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Optical hole burning with finite excitation time

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This paper extends earlier calculations [M. Yamanoi and J. H. Eberly, Phys. Rev. A **34**, 1609 (1986)] of hole burning using continuous excitation to the finite excitation time used in experiments on $Pr:LaF_3$. Both Gaussian-Markov (GM) and random-telegraph (RT) dephasing models are studied. For the GM model, a new feature is the appearance of nutation structure near the center of the hole for a 400-μsec excitation time. In contrast for RT, a similar, although much weaker, structure appears in the wings of the hole. It is noted that for finite excitation times, a *detuning* as well as the usual intensity dependence of the dephasing time appears.

INTRODUCTION

The failure of the conventional optical Bloch equations[1] (OBE) to describe measurements[2] of saturation in a low-temperature solid has led to many theoretical studies.[3–10] These studies have used various models of atomic frequency fluctuations that lead to dephasing and have concentrated on a time-domain description of saturation, i.e., free-induction decay (FID). So far, however, there is no clear unanimity concerning which model, if any, best describes the data (see, e.g., discussion by Berman[10]). It has recently been suggested[11] that studies in the frequency domain may be useful in clarifying the correspondence between the various theories and experiment, and some hole-shape calculations have been presented. In these calculations, as well as in earlier FID work, an infinite excitation period was assumed. Experimentally, however, a finite excitation period is necessary because of optical-pumping effects and the limited time during which a narrow laser linewidth (< 1 kHz) can be maintained.

In this paper we present calculations on the effects of a finite excitation time on hole burning for both Gaussian-Markov[3–8] (GM) and random-telegraph[9] (RT) dephasing models. A principal new result is that in addition to the usual intensity dependence often noted for the various models, a detuning dependence appears for finite excitation times. In particular, for GM, a nutation structure appears near the center of the hole and persists for excitation times as long as 400 μsec in spite of a much shorter (21.7 μsec) dephasing time.

THEORY

The general form of the reduced[8] OBE is given by the matrix equation

$$d\underline{\beta}/dt=\underline{M}\,\underline{\beta}+\underline{L}\ , \tag{1}$$

where $\underline{\beta}$ is the Bloch vector expressed as a three-component column vector, $\beta(1)=u$, $\beta(2)=v$, $\beta(3)=w$, using the usual notation, and $L(1)=L(2)=0$, $L(3)=2\gamma w_{\rm eq}$. We use the equilibrium value $w_{\rm eq}=-1$ and assume a closed two-level system so that $2\gamma=1/T_1$, where T_1 is the upper-state lifetime. The 3×3 matrix $\underline{M}$ can be written as

$$\underline{M}=-\begin{bmatrix}\gamma & 0 & 0\\ 0 & \gamma & 0\\ 0 & 0 & 2\gamma\end{bmatrix}+\begin{bmatrix}0 & -\Delta & 0\\ \Delta & 0 & \Omega\\ 0 & -\Omega & 0\end{bmatrix}-\begin{bmatrix}\Gamma_{11} & \Gamma_{12} & \Gamma_{13}\\ \Gamma_{21} & \Gamma_{22} & \Gamma_{23}\\ 0 & 0 & 0\end{bmatrix}, \tag{2}$$

where the first matrix accounts for lifetime damping, the second is the coherent driving matrix, and the third is a generalized damping matrix.

In the calculation we consider three cases: (1) the conventional OBE for which $\Gamma_{12}=\Gamma_{21}=\Gamma_{13}=\Gamma_{23}=0$ and $\Gamma_{11}=\Gamma_{22}=T_2^{-1}-\gamma$; (2) the Gaussian-Markov dephasing model, where for $\gamma'\tau_c \ll 1$,

$$\begin{aligned}&\Gamma_{11}=\Gamma_{22}=\gamma'[1+(\Delta\tau_c)^2]/[1+(\Omega'\tau_c)^2]\ ,\\ &\Gamma_{13}=\gamma'\Omega\Delta\tau_c^2/[1+(\Omega'\tau_c)^2]\ ,\\ &\Gamma_{23}=-\gamma'\Omega\tau_c/[1+(\Omega'\tau_c)^2]\ ,\\ &\Gamma_{12}=\Gamma_{21}=0\ ,\end{aligned} \tag{3}$$

where $\gamma'=(\delta\omega)^2\tau_c$, $\Omega'=(\Delta^2+\Omega^2)^{1/2}$, $\Omega/2\pi$ is the Rabi frequency, $\Delta/2\pi$ is the detuning frequency, and $\delta\omega$ and τ_c are parameters of the frequency fluctuation model described in Ref. 4 and equivalently in Ref. 8; and (3) random-telegraph dephasing model where

