Low-temperature breakdown of coherent tunneling in amorphous solids induced by the nuclear quadrupole interaction

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We consider the effect of the internal nuclear quadrupole interaction on quantum tunneling in complex multiatomic two-level systems. Two distinct regimes of strong and weak interactions are found. The regimes depend on the relationship between a characteristic energy of the nuclear quadrupole interaction λ_* and a bare tunneling coupling strength Δ_0. When $\Delta_0 > \lambda_*$, the internal interaction is negligible and tunneling remains coherent determined by Δ_0. When $\Delta_0 < \lambda_*$, coherent tunneling breaks down and an effective tunneling amplitude decreases by an exponentially small overlap factor $\eta^* \ll 1$ between internal ground states of left and right wells of a tunneling system. This affects thermal and kinetic properties of tunneling systems at low temperatures $T < \lambda_*$. The theory is applied for interpreting the anomalous behavior of the resonant dielectric susceptibility in amorphous solids at low temperatures $T \lesssim 5$ mK, where the nuclear quadrupole interaction breaks down coherent tunneling. We suggest the experiments with external magnetic fields to test our predictions and to clarify the internal structure of tunneling systems in amorphous solids.

I. INTRODUCTION

A transition between two energy minima separated by a potential barrier U occurs in different ways at high and low temperatures. At high temperatures the motion between the energy minima is classical, i.e., thermally activated, suggesting that the particle acquires the energy U from the environment to overcome the barrier. This results in an exponential Arrhenius factor $\exp(-U/T)$ for the transition rate. The classical transition rate decreases rapidly with decreasing the temperature. At low temperature the above-described transport crosses over into the weakly temperature-dependent quantum tunneling. A tunneling transition rate is essentially governed by the exponentially small and temperature-independent factor $\exp[-2S(U)/\hbar]$, where $S(U)$ describes the classical action for the motion through the inverted potential barrier. The strong sensitivity of the tunneling exponent against particle mass makes quantum tunneling more favorable for light particles as electrons, while the motion of heavy nuclei is classical down to temperatures of order of 1 K. However, for $T \lesssim 1$ K quantum tunneling displays itself in the thermodynamic and kinetic properties of atomic systems. One impressive example is amorphous solids, in which the low-temperature properties are governed by the two-level systems (TLS's). These TLS's are made of atoms or groups of atoms, experiencing a tunneling motion between pairs of energy minima separated by potential barriers.[1] They contribute to the universal thermodynamic and kinetic properties in all known glasses as well as some other disordered materials for $T \lesssim 1$ K (see Refs. 2–4).

A TLS is described by the standard pseudospin 1/2 Hamiltonian

$$\hat{h}_{\text{TLS}} = -\Delta_0 s^x - \Delta s^z. \tag{1}$$

Here Δ_0 is a tunneling amplitude coupling two energy minima and Δ is a level asymmetry. The quantity Δ_0 is exponentially sensitive to external and internal parameters of the system. This results in a logarithmically uniform distribution of tunneling amplitudes for various TLS's

$$P(\Delta, \Delta_0) = \frac{P}{\Delta_0}, \quad P = \text{const}. \tag{2}$$

This distribution of TLS's leads to the universal temperature and time dependencies for various physical characteristics of amorphous solids, thus making "glassy" behavior very easily recognizable. For instance, this includes the logarithmic relaxation in time of the specific heat[1–4] and the nonequilibrium dielectric susceptibility.[5] In addition, dielectric and acoustic (a sound velocity) susceptibilities in glasses show a logarithmic temperature dependence also associated with the distribution Eq. (2). Note that the exponential sensitivity of a tunneling strength Δ_0 to environmental interactions also broadens noticeably the distribution of tunneling amplitudes and relaxation rates in other systems, e.g., tunneling of a large electronic spin in magnetic molecule $Mn_{12}Ac$. The latter system shows a broad spectrum of relaxation times due to interaction of electronic spins with nuclear spins.[6,7] A broad spectrum of relaxation times also has been reported in new disordered magnetic alloys.[8]

