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Random-telegraph-signal theory of optical resonance relaxation with applications to free induction decay

K. Wódkiewicz
*Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627 and Institute of Theoretical Physics, Warsaw University, PL-00-681 Warsaw, Poland**

J. H. Eberly
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627

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We derive the effects of random telegraph noise on the equations of optical resonance. The random-telegraph-signal model allows some results to be found exactly, independent of the strength of the noise or the strength of the field that induces the resonant transitions. Our optical resonance equations depart from the well-known empirical optical Bloch equations in several ways. We obtain excellent agreement at high, low, and intermediate Rabi frequencies with the data of DeVoe and Brewer [Phys. Rev. Lett. **50**, 1269 (1983)]. This is the first such agreement that has been reported with a value of the noise coherence time smaller than about 20 μsec.

I. INTRODUCTION

In magnetic[1] or optical[2] resonance the effective dynamical equations in universal use are the Bloch equations[3] which are conventionally written using the rotating frame and the rotating-wave approximation as follows for any two-level or spin system:

$$\frac{d}{dt}\begin{bmatrix} u \\ v \\ w \end{bmatrix} = \left[\begin{bmatrix} 0 & -\Delta & 0 \\ \Delta & 0 & \chi \\ 0 & -\chi & 0 \end{bmatrix} - \begin{bmatrix} 1/T_2 & 0 & 0 \\ 0 & 1/T_2 & 0 \\ 0 & 0 & 1/T_1 \end{bmatrix} \right] \begin{bmatrix} u \\ v \\ w \end{bmatrix} + \begin{bmatrix} 0 \\ 0 \\ w_{\text{eq}}/T_1 \end{bmatrix} . \tag{1.1}$$

Here χ and Δ are the Rabi frequency and detuning, and u, v, and w are defined according to the optical convention[2] ($w=-1$ indicates the lower energy state). These equations have been applied to a very wide range of different effects[2] with enormous success for more than three decades.

The Bloch equations[3] include, in addition to the coherent evolution matrix, a *diagonal* relaxation matrix containing the two homogeneous lifetimes T_1 and T_2. These two phenomenological lifetimes were added by Bloch to take into account transverse (T_2) and longitudinal (T_1) losses caused by the interaction of a spin system with all additional degrees of freedom that are not accounted for by the spin's coherent evolution matrix.

For optical transitions in completely isolated atoms or molecules the dominant contribution to T_1 and T_2 comes from radiative effects associated with radiation reaction or zero-point fluctuations of the free electromagnetic field. But in most realistic situations the radiative effects are certainly not the only source and usually not the dominant source of relaxation fluctuations and incoherence. In other words, additional nonradiative damping is understood to be included in the definitions of T_1 and T_2. Because of the very complicated, and in many cases even obscure, microscopic nature of all the relevant relaxation fluctuations, it is well established to model them by classical random processes.[4] Depending on the source of the fluctuations, different modifications of T_1 and T_2 occur.[5]

In some cases it has been shown that external fluctuations lead to so-called "substitution" rules.[5] In such situations we can derive certain rules explaining how to add "by hand" (but exactly) the nonradiative contributions to T_1 and T_2. Even if it is rather difficult to give a very general criterion for the substitution rules to hold, it appears that in most situations when the noise enters the dynamical equations of motion in a multiplicative way, and has an infinitely short coherence time τ_c, substitution rules are to be expected.

However, realistic noise fluctuations have a finite coherence time, perhaps comparable in magnitude with other typical lifetimes. What is the impact of such fluctuations on the empirical lifetimes in the Bloch equations? Are the substitution rules still valid? Do the Bloch equations themselves remain valid and in the same form?

Some attacks on such questions were undertaken in the past.[6,7] In these studies the focus was on high power of the exciting resonant field. We can thus regard them as studies of the case $\tau_c > 1/\chi$, i.e., the case where the noise coherence time is a substantial fraction of one Rabi cycle or greater. The result of Redfield's work is particularly notable because it predicts accurately (verified repeatedly in magnetic resonance observations) that T_2 is dependent on χ in such a way that $T_2 \rightarrow 2T_1$ as $\chi \rightarrow \infty$. That is, at the highest powers, T_2 approaches its purely radiative relation with T_1. However, in none of these past studies was a formula given appropriate to intermediate-field strengths. Also, one can criticize reliance on the concept of entropy, which should not be necessary in a properly dynamic theory.

Just recently the first optical experiments involving measurements of effective free-induction-decay (FID) lifetimes at large, small, and intermediate values of χ have been reported.[8–11] The quality of data obtained in these experiments calls for a much more systematic and quantitative approach to the problem. The observed data show that we have to deal with a completely new regime where the usual substitution rules are not valid any longer.

In this paper we give a detailed theory of optical resonance relaxation in a format that avoids many of the technical problems associated with other approaches. It is based on a random-telegraph-signal description of noise fluctuations. The telegraph model is simple enough to allow a full exact analytic solution to the steady-state optical resonance equations (ORE) for any χ or τ_c.

This paper is organized as follows. In Sec. II we introduce different models of noise fluctuation based on the random telegraph signal (RTS). Phase, frequency, and amplitude fluctuations are then defined. In Sec. III we show how it is possible to derive the generalized relaxation structure that replaces the purely "diagonal" damping of the Bloch equations.

In Secs. IV—VI we apply this method to the explicit derivation of the effective resonance relaxation dynamics arising from frequency, amplitude, and phase fluctuations, respectively. We define effective linewidths in all cases.

In Sec. VII we discuss free induction decay (FID). In Sec. VII A we give a simplified theory of the absorption line shape and we discuss in this framework the impact of various noises on FID. In Sec. VII B we make a detailed quantitative discussion of the recent experiments and our theory and show that good agreement exists between them. This agreement is obtained for the first time with a value of τ_c smaller than about 20 μsec. Finally, Sec. VIII gives some concluding remarks.

II. RTS MODEL OF EXTERNAL FLUCTUATIONS

We are going to assume that the environment of a two-level system acts as a reservoir and gives rise to fluctuations in various system parameters. The Rabi frequency, its phase, and the detuning frequency are examples of these parameters. In each case we will use a random telegraph signal (RTS) as a working model for the way these parameters fluctuate.

We shall assume that these fluctuations can be decorrelated from purely radiative vacuum fluctuations characterized by the lifetime $1/2\gamma$ and linewidth γ, where 2γ is the spontaneous Einstein A coefficient. Because the coherence time or the "memory" of spontaneous emission is extremely short, we believe that this is well justified. This approach is in agreement with the point of view that certain types of fluctuations can be treated or modeled at the so-called mesoscopic level, where a partial reduction of some other degrees of freedom has already been performed. A full theory would of course require that all kinds of fluctuations are treated on equal footing using a complete microscopic description.

