Optical linewidths and photon-echo decays of impurities in glasses

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Using a stochastic model, we calculate the temperature dependence of the homogeneous linewidth and the two-pulse photon-echo decay rate of optically active impurities in glasses. The theory applies to the low-temperature regime $T \lesssim 10$ K, where it is assumed that the dephasing arises from an elastic dipole interaction of the impurity with an array of thermally excited tunneling systems. Averaging over the tunneling parameters and using relaxation rates appropriate to vitreous silica, we find that the photon-echo decay rate varies as $T^{1.1}$ and the homogeneous linewidth as $T^{1.2}$ for $0.1 \leq T \leq 1$ K, assuming a constant density of states for the tunneling systems. Both the photon-echo amplitude and the optical dipole-moment correlation function decay exponentially in time. The rates, however, are different, with a ratio of photon-echo dephasing time to optical-absorption dephasing time of 6.9 at $T = 1$ K, assuming a constant density of states. The relation of our results to various experimental studies is discussed.

I. INTRODUCTION

In a recent paper[1] one of us outlined a stochastic model for optical dephasing in glasses. The model, which is appropriate to the low-temperature regime $T \leq 10$ K, is based on the assumption that the dephasing arises from the interaction of the optically active impurity with an array of thermally excited tunneling systems. In Ref. 1 formal results were presented for both the optical dipole moment correlation function, which characterizes optical absorption and emission, and the two-pulse photon-echo amplitude. A detailed analysis was given for the high-temperature limit, $kT \gg$ energy splitting of the tunneling system, assuming all systems had the same relaxation rate. In this paper, we apply the theory to a more realistic situation in which we allow for a distribution of relaxation rates and do not make the high-temperature approximation. Detailed results are given for Nd^{3+} in vitreous silica where we make contact with prior investigations.[2–4]

One of the surprising features to emerge from the calculations is that the dephasing time characterizing the decay of the photon echo differs by a factor of 7–10 from the dephasing time inferred from the homogeneous linewidth. We comment on the result in relation to recent comparative studies of nonphotochemical hole burning and photon-echo decay in organic glasses.[5]

The remainder of this paper is divided into three sections. In Sec. II we review the formal aspects of the model; the results of numerical calculations are presented in Sec. III, while Sec. IV is devoted to a discussion of our findings and their relation to the experimental studies cited above. We also make a brief comparison between our work and previous theoretical treatments of the problem.

II. MODEL

The model outlined in Ref. 1 is based on the hypothesis that the dephasing arises from a modulation of the optical transition frequency brought about by the interaction of the optically active impurity with an array of thermally excited tunneling systems. The analysis of Ref. 3 shows that the exponential time dependence of the photon echo and the near-linear temperature dependence of the decay rate is an indication that the dominant coupling between the impurity and the tunneling systems varies as r^{-3}, i.e., a dipole-dipole interaction. The order-of-magnitude calculations in Ref. 3 lead one to infer that in the case of Nd^{3+} (and probably other rare earth and transition metal impurities, as well) it is the *elastic* dipole interaction rather than the *electric* dipole interaction which is dominant.

In Ref. 1 it was pointed out that the stochastic model is based on a physical picture for the dephasing similar to that underlying the model calculations of Hu and Hartmann[6] and of Hu and Walker.[7] Although our analysis differs in detail from that in both Refs. 6 and 7, we obtain identical results in the high-temperature limit and qualitative agreement with Ref. 7 at finite temperatures.

Apart from inhomogeneous effects, the optical absorption line shape $I(\omega)$ is given by

$$I(\omega) = \int_0^\infty e^{-i\omega t} C(t) dt \ , \tag{1}$$

where $C(t)$ is the optical dipole-moment correlation function, and Re denotes real part. In the theory of Ref. 1 the dipole-moment correlation function is written

$$C(t) = \left\langle\left\langle\left\langle e^{i\omega_0 t} \prod_v \exp\left[i \int_0^t \Delta\omega_v(t') dt' \right]\right\rangle\right\rangle\right\rangle \ . \tag{2}$$

Here ω_0 is the bare transition frequency, and $\Delta\omega_v(t)$ is the fluctuation in frequency coming from the interaction between the impurity and the vth tunneling system. This we take to be of the form

$$\Delta\omega_v = (A / r_v^3) s_v(t) \ , \tag{3}$$

where $s_v(t)$ is a random variable taking on the values ± 1 with probabilities consistent with the Boltzmann factors

for the two levels of the tunneling system.