The nature of the tunneling systems in amorphous solids remains unclear in spite of the theoretical efforts attempting various models.[9–13] The main problem in the theory is a lack of experiments which are capable of proving the advantage of a specific model against the original phenomenological
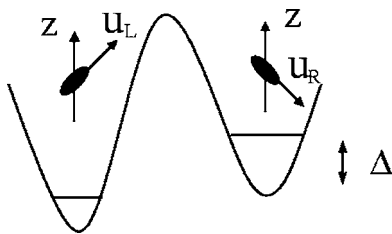
FIG. 1. The two level system having different nuclear quadrupole quantization axes u_L and u_R defined by the local electric field gradient in the left and right wells.

model[1] based on Eq. (2). The phenomenological approach Eq. (2) can be used to explain a variety of experimental data, while the interaction between TLS can successfully be treated as a weak correction.[13,14] Even the internal TLS structure is unclear yet. For instance, nobody knows how many atoms do participate in a single tunneling event. We hope that understanding the nuclear quadrupole interaction effects in glasses, which are sensitive to the internal TLS structure, will help to resolve this question.

Recent experimental investigations of amorphous solids at very low temperatures have revealed a number of qualitative deviations from the predictions of the standard tunneling model Eq. (2). In particular, it is demonstrated in several works[13,15,16] that, for $T \lesssim 5$ mK, the expected logarithmic temperature dependence of the dielectric constant breaks down and the dielectric constant becomes approximately temperature independent. This result conflicts with the logarithmically uniform distribution Eq. (2) of TLS's over their tunneling amplitudes. To resolve the problem, one can assume that the distribution of TLS's has a low-energy cutoff at $\Delta_{0,min} \simeq 5$ mK. This assumption, however, contradicts the observation of very long relaxation times in all glasses. These times (a week or longer) require much smaller tunneling amplitudes[5] than 5 mK (remember that the TLS relaxation time is inversely proportional to its squared tunneling amplitude).

We suggest the explanation of this controversial fact by using the recent model of Würger, Fleischmann, and Enss[17] who proposed that the nuclear quadrupole interaction affects the properties of TLS's at low temperatures by the mismatch of the nuclear quadrupole states in different potential wells (see Fig. 1). This is very similar to the electronic spin tunneling suppression by the nuclear spin interaction in magnetic molecules.[7] The significance of the nuclear quadrupole interaction has recently been proven experimentally in glycerol glass.[18] This interaction helps to understand the anomalous magnetic field dependence of dielectric properties in entirely nonmagnetic dielectric glasses.[16,19,20]

We show that an effective tunneling amplitude of TLS having an energy less than its nuclear quadrupole interaction is remarkably reduced due to the mismatch of TLS nuclear quadrupole states in its two energy minima, similarly to the polaron effect. Consequently the spectrum of TLS tunneling amplitudes Δ_0 possesses a pseudogap below the nuclear quadrupole interaction energy λ_*. Dielectric and acoustic susceptibilities of glasses become temperature independent for temperatures belonging to this pseudogap because there is no

TLS with $\Delta_0 \sim T$ to contribute. Thus our model explains the experimental observations. In addition, we predict that the application of a strong external magnetic field will reduce the mismatch of different nuclear quadrupole states and thus restore the logarithmic temperature dependence of TLS susceptibilities. According to our theory dielectric glasses having no nuclear quadrupole interaction should obey the predictions of the tunneling model, which mostly agrees with the experiment.

Since a low-temperature dielectric constant and a speed of sound in glasses have a similar physical nature they should have a similar behavior at low temperatures. Therefore the saturation in the logarithmic temperature dependence of a sound velocity should be seen at $T < 10$ mK in materials possessing nonvanishing nuclear quadrupole moment. The situation with sound velocity measurements is, however, more complicated because it is more difficult to perform the low-temperature measurements for a sound velocity then for a dielectric constant. There exists a few measurements that we discuss together with the measurements of the dielectric constant. The present theory is developed for the dielectric constant, although our conclusions can be extended to the velocity of sound without major changes.