With this important simplification of the vacuum reservoir we shall assume that the stochastic incoherences due to the fluctuating environment are going to be described by a two-step random telegraph signal $x(t)$ jumping between two states a and $-a$. In this respect our approach is qualitatively different from several other discussions that have appeared,[12–15] and, as we shall show, it has a number of advantages. The jump process is characterized by the frequency $1/T$ that the telegraph signal changes its state. This very simple well-known dichotomic Markov process is fully defined by the following correlations:[16(a)]

$$\langle x(t)\rangle=0;\;\; \langle x(t+\tau)x(t)\rangle=a^2e^{-|\tau|/\tau_c}\,, \tag{2.1}$$

where the coherence time τ_c is related to the telegraph jump time by $\tau_c=T/2$. It is well known that a random telegraph signal is not a Gaussian stochastic process, though it is entirely defined by its correlation functions up to second order, as given by Eq. (2.1).

We shall not give here any deep justification or physical reasons for choosing a random telegraph stochastic description of the external noises. Basically one adopts a stochastic description in the first place only when the physical fundamentals are obscure or too difficult. Thus convenience and flexibility of the model are more than sufficient motivation. The flexibility of the telegraph model is great enough, for example, to admit a finite coherence time τ_c. This will be fundamental to our study of the relaxation dynamics of the optical resonance equations.

We shall include these RTS incoherences into Eq. (1.1), assuming that the atomic detuning, the amplitude, and the phase of the Rabi oscillation perform independent random telegraph jumps. For detuning fluctuations (sometimes called frequency fluctuations) the detuning Δ is replaced by $\Delta+\delta\Delta(t)$ where

$$\langle \delta\Delta(t)\rangle=0;\;\; \langle \delta\Delta(t+\tau)\,\delta\Delta(t)\rangle=a^2e^{-|\tau|/\tau_c}\,. \tag{2.2}$$

Rabi amplitude fluctuations will be described by a deviation $\delta\chi(t)$ from the constant coherent value χ, and this deviation is a random telegraph signal:

$$\langle \delta\chi(t)\rangle=0;\;\; \langle \delta\chi(t+\tau)\,\delta\chi(t)\rangle=a^2e^{-|\tau|/\tau_c}\,. \tag{2.3}$$

The phase fluctuations correspond to a noise in which the phase $\phi(t)$ performs random jumps with statistics characterized by

$$\langle \delta\phi(t)\rangle=0;\;\; \langle \delta\phi(t+\tau)\,\delta\phi(t)\rangle=a^2e^{-|\tau|/\tau_c}\,. \tag{2.4}$$

In each of the three cases (2.2)—(2.4) we have used the same symbols a and τ_c to denote the strength and the coherence of the noise. Obviously in each case the physical meaning and the interpretation of these parameters is different but because we are going to treat all these cases separately and independently we shall preserve this uniform notation in this paper.

We close this section by giving one simple example that illustrates the difficulties one encounters in making connections between reasonably flexible noise models and well-known relaxation formulas. We consider free induction decay of a collection of linear oscillators. We want to compute the macroscopic polarization at times following application of a saturating laser field that is turned off at $t=0$.

The relevant equations are just those for u and v given in (1.1) with $w=-1$. We choose detuning fluctuations for illustration and so have, for $t\geq 0$,

$$\frac{d}{dt}(u-iv)=-[\gamma+i\Delta+i\,\delta\Delta(t)](u-iv)\ . \qquad (2.5)$$

The solution to (2.5) is of course trivial:

$$(u-iv)_t=(u-iv)_0 e^{-(\gamma+i\Delta)t}\exp\left[-i\int_0^t ds\,\delta\Delta(s)\right]\ , \qquad (2.6)$$

and $(u-iv)_0$ is determined completely by Δ and the field χ that saturated the dipole prior to $t=0$.

To obtain the FID polarization we must average over the distribution of detunings $g(\Delta)$ and over the ensemble of fluctuations $\delta\Delta(t)$. The result can be written

$$P(t)=\mathrm{Re}\int d\Delta\, g(\Delta)(u-iv)_0 e^{-(\gamma+i\Delta)t}C(t)\ , \qquad (2.7)$$

where

$$C(t)=\left\langle \exp\left[-i\int_0^t ds\,\delta\Delta(s)\right]\right\rangle\ . \qquad (2.8)$$

For a telegraph noise model of $\delta\Delta(t)$ we have[16(b)]

$$C(t)=(1/2\lambda)(\lambda+1/2\tau_c)e^{(\lambda-1/2\tau_c)|t|}+(1/2\lambda)(\lambda-1/2\tau_c)e^{(-\lambda-1/2\tau_c)|t|}\ , \qquad (2.9)$$

where $(1/2\tau_c)^2-\lambda^2=a^2$. In contrast, a Gaussian Ornstein-Uhlenbeck model of $\delta\Delta(t)$ gives[5]

$$C(t)=\exp\{-a^2\tau_c[t+\tau_c(e^{-|t|/\tau_c}-1)]\}\ . \qquad (2.10)$$

As is clear from expressions (2.9) and (2.10), FID in general cannot be described by a single parameter. The two different models' characteristic functions (2.9) and (2.10) depend explicitly on the two parameters a and τ_c in different ways.

This situation has already been encountered in measurements of multiphoton-absorption line shape in the presence of laser fluctuations. In that case, the strength as well as the correlation time must be fixed separately.[17] This is the same as saying that the correlation function simply does not decay as a single exponential. In particular, for $t\ll\tau_c$ the Ornstein-Uhlenbeck correlation function always strongly deviates from an exponential, as does the telegraph correlation[16] if $2a=1/\tau_c$.

These obvious remarks are nevertheless important because they show that there is not a unique definition of Bloch's transverse relaxation time T_2, *even in the weak-field limit.* That is, one identifies T_2 as the overall lifetime for homogeneous polarization decay [the $d\Delta$ integral in (2.7) is irrelevant], so if $C(t)$ is characterized by the single exponential decay rate γ_c, then

$$1/T_2=\gamma+\gamma_c\ . \qquad (2.11)$$

But if $C(t)$ is not a single exponential, then T_2 does not have a meaning in any usual sense.

Note that an assumption about the telegraph model, namely $a\tau_c\ll 1$, leads to very-nearly-exponential decay with

$$\gamma_c=a^2\tau_c\ . \qquad (2.12)$$

In order to make contact with reported data, which appears to fit a single exponent, we will use (2.12) in (2.11) to determine the value of T_2 when necessary for numerical purposes. However one can keep in mind that a possible result of future experiments, which will assist theoretical developments greatly, will be careful observations of line shapes. These will help discriminate among various possible $C(t)$ functions proposed in theoretical models.

