Optical dephasing by a random telegraph frequency modulation

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Using the random telegraph model, we investigate optical dephasing processes arising from Poisson stochastic modulation, we obtain the analytic expression for free-induced decay (FID) and photon-echo decay in the whole range of the stochastic parameters. In the small modulation range, FID or echo-intensity decay varies from single exponential to multiexponential with the increase of frequency modulation. In the large modulation range, FID is a damping oscillation, and the echo intensity decays exponentially with periodical fluctuations. When the modulation is very large the echo-intensity decay tends to go back to an exponential. The $R_1(-\frac{3}{2})$ echo-intensity decay in ruby at high magnetic fields is calculated based on the analytic expression; the result is compared with experiments and computer simulations. [S1050-2947(96)05609-0]

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I. INTRODUCTION

In many systems, a common source of dephasing, both for optical and magnetic resonance in solids, is magnetic fluctuations at the impurity site due to nuclear spin flipping in the host lattice and/or electron-spin flipping of the dopant ion [1,2]. For low magnetic fields, the photon echo has been observed to be exponential up to four orders in intensity [3]. It can be described by the phenomenological optical Bloch equation. However, more recent experiments showed that the photon echo displays a series of nonexponential decay for different magnetic fields or samples [4–6]. Many theories using various statistical models attempted to explain the experimental results [7–10]. All these theories predict that spin-flip-induced echo decay is expected to be nonexponential having a temporal form $I=I_0\exp[-(4t_{21}/T_m)^x]$, where t_{21} is the pulse separation and T_m is the phase storage time. The parameter x may take the values 0.5,1,2,3 depending on the stochastic models. In general, the experimental decay curves seem to take those forms with x changing from 1 to 2.6 [5,6]. It is not satisfactory that the previous theories cannot give a clear analytic expression in the continuous range of the stochastic parameter. Gaussian and Poisson processes are two important stochastic processes in practical systems. Gaussian modulation has been extensively studied [10,11]; using the central limit theorem, an analytic solution has been given. In the slow ($\Delta\tau_c \gg 1$, here Δ is the magnitude of frequency flip, τ_c the correlation time) or fast ($\Delta\tau_c \ll 1$) modulation region, x takes the values 3,1 respectively.

In this paper, we investigate the other important stochastic process, i.e., Poisson modulation. Using the simple random telegraph model we give the analytic expressions for free-induced decay and photon-echo decay in the continuous range of the stochastic parameters. The results are compared with previous theories, especially with those concerning Gaussian modulation. Furthermore, we calculate the $R_1(-\frac{3}{2})$ echo-intensity decay in ruby at high magnetic fields, and compare the result with experimental and computer simulated results.

II. FREE-INDUCED DECAY

It is well known that the decay due to frequency flip ($\delta\omega$) can be described by a relaxation function [12]. For free induced decay (FID), the relaxation function $\langle\exp(\phi_1)\rangle$ is equal to the field average over random frequency flip perturbations, and can be expressed as

$$\langle E(t)\rangle=\langle e^{i\int_0^t\delta\omega(t)dt}\rangle=\left\langle \sum_{n=0}^{\infty}\frac{(i)^n}{n!}\left(\int_0^t\delta\omega(t)dt\right)^n\right\rangle$$
$$=\sum_{n=0}^{\infty}\frac{(i)^n}{n!}\int_0^t\int_0^{t_n}\cdots\int_0^{t_2}$$
$$\times\langle\delta\omega(t_1)\delta\omega(t_2)\cdots\delta\omega(t_n)\rangle dt_n dt_{n-1}\cdots dt_1. \tag{2.1}$$

We consider $\delta\omega(t)$ flips between two possible frequency values Δ and $-\Delta$, the probabilities of taking Δ or $-\Delta$ are both $\frac{1}{2}$. The distribution of time intervals between two adjacent flips is given by $p(t)=W\exp(-Wt)$, and k, the number of flips within T satisfies the Poisson distribution $p_k=\exp(-WT)(WT)^k/k!$. This model describes the bivalued random telegraph process.