The paper is organized as follows. In Sec. II the model of two-level systems affected by the nuclear quadrupole interaction is introduced. Then, we discuss qualitatively the renormalization of tunneling amplitude by the nuclear quadrupole interaction depending on the relation between tinneling splitting Δ_0 and a quadrupole interaction λ_*. In Sec. III the expression for the resonant dielectric susceptibility is obtained and the influence on the tunneling amplitude renormalization on the resonant part of the TLS dielectric constant is described in the qualitative level. In Sec. IV we use a perturbation theory to characterize quantitatively the temperature dependence of the dielectric constant in the presence of nuclear quadrupole interactions in the high- and low-temperature limits. In Sec. V we consider a solvable model of TLS's coupled to harmonic oscillators, conveniently replacing nuclear spins. The solution of this problem permits us to simulate the influence of the nuclear quadrupole interaction on a TLS dielectric constant at all temperatures of interest. In Sec. VI the effect of the external magnetic field on the dielectric constant is considered within the simplified model of Sec. V. In Sec. VII the parameters of our model are compared with the experimental data. In final Sec. VIII the conclusions are formulated and the suggestions for an experimental verification of our theory are made. The short version of the manuscript can be found in Ref. 21

II. MODEL

A. Nuclear quadrupole interaction

How does the nuclear quadrupole interaction affect tunneling? Consider a tunneling system formed by n atoms all possessing a nuclear spin $I \geq 1$ and consequently a nuclear electrical quadrupole moment. The total tunneling Hamiltonian $\hat{h}$ can be described by the standard TLS pseudospin Hamiltonian $\hat{h}_{TLS}$ (1) and the quadrupole interactions $\hat{H}_r$ in

the right well ($s^z=1/2$) and $\hat{H}_l$ in the left well ($s^z=-1/2$) as follows:

$$\hat{h} = \hat{h}_{\text{TLS}} + \frac{\hat{H}_r + \hat{H}_l}{2} + (\hat{H}_r - \hat{H}_l)s^z. \tag{3}$$

The local nuclear quadrupole Hamiltonians $\hat{H}_{r,l}$ can be expressed as a sum of interactions of all n nuclear spins $\hat{\mathbf{I}}_i$ over all n atoms that simultaneously participate in its tunneling motion with the local electric field gradient tensors $\partial F_a^{(l,r)}/\partial x_b$ different in general for the right and left wells

$$\hat{H}_{r,l} = \sum_{i=1}^{n} \hat{h}_i^{(r,l)},$$

$$\hat{h}_i^{(r,l)} = -\frac{Q}{2} \sum_{a,b=x,y,z} \left(\hat{I}_i^a \hat{I}_i^b + \hat{I}_i^b \hat{I}_i^a - 2\delta_{ab}\frac{I(I+1)}{3} \right) \frac{\partial F_a^{(l,r)}}{\partial x_b}. \tag{4}$$

Here $\hat{I}_i^a$ is a nuclear spin projection onto the a-axis and Q is the electrical quadrupole moment.

In what follows, we consider a simplified model for the nuclear quadrupole interaction (4) possessing the axial symmetry (see Fig. 1)

$$\hat{H}_{l,r} = b\left((I_{l,r}^{u_{l,r}})^2 - \frac{I(I+1)}{3} \right). \tag{5}$$

Here

$$b \approx Q|\partial F/\partial x|, \tag{6}$$

and u_L and u_R define the directions of the electric field gradient in the left and right wells, respectively.

There exists also the magnetic-dipolar interaction of nuclear spins. It is usually by several orders of the magnitude smaller than the quadrupole interaction and can be neglected.

B. TLS ground state and its effective tunneling amplitude

It will be shown below that the resonant dielectric susceptibility of a TLS ensemble is due to the ground state of tunneling systems affected by the external electrical field. Only tunneling systems for which $\Delta_0 \sim \Delta$ can contribute to the dielectric constant. These two parameters must exceed the thermal energy, i.e., $\Delta_0 \gtrsim T$ (see Ref. 3 and 4 and Sec. III for details). The structure of the ground state of the Hamiltonian (3) depends on the relation between typical tunneling amplitude $\Delta_0 \sim T$ and characteristic value of the quadrupole interaction nb. If $\Delta_0 > nb$, the nuclear quadrupole interaction can be treated as a small perturbation and the TLS behavior obeys the standard tunneling model.