$$\begin{aligned}&\Gamma_{11}=a^2\frac{(1/\tau_c+\gamma)(1/\tau_c+2\gamma)}{P}\ ,\\ &\Gamma_{22}=a^2\frac{(1/\tau_c+\gamma)(1/\tau_c+2\gamma)+\Omega^2}{P}\ ,\\ &\Gamma_{12}=-\Gamma_{21}=-a^2\frac{\Delta(1/\tau_c+2\gamma)}{P}\ ,\\ &\Gamma_{13}=\Gamma_{23}=0\ ,\end{aligned} \tag{4}$$

and

$$P=[(1/\tau_c+\gamma)^2+\Delta^2](1/\tau_c+2\gamma)+\Omega^2(1/\tau_c+\gamma) .$$

If we limit our consideration only to the values of parameters which lead to nearly exponential decay (i.e., $a\tau_c \ll 1$),[9] then the parameter a is related to the dephasing time T_2 obtained from photon echoes by

$$T_2^{-1}=\gamma+a^2\tau_c . \tag{5}$$

The form of the solution to Eq. (1) depends on the roots of the characteristic equation (CE) for $\underline{M}$; $\det(\underline{M}+\lambda\underline{I})=0$, where $\underline{I}$ is the unit matrix. There are two cases to consider.

Case 1: CE roots a and $b\pm is$

In the case of one real and two complex-conjugate roots, the solution for $\underline{\beta}$ is of the form

$$\underline{\beta}(\Delta,t)=\underline{A}\exp(-at)+\exp(-bt)\times\left[\underline{B}\cos(st)+\underline{C}\left[\frac{\sin st}{s}\right]\right]+\underline{\beta}_\infty , \tag{6}$$

where $\underline{A}$, $\underline{B}$, $\underline{C}$, and $\underline{\beta}_\infty$ are three-component column vectors given by

$$\begin{aligned}\underline{A}&=[(a-b)^2+s^2]^{-1}[(b^2+s^2)\underline{E}+2b\underline{F}+\underline{G}] ,\\ \underline{B}&=\underline{E}-\underline{A} ,\\ \underline{C}&=a\underline{A}+b\underline{B}+\underline{F} ,\\ \underline{\beta}_\infty&=\underline{M}^{-1}\underline{L} ,\end{aligned} \tag{7}$$

where $\underline{\beta}_\infty$ is the value of the Bloch vector with cw excitation and $\underline{E}=\underline{\beta}_0-\underline{\beta}_\infty$, where $\underline{\beta}_0$ is the initial value of $\underline{\beta}$. Also, $\underline{F}=\underline{M}\,\underline{\beta}_0+\underline{L}$ and $\underline{G}=\underline{M}\,\underline{F}$. Written in this form, the coefficients may then be easily evaluated by a computer.

Case 2: CE roots a, b, and c

In the case of three real roots,

$$\underline{\beta}(\Delta,t)=\underline{A}\exp(-at)+\underline{B}\exp(-bt)+\underline{C}\exp(-ct)+\underline{\beta}_\infty ,$$

where

$$\begin{aligned}\underline{A}&=[(b-c)/D][bc\underline{E}+(b+c)\underline{F}+\underline{G}] ,\\ \underline{B}&=[(c-a)/D][ca\underline{E}+(c+a)\underline{F}+\underline{G}] ,\\ \underline{C}&=\underline{E}-\underline{A}-\underline{B} ,\\ D&=ab(a-b)+bc(b-c)+ca(c-a) .\end{aligned} \tag{8}$$

It may be readily verified that the vectors $\underline{A}$, $\underline{B}$, and $\underline{C}$ for cases 1 and 2 become equivalent as $s\to 0$ and $c\to b$.

RESULTS AND DISCUSSION

We present calculations using parameters appropriate to the DeVoe-Brewer experiment (Pr:LaF$_3$, $T_1=500$ μsec, $T_2=21.7$ μsec). Also, for comparison with earlier[11] cw hole-burning calculations (GM model), we choose a correlation time $\tau_c=9$ μsec.

Figure 1 shows plots of the normalized population $h(\Delta)=[1+w(\Delta)]/[1+w(0)]-0.5$ for the GM model using the relations $1/T_1=2\gamma$ and $1/T_2=\gamma+\gamma'$. All of the plots show four curves, C_{cw} is the conventional OBE with cw excitation, C_p is the conventional OBE with 400-μsec pulse excitation, and similary M_{cw} and M_p for the modified OBE. For Rabi frequencies up to 10 kHz, it is evident that, aside from an expected slight decrease of the holewidth, the pulsed and cw results are similar. However, at $\Omega/2\pi=30$ kHz a new feature is the appearance of nutation structure near the center of the hole (curve M_p). This structure becomes more prominent as $\Omega/2\pi$ increases, as shown in Fig. 2, where, for clarity, we plot directly the Bloch vector $w(\Delta)$. At first sight this is a surprising result since the 400-μsec pulse is about 20 times longer than the dephasing time of 21.7 μsec.

