Dielectric rotary echoes in vitreous silica

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Dielectric rotary echo experiments at microwave frequencies have been performed on vitreous silica at temperatures between 11 and 70 mK. The relaxation time T_1 of the tunneling states has been measured directly and found to be distributed in agreement with theoretical predictions. From a Fourier transformation of the echo envelope, the absolute value and the distribution of the dipole moments could be deduced. Significant differences between intrinsic tunneling states and those associated with OH^- impurities were observed.

I. INTRODUCTION

It has long been known that the low-temperature thermal[1] and acoustic[2] properties of amorphous materials are fundamentally different from those of their crystalline counterparts. These anomalies are well described by the tunneling model proposed by Anderson *et al.*[3] and Phillips[4] and, in a more detailed version, by Karpov *et al.*[5] This model assumes that in an amorphous material microscopic entities in double-well potentials can tunnel between the two potential minima and thus form two-level states (TLS). The microscopic nature of the tunneling states, however, is still unknown.

At sufficiently low temperatures the lifetimes of these states become long enough to render coherent phenomena observable. Under such conditions acoustic[6,7] and dielectric[8–10] echo experiments can be performed which are an analog of spin[11] and photon[12] echoes. Another type of echo experiment is the rotary echo which is known from spin[13] and optical resonance.[14] Rotary-echo experiments can also be performed on the TLS in amorphous materials. For the acoustic rotary echo this has been done previously.[15]

In the present work we report on the generation of dielectric rotary echoes and their application to measure the phase coherence time T_2, the lifetime T_1, and the dipole moment p of the TLS. We want to point out that our method—unlike other types of echo experiments—allows us to determine T_1 without the necessity of taking into account the disturbing influence of spectral diffusion.[6,16,17]

Our measurements were performed on two types of vitreous silica, Suprasil W and Suprasil I, which differ by their content of OH^- groups of <5 ppm and about 1200 ppm, respectively. Our choice of these samples had two reasons: first of all, it is known[8,18] that the addition of OH^- causes the appearance of a tunneling species in addition to the "intrinsic" ones observed in both Suprasil W and Suprasil I. Secondly, we wanted to compare our results with those derived for the same materials in previous echo experiments and thus to assess the capabilities of our method.

II. THEORY

Because of the analogy between a two-level tunneling state and a spin-$\frac{1}{2}$ system in a magnetic field, the formalism derived for the magnetic case can be employed for our problem as well. At first, however, let us introduce the parameters describing the behavior of a two-level state in an electrical field. A particle with mass m is assumed to tunnel through the barrier V between two overlapping harmonic potentials with ground-state energy $\hbar\Omega/2$ spaced by a distance d. This causes an energy splitting $\Delta_0=\hbar\Omega e^{-\lambda}$ with $\lambda=d(2mV/\hbar^2)^{1/2}$. An additional parameter is an energy difference Δ between the two potential minima. The tunneling model assumes a distribution of Δ and λ such that the density of states $P(\Delta,\lambda)$ is a constant $\bar{P}$. The TLS are coupled to external electrical fields $\mathbf{F}$ by a dipole moment $\mathbf{p}$ which is also assumed to be constant. For the Hamiltonian of the system in the energy representation we then have

$$H=H_0+H_1=\frac{1}{2}\begin{bmatrix}E & 0\\ 0 & -E\end{bmatrix}+\mathbf{p}\cdot\mathbf{F}\begin{bmatrix}\Delta/E & \Delta_0/E\\ \Delta_0/E & -\Delta/E\end{bmatrix}, \tag{1}$$

with $E=(\Delta^2+\Delta_0^2)^{1/2}$. The off-diagonal elements cause resonant transitions and the diagonal elements a variation of the energy splitting E if an electrical field is applied.