In the opposite case $\Delta_0 < nb$, the tunneling term can be treated as a weak perturbation. In a zero-order perturbation theory the states in the left and right well can be considered separately. These states form a proper basis for eigenstates of a tunneling system. In particular, the actual ground state is a linear superposition of the ground states in the isolated left and right wells. The energies corresponding to these ground states are $E_{gl}=-\Delta/2+E_{g,l}$, $E_{gr}=\Delta/2+E_{g,r}$, where $E_{g,r}$, $E_{g,l}$

are the ground-state energies of the quadrupole interaction Hamiltonians $\hat{H}_{r,l}$ [Eq. (3)] in the right or left wells, respectively.

The effective tunneling amplitude Δ_{0*}, which couples any pair of levels in the left and right wells of TLS's with $\Delta_0 < nb$ decreases due to a mismatch of nuclear quadrupole ground states in two potential wells. Obviously, when $H_r = H_l$, the nuclear quadrupole interaction does not affect tunneling. This mismatch can be expressed in terms of a characteristic overlap integral between the nuclear spin ground states in the two wells. In the absence of tunneling one has

$$\eta_* = \eta^n, \quad \eta = \langle lg|rg \rangle, \tag{7}$$

where the symbols $|lg\rangle$ ($|rg\rangle$) stand for the ground-state wave function of the nuclear spin of a single tunneling atom in the left (right) well. This overlap integral enters directly into the effective tunneling amplitude. In fact, at very low temperatures and small tunneling amplitude $\Delta_0 < nb$ one may treat the TLS in terms of the ground states in the right and left wells. The tunneling matrix element Δ_{0*} between these two states resulting from the perturbation $-\Delta_0 s^x$ is

$$\Delta_{0*} = \eta_* \Delta_0. \tag{8}$$

Thus, the overlap integral η_* determines the reduction of the tunneling amplitudes due to the nuclear quadrupole interaction.

The specific value of the overlap integral depends on the absolute value of the nuclear spin, nature of the nuclear quadrupole interaction, the number of tunneling atoms per one TLS, and the difference in the electric field gradients in the right and left wells. Currently, reliable information about all these parameters is not available. In principle the value of the overlap integral η fluctuates from atom to atom. This will lead to the log-normal distribution of the overall overlap integral η_* in the large n limit. As we will see below [e.g., Eq. (56)] the behavior of the dielectric constant is mostly sensitive to $\ln(\eta_*)$, which possesses the gaussian distribution, so we can use its average value with the logarithmic accuracy of our consideration.

To estimate typical value η_* let us turn to a simplified model for the nuclear quadrupole interaction Eqs. (4) and (6), possessing the axial symmetry. Let ϕ be the angle between the directions of the electrical field gradient in the left and right wells.

First, consider the case of an integer nuclear spin. If the interaction constant is positive, i.e., $b>0$, the energy minimum is obtained when the projection $I_{l,r}^\alpha=0$. In this case the ground state is nondegenerate. This simplifies our consideration and makes it possible to calculate the overlap integral of the ground state wavefunctions depending on the mismatch angle ϕ (see Fig. 1). We confine ourselves with two simple cases of integer nuclear spins, namely, $I=1,2$. Then the overlap integral between the left and right ground states can be expressed as

$$\eta = \cos(\phi), \quad I = 1,$$

$$\eta = |\cos^2(\phi) - \sin^2(\phi)/2|, \quad I = 2. \quad (9)$$

We will pay most attention to these two nondegenerate cases because they are very convenient for the investigation of the effect of an external magnetic field on the overlap integral (see Sec. VI.)

The value of the rotational angle ϕ in glasses is unknown. We expect that it can be estimated by extended molecular dynamics simulations.[22] On the other hand, in the orientational glass $(KBr)_{1-x}(KCN)_x$ (see Ref. 23) the CN group rotates between different equilibrium positions by angle $\phi = \cos^{-1}(1/3)$ (see Ref. 3). We assume that in glasses the rotational angle ϕ is the same and neglect its fluctuations due to a structural disorder. Then, for spin $I=1$, one can estimate the overlap integral Eq. (9) as

$$\eta \approx \cos(\phi) \approx 0.33. \quad (10)$$

When the TLS contains n atoms tunneling simultaneously, the characteristic overlap integrals becomes

$$\eta_* = \langle l | r \rangle \approx \eta^n. \quad (11)$$

To understand and interpret experimental data, we assume that the total overlap integral is small

$$\eta_* \ll 1. \quad (12)$$

This assumption is justified by the exponential decrease of the overlap integral with the number of atoms n participating in a single tunneling system (TLS).