The triple brackets in Eq. (2) designate three averages: a configurational average over the spatial distribution of tunneling systems, which are assumed to be distributed at random, an ensemble average of exponential of the integral of $\Delta\omega_v(t)$, and an average over the energy splittings and relaxation rates of the tunneling systems. (The latter average was omitted in Ref. 1 since all tunneling systems were assumed to be identical.) As discussed in Ref. 6, the configurational average takes the form

$$C(t)=e^{i\omega_0 t}\exp\left\{-N_0\int d\mathbf{r}_v\left\langle\left[1-\left\langle\exp\left[i\int_0^t\Delta\omega_v(t')dt'\right]\right\rangle\right]\right\rangle\right\}, \tag{4}$$

where N_0 is the spatial density of thermally excited tunneling systems.

As shown in Ref. 1, the ensemble average of the exponential can be written

$$\left\langle\exp\left[i\int_0^t\Delta\omega_v(t')dt'\right]\right\rangle=e^{-Rt}\mathbf{P}\cdot[\cos(Yt)\mathbb{1}+i\hat{\mathbf{n}}\cdot\boldsymbol{\sigma}\sin(Yt)]\mathbf{U}, \tag{5}$$

where $\mathbb{1}$ denotes the unit matrix, $\boldsymbol{\sigma}=(\sigma_x,\sigma_y,\sigma_z)$ is a vector whose components are the Pauli spin matrices; $\mathbf{P}$ is a row vector with components equal to the occupation probabilities of the two levels of the vth tunneling system, and $\mathbf{U}$ is a column vector with both components equal to unity. The parameters R and Y and the unit vector $\hat{\mathbf{n}}$ are expressed in terms of the relaxation rates $+\rightarrow-$ and $-\rightarrow+$ of the upper and lower levels of the tunneling system through the equations

$$R=\tfrac{1}{2}(R_{+-}+R_{-+}), \tag{6}$$

$$Y=[\epsilon^2-R^2+(A/r_v^3+i\epsilon)^2]^{1/2}, \tag{7}$$

and

$$\hat{\mathbf{n}}=(-iR,\ \epsilon,\ A/r_v^3+i\epsilon)/Y, \tag{8}$$

with

$$\epsilon=\tfrac{1}{2}(R_{+-}-R_{-+}). \tag{9}$$

The decay of the two-pulse photon echo can also be modeled following the approach outlined above. The amplitude of the echo at time $2t$ following a π pulse at time t is given by

$$E(2t)=\left\langle\left\langle\left\langle\prod_v\exp\left[i\int_0^t\Delta\omega_v(t')dt'-i\int_t^{2t}\Delta\omega_v(t')dt'\right]\right\rangle\right\rangle\right\rangle. \tag{10}$$

The ensemble average over the exponential factor takes the form

$$\left\langle\exp\left[i\int_0^t\Delta\omega_v(t')dt'-i\int_t^{2t}\Delta\omega_v(t')dt'\right]\right\rangle=\mathbf{P}\cdot\exp\{-[R\mathbb{1}-R\sigma_x-i\epsilon\sigma_y-(iA/r_v^3-\epsilon)\sigma_z]t\}$$

$$\times\exp\{-[R\mathbb{1}-R\sigma_x-i\epsilon\sigma_y+(iA/r_v^3+\epsilon)\sigma_z]t\}\cdot\mathbf{U}, \tag{11}$$

where the symbols have the same meaning as before.

The evaluation of the rhs of (11) is facilitated by the identity

$$\exp(ix\mathbb{1}+iy\hat{\mathbf{m}}\cdot\boldsymbol{\sigma})=e^{ix}(\mathbb{1}\cos y+i\hat{\mathbf{m}}\cdot\boldsymbol{\sigma}\sin y), \tag{12}$$

which is valid for arbitrary x,y, and unit vector $\hat{\mathbf{m}}$ ($\hat{\mathbf{m}}\cdot\hat{\mathbf{m}}=1$), along with the property $\sigma_\alpha^2=1$, $\alpha=x,y,z$, and the commutation relations

$$\boldsymbol{\sigma}\times\boldsymbol{\sigma}=2i\boldsymbol{\sigma}. \tag{13}$$

After using (12) and carrying out the matrix multiplication the right-hand side of (11) takes the form

$$\mathbf{P}\cdot\exp(\cdots)\exp(\cdots)\mathbf{U}=e^{-2Rt}\mathbf{P}\cdot[\mathbb{1}\cos(Yt)\cos(Y't)+i\mathbf{n}\cdot\boldsymbol{\sigma}\sin(Yt)\cos(Y't)$$