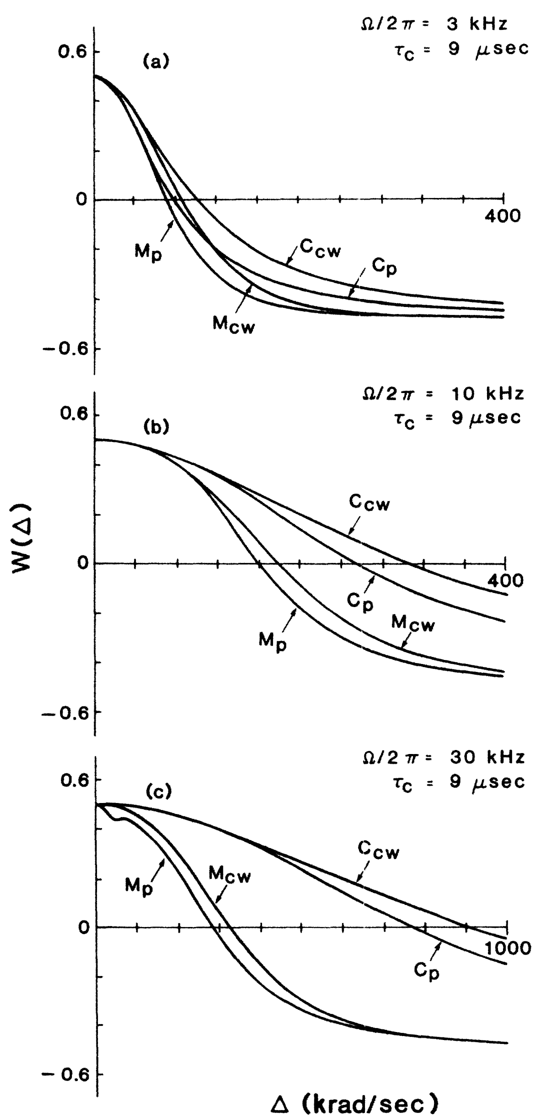

FIG. 1. Normalized inversion $h(\Delta)$ vs detuning Δ for the Gaussian-Markov dephasing model. A correlation time $\tau_c=9$ μsec is assumed. See text for curve descriptions.

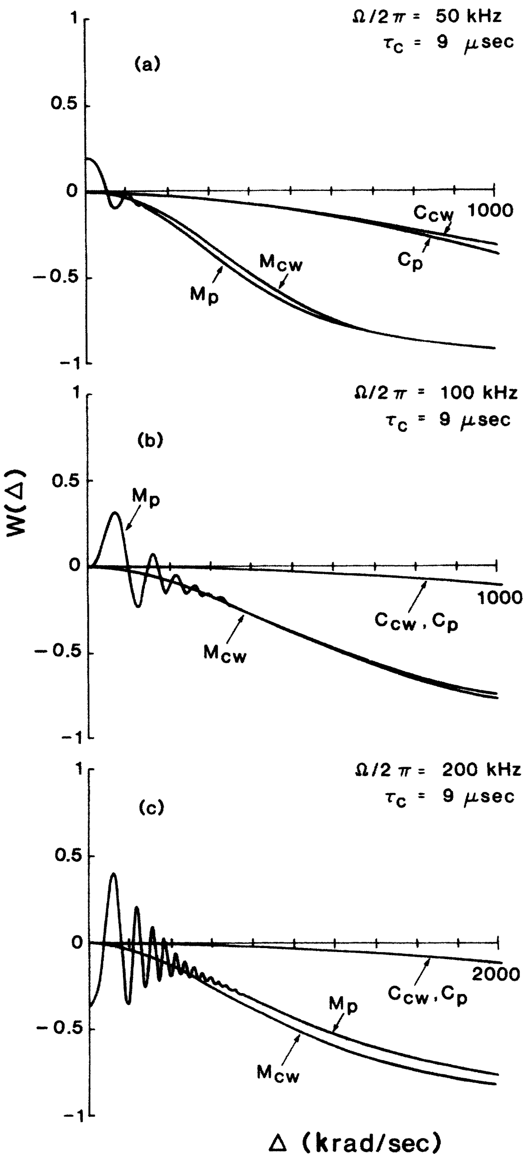

FIG. 2. Unnormalized inversion $w(\Delta)$ vs detuning Δ for the Gaussian-Markov dephasing model at high Rabi frequencies. Correlation time $\tau_c = 9$ μsec.