III. EFFECTIVE OPTICAL RESONANCE EQUATIONS

In the case of optical resonance, the dynamical equations of motion, which have their origin in the basic Liouville or Heisenberg equations, can be written in the following general form:

$$\frac{dV(t)}{dt}=-iM(x(t))V(t) \qquad (3.1)$$

with a given operator M which depends possibly nonlinearly on the external arbitrary random telegraph noise. In the most familiar case, which is to be treated here, there are four independent Heisenberg operators or density matrix elements, corresponding to the populations of the two relevant levels and the real and imaginary (dispersive and absorptive) parts of the transition dipole moment. Thus in this case M is a 4×4 matrix and V is a four-component vector. If transitions out of the two-level subspace are assumed absent (automatically the case for spins, but not for atoms), then one of the four independent dynamical elements corresponds to the conserved total population and can be removed from consideration. The small price is the familiar inhomogeneous term shown in (1.1). Temporarily we will work without the inhomogeneous term in the more convenient 4×4 format and revert to the 3×3 format at the end.

It is well known that the stochastic expectation value of V is given by

$$\langle V(t)\rangle=\tfrac{1}{2}[V_a(t)+V_{-a}(t)]\ , \qquad (3.2)$$

where the marginal average $V_a(t)$ can be calculated from the following master equation:[16]

$$\frac{dV_a(t)}{dt}=\left[-iM(a)-\frac{1}{2\tau_c}\right]V_a(t)+\frac{1}{2\tau_c}V_{-a}(t)\ . \qquad (3.3)$$

We now use (3.3) to write the equations for the symmetric and antisymmetric superpositions of $V_a(t)$. Forming the solutions of Eq. (3.3) we can write the following exact and closed system of equations involving the stochastic expectation value of V:

$$\frac{dV_S(t)}{dt}=-iM_S V_S(t)-iM_A V_A(t)\ , \qquad (3.4a)$$

$$\frac{dV_A(t)}{dt}=(-iM_S-1/\tau_c)V_A(t)-iM_A V_S(t)\ , \qquad (3.4b)$$

where

$$V_{S,A}(t)=\tfrac{1}{2}(V_a\pm V_{-a})\ , \qquad (3.4c)$$

$$M_{S,A}=\tfrac{1}{2}[M(a)\pm M(-a)]\ , \qquad (3.4d)$$

and following Eq. (3.2),

$$\langle V(t)\rangle = V_S(t) \ . \tag{3.4e}$$

Note that the evolution of $V_S(t)$ in Eq. (3.4a) does not involve the coherence time τ_c of the noise. This jump time appears in an explicit way only in the evolution of $V_A(t)$.

We can easily derive the following integro-differential equation for $\langle V(t)\rangle$ along from Eqs. (3.4a) and (3.4b):

$$\frac{d\langle V(t)\rangle}{dt} = -iM_S\langle V(t)\rangle - M_A \int_0^t ds\, e^{(-iM_S-1/\tau_c)(t-s)} \times M_A\langle V(S)\rangle \ . \tag{3.5}$$

This equation has a convenient Laplace transform and the solution has the form

$$\langle V(t)\rangle = \int \frac{dz}{2\pi i} \frac{e^{zt}}{z+iM_S+M_A\left[\dfrac{1}{z+iM_S+1/\tau_c}\right]M_A} V(0) \tag{3.6}$$

which can be simplified if we focus our attention on long times by using the "pole approximation" (taking $z=0$ in the term proportional to M_A^2). In this approximation (valid, roughly speaking, when the early transients have died away) the denominator takes the form $z+iMs+\Sigma$, where the "line broadening" or "relaxation" matrix Σ is given by

$$\Sigma = M_A(iM_S+1/\tau_c)^{-1}M_A \ . \tag{3.7}$$

In this long time approximation Eq. (3.5) is strictly equivalent to the simpler equation

$$d\langle V(t)\rangle/dt = (-iM_S-\Sigma)\langle V(t)\rangle \tag{3.8}$$

which has a time-independent damping matrix. *It is this equation that we define to be the random telegraph equation of optical resonance,* or the optical resonance equations (ORE), and which can be taken as the substitute for the Bloch equations. That is, the ORE in (3.8) are in the spirit of the Bloch equations, because the relaxation matrix is specified and time independent. But, as we discuss below, the ORE are not even approximately, except in a very loose sense, the Bloch equations themselves.

It is very important to note that a steady-state solution of Eqs. (3.4) leads to the following nontrivial formula:

$$(-iM_S-\Sigma)V_S(\infty)=0 \ . \tag{3.9}$$

Equation (3.9) shows that Σ not only determines the relaxation time scale of the (necessarily approximate) ORE defined in (3.8) but also defines a power- and noise-dependent effective linewidth matrix, which is *exact* in steady state.

In the 3×3 Bloch-equation case, given by Eq. (1.1), the matrix M_S contains on its diagonal part the radiative dampings characterized by γ. This means that regardless of the character of the noise (collisional, phase, etc.) we can rewrite the matrix M_S in the following form:

$$-iM_S = -iM_{\rm coh} - \Gamma_r \ , \tag{3.10}$$

where Γ_r is the diagonal spontaneous emission matrix containing only the Einstein A coefficient and $M_{\rm coh}$ depends only on the detuning and Rabi frequency. Thus the optical resonance equations can also be written in the more explicit form

$$\frac{d}{dt}\begin{bmatrix} u \\ v \\ w \end{bmatrix} = (-iM_{\rm coh}-\Gamma_r-\Sigma)\begin{bmatrix} u \\ v \\ w \end{bmatrix} + \begin{bmatrix} 0 \\ 0 \\ 2\gamma w_{\rm eq} \end{bmatrix} , \tag{3.11}$$

where we have recovered the inhomogeneous term missing from Eq. (3.1), by returning to 3×3 matrices, as discussed there.

IV. ORE WITH ATOMIC-FREQUENCY FLUCTUATIONS

For atomic-frequency fluctuations[16(c)] described by Eq. (2.2) we have the following relations:

$$-iM_{\rm coh} = \begin{bmatrix} 0 & -\Delta & 0 \\ \Delta & 0 & \chi \\ 0 & -\chi & 0 \end{bmatrix}, \quad \Gamma_r = \begin{bmatrix} \gamma & 0 & 0 \\ 0 & \gamma & 0 \\ 0 & 0 & 2\gamma \end{bmatrix} \tag{4.1}$$

and from Eq. (3.7) we calculate

$$\Sigma = \begin{bmatrix} \Gamma_{11} & \Gamma_{12} & 0 \\ -\Gamma_{12} & \Gamma_{22} & 0 \\ 0 & 0 & 0 \end{bmatrix} , \tag{4.2}$$

where

$$\Gamma_{11} = a^2\frac{(1/\tau_c+\gamma)(1/\tau_c+2\gamma)}{P} , \tag{4.3a}$$

$$\Gamma_{22} = a^2\frac{(1/\tau_c+\gamma)(1/\tau_c+2\gamma)+\chi^2}{P} , \tag{4.3b}$$

$$\Gamma_{12} = -a^2\Delta\frac{(1/\tau_c+2\gamma)}{P} , \tag{4.3c}$$

and

$$P = [(1/\tau_c+\gamma)^2+\Delta^2](1/\tau_c+2\gamma)+\chi^2(1/\tau_c+\gamma) \ . \tag{4.3d}$$