The case of a half-integer spin is more complicated because of the Kramers degeneracy. The quadrupole spin Hamiltonian of a tunneling atom i has two orthogonal ground states $|il+\rangle$, $|il-\rangle$ in the left well and two orthogonal ground states $|ir+\rangle$, $|ir-\rangle$ in the right well. As a result, the ground state of n tunneling atoms is 2^n-fold degenerate. In order to reduce the problem to a single pair of levels (one in the left well and the other in the right one), we divide the degenerate ground state into 2^n pairs of left $|il\rangle$ and right $|ir\rangle$ ground states coupled only with each other. In fact, tunneling amplitude between left and right states of a tunneling atom i is directly proportional to the overlap integral of the two states involved $\langle il | ir \rangle$. Let us introduce two superposed states in the left well as follows:

$$|l1\rangle = \cos(\alpha)|il+\rangle + \sin(\alpha)|il-\rangle,$$

$$|l2\rangle = -\sin(\alpha)|il+\rangle + \cos(\alpha)|il-\rangle. \quad (13)$$

The angle α is defined by the condition of orthogonality

$$0 = \langle ir+ |l2\rangle = \langle ir- |l1\rangle,$$

$$= -\sin(\alpha)\langle ir+ |il+\rangle + \cos(\alpha)\langle ir+ |il-\rangle,$$

$$= \cos(\alpha)\langle ir- |il+\rangle + \sin(\alpha)\langle ir- |il-\rangle. \quad (14)$$

This equation can be solved for α if the equation determinant is equal to zero, i.e.,

$$\langle il+ |ir+\rangle\langle il- |ir+\rangle + \langle il+ |ir-\rangle\langle il- |ir-\rangle = 0. \quad (15)$$

This condition is satisfied when pairs of eigenstates $|ir+\rangle$, $|ir-\rangle$ and $|il+\rangle$, $|il-\rangle$ are orthogonal to each other. One can verify the validity of Eq. (15) by projecting the left vectors $|il+\rangle$, $|il-\rangle$ onto the right vector subspace $(|ir+\rangle, |ir-\rangle)$. Choosing the basis for the left well from the pairs of states Eq. (13), we can construct 2^n possible states of nuclear spins from the products of different left states (two choices for each atom). The basis states in the right well can be constructed similarly. Then each of the 2^n left basis states possesses a nonzero overlap integral with the only single state from the 2^n right basis states.

Thus, the degenerate states form groups of pairs of states connected by tunneling, while the states belonging to different pairs are uncoupled. Since all 2^n pairs possess identical properties with respect to tunneling, one can treat them as independent pairs of states. Then, our considerations become similar to those for integer nuclear spins with degenerate ground states of nuclear quadrupoles.

III. RESONANT SUSCEPTIBILITY

The main attention of this work is paid to the TLS resonant dielectric susceptibility ε_{res} which shows the large deviation from the conventional tunneling model for $T \leqslant 5$ mK. We begin from reviewing the nature of the contribution of tunneling systems to the dielectric constant. Although the resonant dielectric susceptibility has the well established behavior (for reviews, see Refs. 3 and 4, and references therein), our analysis will be helpful for understanding the effect of the nuclear quadrupole interaction. It is also useful for readers not too familiar with the TLS dielectric response.

Consider a single TLS polarization due to the external electric field $\mathbf{F}$. We suppose that tunneling atoms possess a nonzero charge. Then tunneling between the right and left wells changes the dipole moment of TLS. The TLS dipole moment operator can thus be expressed in the terms of pseudospin [see Eq. (3)]

$$\hat{\boldsymbol{\mu}} = \boldsymbol{\mu} s^z. \quad (16)$$

Here $\boldsymbol{\mu}$ is a dipole moment of a tunneling system. The interaction of the external field $\mathbf{F}$ with a TLS can be written as

$$\hat{V} = -\mathbf{F}\boldsymbol{\mu} s^z. \quad (17)$$

In general, there can be a contribution to the dipole moment proportional to the off-diagonal operator s^x. This results from a change of the barrier due to electric field. Employing the experimental data[3] and theoretical estimates,[24] we can argue that such term leads to much smaller effect than that from the "diagonal" term Eq. (16). Therefore we neglect it.