$$+i\hat{\mathbf{n}}'\cdot\boldsymbol{\sigma}\sin(Y't)\cos(Yt)+\hat{\mathbf{n}}\cdot\hat{\mathbf{n}}'\sin(Yt)\sin(Y't)$$

$$+i\hat{\mathbf{n}}\times\hat{\mathbf{n}}'\cdot\boldsymbol{\sigma}\sin(Yt)\sin(Y't)]\mathbf{U}, \tag{14}$$

where Y and $\hat{\mathbf{n}}$ are given by Eqs. (7) and (8) and Y' and $\hat{\mathbf{n}}'$ by similar equations with A/r_v^3 replaced by $-A/r_v^3$.

III. NUMERICAL RESULTS

Having dealt with the ensemble and configurational averages in Sec. II we are left with the averaging over the energy splittings and relaxation rates of the tunneling systems. The former appear implicitly in the expressions for the relaxation rates and explicitly in the components of the vector $\mathbf{P}$ (cf. Sec. II)

$$\mathbf{P}=(1/(1+e^{\beta E}),\ e^{\beta E}/(1+e^{\beta E}))\ , \tag{15}$$

where E is the splitting and $\beta=1/kT$. The relaxation of the tunneling system occurs through the absorption and emission of phonons.[8] With this mechanism the two rates take the form

$$R_{+-}=WE^3\,|\langle +|V|-\rangle|^2 e^{\beta E}(e^{\beta E}-1)^{-1}\ , \tag{16}$$

and

$$R_{-+}=WE^3\,|\langle +|V|-\rangle|^2 (e^{\beta E}-1)^{-1}\ . \tag{17}$$

Here W is a constant and $\langle +|V|-\rangle$ is the matrix element of the deformation potential characterizing the interaction of the tunneling system with the phonons.[9]

In addition to averaging over E one must also average $|\langle +|V|-\rangle|^2$. The variation in the matrix elements arises primarily through the tunneling parameter traditionally denoted by λ. As a first approximation we will neglect all other variation and take[9]

$$|\langle +|V|-\rangle|^2=Be^{-2(\lambda-\lambda_{\min})}\ , \tag{18}$$

where B, the maximum value of $|\langle +|V|-\rangle|^2$, is assumed to be the same for all systems. We also make the standard assumption that λ is distributed approximately uniformly over the interval $\lambda_{\min}\le\lambda\le\lambda_{\max}$.[10,11] In addition, we neglect any correlation between the matrix element and the energy and average over the two variables separately.

After making these approximations, the averaging over the parameters of the tunneling system takes the form

$$\left\langle\left[1-\left\langle\exp\left[i\int_0^t\Delta\omega_v(t')dt'\right]\right\rangle\right]\right\rangle=N_0^{-1}(\lambda_{\max}-\lambda_{\min})^{-1}\int_0^\infty\rho(E)dE\int_{\lambda_{\min}}^{\lambda_{\max}}d\lambda\left[1-\left\langle\exp\left[i\int_0^t\Delta\omega_v(t')dt'\right]\right\rangle\right]\ , \tag{19}$$

for the optical dipole-moment correlation function, with an equivalent result for the photon echo. Here $\rho(E)$ is the density of states of the tunneling systems, and the angular brackets denote the ensemble average. Although the integral over E extends to infinity, only the thermally excited systems contribute to the dephasing.

As noted in the Introduction we are particularly interested in making contact with the measurements of the echo decay associated with the $^4F_{3/2}(1)$ level of Nd^{3+} in vitreous silica. In this system the relaxation rates are known from ultrasonic studies[3,4] so that the only adjustable parameters are $\lambda_{\min}$, $\lambda_{\max}$, and an overall multiplicative constant involving the product of the density of states and the parameter A characterizing the interaction between the Nd ion and the tunneling systems. With a uniform distribution of λ our results depend only on $\lambda_{\max}-\lambda_{\min}$, which we have set equal to 5.5. Further increases in $\lambda_{\max}-\lambda_{\min}$ produce no change in the temperature dependence of the dephasing rates. (Note that with $\lambda_{\max}-\lambda_{\min}=5.5$, $|\langle +|V|-\rangle|^2$ varies by a factor of 6×10^4.)