For this model, we have the expectation value $\langle\delta\omega(t)\rangle=0$, and the autocorrelation $\langle\delta\omega(t)\delta\omega(t')\rangle=\Delta^2\exp(-2W|t-t'|)$. It is derived as follows: if there is an even number of flips within $|t-t'|$ then $\delta\omega(t)\delta\omega(t')=\Delta^2$; if there is an odd number of flips then $\delta\omega(t)\delta\omega(t')=-\Delta^2$. Hence

$$\langle\delta\omega(t)\delta\omega(t')\rangle=\sum_{k=0}^{\infty}\Delta^2[p_{2k}(|t-t'|)-p_{2k+1}(|t-t'|)]$$
$$=\Delta^2e^{-2W|t-t'|}. \tag{2.2}$$

By a similar consideration, we have

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$$\langle \delta\omega(t_1)\delta\omega(t_2)\cdots\delta\omega(t_{n-1})\delta\omega(t_n)\rangle = \begin{cases} 0 \quad \text{if } n=\text{odd} \\ \Delta^n e^{-2W[(t_n-t_{n-1})+\cdots+(t_2-t_1)]} & \text{if } n=\text{even}, \quad \text{and } t_n \geqslant t_{n-1} \geqslant \cdots \geqslant t_2 \geqslant t_1. \end{cases} \tag{2.3}$$

Substituting Eq. (2.3) into Eq. (2.1), and for shortening using $2W$ as frequency unit and $1/2W$ as time unit, we have

$$f(t)=\langle E(t)\rangle = \sum_{n=0}^{\infty} \frac{(-1)^{2n}}{(2n)!} \int_0^t \int_0^{t_{2n}} \cdots \int_0^{t_2} \langle \delta\omega(t_1)\delta\omega(t_2)\cdots\delta\omega(t_{2n})\rangle dt_1 dt_2 \cdots dt_{2n}$$
$$= \sum_{n=0}^{\infty} (-1)^n \Delta^{2n} \int_0^t dt_{2n} \int_0^{t_{2n}} dt_{2n-1} e^{-(t_{2n}-t_{2n-1})} \cdots \int_0^{t_3} dt_2 \int_0^{t_2} dt_1 e^{-(t_2-t_1)}. \tag{2.4}$$

The integral in the first step is performed in the space $t \geqslant t_k \geqslant 0$ $(k=1,2,\ldots,2n)$, the space is divided into $(2n)!$ subspaces, and the $2n$ variables are rearranged such that $t_{2n} \geqslant t_{2n-1} \geqslant \cdots \geqslant t_2 \geqslant t_1$. Integrals in every subspace are equal. We define operators I and I' as follows:

$$I = \int_0^t dt\ e^{-t} \cdots, \quad I' = \int_0^t dt\ e^{t} \cdots, \tag{2.5}$$

and abbreviate Eq. (2.4) as

$$f(t) = \sum_{n=0}^{\infty} (-1)^n \Delta^{2n} [II']^n(1). \tag{2.6}$$

Then we calculate the first and second derivatives of $\langle f(t)\rangle$:

$$\frac{df(t)}{dt} = \sum_{n=1}^{\infty} (-1)^n \Delta^{2n} e^{-t} I' [II']^{n-1}(1)$$
$$= -\Delta^2 \sum_{n=0}^{\infty} (-1)^n \Delta^{2n} e^{-t} I' [II']^n(1),$$

$$\frac{d^2 f(t)}{dt^2} = -\sum_{n=1}^{\infty} (-1)^n \Delta^{2n} e^{-t} I' [II']^{n-1}(1) + \sum_{n=1}^{\infty} (-1)^n \Delta^{2n} [II']^{n-1}(1)$$
$$= \Delta^2 \sum_{n=0}^{\infty} (-1)^n \Delta^{2n} e^{-t} I' [II']^n(1) - \Delta^2 \times \sum_{n=0}^{\infty} (-1)^n \Delta^{2n} [II']^n(1). \tag{2.7}$$

Clearly, $f(t)$ satisfies the differential equation:

$$\Delta^2 f(t) + \frac{df(t)}{dt} + \frac{d^2 f(t)}{dt^2} = 0, \qquad f(0)=1, \quad \left.\frac{d}{dt} f(t)\right|_{t=0} = 0. \tag{2.8}$$

We define the dimensionless parameter Δ/W as p(modulation), with $2W$ as frequency unit $p=2\Delta$, and the solution of this equation is

$$f(t) = \begin{cases} \frac{1}{2}\left(1+\frac{1}{\sqrt{1-p^2}}\right) e^{-(t/2)(1-\sqrt{1-p^2})} - \frac{1}{2}\left(\frac{1}{\sqrt{1-p^2}}-1\right) e^{-(t/2)(1+\sqrt{1-p^2})}, & p<1 \\ \left(1+\frac{t}{2}\right) e^{-t/2}, & p=1 \\ e^{-t/2} \frac{p}{\sqrt{p^2-1}} \cos\left[\frac{t}{2}\sqrt{p^2-1} - \sin^{-1}\left(\frac{1}{p}\right)\right] & p>1. \end{cases} \tag{2.9}$$