These relations give an *exact* expression for the effective linewidths of the *steady-state* Bloch vector $(u(\infty,\Delta)$, $v(\infty,\Delta)$, $w(\infty,\Delta))$. In order to get a rough idea of the power dependence of these effective linewidths we temporarily simplify the relaxation matrix (4.2) by assuming that $1/\tau_c > \gamma, \Delta$. As a result we obtain

$$\Gamma_{11} \simeq a^2\frac{\tau_c}{1+\chi^2\tau_c^2}; \quad \Gamma_{22} \simeq a^2\tau_c; \quad \Gamma_{12} \simeq 0 \ . \tag{4.4}$$

Note that Γ_{11} and Γ_{22} are proportional to the strength of the noise a^2 and Γ_{11} depends on the Rabi frequency χ of the driving field in such a way that the damping forms of u and v are different and this difference is power dependent. This asymmetry between Γ_{11} and Γ_{22} is most pronounced in the very-strong-field regime $(\chi\tau_c \gg 1)$ where

$$\Gamma_{11} \simeq 0; \quad \Gamma_{22} \simeq a^2\tau_c \ . \tag{4.5}$$

Expressions for the conventional transverse and longitudinal damping times are associated with the weak-field limit. We can obtain these very simply now from (4.4). For a weak incident field (or, equivalently, a very short fluctuation coherence time) we have $\chi\tau_c \ll 1$ and we obtain from Eqs. (4.4), $\Gamma_{11} \simeq \Gamma_{22} \simeq a^2\tau_c$. As a result the following expressions are found for the usual homogeneous relaxation rates $1/T_2$ and $1/T_1$ of a two-level system exposed to telegraph frequency noise:

$$1/T_2 = \gamma + a^2\tau_c \ , \tag{4.6a}$$

$$1/T_1 = 2\gamma \ . \tag{4.6b}$$

This means that at zero power, frequency noise influences only the value of the transverse rate $1/T_2$, leaving an unchanged longitudinal rate. From Eqs. (4.6) it follows that one can fix the noise parameters a and τ_c (or rather their combination) by measuring the difference between the two homogeneous lifetimes at zero value of the incident field:

$$1/T_2 - 1/2T_1 = a^2\tau_c \ . \tag{4.7}$$

The appearance of this particular combination of the parameters a and τ_c when $\chi \to 0$ can be easily understood for the frequency noise in the limit of $\tau_c \to 0$ but with a fixed value of $a^2\tau_c = D$. In this limit the random telegraph has all of the mathematical properties of Gaussian white noise with diffusion constant D and accordingly we obtain from Eqs. (4.3) the well-known result

$$\Gamma_{11} = \Gamma_{22} = D \quad \text{and} \quad \Gamma_{12} = 0 \ ,$$

i.e., the usual substitution rules for the natural linewidths in the Bloch equations. For fluctuations with a finite τ_c, these substitution rules are not valid any longer and the effective linewidths become power dependent.

V. ORE WITH LASER INTENSITY FLUCTUATIONS

Intensity fluctuations of the laser give rise in the ORE to fluctuations in χ. For Rabi-frequency fluctuations described by Eq. (2.3) we have the coherent and the radiative evolution given by the same matrices (4.1) as in the case of atomic-frequency fluctuations. What is different is the matrix M_A and accordingly different is the damping matrix Σ. For such fluctuations we have

$$\Sigma = \begin{bmatrix} 0 & 0 & 0 \\ 0 & \Gamma_{22} & \Gamma_{23} \\ 0 & -\Gamma_{23} & \Gamma_{33} \end{bmatrix} , \tag{5.1}$$

where

$$\Gamma_{22} = a^2 \frac{(1/\tau_c + \gamma)^2 + \Delta^2}{P} , \tag{5.2a}$$

$$\Gamma_{33} = a^2 \frac{(1/\tau_c + 2\gamma)(1/\tau_c + \gamma)}{P} , \tag{5.2b}$$

$$\Gamma_{23} = a^2 \chi \frac{(1/\tau_c + \gamma)}{P} , \tag{5.2c}$$

and the expression for P is again given by Eq. (4.3d).

In the transient regime, from Eqs. (5.2) we obtain the following expressions for the effective linewidths:

$$\Gamma_{22} = \Gamma_{33} = a^2 \frac{\tau_c}{1+\chi^2\tau_c^2} , \quad \Gamma_{23} \simeq a^2 \frac{\chi\tau_c^2}{1+\chi^2\tau_c^2} \ . \tag{5.3}$$

In the strong-field limit $\chi\tau_c \gg 1$ all these additional noise-dependent dampings go to zero, leaving the Bloch vector damped only by its radiative rate γ. This means that in the super-strong-field case there is no impact of the fluctuations on the Bloch-vector evolution, in sharp contrast to the previous case of atomic-frequency fluctuations where a nontrivial saturation of Γ_{22} occurred. Note that a different conclusion would be obtained if a denoted relative rather than absolute jumps of Rabi frequency.

If the incident field fluctuates about a zero average ($\chi = 0$ but $a \neq 0$), we obtain from Eq. (5.2) the following lifetimes:

$$1/T_1 = 2\gamma + a^2\tau_c \tag{5.4a}$$

and

$$1/T_2^v = \gamma + a^2\tau_c \ , \tag{5.4b}$$

where T_2^v indicates that only the damping of the v component of the Bloch vector is affected. Note that the damping term of u is not influenced by the noise because $\Gamma_{11} = 0$ and a single universal transverse lifetime $1/T_2$ cannot any longer describe the damping of the u and v components of the Bloch vector.

In the limit of Gaussian white noise, i.e., when $\tau_c \to 0$ with constant $D = a^2\tau_c$, we recover the known result

$$\Gamma_{22} = \Gamma_{33} = D \ \text{and} \ \Gamma_{23} = 0 \ \text{ for } \tau_c \to 0 \ , \tag{5.5}$$

and, again, simple substitution rules with power-independent lifetimes hold true. This case gives an interesting example of a "partial" substitution rule for the ORE. For $\chi = 0$, Rabi-frequency fluctuations differentiate the transverse dampings of the u and v component of the Bloch vector. This asymmetry follows, of course, from the obvious fact that for $\chi = 0$ the dynamical evolution of u can be fully decoupled from the v and the w components.