The dynamics of the TLS can be described by the Bloch equations[19] which were originally derived for the magnetic case. If we define a pseudopolarization vector $\mathbf{P}=\mathrm{tr}(\rho\cdot\sigma)=(u,v,w)$, where ρ is the density matrix and σ the Pauli matrices, the Bloch equations in a frame rotating about the z axis at frequency ω_0 can be written as[20]

$$\begin{aligned}\dot{u}&=-u/T_2-(\omega-\omega_0)v\ ,\\ \dot{v}&=(\omega-\omega_0)u-v/T_2+\omega_1 w\ ,\\ \dot{w}&=-\omega_1 v-(w-w_0)/T_1\ ,\end{aligned} \tag{2}$$

where $\hbar\omega$ corresponds to the energy of the TLS and w_0 denotes their thermal occupation difference. In the case

of an electrical field $\mathbf{F}(t)=\mathbf{F}_0\exp(-i\omega_0 t)$ the Rabi frequency ω_1 is given by

$$\omega_1 = rpF/\hbar$$

with (3)

$$r=(\Delta_0/E)\cos(\varphi) \quad \text{and} \quad \varphi \sphericalangle (\mathbf{p},\mathbf{F}) .$$

The frequency ω_0 of the external field selects the energy of the TLS which mainly contribute to the experiment. The component of the pseudopolarization contributing to the echo is given by $u+iv$.

The relaxation times T_1 and T_2 have been introduced into the Bloch equations as phenomenlogical quantities. T_1 describes the return of the TLS into thermal equilibrium. For temperatures below 1 K only the one-phonon process is relevant for T_1 resulting in[21]

$$T_1^{-1}=\left[\frac{\gamma_l^2}{c_l^5}+2\frac{\gamma_t^2}{c_t^5}\right]\frac{\Delta_0^2 E}{2\pi\rho\hbar^4}\coth\left[\frac{E}{2kT}\right], \tag{4}$$

where ρ is the mass density, γ_i and c_i the elastic deformation potentials (i.e., the coupling constant between the TLS and phonons), and the velocity of sound for longitudinal and transverse phonons, respectively. Because of the distribution of Δ and λ (and thus of Δ_0), T_1 will be distributed as well:

$$P(E,T_1^{-1})=\bar{P}\frac{T_1}{2(1-T_{1,m}/T_1)^{1/2}}, \tag{5}$$

where $T_{1,m}$ is the minimum relaxation time for states with $\Delta_0=E$ in (4).

T_2 describes the phase memory of the TLS. In our case it is mainly determined by fluctuations of the energy of the TLS caused by the interaction with neighboring TLS undergoing thermal transitions. This so-called "spectral diffusion"[17,22] leads to more complicated behavior of the phase relaxation than a single relaxation time assumed in (2). Nevertheless, a decay-time constant can be defined for limited time windows, and usually the designation T_2 is retained. For times short compared to a characteristic time $\tau_0=T_{1,m}(E\approx kT)$ (which, in this experiment, is close to $T_{1,m}$), a T^{-2} temperature dependence of T_2 is expected.[22]

The Bloch equations (2) can be solved to give[14]

