= \left[\frac{E}{2} + K_z\right]a + [K_x - iK_y]b, \\
\frac{db}{dt} &= -\left[\frac{E}{2} + K_z\right]b + [K_x + iK_y]a.
\end{align*}
$$

(2)

The issues are to find out the spin relaxation and decoherence time for a given fluctuating field. Spin-relaxation time $T_1$ can be obtained by examining how $\langle |a(t)|^2 \rangle$ changes with time under an initial condition, where $\langle \cdots \rangle$ denotes the average over the system state. $T_1$ is the characteristic decay time extracted from $\langle |a(t)|^2 \rangle$.

To evaluate $T_2$ or $T_2^*$, it is useful to notice that a spin state along a particular direction, specified by the polar and azimuthal angles $\beta$ and $\alpha$ [inset of Fig. 1(a)], can also be written as $\psi(t) = \exp(\alpha/\omega)\cos(\beta/2)\exp(\alpha/2)|+⟩ + \sin(\beta/2)\exp(-\alpha/2)|-⟩$. If one can find $\alpha(t)$ by solving Eq. (2), then $T_2$ for a single spin or $T_2^*$ for an ensemble of spins corresponds to the time at which the deviation of $\alpha$ equals $\pi$, i.e., $\langle \Delta \alpha^2(T_2) \rangle = \langle \alpha(T_2)^2 \rangle - \langle \alpha(T_2) \rangle^2 = \pi^2$.

**Ensemble spin relaxation time $T_1^*$**. In order to demonstrate the necessity of introducing an ensemble spin relaxation time $T_1^*$, we consider an ensemble of noninteracting spin-1/2 particles each of which is described by a Hamiltonian similar to $H$ with $K_x = 0$ and $K_x + iK_y = Ke^{i\omega t}$. Then Eq. (2) can be solved analytically with solution

$$
\begin{align*}
& a_k(t) = e^{-i(\omega/2)t}\left[|a_k(0)\cos(\Omega t)\right] + i\Delta \omega a_k(0) - 2K_k b_k(0) \sin(\Omega t)], \\
& b_k(t) = e^{i(\omega/2)t}\left[|b_k(0)\cos(\Omega t)\right] - i\Delta \omega b_k(0) + 2K_k a_k(0) \sin(\Omega t)],
\end{align*}
$$

(3)

where $\Delta \omega = \omega - E$ and $4\Omega^2 = 4K_k^2 + \delta \omega^2$.

To make following analysis simple, we shall first assume $\omega = E$ for all spins, and $K = K_k = 0$ a random real constant for $k$th spin. Thus, each spin is nothing but the famous Rabi problem at resonance. For the $k$th spin, Eq. (3) gives $a_k(t) = e^{-i(\Omega/2)t}\left[|a_k(0)\cos(K_k t) - b_k(0)\sin(K_k t)\right]$ and $b_k(t) = e^{i(\Omega/2)t}\left[|b_k(0)\cos(K_k t) - a_k(0)\sin(K_k t)\right]$. If one measures the expectation value of $S_k$, the $k$th spin defined as $(S_{k, z}) = \langle |a_k(t)|^2 \rangle - |b_k(t)|^2$ (his set to 1), one finds $(S_{k, z}) = ||a_k(0)||^2 - |b_k(0)|^2||/2 \cos(2K_k t) + 3|a_k(0)|^2b_k(0)\sin(2K_k t)$, which oscillates periodically in time instead of decay. One then should conclude $T_1 = \infty$ as it should be. However, if we measure $S_{k, z}$ of the whole system, we need to include contributions from all spins. Since different spins have different $K$ values, they will oscillate with different periods, and the spins initially in phase will be out of phase during the evolution, as is illustrated in Fig. 2. Thus, even all spins are fully polarized along $z$ direction initially, $S_{k, z}$ will decrease with time. If $K$ has a distribution function of $\mathcal{P}(K) = (2K/\sigma)e^{-K^2/\sigma}$, where $\sigma$ measures the width of the distribution, by using Eq. (3), we have $\langle (S_k) \rangle = (1/N)\sum(S_{k, z}) = 1 - \sqrt{\pi}e^{-\alpha^2}erfi(\sqrt{\pi}t)$, where $erfi(x)$ is the imaginary error function, and $[\cdots]$ is an average over all spins. This time dependence is plotted in the inset of Fig. 2. Thus, one finds the polarization of the system relaxes after a characteristic time
$T_1^* = 1/\sqrt{\sigma}$, although there is no relaxation for individual spin. It is also interesting to evaluate $T_2$ and $T_2'$ of this toy model. Set $a_k(0) = b_k(0) = 1/\sqrt{2}$ for all $k$ and compare $a_k(t)$ and $b_k(t)$ with their expressions in terms of the polar and azimuthal angles mentioned in the early paragraphs, we find $a_k(t) = Et$. All spins precess at the same rate and in phase. Thus, both $T_2$ and $T_2'$ are infinite. This is a peculiar case in which each spin follows a deterministic motion, and there is no relaxation and no decoherence. But if one measures the polarization of the whole system, the system appear to have a relaxation time $T_1^*$ that is fundamentally different from $T_1$.