VI. ORE WITH PHASE FLUCTUATIONS

For phase fluctuations[16(b)] described by Eq. (2.4) the radiative damping matrix Γ_r is the same as in Eq. (4.1), but the coherent evolution is modified [see the definition of M_S given by Eq. (3.4d) and the relation (3.10)]:

$$-iM_{\text{coh}} = \begin{bmatrix} 0 & -\Delta & 0 \\ \Delta & 0 & \chi\cos a \\ 0 & -\chi\cos a & 0 \end{bmatrix} , \tag{6.1}$$

where a is now the jump size of the fluctuating phase.

From Eq. (3.6) we calculate the following expression for the damping matrix:

$$\Sigma = \begin{bmatrix} \Gamma_{11} & 0 & \Gamma_{13} \\ 0 & 0 & 0 \\ -\Gamma_{13} & 0 & \Gamma_{33} \end{bmatrix} , \tag{6.2}$$

where

$$\Gamma_{11}=\chi^2 \sin^2 a \frac{(1/\tau_c+\gamma)^2+\Delta^2}{Q} , \qquad (6.3a)$$

$$\Gamma_{33}=\chi^2 \sin^2 a \frac{(1/\tau_c+\gamma)(1/\tau_c+2\gamma)+\chi^2\cos^2 a}{Q} , \qquad (6.3b)$$

$$\Gamma_{13}=-\chi^2 \sin^2 a \frac{\Delta\chi \cos a}{Q} , \qquad (6.3c)$$

and

$$Q=[(1/\tau_c+\gamma)^2+\Delta^2][(1/\tau_c+2\gamma)] +\chi^2\cos^2 a\,(1/\tau_c+\gamma) . \qquad (6.4)$$

Note that the expression for P introduced in Secs. IV and V can be obtained from Q simply by taking $a=0$: $P=Q(a=0)$. As in the case of atomic-frequency fluctuations, the off-diagonal damping Γ_{13} vanishes for resonant interactions. The damping terms in the matrix (6.2) depend in a nonlinear way on the jump size a of the phase fluctuations in obvious contrast to the two previously discussed cases. This follows from the fact that laser phase fluctuations cannot be incorporated in the ORE as a linear multiplicative noise.

In the transient-time regime we obtain from Eqs. (6.3) and (6.4) the following expression for the effective linewidths:

$$\Gamma_{11}\simeq\chi^2 \sin^2 a \frac{\tau_c}{1+\chi^2\tau_c^2\cos^2 a} ,$$
$$\Gamma_{33}\simeq\chi^2\tau_c \sin^2 a ,\quad \Gamma_{13}\simeq 0 . \qquad (6.5)$$

In the strong-field limit ($\chi\tau_c>1$) the damping Γ_{11} becomes power independent. Because the phase fluctuations accompany the incident field χ it is not surprising at all that in the limit $\chi=0$ there is no impact of the fluctuations on the homogeneous lifetimes and as a result in that limit we have $1/T_1=2\gamma$, $1/T_2=\gamma$. In this case the parameters of the noise a and τ_c should be fixed by an independent measurement of the laser-field correlations. Here it is useful to recall the appropriate autocorrelation function[16(b)]

$$\langle e^{-i\phi(t+\tau)}e^{i\phi(t)}\rangle=\cos^2 a+\sin^2 a \exp(-|\tau|/\tau_c) \qquad (6.6)$$

which is needed for the calculation of the power spectrum of the driving laser light.

VII. FREE INDUCTION DECAY OF THE EFFECTIVE ORE

One possible way of testing the behavior of the effective ORE is the measurement of the optical free induction decay (FID) of the Bloch vector for different values of the incident field and for different sources of noise. The first experiments of this type have been performed recently,[10] and they show quite clearly that the approach via the standard Bloch equations to FID fails to describe properly the observed experimental data. Several theoretical analyses of these experiments have been constructed recently with power-dependent lifetimes induced by weak Gaussian fluctuations of the atomic frequency.[12–14]

In this paper we reconsider this problem in the framework of telegraph noise with no constraints on the power of the noise. In order to give better insight into the theory of the effective ORE with intensity-dependent lifetimes we shall present in this section two approaches to this problem.

The first approach, contained in Sec. VII A, is based on simplified expressions ($1/\tau_c \gg \gamma,\Delta$) for the effective linewidths derived in the previous sections. This simplified theory will give us approximate expressions for the absorption line shape of the optical Bloch vector under the influence of relaxation fluctuations and a strong incident field. These linewidths could be determined for a two-level system strongly driven by external stochastic perturbations in a variety of measurements.

This simplified approach need not give a good quantitative agreement with the observed data, but it should contain the essential ingredients of the effective line shapes, and give at the same time a qualitative explanation of the observed effects.

In our second approach, contained in Sec. VII B, we perform a more careful and a more detailed discussion of FID with random telegraph frequency fluctuations. We will use the *exact* form of the *steady state* effective linewidths derived in Sec. IV in order to derive the FID signal observed in an optical transition in an impurity-ion solid. We obtain a good quantitative agreement between our theory and the recently published experiments. We postpone such a detailed discussion of phase and amplitude fluctuations because present experiments do not appear to require them.

A. Absorption line shape

We shall now present a simplified version of the absorption line shape based on the approximate values of the effective linewidths in the ORE derived in the previous sections under the condition that $1/\tau_c>\gamma,\Delta$. Note that for all three cases (frequency, amplitude, and phase fluctuations) the only nonvanishing elements of the effective damping matrix Σ are the following [see Eqs. (4.4), (5.3), (6.5)]:

$$\Sigma=\begin{vmatrix}\Gamma_{11} & 0 & 0\\ 0 & \Gamma_{22} & \Gamma_{23}\\ 0 & \Gamma_{32} & \Gamma_{33}\end{vmatrix} , \qquad (7.1)$$

where the analytic form of any of these Γ_{ij} can be found in Secs. IV–VI. Due to our approximations ($1/\tau_c>\gamma$, and Δ) the matrix elements of Σ are dependent only on the noise and on χ.

With this form of the damping matrix we calculate that the steady-state solutions of Eq. (3.11) for the two real components of the atomic dipole moment (u and v) are the following:

$$u(\infty)=-2w_{\rm eq}\gamma\frac{\Delta(\Gamma_{23}-\chi)}{D} , \qquad (7.2a)$$

$$v(\infty)=-2w_{\rm eq}\gamma\frac{(\gamma+\Gamma_{11})(\Gamma_{23}-\chi)}{D} , \qquad (7.2b)$$

where

$$D=(\gamma+\Gamma_{11})(\gamma+\Gamma_{22})(2\gamma+\Gamma_{33})-(\gamma+\Gamma_{11})(\Gamma_{32}+\chi)(\Gamma_{23}-\chi)+(\Gamma_{23}+2\gamma)\Delta^2 . \quad (7.2c)$$