This solution has been obtained by Wodkiewicz *et al.* in their work on noise in strong laser-atom interaction using different methods [13]. Some examples are shown in Fig. 1, in a semilogarithmic plot. One can find that in the small modulation region ($p \leqslant 0.2$), FID is a single-exponential decay process. When the modulation is increased, it becomes a multiexponential one. For $p>1$, the decay is an oscillation enveloped by an exponential. It is convenient comparing our results with Gaussian modulation from a spectral point of view. The Fourier transform of the relaxation function $\langle \exp(\phi_1)\rangle$ gives the single-site absorption spectrum. For Gaussian modulation, the spectrum varies from Lorentzian to Gaussian as the magnitude of the frequency modulation p increases [10,11]. For Poisson modulation (our results), one can find that the absorption spectrum corresponds to two Lorentzian line shapes in the complete range of the frequency modulation [see Eq. (2.9)]. When $p<1$, the two Lorentzian lines have the same central frequency, but differ

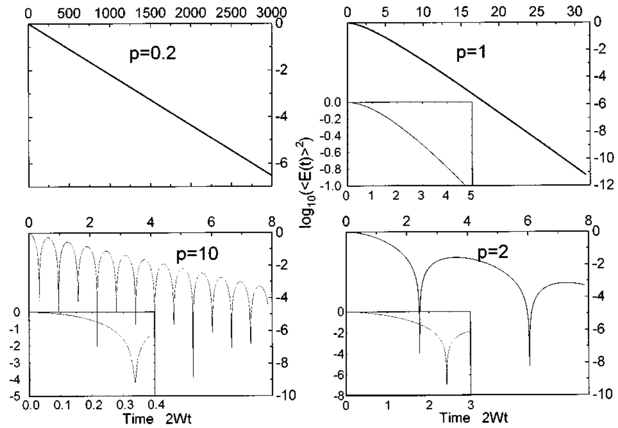

FIG. 1. Free-induced decay curves for different values of the frequency modulation p.

in their linewidth and magnitude. For very small p, one of them is dominated. So the spectrum is a Lorentzian line, like that predicted by Gaussian modulation. In the range of $p>1$, two lines with the same linewidth and central frequencies are separated by a value of $(p^2-1)^{1/2}$. In a practical system, all spin flips with various values of $\delta\omega$ can induce the dephasing; they all contribute to the absorption spectrum. Therefore the practical line is an overlap of all the two separated lines; it is no longer separate when the number of the spin flips is large, so FID will have no oscillating form.

III. PHOTON ECHO

For a two-pulse photon-echo process, the relaxation function that describes the echo decay arising from the stochastic frequency flip can be expressed as

$$\langle \exp(\phi_2)\rangle = \left\langle \exp\left[i\int_0^{t_{21}} \delta\omega(t)dt - i\int_{t_{21}}^{2t_{21}} \delta\omega(t')dt' \right]\right\rangle. \tag{3.1}$$

For the extremely broad inhomogeneous spectrum, the echo field $\langle E(t_{21})\rangle$is equal to $\langle \exp(\phi_2)\rangle$. Taking a Taylor expansion of $\langle E(t_{21})\rangle$, using the same consideration of the integration range of t and t' as in Sec. II and noting that t' is always greater than or equal to t, we have

$$\begin{aligned}\langle E(t_{21})\rangle &= \left\langle \sum_{n=0}^{\infty} \frac{(i)^n}{n!}\left[\int_0^{t_{21}} \delta\omega(t)dt - \int_{t_{21}}^{2t_{21}} \delta\omega(t')dt' \right]^n \right\rangle \\ &= \sum_{n=0}^{\infty}\sum_{m=0}^{\infty} (-1)^{n+m}\Delta^{2(n+m)}[II']^n(1)[II']^m(1) \\ &\quad - \sum_{j=0}^{\infty}\sum_{k=0}^{\infty} (-1)^{j+k+1}\Delta^{2(j+k+1)}e^{-t_{21}}I'[II']^j(1)[II']^k I(1) \\ &= f^2 + \sum_{j=0}^{\infty}\sum_{k=0}^{\infty} (-1)^{j+k}\Delta^{2(j+k+1)}e^{-2t_{21}}I'[II']^j(1)I'[II']^k(1) = f^2 + \frac{1}{\Delta^2}\left(\frac{df}{dt_{21}}\right)^2 .\end{aligned} \tag{3.2}$$