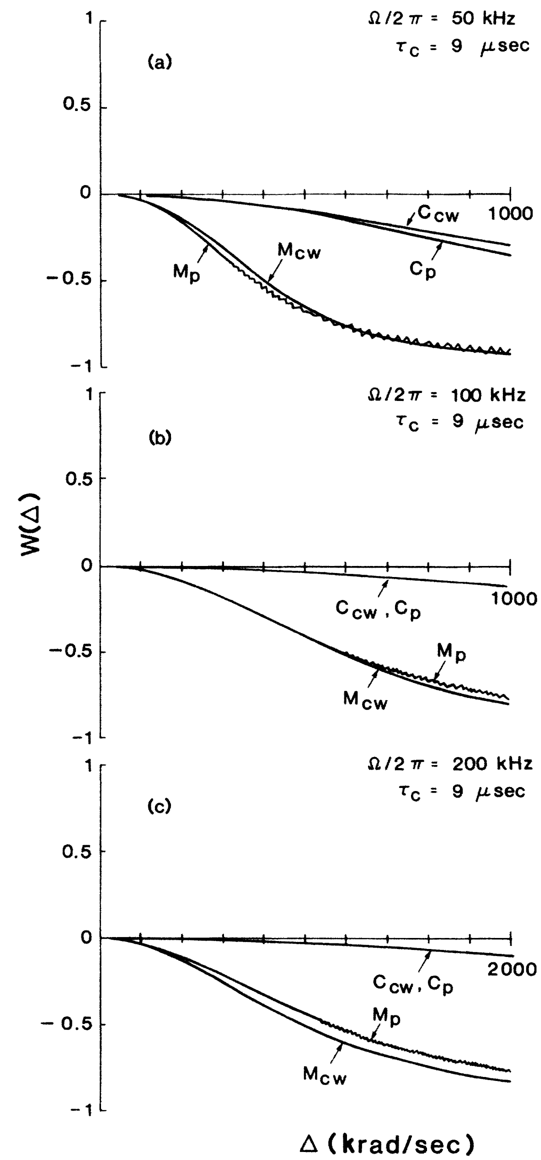

FIG. 3. Unnormalized inversion $w(\Delta)$ vs detuning Δ for the random-telegraph dephasing model at high Rabi frequencies. Correlation time $\tau_c = 9$ μsec.

Indeed, as expected, no such structure appears for the conventional OBE holes. The reason is that, from Eq. (3), the effective dephasing time Γ_{11}^{-1} depends both on the detuning Δ as well as the usual light intensity. In particular, Γ_{11}^{-1} is longest near the center of the hole. For the cw case (curves M_{cw}), the nutation structure is absent, since the nutation must decay eventually, given enough time.

It is of interest to compare these results with those using other dephasing models. We show in Fig. 3 calculations for the RT model similar to those of Fig. 2. It is evident that the pulsed hole shapes are quite different from those of the GM model. In particular, nutation structure is now absent near the center of the hole, and a very weak fine-structured nutation appears in the wings, as expected from Eq. (4).

CONCLUSIONS

While various theoretical treatments appear to successfully describe the optical saturation behavior in the DeVoe-Brewer[2] experiment (using a range of correlation times $\tau_c = 5-37$ μsec), Berman [10] as well as Javanainen[5] have noted that these descriptions are invalid either because of a violation of the range of τ_c for which the theory is valid and/or because the theories predict nonexponential decay, a result that disagrees with the observed exponential decay of both FID and photon echoes. It appears that the random-telegraph theory (with $\tau_c = 8$ $\mu\text{sec} < T_2 = 21.7$ μsec) comes closest to explaining all the data in a consistent manner. However, more complex formulations[10] of dephasing which cannot be simply expressed as modified Bloch equations remain to be numerically investigated.

It is evident that additional experiments using both frequency and time-domain techniques would be useful to help identify which of the various dephasing models (if any) consistently describe optical saturation behavior in low-temperature solids. In particular, this paper demonstrates that there are striking differences in hole-burning line shapes for cw versus finite excitation times. In the latter case, the holes display a DETUNING as well as the usual intensity dependence of the effective dephasing time, the nature of which is strongly model dependent.

Finally, as noted by Schenzle *et al.*,[7] studies of nutation or rotary echoes[12,13] should provide a further powerful test of the various models which in general predict an intensity-dependent echo decay time. Experiments using an ultranarrow ($\sim$ 1-kHz linewidth) dye laser are presently in progress and will be reported on later.

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