From the resonance form of $u(\infty)$ and $v(\infty)$ we obtain immediately the following expression for the absorption linewidth of the strongly driven two-level system:

$$\Gamma_A=\left[\frac{(\gamma+\Gamma_{11})(\gamma+\Gamma_{22})(2\gamma+\Gamma_{33})-(\gamma+\Gamma_{11})(\Gamma_{32}+\chi)(\Gamma_{23}-\chi)}{\Gamma_{33}+2\gamma}\right]^{1/2} . \quad (7.3)$$

In the absence of external fluctuations, i.e., when $\Sigma=0$, the absorption line shape has the well-known Lorentzian form with linewidth

$$\Gamma_A=[(1/T_2^r)^2+\chi^2T_1^r/T_2^r]^{1/2} , \quad (7.4)$$

where $1/T_2^r=\gamma$ and $1/T_1^r=2\gamma$ are the purely radiative lifetimes and where, for strong excitations, we have

$$\Gamma_A\simeq\chi(T_1^r/T_2^r)^{1/2}=\chi/\sqrt{2} \;\text{ for } \chi\gg\gamma . \quad (7.5)$$

With external noise we have $\Sigma\neq0$ and as a result the absorption line shape Γ_A depends on the power of the driving field. For some values of the characteristic parameters of the noise, τ_c and a, we can have a significant modification of Σ as a function of χ compared to the noise-free case.

For detuning fluctuations we have only Γ_{11} and Γ_{22} not equal to zero [see Eq. (4.4)] and as a result, the absorption line shape (7.3) takes the following form:

$$\Gamma_A^\Delta=\left[\left[\gamma+a^2\frac{\tau_c}{1+\chi^2\tau_c^2}\right](\gamma+a^2\tau_c+\chi^2/2\gamma)\right]^{1/2} , \quad (7.6a)$$

where

$$\Gamma_A^\Delta(\chi=0)=1/T_2=\gamma+a^2\tau_c \quad (7.6b)$$

and

$$\Gamma_A^\Delta\simeq\chi/\sqrt{2} \;\text{ for } \chi>\gamma,1/\tau_c . \quad (7.6c)$$

Note that in the limit of very high power, Γ_A^Δ reaches the noise-free limit given by Eqs. (7.6). For lower power, Γ_A^Δ is considerably modified by χ, and the transverse lifetime $1/T_2$, which does not show up for high intensity, fixes the value of Γ_A^Δ at $\chi=0$. A fixed value of T_2 for arbitrary χ would lead to the standard prediction that $\Gamma_A^\Delta\sim\chi(1/2\gamma T_2)^{1/2}$ for large values of χ. This behavior has been shown to be in disagreement with the observed data.[10] The experiments have shown that the saturated Γ_A^Δ behaves according to Eqs. (7.6).

For Rabi-frequency fluctuations $\Gamma_{22}=\Gamma_{33}$ and Γ_{23} are not equal to zero [see Eq. (5.3)] and as a result we have

$$\Gamma_A^\chi=\left[\gamma^2+\frac{a^2\tau_c\gamma}{1+\chi^2\tau_c^2}+\frac{\gamma\chi^2[1-a^2\tau_c^2/(1+\chi^2\tau_c^2)]^2}{2\gamma+a^2\tau_c/(1+\chi^2\tau_c^2)}\right]^{1/2} \quad (7.7)$$

with

$$\Gamma_A^\chi(\chi=0)=\gamma(\gamma+a^2\tau_c) , \quad (7.8)$$

where, because the impact of the noise is different on v than on u, no simple relation connecting Γ_A^χ at $\chi=0$ with $1/T_2$ exists.

Again, for very strong excitations we have

$$\Gamma_A^\chi\simeq\chi/\sqrt{2}$$

as in the case of frequency fluctuations. For very large powers the saturated line for detuning and amplitude fluctuations is noise free. This can be easily understood on the grounds that for $\chi>\gamma,\Delta$ a standard perturbation theory can be applied, treating the noise as a small correction to the dominant term due to χ.

For phase fluctuations only Γ_{11}, Γ_{33}, and Γ_{13} are not equal to zero [see Eq. (6.5)] and as a result we obtain the following expression for the absorption line shape:

$$\Gamma_A^\phi=\left[\left[\gamma+\frac{\chi^2\tau_c\sin^2a}{1+\chi^2\tau_c\cos^2a}\right]\left[\gamma+\frac{\chi^2}{2\gamma+\chi^2\tau_c\sin^2a}\right]\right]^{1/2} . \quad (7.9)$$

For $\chi=0$ this reduces to

$$\Gamma_A^\phi(\chi=0)=\gamma=1/T_2$$

and there is no impact of the noise on $1/T_2$. In the strong-field limit ($\sin a\neq0$ and $\cos a\neq0$) we have from Eq. (7.9) the limiting form

$$\Gamma_A^\phi=\{[\gamma+(1/\tau_c)\tan^2a][\gamma+(1/\tau_c)\sin^2a]\}^{1/2} . \quad (7.10)$$

Here, contrary to the two previously discussed cases, the saturated absorption line shape depends on the phase noise. This is due to the fact that, for high power, the phase fluctuations cannot be cast as a small perturbation to the coherent dynamics. As a result, phase fluctuations will have a persistent effect on any effective linewidth.[16(b)]

We note here that the familiar "phase-diffusion" or Gaussian Wiener-Levy model is based on frequency, not phase, fluctuations. This is important to keep in mind if connections are sought between results obtained here and results available in the literature for short-memory fluctuations (see, for example, Ref. 5).

B. FID experiments

Experiments by DeVoe and Brewer[10] were carried out on the Pr^{3+} optical resonance in samples of Pr^{3+}:LaF_3. The flip-flop motion of the F nuclear spins is expected to give a time-dependent magnetic field at each Pr^{3+} even at very low temperatures. We assume that this magnetic field creates a random fluctuation $\delta\Delta(t)$ in the resonance detuning of the ions. We shall assume that $\delta\Delta(t)$ performs a random telegraph motion with jump size a and switching rate $1/T$ to be determined later.

In the experimental situation two states of the Pr^{3+} ion, which make up a two-level system, interact resonantly with the incident laser field. The macroscopic polarization $P(t,\omega_0)$ due to such a two-level system is allowed to reach its steady-state value. Next the laser is effectively switched off at $t=0$ by detuning the resonance by Stark shifting the atomic frequency from ω_0 to $\omega_0+\delta\omega_S$.

The FID signal is proportional to the real part of $(u-iv)e^{-i\Delta t}$ averaged over the detunings associated with whatever inhomogeneous broadening is present. We assume the inhomogeneous linewidth is far larger than any other, as is typically the case for low-temperature solids. The sample polarization can be written

$$P_{\rm FID}(t)\sim e^{-i(\omega_L+\delta\omega_S)t}g(0)\times\int d\Delta\, e^{-i\Delta t}e^{-t/T_2}[u(\infty,\Delta)-iv(\infty,\Delta)]+\text{c.c.}\ , \tag{7.11}$$

where the field-free transverse lifetime T_2 is defined as follows [see Eq. (2.12)]:

$$1/T_2=\gamma+a^2\tau_c \tag{7.12}$$

and the steady-state values of the atomic dipoles $u(\infty,\Delta)$ and $v(\infty,\Delta)$ are given as solutions of the *exact* equation (3.9) with the damping matrix defined by Eqs. (4.2) and (4.3).