Substituting (2.9) into (3.2), we have

$$\langle E(t_{21})\rangle = \begin{cases} \dfrac{1}{1-p^2}\left[\tfrac{1}{2}(1+\sqrt{1-p^2})e^{-t_{21}(1-\sqrt{1-p^2})} + \tfrac{1}{2}(1-\sqrt{1-p^2})e^{-t_{21}(1+\sqrt{1-p^2})} - p^2 e^{-t_{21}}\right], & p<1 \\ \left(1+t_{21}+\dfrac{t_{21}^2}{2}\right)e^{-t_{21}}, & p=1 \\ \dfrac{p^2}{p^2-1}\, e^{-t_{21}}\left[1+\dfrac{1}{p}\sin\left(t_{21}\sqrt{p^2-1}-\sin^{-1}\dfrac{1}{p}\right)\right], & p>1. \end{cases} \tag{3.3}$$

Echo intensities versus t_{21} for various values of p are given in Fig. 2. When $p<1$, the echo decay contains several different exponential decay terms. In the small modulation case ($p<0.2$), only one of these terms is dominant, so the decay is exponential [see Fig. 2(a)], like the results of conventional optical Bloch equation with $T_2\sim 2/Wp^2$. With the increase of the modulation, the contributions of the other terms become larger and the decay tends to a multiexponential process [Fig. 2(b)]. When $p>1$, the echo decays exponentially and is modulated by a triangle function [Fig. 2(c)]. In the range $1<p<2$, the period of the triangle function is long; in the first few decades, owing to the exponential envelope, the oscillation does not appear. As p increases, the period becomes shorter and shorter and the oscillation is no-

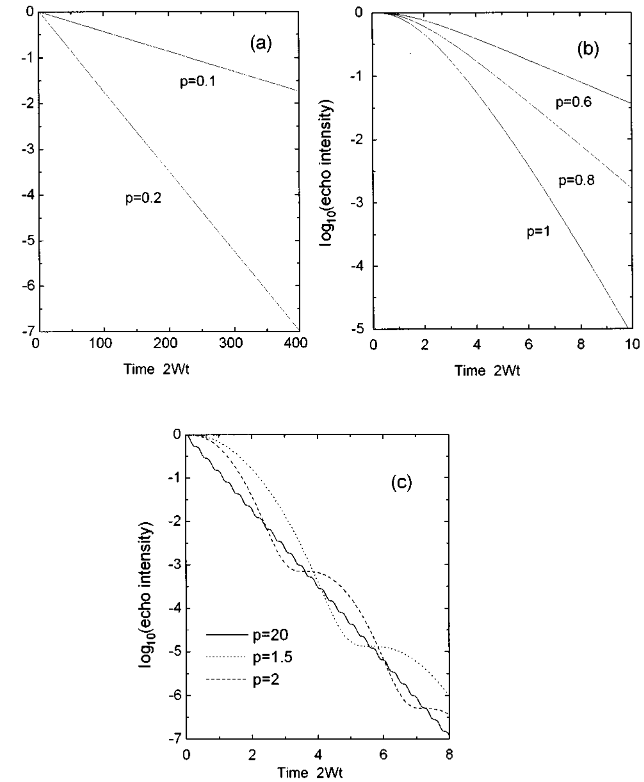

FIG. 2. Echo-intensity decay curves for different values of frequency modulation p. (a) $p=0.1$, 0.2; (b) $p=0.6$, 0.8, 1; (c) $p=1.5$, 2, 20.

table. For very large modulation, however, the factor of the triangle function term is very small, so echo decay tends to go back to an exponential with $T_2 \sim 1/W$.

It is interesting to compare the echo-intensity decay by Gaussian and Poisson processes. With our notation, both processes lead to an $\exp[-p^2 t]$ decay for small modulation p, while for large p, they take the form $\exp[-p^2 t^3/3]$ and $\exp(-2t)$, respectively. Different from the Gaussian process, the Poisson process results in a comparable slow and Δ-independent decay. From Eq. (3.2), we calculate the derivatives of $\langle E(t_{21})\rangle$ with respect to t_{21}. The first nonzero derivative is $[d^3\langle E(t_{21})\rangle/dt_{21}^3] = -p^2$ at $t_{21}=0$, so when $t_{21}\rightarrow 0$, the asymptote of the echo intensity is the same as $\exp[-p^2 t^3/3]$, similar to the Gaussian process.