As will be shown in the next section, we obtain asymptotic exponential decay of the photon echo, $E(2t)\sim\exp(-2t/{}^{\rm PE}T_2)$, from which we can extract a dephasing time ${}^{\rm PE}T_2$. In order to make a quantitative comparison with the data of Ref. 2, we fix the overall multiplicative constant characterizing the theoretical dephasing rate so as to obtain agreement between theory and experiment at 1 K.

In Fig. 1 we display our results for ${}^{\rm PE}T_2$ making the standard assumption of a constant density of states for the tunneling systems.[10,11] From the curve it is found that $(\pi\,{}^{\rm PE}T_2)^{-1}$ varies as $T^{1.1}$ over the interval $0.1\le T\le1$ K and as $T^{1.0}$ over the interval $1\le T\le10$ K.

Having fixed the multiplicative constant to give agreement with the measured photon-echo decay rate at 1 K, one can use this value to make a prediction for the dephasing time characterizing the optical absorption ${}^{\rm OA}T_2$. The latter is inferred from the asymptotic decay of the optical dipole moment correlation function $C(t)\sim\exp(-1/{}^{\rm OA}T_2)$. Our results for ${}^{\rm OA}T_2$ are shown in Fig. 2, again for a constant density of states. From this figure we infer that $(\pi\,{}^{\rm OA}T_2)^{-1}$ varies as $T^{1.2}$ in the interval $0.1\le T\le1$ K and as $T^{1.1}$ in the interval $1\le T\le10$ K.

Although the temperature dependences of the two dephasing times are similar, there are significant differences in overall magnitude. This is shown in Table I where we list values of ${}^{\rm PE}T_2/{}^{\rm OA}T_2$ obtained by making various assumptions about the density of states. With a constant density of states the ratio varies between 6.8 and 7.6 over the interval $0.1\le T\le10$ K. These results will be discussed in the next section along with the corresponding results obtained with energy-dependent densities of states.

IV. DISCUSSION

The results presented in Sec. III were obtained assuming a constant density of states for the tunneling systems and averaging the square of the matrix element over the tunneling parameter λ. The average over λ is an essential step in the calculation. As can be seen from Fig. 3, where we plot results for the echo amplitude obtained with and without the λ average, only by averaging over λ do we obtain the asymptotic exponential decay that is seen experimentally.[2,4,5]

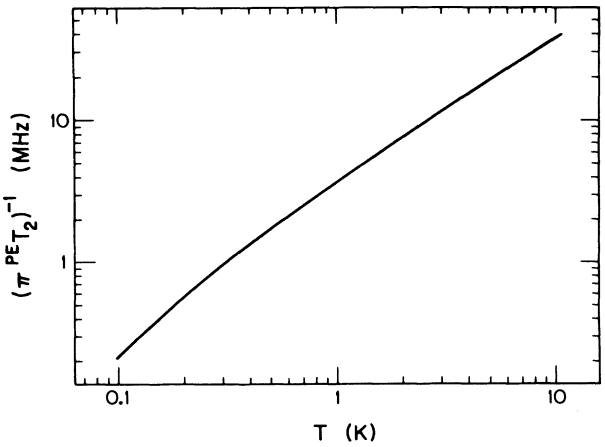

FIG. 1. $(\pi\,^{PE}T_2)^{-1}$ vs T. $^{PE}T_2$ is the dephasing time inferred from photon-echo decay $E(2t)\sim\exp(-2t/^{PE}T_2)$. Constant density of states for the tunneling systems. Parameters have been chosen to give agreement at 1 K with the experimental results reported in Ref. 2.

It was pointed out in Ref. 1 that one can separate the contributions to the correlation functions coming from interactions with "nearby" and distant tunneling systems, where "nearby" systems are ones for which $A/r_v^3 \geq R$. It was found that for times $Rt \gtrsim 5$ the decay of both the optical dipole-moment correlation function and the photon-echo amplitude was dominated by the contribution arising from distant tunneling systems, where $A/r_v^3 \ll R$. This result is relevant to the applicability of the stochastic model and continuum approximation on which our approach is based. Generally speaking, both the model and the approximation are not appropriate for tunneling systems which are located close to the optically active impurities. However, interactions with systems where $A/r_v^3 \gg R$ affect only the early part of the decay; the asymptotic behavior, $Rt \gg 1$, arises from interactions with distant systems.

TABLE I. Ratios of photon-echo dephasing time to optical-absorption dephasing time.