After some simple algebra we obtain from Eqs. (3.9) and (4.2) the result

$$u(\infty,\Delta)-iv(\infty,\Delta)=-2w_{\rm eq}\gamma\frac{\chi\Delta+i\chi(\gamma+\Gamma_{11})}{(\gamma+\Gamma_{11})(\gamma+\Gamma_{22})2\gamma+\chi^2(\gamma+\Gamma_{11})+2\gamma\Delta^2(1-\Gamma_{12})^2}\ , \tag{7.13}$$

where Γ_{11}, Γ_{22}, and Γ_{12} are defined by Eqs. (4.3). By performing the Fourier transform of this exact expression, we can obtain an FID rate R:

$$R=1/T_2+\Gamma_e\ , \tag{7.14}$$

where $1/T_2$ comes from the field-free contribution and Γ_e is totally determined by the Δ integration of $u(\infty,\Delta)-iv(\infty,\Delta)$.

From Eq. (7.13) it is quite clear that the atomic dipole moments can be written as the ratio of two rational functions of the atomic detuning

$$u(\infty,\Delta)-iv(\infty,\Delta)=\frac{N(\Delta)}{D(\Delta)}\ , \tag{7.15a}$$

$$D(\Delta)=d_6\Delta^6+d_4\Delta^4+d_2\Delta^2+d_0\ , \tag{7.15b}$$

where the coefficients can be evaluated at length from Eq. (7.1) if necessary. The rates R and Γ_e can be evaluated numerically by an integration over Δ.

In order to obtain an analytical expression for the FID signal we shall adopt a simplified approach based on the so-called single- (slowest-) rate approximation.[18,19] In this case the FID can be described by a single rate obtained from the linearization of the polynomial $D(\Delta)$:

$$D(\Delta)\simeq d_2\Delta^2+d_0\ . \tag{7.16}$$

This approximation of D by the smallest eigenvalue (the slowest rate) has been used and tested in several physical situations, but must usually be justified *a posteriori.*[19] Equation (7.17) allows a trivial calculation of the FID rate:

$$\Gamma_e=\left[\frac{d_0}{d_2}\right]^{1/2}\ . \tag{7.17}$$

After some simple algebra one can easily calculate the two coefficients d_0 and d_2 from Eq. (7.13). As a result we obtain

$$\Gamma_e=\left[\frac{f_1(2\gamma f_1+\chi^2 f_2)}{4\gamma^2\tau_c^2[(1+2\gamma\tau_c)/(1+\gamma\tau_c)]f_1+[(1+2\gamma\tau_c)/(1+\gamma\tau_c)]\chi^2\tau_c^2(f_1+\gamma f_2)+2\gamma f_3^2}\right]^{1/2}\ . \tag{7.18}$$

Here the functions f_1, f_2, and f_3 are given by

$$f_1=(1+2\gamma\tau_c)(\gamma^2\tau_c+\gamma+a^2\tau_c)+\gamma\tau_c^2\chi^2\ , \tag{7.19a}$$

$$f_2=(1+2\gamma\tau_c)(1+\gamma\tau_c)+\chi^2\tau_c^2+\frac{2\gamma\tau_c^3 a}{1+\gamma\tau_c}\ , \tag{7.19b}$$

$$f_3=(1+\gamma\tau_c)(1+2\gamma\tau_c)+\chi^2\tau_c^2-a^2\tau_c^2\frac{(1+2\gamma\tau_c)}{1+\gamma\tau_c}\ . \tag{7.19c}$$

It is now easy to check the limit of a "white-noise" telegraph, that is, to compute the value of the effective linewidth Γ_e when the telegraph correlation time scale τ_c is shorter than any other time scale. Thus we take $1/\tau_c\gg\chi,\gamma,\Gamma_e(\tau_c)$ and find

$$\Gamma_e\rightarrow(\gamma^2+\chi^2/2)^{1/2}\quad\text{as } \tau_c\rightarrow 0\ . \tag{7.20}$$

Clearly only radiative relaxation and power broadening remain in this limit, and at high powers, (7.20) is in agreement with observation.

Another short-coherence-time limit also exists. It is familiar in other physical problems as the white-noise diffusion limit. If $\tau_c\rightarrow 0$, while $a^2\tau_c$ is held fixed at the value D, then the telegraph tends to Gaussian white noise, and expression (7.18) takes the following form:

$$\Gamma_e\rightarrow[(\gamma+D)^2+\chi^2(\gamma+D)/2\gamma]^{1/2}\quad\text{as } \tau_c\rightarrow 0\ . \tag{7.21}$$

For high power of the laser the "white-noise" formula leads to the prediction that $\Gamma_e \to \chi[(\gamma+D)/2\gamma]^{1/2} \neq \chi/\sqrt{2}$. This limit does not agree with the experimental data obtained by DeVoe and Brewer, indicating directly that the noise fluctuations in the materials studied cannot be described by white-noise phase diffusion.

The physical reason for the distinction between these two white-noise limits (7.20) and (7.21) can be explained easily. The amount of dipole phase accumulated between two successive telegraph jumps goes rapidly to zero in the first case, being roughly equal to $a\tau_c$. But in the second case since $a^2=D/\tau_c$, the same single-jump phase accumulation is roughly equal to $(D\tau_c)^{1/2}$, which is much less sensitive to the limit, allowing a mean-square finite phase drift. A qualitative argument based on a Bloch-vector diagram given by Yodh *et al.*[11] is in complete agreement with this conclusion. They indicate clearly why dipole phase accumulation in optical resonance must be quenched at high powers.

In order to obtain quantitative comparisons of formula (7.18) with the experimental data we shall reexpress $a^2\tau_c$ by the zero-power linewidths following Eq. (2.12). Both T_2 and γ but not $a^2\tau_c$ can be measured in an independent way. For finite values of τ_c, the effective linewidths Γ_{ij} in the ORE are power dependent and as a result the FID rate is described by the full expression (7.18) with $a^2\tau_c$ fixed by T_2 and γ. In the actual FID experiment[10] on Pr^{3+}:LaF_3 at low temperatures, the following lifetimes have been reported: $T_2=21.7$ μsec and $T_1=10^{-3}$ sec.