$$\begin{aligned}
u(t)&=u(0)\left[\frac{\omega_1^2}{\beta^2}e^{-\alpha_1 t}+\frac{\omega'^2}{\beta^2}e^{-\alpha_2 t}\cos(\beta t)\right]-v(0)\frac{\omega'}{\beta}e^{-\alpha_2 t}\sin(\beta t)\\
&\quad -w(0)[e^{-\alpha_1 t}-e^{-\alpha_2 t}\cos(\beta t)]+w_0\frac{\omega'\omega_1}{T_1\alpha_1\beta^2}(e^{-\alpha_1 t}-1),\\
v(t)&=u(0)\frac{\omega'}{\beta}e^{-\alpha_2 t}\sin(\beta t)+v(0)e^{-\alpha_2 t}\cos(\beta t)+w(0)\frac{\omega_1}{\beta}e^{-\alpha_2 t}\sin(\beta t)\\
&\quad +w_0\left[\frac{\omega_1}{T_1T_2\alpha_1\beta^2}+\frac{\alpha_1-T_2^{-1}}{T_1\alpha_1\beta^2}-\frac{\omega_1}{T_1\beta^2}e^{-\alpha_2 t}\cos(\beta t)\right],\\
w(t)&=u(0)\frac{\omega'\omega_1}{\beta^2}[-e^{-\alpha_1 t}+e^{-\alpha_2 t}\cos(\beta t)]-v(0)\frac{\omega_1}{\beta}e^{-\alpha_2 t}\sin(\beta t)\\
&\quad +w(0)\left[\frac{\omega'^2}{\beta^2}e^{\alpha_1 t}+\frac{\omega_1^2}{\beta^2}e^{-\alpha_2 t}\cos(\beta t)\right]+w_0\left[\frac{\omega'^2}{T_1\alpha_1\beta^2}(1-e^{-\alpha_1 t})+\frac{\omega_1^2}{T_1\beta^3}e^{\alpha_2 t}\sin(\beta t)\right],
\end{aligned} \tag{6}$$

with abbreviations

$$\omega'=\omega-\omega_0, \quad \beta=(\omega_1^2+\omega'^2)^{1/2}, \tag{7}$$

$$\alpha_1=\frac{1}{\beta^2}\left[\frac{\omega'^2}{T_1}+\frac{\omega_1^2}{T_2}\right], \quad \alpha_2=\frac{1}{\beta^2}\left[\frac{\omega'^2}{T_1}+\frac{\omega_1^2}{2T_1}+\frac{\omega_1^2}{2T2}\right].$$

If a rf electrical field $F(t)=F_0\cos(\omega_0 t+\vartheta)$ is applied to the sample at $t=0$, and if the phase of this field is reversed at $t=\tau_p$, an echo will be superimposed to the driving rf field at $t=2\tau_p$. This situation is described by (6) for $t>\tau_p$:

$$v(t)=w(0)e^{-\alpha_2 t}\frac{\omega_1^3}{\beta^3}\sin[\beta(t-2\tau_p)] \tag{8}$$

if only terms contributing to the echo are retained. In order to find the shape of the echo one has to integrate over ω_1 and β weighted with the appropriate distribution functions. Because of the flat distribution of ω (corresponding to that of the energy of the TLS), the u component vanishes after integration over ω'. For $t=2\tau_p$ the echo amplitude will pass through zero (Fig. 1). Due to the ω_1^3/β^3 term in (8) only small values of ω' contribute and the decay-time constant α_2 can be approximated by $(1/T_1+1/T_2)/2$. Experimentally it is found that $T_1 \gg T_2$ so that for the above pulse sequence the amplitude of the rotary echo should decay with a time constant $2T_2$—twice the value of the spontaneous echo.

For a measurement of T_1 we have used a different pulse sequence: The rf field is turned on for a period τ_f, then switched off for a pause τ_{12} and switched on again. In this case the terms contributing to the echo amplitude are

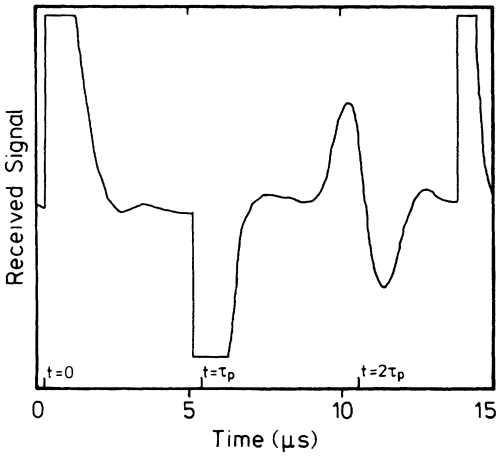

FIG. 1. Typical signal pattern for a dielectric rotary echo. Switching transients are observed when the rf field is turned on at $t=0$, at the 180° phase change at $t=\tau_p$, and when the field is turned off at $t>2\tau_p$. At $t=2\tau_p$ the rotary echo appears. Because of phase-sensitive detection a bipolar signal is observed.