The external field effect application can be taken into account by introducing the field-dependent asymmetry energy

$$\Delta(\mathbf{F}) = \Delta + \mathbf{F}\hat{\boldsymbol{\mu}}. \quad (18)$$

Thus, the energies E_α, $\alpha = 1, 2, \ldots Z$, of all $Z = 2 \cdot (2I+1)^n$ eigenstates of a tunneling system become dependent on the

external field. Remember that $Z=2$ in the absence of the nuclear quadrupole interaction.

We are interested in a *linear* response of a TLS ensemble, i.e., response to an infinitesimal electric field. Then the dipole moment of the eigenstate α can be expressed as

$$\boldsymbol{\mu}_\alpha = -\frac{\partial E_\alpha}{\partial \mathbf{F}} = -\boldsymbol{\mu}\frac{\partial E_\alpha}{\partial \Delta}. \tag{19}$$

The total TLS dipole moment can be expressed as a sum of contributions of all the Z eigenstates α, weighed by the Gibbs population factors P_α

$$\bar{\boldsymbol{\mu}} = \sum_{\alpha=1}^{Z} \boldsymbol{\mu} P_\alpha \frac{\partial E_\alpha}{\partial \Delta}. \tag{20}$$

The population factor P_α is given by the equilibrium distribution for *unperturbed* TLS's

$$P_\alpha(\Delta_0,\Delta;T) = \frac{\exp\left(-\dfrac{E_\alpha}{T}\right)}{\displaystyle\sum_{\gamma=1}^{Z} \exp\left(-\dfrac{E_\gamma}{T}\right)}. \tag{21}$$

Finally, the susceptibility of a given TLS is determined by

$$\chi_{ab} = \frac{\partial \bar{\mu_a}}{\partial F_b} = \sum_{\alpha=1}^{Z} \left(-\mu_a\mu_b P_\alpha \frac{\partial^2 E_\alpha}{\partial \Delta^2} - \mu_a \frac{\partial P_\alpha}{\partial F_b}\frac{\partial E_\alpha}{\partial \Delta} \right). \tag{22}$$

Here the indexes a and b denote the Cartesian coordinates of the corresponding vectors and tensors. The first term is associated with the *adiabatic* excitation of the TLS due to the field-induced change in its eigenenergies. This contribution reaches its maximum when TLS has an asymmetry Δ smaller than its tunneling amplitude Δ_0. It is called a resonant contribution.[3] The remaining term is associated with the change in populations of TLS energy levels induced by the external electric field. Such changes take place by the TLS transitions between different quantum states. Therefore, this contribution corresponds to the relaxational term.[3]

For temperatures $T<50$ mK and an external alternating field $\mathbf{F}$ with frequency $\nu>100$ Hz, the relaxation contribution is negligibly small[3,5,15] because a relaxation rate of TLS populations becomes much smaller than the field oscillation rate ν. Therefore, the relevant TLS's cannot adjust their thermal populations to the rapidly changing field. In the range of interest $T\sim 5$ mK we can neglect the relaxational contribution and restrict our consideration to the first term on the right-hand side of Eq. (22).

The contribution of a single TLS should be summed over all TLS's belonging to the system. This is equivalent to averaging the susceptibility χ_{ab} in Eq. (22) over energies, tunneling amplitudes, dipole moments, and other possible relevant parameters. Averaging over the directions and absolute values of TLS dipole moments is straightforward and we can rewrite the resonant TLS contribution in the form

$$\chi_{ab}^{\mathrm{res}} = \delta_{ab}\chi,$$

$$\chi = \frac{\mu_0^2}{3}\left\langle \sum_{\alpha=1}^{Z} P_\alpha(\Delta_0,\Delta;T)\chi_\alpha(\Delta,\Delta_0) \right\rangle,$$

$$\chi_\alpha(\Delta,\Delta_0) = -\frac{\partial^2 E_\alpha}{\partial \Delta^2}. \tag{23}$$

Here μ_0^2 is the average square of the TLS dipole moment. Also, we have ignored the correlations between TLS dipole moments with the other tunneling parameters. This is justified by the available experimental data.[3] The average $\langle\cdots\rangle$ implies the integration of the single TLS response over $d\Delta d\Delta_0/\Delta_0$ in accordance with the postulated TLS distribution Eq. (2).