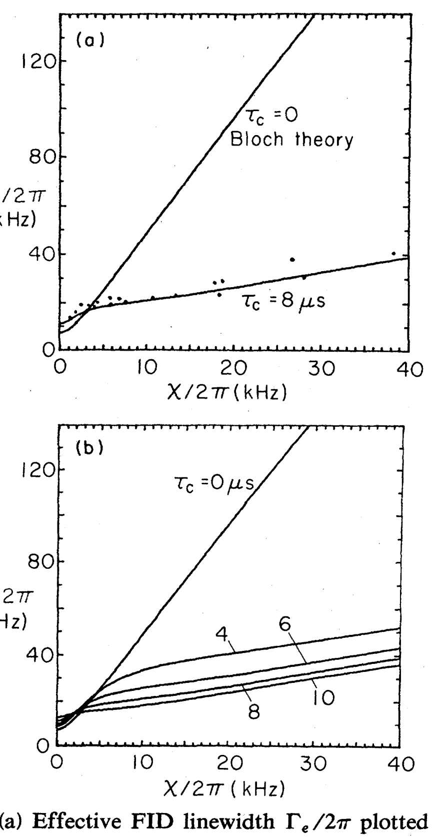

FIG. 1. (a) Effective FID linewidth $\Gamma_e/2\pi$ plotted for both the Bloch theory and for our random-frequency telegraph theory of optical resonance Eq. (7.18) as a function of Rabi frequency (in Hz). The free parameter τ_c in our theory has been fixed at the value 8 μsec. The data points are taken from Ref. 10. (b) Effective FID linewidth $\Gamma_e/2\pi$ predicted by Eq. (7.18) plotted as a function of Rabi frequency (in Hz) for several values of the free parameter τ_c, showing the relative stability of our telegraph theory near to the value 8 μsec shown in (a).

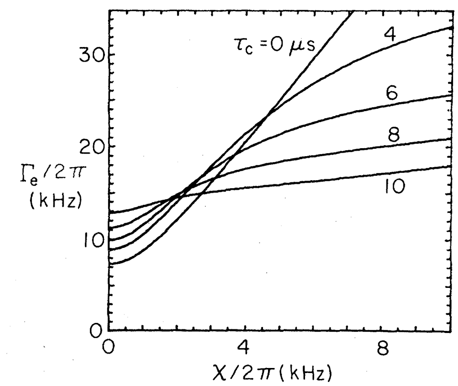

FIG. 2. Effective FID linewidth $\Gamma_e/2\pi$ predicted by Eq. (7.18) and shown in Fig. 1(b) replotted to clarify the τ_c dependence at low values of the Rabi frequency.

Figure 1(a) shows the FID rate $\Gamma_e/2\pi$ as a function of $\chi/2\pi$ for $\tau_c=0$ and $\tau_c=8$ μsec. The figure shows that for $\tau_c=0$ we have the conventional Bloch theory which disagrees with experiments for large values of χ. The curve with $\tau_c=8$ μsec shows good agreement with the experimental data for almost all values of $\chi/2\pi$. Figure 1(b) shows that the theory is stable. That is, the agreement with experiment is good for values in a neighborhood ($\pm 10\%$) of $\tau_c=8$ μsec.

Note the "kink" structure of $\Gamma_e/2\pi$ around $\chi/2\pi \sim 3$ kHz. We speculate that in this region the "identity" of the dominant FID rate is changing. That is, of the three solutions of the sixth-order polynomial in Eq. (7.15), which one happens to be the slowest rate may change as χ increases. A single rate approximation leading to the expression (7.18) still gives an excellent agreement with the observed data. Note that the kink occurs in the weak-field region of Fig. 1, which is expanded in Fig. 2. Recall the discussion of weak-field correlations at the end of Sec. II.

In closing this section, we note that the value of $\tau_c=8$ μsec has already been obtained from a partially numerical evaluation of the three rates obtained from an approximate second-cumulant solution of this optical resonance problem, using an Ornstein-Uhlenbeck model of the detuning fluctuations.[13(b)]

VIII. CONCLUSIONS

In this paper we have derived the effective optical resonance equations (ORE), with power-dependent lifetimes, for a two-level system subjected to jump-type fluctuations of the detuning or of the laser phase or amplitude. Our approach goes beyond the standard substitution rules. It shows the influence of the driving field on the transverse and longitudinal lifetimes whenever the external noise has a finite coherence time τ_c. We predict departures from the familiar Bloch-equation format as well, particularly in the nondiagonal form of the ORE relaxation matrix, for all types of fluctuation considered.

We have applied the theory to explain qualitatively and quantitatively recent FID experiments of ion impurities in solids. We can contrast our results with several other theoretical predictions made recently as a response to the DeVoe-Brewer experiments by focusing attention on the values chosen in those theories for τ_c, the single fitting parameter in recent theoretical discussions. A purely numerical, but microscopically modeled, Monte Carlo calculation,[15] including a large number of neighboring interacting spins, leads to the selection of $\tau_c \simeq 70$ μsec, which seems too high (because then $\tau_c > T_2$). Numerical investigation of the full hierarchy of equations[14] associated with an Ornstein-Uhlenbeck model of fluctuations leads to a value of $\tau_c \simeq 27$ μsec, which again seems high. An analytic theory which includes numerical evaluation of the inhomogeneous broadening integration, developed in Ref. 15, leads to $\tau_c \simeq 20$ μsec, which is also unsatisfactory. Only the second-cumulant approximate analytic theory of Ref. 13(b) shares the property of our solution that τ_c can be chosen significantly smaller than T_2, and still obtain good agreement with the experimental data. Interestingly, the theories of Ref. 13 also make our prediction that the relaxation matrix is nondiagonal, and thus departs from the Bloch form, but they do not find the same off-diagonal elements to be nonzero as given here. The solution of Ref. 13(b) is restricted to weak noise.

Compared to most of these theories, our RTS approach to detuning fluctuations is very simple. The model is exactly soluble in steady state, leading to exact expressions for the power-dependent effective lifetimes. The principal approximation involved in our calculations has been the slowest-rate approximation.[18,19] It is remarkable that such a very simple theory leads to good agreement with the experimentally observed data and also predicts a quite sensible value for τ_c. The supersimplified version of the theory explained in the first part of Sec. VII leads, however, to a value of τ_c which is too high (in this case $\tau_c \simeq 25$ μsec).

From all the theoretical discussions given, beginning with that of Redfield,[6] it is easy to see that there is no basic problem to fit very accurately the data at very high values of χ. The situation is actually more complicated for small or moderate values of χ. All the theoretical calculations (with the exception of the simplified theories which neglect detuning in the expressions for Σ), including ours, indicate that the value of Γ_e at $\chi=0$ is different from $1/T_2$ and this difference is dependent on the value of τ_c. This indicates that the relation of a and τ_c to T_2 can be more complicated than the one suggested by Eq. (7.12) and similar relations obtained in other papers. Our discussion of the linearized theory in Sec. II illustrates some of the possibilities. In order to clarify this point, more experiments with a careful measurement of the shape of the FID signal at low power would be most useful.

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*Permanent address.

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