In fact, one may also assume that $k$th spin has $\omega_k$ and different level spacing $E_k$. Since $E_k$, $\omega_k$, and $K_k$ are constants for a given spin, $a_k(t)$ and $b_k(t)$ are well-defined periodic functions according to Eq. (3). Thus its precession angle and expectation value of $S_{k,z}$, which can be obtained after some algebra

$$\langle S_{k,z}(t) \rangle = \frac{|a_k(0)|^2 - |b_k(0)|^2}{2\Omega^2} \{\Delta\omega_k^2/4 + K_k^2 \cos(2\Omega t)\},$$

which oscillates periodically in time. Therefore, both $T_1$ and $T_2$ are infinite. However, if one measures $S_z$ or $S_x - iS_y$ (which is a measure of precession) of the whole system, we need to sum over all spins. This corresponds to a sum of many periodic functions with different periods. Similar to the case of random $K_k$ discussed early, it leads to a decay function, and $T_1^*$ and $T_2'$ are finite because different spins precess with different speeds in this case.

![FIG. 2: The time dependence of occupation probability of different spin in its upper level. All spins are initially in their upper levels. Due to the random coupling strength, their Rabi frequencies are also random. Inset: The time dependence of $\langle |S_k| \rangle$.](image)

Away from the resonance $\omega \neq E$, a spin initially at the upper level will not evacuate completely from the level. The occupation probability for a spin in the upper level is given by the well-known Rabi solution

$$|a(t)|^2 = 1 - A(\omega) \sin^2(\Omega t),$$

where $4\Omega^2 = 4K^2 + (\omega - E)^2$, and $A(\omega) = 4K^2/[4K^2 + (\omega - E)^2]$. Amplitude $A$ at the resonance takes the maximal value 1. $A$ decreases fast when $\omega$ deviates from $E$. Thus, the mode of the fluctuating field near the resonance is expect to relax the spin more effectively. We shall see that the diagonal coupling behaves differently.

**Effects of diagonal coupling on $T_1$ and $T_2$.** We would like to discuss how a diagonal coupling affects the spin decoherence and the spin relaxation. Consider a case in which $K_z$ in Eq. (1) is a randomly fluctuating field and $K_z = K_y = 0$, this is an exactly solvable problem because there is no coupling between different levels, and $K_z$ causes only the energy levels to move up and down randomly. The consequence is the accumulation of a dynamical random phase for each level. The solutions for the expansion coefficients are $a(t) = a(0) \exp[-i \int_0^t (E/2 + K_z(t')) dt']$ and $b(t) = b(0) \exp[i \int_0^t (E/2 + K_z(t')) dt']$, where $a$ and $b$ are separated because of the absence of off-diagonal matrix elements. Thus, a spin in either upper or lower level will not relax, and $T_1$ is infinite. However, for a spin in superposition state, each of the two expansion coefficients will obtain a random phase due to random field $K_z$ leading to a random precession angle $\alpha(t) = Et + 2 \int_0^t K_z(t') dt'$. Thus, the expectation of the deviation of the precession angle is

$$\langle \Delta \alpha^2(t) \rangle = 4 \int_0^t dt_1 dt_2 \langle K_z(t_1) K_z(t_2) \rangle.$$