Let us first consider the behavior of the dielectric constant in the zero-temperature limit. In this case it suffices to take into account the ground state only in Eq. (22), calculating the resonant dielectric constant

$$\chi = -\frac{P\mu_0^2}{3}\int_0^W \frac{d\Delta_0}{\Delta_0}\int_{-\infty}^{\infty} \frac{\partial^2 E_g}{\partial \Delta^2}d\Delta. \tag{24}$$

The upper integration limit W represents the maximum tunneling amplitude, while the integration limits for an asymmetry parameter Δ are set to $\pm\infty$ because its absolute value can be much larger than the tunneling splitting Δ_0. The integral over asymmetries can be evaluated as

$$-\frac{\partial E_g(\Delta_0,\Delta)}{\partial \Delta}\Bigg|_{\Delta=-\infty}^{\Delta=\infty}. \tag{25}$$

For a large asymmetry $|\Delta|\gg\Delta_0$, the ground state is determined by the minimum energy state of a particle in the left $\Delta>0$ (see Fig. 1) or the right well $\Delta<0$. So, for large Δ the ground-state energy behaves as $E_g\approx-|\Delta|/2$, while the tunneling amplitude and nuclear interaction give rise only to the small correction. Then the expression in Eq. (25) equals unity. Therefore, Eq. (24) results in the divergent integral

$$\chi \approx \frac{P\mu_0^2}{3}\int_0^W \frac{d\Delta_0}{\Delta_0}. \tag{26}$$

It is remarkable that this result does not depend on whether or not the quadrupole interaction exists. So, the question remains: what happens at finite temperature?

The answer is clear for TLS's without a nuclear quadrupole interaction. In this case each TLS has only the two states, i.e., the ground state g and the excited state e with energies $E_{g,e}=\mp(1/2)\sqrt{\Delta^2+\Delta_0^2}$ [Eq. (1), see Ref. 3], respectively. The susceptibilities of excited and ground states differ by the sign since

$$\frac{\partial^2 E_e}{\partial \Delta^2} = -\frac{\partial^2 E_g}{\partial \Delta^2} = -\frac{1}{2}\frac{\Delta_0^2}{(\Delta^2+\Delta_0^2)^{3/2}}. \tag{27}$$

The response comes mainly from resonant TLS's having $|\Delta|\lesssim\Delta_0$. Therefore, both levels of a TLS with $\Delta_0<T$ are approximately equally populated. For this reason, the contribution of the excited state to the dielectric constant cannot be
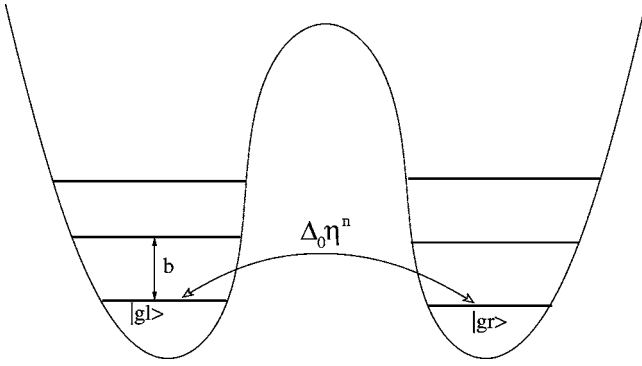
FIG. 2. Two level configuration with splitted energy levels.

neglected. Moreover, the contributions from the excited and ground state nearly cancel each other if $\Delta_0 < T$.