$$v(t)=w(0)e^{-\alpha_2\tau_f}\frac{\omega_1^3}{2\beta^3}\sin[\beta(t-\tau_{12}-2\tau_f)] \times[e^{-\tau_{12}/T_1}-\cos(\omega'\tau_{12})e^{-\tau_{12}/T_2}] . \quad (9)$$

Now the echo amplitude passes through zero at $t=\tau_{12}+2\tau_f$. The second term in Eq. (9) predicts an oscillation of the echo amplitude with ω'. Because of the distribution of ω' this oscillation vanishes rapidly. Therefore, after a few microseconds the echo amplitude should decay only with the pulse length τ_f and the length of the pause τ_{12} with corresponding time constants T_2 and T_1.

For a calculation of shape and amplitude of the echo Eqs. (8) and (9), respectively, must be multiplied with the off-diagonal matrix element [Eq. (1)]. This takes into account the dependence of the echo amplitude induced in the microwave cavity on the dipole moment of the TLS (the electrical properties of the cavity in which the experiment is performed are disregarded here). Then an integration has to be performed over ω_1 and β. This integral has to be weighted with the density of states of the tunneling model supplemented with the isotropic distribution of the dipolar orientation against the external field:

$$P(E,\Delta_0/E,\varphi)=\bar{P}\frac{\sin\varphi}{\frac{\Delta_0}{E}\left[1-\frac{\Delta_0^2}{E^2}\right]^{1/2}} \quad (10)$$

which, after substituting with $r=(\Delta_0/E)\cos\varphi$ gives

$$P(E,r)=\bar{P}(1/r^2-1)^{1/2} . \quad (11)$$

With (3) we therefore expect a distribution of Rabi frequencies

$$P(E,\omega_1)=\bar{P}(\omega_1^{-2}-\omega_{1,\max}^{-2})^{1/2} , \quad (12)$$

where $\omega_{1,\max}=\omega_1(\Delta_0=E)$. Unfortunately, a Fourier transformation of the echo envelope will not directly yield this distribution, but that of an effective Rabi frequency β [Eq. (7)] which, because of the cubic prefactor in Eqs. (8) and (9), is close to that of ω_1. A correct fit to the echo spectrum must therefore be based on the distribution (12) and a constant distribution for ω' (corresponding to the constant energy density of the TLS). This function has to be integrated over ω' and ω_1 weighted with the prefactor $(\omega_1/\beta)^3$ and corrected for the r dependence of the coupling of the dipoles to the electrical field.

In the course of this evaluation it turned out that the frequency spectrum of the experimental echo envelope was still broader than predicted. Experimental artifacts like field inhomogeneities could be ruled out as being the cause. Therefore we had to assume the dipole moment p to be distributed as well. For simplicity a Gaussian distribution was chosen.

III. EXPERIMENT

In our rotary-echo experiment T_2 has to exceed the time scale of a few microseconds imposed by the limited time resolution of the experiment. Therefore, and in order to achieve a sufficient occupation difference w_0, the experiment had to be performed at temperatures between 80 mK and the minimum temperature of 10 mK of our dilution refrigerator. Our samples, disks with a diameter of 14 mm and a thickness of 2 mm, were placed in the uniform electric field region of a reentrant microwave resonator operating at 720 MHz. This resonator was attached to the outside of the mixing chamber of a dilution refrigerator. Temperature was monitored with a carbon thermometer at the resonator.

The field strength in the sample was calculated from the injected power and the filling factor of the cavity. The filling factor was determined by the perturbation method: to simulate a variation of the dielectric constant, a hole was drilled into a sample and the associated change of the resonant frequency was evaluated.