For a stochastic field with autocorrelation,

$$\langle K_z(t_1) K_z(t_2) \rangle = \frac{\Delta}{2\tau_c} \exp\left(-\frac{|t_1 - t_2|}{\tau_c}\right),$$

(6a)

where $\tau_c$ and $\Delta$ are the correlation time and strength, we have

$$\langle \Delta \alpha^2(t) \rangle = 4\Delta [t + \tau_c(e^{-\frac{t}{\tau_c}} - 1)].$$

(6b)

Thus, the spin-decoherence time $T_2$ is given by

$$T_2 + \tau_c[\exp(-T_2/\tau_c) - 1] = \pi^2/(4\Delta).$$

(7)

In the limit of $\tau_c \to 0$, the fluctuating field is a white noise $\langle K_z(t_1) K_z(t_2) \rangle = \Delta \delta(t_1 - t_2)$, and the decoherence time is simply $T_2 = \pi^2/(4\Delta)$, inversely proportional to the correlation strength of fluctuating field and independent from the level spacing. Therefore, if the amplitude of the fluctuating field increases $n$ times, $T_2$ decreases by $n^2$ times.

Solution (7) exhibits three regimes. In the white noise regime where $\tau_c \Delta \ll 1$, the fluctuating field behaves like an independent force on the spin precession. The spin precession angle undergoes a random walk around its mean rate $d\alpha/dt = E$, and $T_2 \approx \frac{\pi^2}{4\Delta}$ insensitive to $\tau_c$. In the long correlation regime where the spin experience a random persistent force, the deviation of the precession angle increases quadratically with time,
and the motion of the spin behaves like a biased random walk. Correspondingly, decoherence time $T_2$ is $T_2 \approx \pi \sqrt{\tau_c/(2\Delta)} \ll \tau_c$. In the intermediate regime of $\tau_c \Delta \sim 1$, the $T_2, \tau_c$ and $\Delta^{-1}$ are in the same order. Unlike the off-diagonal coupling that affects the spin relaxation most when it is at the resonance, all fluctuations of the diagonal coupling contribute to the deviation of the precession angle as it is shown in Eq. (6). Thus, no matter whether there are fast (white noise) or slow (long correlation time) fluctuations, $K_2$ will lead to an effective decoherence. Of course, it does not mean that all fluctuations will play an equal role.

Our analysis so far does not rely on the sources of the fluctuating field. Thus, we expect our results to be applicable to all two level systems as long as they can be described by a Hamiltonian similar to (1). Although $T_1^*$ in our toy-model is more a definition in a thought experiment than a measure of a true spin relaxation, it makes the point that the relaxation time for a single spin and for an ensemble of spins could be totally different. We present an extreme case where $T_1^*$ is finite while $T_1, T_2$ and $T_2^*$ are infinite. In many realistic two-level systems, one may find another extreme where the distinction between $T_1$ and $T_1^*$ is not necessary\textsuperscript{8}, but the existence of realistic systems between the two extremes so that one has to use two relaxation time to separately describe the relaxation for a single spin and for an ensemble of spins, just as $T_2$ and $T_2^*$ for spin decoherence, cannot be ruled out\textsuperscript{3,8,9}.