T (K)	R^a	R^b	R^c
0.1	6.9	7.1	7.8
0.2	6.8	7.1	7.6
0.4	6.8	7.5	8.2
0.6	6.8	7.7	8.3
0.8	6.8	7.5	8.1
1.0	6.9	7.5	8.1
2.0	6.9	7.7	8.2
3.0	7.1	7.8	8.5
4.0	7.2	7.9	8.6
5.0	7.3	8.0	8.8
6.0	7.3	8.1	8.9
7.0	7.3	8.2	9.2
8.0	7.6	8.3	9.4
9.0	7.6	8.5	9.6
10.0	7.6	8.6	9.7

[a] $^{PE}T_2/^{OA}T_2$ calculated with a constant density of states for the tunneling systems.
[b] $^{PE}T_2/^{OA}T_2$ calculated with a density of states varying as $E^{0.15}$.
[c] $^{PE}T_2/^{OA}T_2$ calculated with a density of states varying as $E^{0.3}$.

As noted, the temperature dependences of the dephasing rates reported in Sec. III pertain to a system with a constant density of states. Since there is evidence from specific heat studies that the density of states in vitreous silica may vary as $E^{0.3}$,[12] we have repeated the calculations with $\rho(E)\sim E^{0.3}$. For the photon echo decay we find that $(\pi\,^{PE}T_2)^{-1}$ varies as $T^{1.5}$ over the interval $0.1 \leq T \leq 1$ K and as $T^{1.3}$ over the interval $1 \leq T \leq 10$ K. In the case of the optical absorption we find $(\pi\,^{OA}T_2)^{-1}$ varying as $T^{1.5}$ over the interval $0.1 \leq T \leq 1$ K and as $T^{1.4}$ over the interval $1 \leq T \leq 10$ K.

The experimental results for the Nd^{3+} echo reported in Ref. 2 correspond to a temperature dependence $(\pi\,^{PE}T_2)^{-1}\sim T^{1.3}$ over the interval $0.1 \leq T \leq 1$ K. This

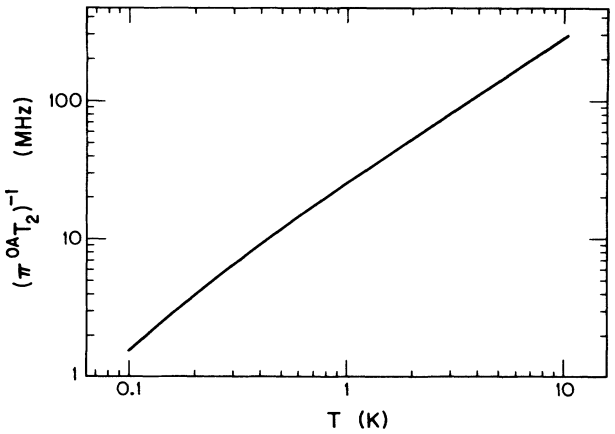

FIG. 2. $(\pi\,^{OA}T_2)^{-1}$ vs T. $^{OA}T_2$ is the dephasing time characterizing the homogeneous linewidth in optical absorption; $C(t)\sim\exp(-t/^{OA}T_2)$. Constant density of states for the tunneling systems. Parameters the same as in Fig. 1. Note that $(\pi\,^{OA}T_2)^{-1}$ is full absorption linewidth at half maximum height.

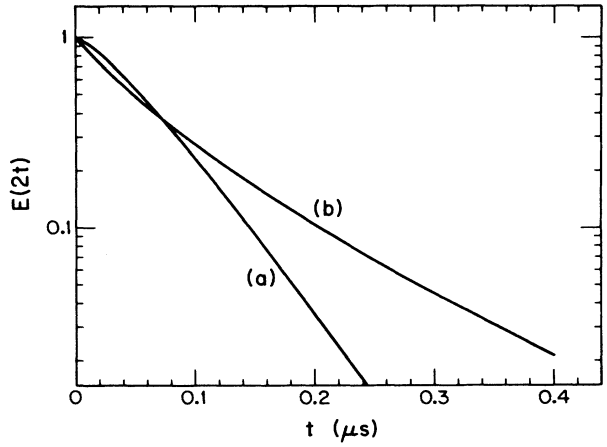

FIG. 3. Time dependence of the photon-echo decay at 1 K. Curve (*a*), $E(2t)$ vs t, parameters the same as in Fig. 1. Curve (*b*), $E(2t)$ vs t, calculated without averaging the matrix element. Strength of the interaction between the Nd^{3+} ion and the tunneling systems have been adjusted so that curve (*b*) coincides with (*a*) when $E(2t)=e^{-1}$.

behavior is compatible with a density of states varying as E^{μ} with $\mu \approx 0.15$. In view of the approximations inherent in the theory and the uncertainties in the experiment, there may be little significance in the differences among results obtained with $\mu=0$, $\mu=0.15$, and $\mu=0.3$.