In order to separate the weak echo signal from the large rf driving field, a microwave bridge circuit was used. The 180° phase jump was accomplished by switching between appropriate lengths of cable with two *p-i-n* diode switches. By digital averaging the sensitivity of the receiving system reached values better than 10^{-16} W.

A. Measurement of T_2

The relaxation time T_2 was determined from the decay time constant of the echo amplitude with increasing time τ_p of the phase reversal. Because the electrical field has to be applied for times long enough to observe the echo, self-heating of the sample at the lowest temperatures turned out to be a problem for times exceeding 50 μs.

In Suprasil W, because of the purity of this material, it is assumed that only "intrinsic" TLS associated with the amorphous SiO_2 matrix are observed. The small dipole

moment of these states leads to a low echo amplitude and thus limits the temperature range of the experiment to below 35 mK. Within this range the temperature dependence of the decay time constant T_2 agrees with the T^{-2} dependence found in previous measurements and thus with theoretical predictions based on spectral diffusion.

The decay of the echo amplitude with increasing τ_p for Suprasil I is shown in Fig. 2. In this material, where TLS associated with OH^- impurities dominate, the experimental results are more difficult to understand. T_2 seems to depend linearly on temperature, a behavior which has also been reported in Ref. 9, whereas a $T^{1.8}$ dependence was observed in Ref. 10. Our absolute value of 25 μs for T_2 at 18 mK agrees with both Refs. 8 and 10. In terms of spectral diffusion a linear temperature dependence of T_2 would indicate that we are in a region where $\tau_p \gg \tau_0$ (Ref. 22) (see Sec. II). On the other hand, estimates of τ_0 indicate that we are always in the regime $\tau_p \ll \tau_0$ and there is no obvious reason why there should be a large difference in τ_0 between Suprasil I and Suprasil W. Finally we should state that we cannot rule out that self-heating of the sample has falsified our data at the lowest temperatures.

B. Measurement of T_1

While our measurement of T_2 does not offer significant advantages over spontaneous echo experiments, the rotary echo is a very good tool to study the relaxation time T_1. Whereas in stimulated echo experiments a dependence of the decay time constant on the separation of the first two pulses is caused by spectral diffusion,[6,16] we did not observe the corresponding effect for the rotary echo. There was no dependence of the echo decay on the width τ_f of the first pulse. The reason lies in the fact that in a rotary-echo experiment nutation is time reversed. If we examine the derivative of the effective Rabi frequency with respect to Δ (the quantity which fluctuates because of spectral diffusion),

$$\frac{d\beta}{d\Delta} = \frac{2\Delta}{\beta E^2}(\omega\omega' - \omega_1^2) , \tag{13}$$

we find that, because of $E = \hbar\omega$, the associated fluctuation of β is reduced by a factor ω_1/ω compared to that of ω' if $\omega' \ll \omega_1$. Since the main contribution to the echo comes from states with $\omega' \ll \omega_1$ (or $\beta \approx \omega_1$), the influence of spectral diffusion onto the amplitude of the rotary echo is strongly reduced compared with the situation in a stimulated echo experiment, where ω' is the relevant quantity.

For Suprasil I the decay of the echo amplitude with pause length τ_{12} is shown in Fig. 3. The decay is clearly nonexponential; the solid line in Fig. 3 has been fitted using Eqs. (4) and (5) and shows how well the distribution of relaxation times [Eq. (5)] predicted by the tunneling model can be fitted to the experimental data. We have verified that the data at higher temperatures agree with the $\tanh(\hbar\omega/kT)$ dependence of T_1 predicted by Eq. (4), although, because of the scatter of the experimental data, the fit is not always as satisfactory as in Fig. 3.

From our data we derive a minimum relaxation time $T_{1,m} = 240$ μs for the OH^- states in Suprasil I at 11 mK and $T_{1,m} = 100$ μs for the intrinsic states in Suprasil W.