IV. OPTICAL DEPHASING IN PRACTICAL SYSTEMS

We have discussed the optical dephasing of an ion at an impurity site arising from the flipping of a single spin. In the practical system, all spin flips contribute to the optical dephasing. If we assume that the effect of the different spins are independent, the total relaxation function can be expressed as: $\langle \exp(\phi)\rangle = \Pi_i \langle \exp(\phi)\rangle_i$, where $\langle \exp(\phi)\rangle_i$ is the relaxation function describing the decay due to the flip of the spin i.

In dilute ruby, Al nuclear spins at the lattice interact with each other and with the Cr^{3+} electron spin by magnetic dipole-dipole interactions. This causes the fluctuation of the Cr^{3+} transition frequency [14], and is the main source of the dephasing at high magnetic fields and low temperature [5,6].

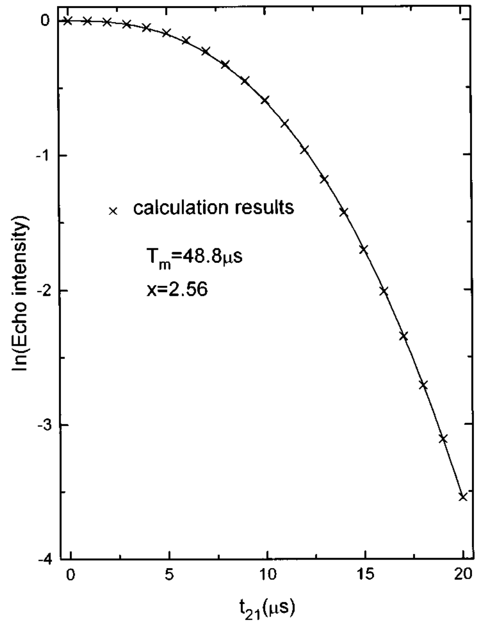

FIG. 3. Calculated results of the echo-intensity decay of $R_1(-\frac{3}{2})$ in ruby. The solid curve is the fitted result using the form $I_0 \exp[-(4t_{21}/T_m)^x]$.

The mutual spin-flip rate of Al spins i and j, W_{ij}, can be calculated as in [15]. The parameters used in the calculation are as follows: Al nucleus gyromagnetic ratio 1.1 kHz/G; the bulk nuclear magnetic resonance linewidth 3.0 kHz [half width at half maximum (HWHM)]; g factors of the ground and excited states of Cr^{3+}, 1.984 and 2.445, respectively; and W_0 the characteristic nearest-neighbor mutual spin-flip rate 5 kHz.

We calculate $\delta\omega_i$ of the Cr^{3+} ion caused by each of the 1100 surrounding Al nuclear spins and mutual flip rate of each Al spin with its 26 neighbors ($W_i = \Sigma_{j=1}^{26} W_{ij}$). The stochastic parameter p_i of the spin i is equal to $W_i \delta\omega_i$, using Eq. (3.3), one can get $\langle \exp(\phi)\rangle_i$. Taking their product, we obtain the echo-intensity decay as shown in Fig. 3. Fitting the result with $I = I_0 \exp[-(4t_{21}/T_m)^x]$, we have $T_m = 48.8\ \mu$s and $x=2.56$, which are very close to the experimental results $T_m = 50\ \mu$s and $x=2.4$ [4,5], and to the computer simulated results $T_m = 54\ \mu$s and $x=2.6$ with the same parameters carried out recently by Huang and Szabo [16]. As mentioned in Ref. [15], x would be rather universal for any paramagnetic ion with a large electron spin magnetic moment, independent of structural details. The result of the simulation reported in Ref. [15] is $x=2.8$ for Er^{3+}:$YLiF_4$ (a little larger value of x arose from the large g factor of the Er^{3+} in the simulation). So, the simple bivalued random telegraph model is in very good agreement with these results.

V. CONCLUSION

In this paper, we have investigated optical dephasing processes arising from Poisson stochastic modulation. Using a simple random telegraph model, we gave the analytic expression for free-induced decay and photon-echo decay in the whole range of the stochastic parameters. In the small modulation range, FID or the echo-intensity decay varies from single exponential ($T_2 \sim 2/Wp^2$) to multiexponential with an increase of frequency modulation. In the large modulation range, FID and the echo decay are damped oscillations. When p is very large, the oscillations can be neglected, the echo-intensity decay is back to exponential $(T_2 \sim 1/W)$. Considering contributions of all the spin flips in a practical lattice, we calculated $R_1(-\frac{3}{2})$ echo-intensity decay in ruby at high magnetic fields. The calculated echo intensity decay is very close to the experimental and computer-simulation results.

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