As is evident from Table I ${}^{\mathrm{PE}}T_2$ and ${}^{\mathrm{OA}}T_2$ differ by a factor of 6.8–9.8 depending on the temperature and the energy dependence of the density of states. This difference appears to be a unique signature of the proposed dephasing mechanism. In the case of conventional mechanisms, e.g., Raman scattering of phonons, one expects to find the same dephasing rates in both optical absorption and photon-echo measurements.

In order to see how this comes about it is useful to consider a simple model for the dephasing in which there is a single fluctuating frequency $\Delta\omega(t)$. The optical dipole-moment correlation function takes the form

$$C(t)=e^{i\omega_0 t}\left\langle \exp\left[i\int_0^t \Delta\omega_v(t')dt'\right]\right\rangle . \tag{20}$$

Treating the fluctuations in the Gaussian approximation,[13] we have

$$C(t)=e^{i\omega_0 t}\exp\left[-\tfrac{1}{2}\int_0^t\int_0^t \langle\Delta\omega(t')\Delta\omega(t'')\rangle dt'dt''\right] , \tag{21}$$

provided $\langle\Delta\omega\rangle=0$. Assuming $\langle\Delta\omega(t)\Delta\omega(0)\rangle$ decays rapidly in comparison with the dephasing time (rapid-modulation limit) Eq. (21) reduces to

$$C(t)=e^{i\omega_0 t}\exp(-t/T_2) , \tag{22}$$

where

$$\frac{1}{T_2}=\int_0^{\infty}\langle\Delta\omega(t)\Delta\omega(0)\rangle dt . \tag{23}$$

In the case of the photon echo one has

$$E(2t)=\left\langle \exp\left[i\int_0^t \Delta\omega(t')dt'-i\int_t^{2t}\Delta\omega(t')dt'\right]\right\rangle . \tag{24}$$

In the rapid-modulation limit one can neglect the correlation between the first and second terms in the exponential in (24) so that after making the Gaussian approximation one obtains

$$E(2t)=\exp(-2t/T_2) , \tag{25}$$

with T_2 given by (23). Thus, the equivalence of the decay rates is seen to be a consequence of the rapid modulation approximation.

Recently, Walsh *et al.*[5] reported the results of a study of the optical dephasing of chromophores in organic glasses. As part of their study, they compared the dephasing time inferred from photon-echo experiments with that given by nonphotochemical hole-burning measurements. Although nonphotochemical hole burning involves more than the optical absorption of the probe beam, our results provide a possible explanation for the differences in the two dephasing times. Since fluorescence line-narrowing measurements provide direct information about the homogeneous linewidth,[14] a more significant test of the theory could be made by carrying out line-narrowing and two-pulse photon-echo studies on the same sample in overlapping temperature ranges.

It should be notes that the two-pulse photon echo is distinct from the three-pulse stimulated photon echo. Neglecting population relaxation (i.e., $T_1=\infty$), the decay of the latter is characterized by an expression of the form[6]

$$\left\langle\left\langle\left\langle \prod_v \exp\left[i\int_0^{\tau}\Delta\omega_v(t')dt'-i\int_{\Lambda+\tau}^{\Lambda+2\tau}\Delta\omega_v(t')dt'\right]\right\rangle\right\rangle\right\rangle ,$$

where Λ is the time between the second and third pulses. It also differs from the accumulated photon echo.[15] In the case of Pr^{3+} in a silicate glass, Macfarlane and Shelby[16] report that the dephasing time inferred from accumulated photon-echo measurements is similar to that given by hole burning experiments in a regime where $1/T_2 \sim T$.

Finally, we comment briefly on the relation of our work ot previous theoretical studies of low-temperature dephasing in glasses. Lyo was the first to point out that a quasilinear temperature dependence in the homogeneous linewidth could arise from a dipolar interaction between the impurity and an array of tunneling systems.[17] The authors of Ref. 3 outlined a general stochastic model for both the photon echo and the linewidth for which the theory presented here provides a concrete realization. Recently, more detailed treatments of the linewidth (but not the photon echo) have appeared.[18] Although some of them give a quasilinear temperature dependence, they do not appear to take proper account of the randomness in the spatial distribution of the tunneling systems.[19]

ACKNOWLEDGMENT

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