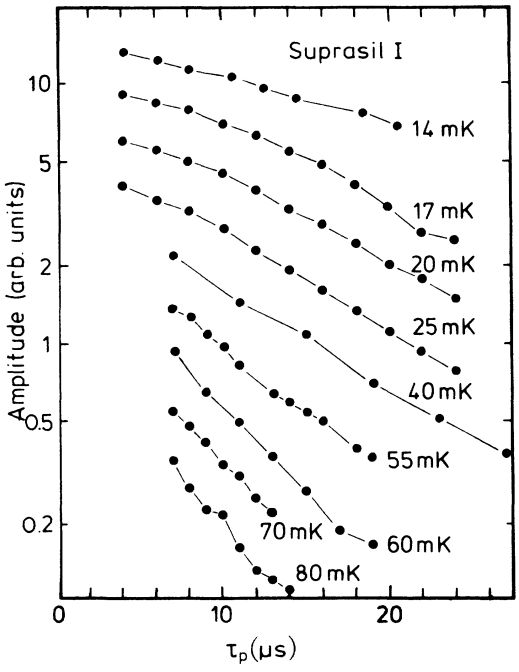

FIG. 2. Amplitude decay of the rotary echo with increasing phase switching time τ_p in Suprasil I. The data for different temperatures are offset for clarity. Lines between data points are a guide to the eye.

IV. THE COUPLING OF TUNNELING STATES TO PHONONS AND ELECTRICAL FIELDS

A. Coupling to phonons

From $T_{1,m}$ the deformation potential γ for the coupling between phonons and the TLS can be calculated with the aid of Eq. (4). We make the usual assumption $\gamma_1^2 = 2\gamma_t^2$. For Suprasil W where the intrinsic TLS are as-

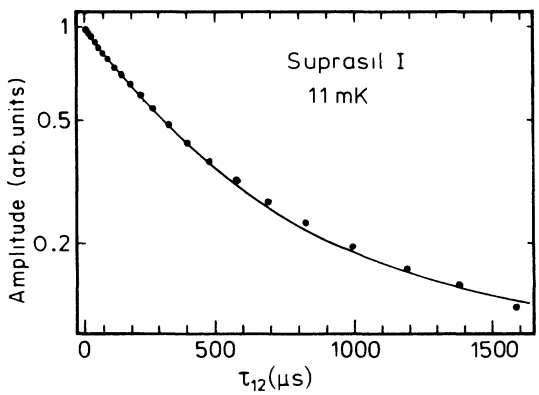

FIG. 3. Amplitude decay of the two-pulse rotary echo in Suprasil I with pause length τ_{12}. The solid line is a fit based on the tunneling model with $T_{1,m} = 240$ μs.

cribed to the motion of SiO_2 units within the amorphous matrix, we find a deformation potential $\gamma_l(\text{int})=1.85 \pm 0.4$ eV. In Suprasil I the OH^- groups seem to be more weakly coupled: $\gamma_l(\text{OH})=1.2\pm0.15$ eV. With the exception of Ref. 9, these values are close to those measured in previous echo experiments.[6,8] Considering that our measurement is not impaired by spectral diffusion, these results should be quite reliable.

B. Electrical dipole moment

Let us now turn to the electric dipole moment of the TLS. It can be calculated from the known field strength in the resonator and from a fit to the Fourier spectrum of the echo. The Fourier transform of a typical echo envelope is shown in Fig. 4 together with a fit based on the assumptions discussed above. It is obvious that the experimental result can be reproduced very well for Suprasil W, the sample containing no OH^- impurities. From our fit we derive a dipole moment of $p=0.7$ D with a width of the distribution of 0.2 D (or 30%) for the intrinsic TLS.

On the other hand, for Suprasil I containing 1200 ppm of OH^-, we find a much higher dipole moment, 3.3 D, which is single-valued or distributed with a width of at most 15%.