It may be proper to explain the processes that contribute to $T_1 (T_1^*)$ and $T_2 (T_2^*)$. $T_1$ and $T_1^*$ are caused by the transitions of a system from one quasi energy level to another. Such a transition is usually irreversible, accompanying a complete change of the wave function. This microscopic view of $T_1$, $T_1^*$ is not in contradiction to the thermodynamic view that $T_1$ is the time of thermal equilibration for spin population\textsuperscript{2} because the time for a system to reach its thermal equilibrium normally requires many jumps between the upper and lower levels. Of course, the numbers in the two definitions may be different, but they should be in the same order. This is similar to the definition of the electron relaxation time scattered by impurities, which is the characteristic time for the electron distribution to return to its equilibrium one; on the other hand, it can be calculated from the Fermi golden rule at the quantum mechanical level. $T_2$ and $T_2^*$ come from the loss of coherence of a state which could be due to a transition for $T_1 (T_1^*)$, but it could also due to the accumulation of random phases to the components of a superposition state without a transition between two levels, as we showed in our toy-models. Thus, in general, $T_2$ and $T_2^*$ must be smaller than $T_1$ and $T_1^*$. In our extreme case, the $T_1^*$ is not caused by true irreversible transitions, but by coherent transitions (Rabi oscillations). In this sense, $T_1^*$ in our example is a pseudorelaxation time, and it should not be surprising to see our $T_1^*$ deviating from $T_2 < T_1^*$. There are misconceptions in the literatures about the difference of $T_2$ and $T_2^*$. It was claimed that $T_2^*$ is referred to as the reversible dephasing\textsuperscript{2}, while irreversible dephasing was attributed to $T_2$. This claim is questionable because the nature of randomness is irreversible. Thus both $T_2$ and $T_2^*$ are due to the irreversible loss of coherence. If the irreversibility in the claim is referred to the transition from one energy level to another, it is again not true because both $T_2$ and $T_2^*$ can be caused by the accumulation of random phases on the expansion coefficients, as revealed in our second toy-model.

Due to the different natures of $T_1$ and $T_2$, the diagonal and off-diagonal coupling play quite different roles on the spin relaxation and the spin decoherence. First, with only diagonal coupling, there is no spin relaxation. But there is a spin decoherence. However, a diagonal coupling can also influence the spin relaxation if an off-diagonal coupling is present. For an off-diagonal coupling, those fluctuation modes near the Rabi resonance affects spin relaxation more. Diagonal coupling could affect the relaxation either through the shifting of resonance point or through the modification of quantum interference by the phase change of wave function.

It may also be useful to comment on the relationships between $T_2$ and $T_2^*$. For a non-interacting spin system subject to the same fluctuating field, we should have $T_2^* < T_2$ because, in addition to the causes of spin decoherence on individual spin, the spatial inhomogeneity can contribute to decoherence of an ensemble of spins. The importance of the inhomogeneity on the decoherence was known in many fields and also in early work in semiconductor community\textsuperscript{3,12} in which the interference effect due to the inhomogeneity of k space or of real space can cause decay. For an interacting spin system, it is more complicated. An interaction can be an extra source of a fluctuating field. On the other hand, the interaction may glue different spins together. The bondings among spins could be so strong that all spins have to move coherently. In this case, one can expect that the dynamics of the system slows down. Also, a system with a single spin and a system with many spins are totally different, and there is no comparison. For example, the spin relaxation and spin decoherence of a Co atom is completely different from that of a bulk Co magnet where all spins are aligned in the same direction. And it is not surprising to have spin decoherence $T_2^*$ of the Co-magnet to be much longer than $T_2$ of one Co-atom. Another example may be superconductor caused by the effective e-e attraction. It is known that coherence time of electrons in a superconducting state could be much longer than that of an electron in a metal.

In conclusion, we demonstrate the necessity of introducing an ensemble spin relaxation time $T_1^*$ that could be orders of magnitude different from $T_1$. We have also shown that all fluctuations should be relevant to the spin
decoherence. There is no too-slow or too-fast fluctuation for $T_2$ and $T_2^*$. This work is supported by UGC, Hong Kong, through RGC CERG grants.

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