In all cases no dependence of the echo spectrum on the type of the experiment (phase reversal or two-pulse) and on the delay time was found. When the Rabi-frequency spectrum was shifted to lower values by reducing the amplitude of the driving field, no influence due to the finite bandwidth of the experiment could be detected. There is, however, a deviation of the experimental spectrum for Suprasil I from the expected shape at high values of β. Here the Fourier amplitude drops faster than theory predicts even for a single-valued dipole moment. This observation can be explained if we assume that the distribution of Δ and λ for the OH^- states does not follow the assumption of the tunneling model and that high values of Δ_0 are less probable. Then, because of the high value of Δ/E, spectral diffusion will suppress the contribution of the states with $\beta \gg \omega_1$

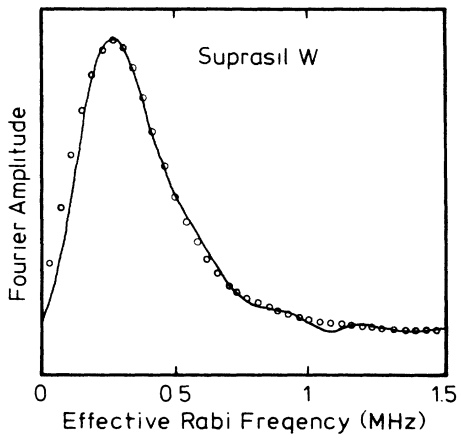

FIG. 4. Fourier transform of the echo envelope of a two-pulse rotary echo in Suprasil W at 10 mK (solid line). The result of a fit with a dipole moment $p=0.7$ D and a width of the distribution of 0.2 D is shown for comparison (open circles).

This result shows for the first time that, apart from the magnitude of the dipole moment, there exists a fundamental difference in the microscopic nature between TLS associated with OH^- impurities and the "intrinsic" states. For the OH^- impurities we see that the TLS practically have a fixed dipole moment, probably given by that of the OH^- molecule. In deed, with the dipole moment of 3.3 D found in this experiment and correcting for local field using the Clausius-Mossotti relation, we arrive at the literature value of 1.66 D (Ref. 23) for OH^- in the gaseous phase. This result strongly supports the assumptions made in Ref. 24 where dielectric experiments on Suprasil I are analyzed in terms of an OH^- molecule rotating in a two- or three-well potential.

For the intrinsic TLS, on the other hand, the distribution of p reflects the distribution of their microscopic parameters. If the charge associated with the dipole moment is assumed to be fixed, then our result indicates that the tunneling distance d is distributed with a typical width of 30%. Considering the fact that λ must be distributed over 1 order of magnitude to agree with the observed spectrum of relaxation times of the TLS, it is obvious that this cannot be caused by the distribution of the tunneling distance d alone. Therefore we conclude that a distribution of the barrier height V, possibly combined with a variation of the mass m, must be mainly responsible for the broad range of λ and thus of the energies and relaxation times of the TLS.

V. CONCLUSION

We have reported on dielectric rotary-echo experiments in pure vitreous silica (Suprasil W) and in samples containing OH^- impurities (Suprasil I). Such experiments are especially suited to study the relaxation behavior of glassy materials in the presence of spectral diffusion. Our measurements of the relaxation time T_1 confirm previous results. We derive elastic deformation potentials of 1.5 and 0.95 eV for the intrinsic TLS in Suprasil W and the OH^- impurity states in Suprasil I, respectively.

From the shape of the echo envelope, the dipole moment of the TLS and the width of its distribution could be derived. For the OH^- impurities we find a dipole moment $p=3.3$ D (uncorrected for local field) which is single valued or has a distribution width $\Delta p/p$ of less than 15%. For the intrinsic TLS the dipole moment is distributed much broader with a width of 30% and a mean value of $p=0.7$ D. This shows for the first time that there exist fundamental microscopic differences between the intrinsic tunneling states in a glass and those associated with impurities.

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