Calculating the finite temperature resonant susceptibility, one should consider only TLS ground states and cutoff the integral Eq. (24) at the lower limit given by $\Delta_0 \sim T$. This leads to the well-known logarithmic temperature dependence

$$\chi = \frac{P\mu_0^2}{3} \int_T^W \frac{d\Delta_0}{\Delta_0} \int_{-\infty}^{\infty} d\Delta \frac{\partial^2 E_g}{\partial \Delta^2} = \frac{P\mu_0^2}{3} \ln(W/T). \quad (28)$$

This result is valid as long as the temperature exceeds the energy of the quadrupole interaction nb. Next, we discuss the case $T \ll b$. TLS's with small tunneling amplitudes $\Delta_0 < nb$ still contribute to the resonant susceptibility. They can be represented by pairs of lowest nuclear quadrupole levels in the right and left wells because the higher levels are separated by the gap $b \gg T$ from these two lowest ones. They are coupled with each other by the tunneling amplitude Δ_0 reduced by the overlap factor Eq. (11) (see Fig. 2), i.e.,

$$\Delta_{0*} \approx \Delta_0 \eta^n. \quad (29)$$

These two lowest levels can be treated as a new TLS. Since only TLS's with $\Delta_{0*} > T$ contribute to the permittivity [see Eq. (28)], this defines the renormalized lower cutoff

$$\Delta_{0l} \sim T\eta^{-n}. \quad (30)$$

Substituting this cutoff into integral Eq. (28) yields in the limit $T \to 0$

$$\chi = \frac{P\mu_0^2}{3} \int_{T\eta^{-n}}^W \frac{d\Delta_0}{\Delta_0} \int_{-\infty}^{\infty} d\Delta \frac{\partial^2 E_g}{\partial \Delta^2} = \frac{P\mu_0^2}{3} [\ln(W/T) - n\ln(1/\eta)].$$

$$(31)$$

Thus, due to the quadrupole interaction, we predict qualitatively a noticeable reduction of the TLS contribution to the dielectric constant at low temperatures. We believe that this reduction can explain the plateau in the temperature dependence of the dielectric constant.

It follows from the above analysis that in the whole energy interval

$$\Delta_{0*} = \begin{cases} \Delta_0, & \Delta_0 \gg nb, \\ \Delta_0 \eta^n, & \Delta_0 \ll nb. \end{cases} \quad (32)$$

This renormalization results in a gap in the distribution of the effective tunneling amplitude Δ_{0*} of tunneling systems

$$P(\Delta_{0*}) = \begin{cases} \dfrac{P}{\Delta_{0*}}, & \Delta_{0*} \gg nb, \\ 0, & nb\eta^n \ll \Delta_{0*} \ll nb, \\ \dfrac{P}{\Delta_{0*}}, & \Delta_{0*} \ll nb\eta^n. \end{cases} \quad (33)$$

Using the latter result to estimate the dielectric constant temperature dependence in the expression similar to Eq. (31), one can obtain the plateau in the temperature dependence of the dielectric constant within the range $nb\eta^n < T < nb$. At $T > nb$ one should use the standard tunneling model result (28), while at $T < nb\eta^n$ the resonant dielectric constant obeys Eq. (31). Thus, our qualitative arguments can explain the behavior observed experimentally. In the following part of this paper we investigate the two regimes of Eqs. (28) and (31) and the crossover between them with the higher accuracy.

IV. PERTURBATION THEORY APPROACH

The renormalization of a tunneling amplitude Δ_0 affects the thermal and kinetic properties of the TLS ensemble and provides a minimum energy splitting of TLS's having zero asymmetry $\Delta = 0$. In particular, the resonant susceptibility is determined by similar resonant TLS's with small asymmetry $|\Delta| < \Delta_0$. Therefore, we will study the most relevant case, i.e., $\Delta = 0$. The resonant permittivity we are interested in here is determined by the ground state of the tunneling systems with the energy difference between the ground and first excited states larger than the thermal energy. In this case the contribution of higher excited states is insignificant because due to an exponentially small probability of their occupation. A positive contribution of a ground state to the dielectric constant reflects the general fact that the ground state minimizes the energy of the system. Thus, the external electrical field aligns the ground state dipole moment along the field direction. The susceptibility of excited states can be negative as in the case of Eq. (27) and also in the case of TLS's is affected by the nuclear quadrupole interaction as we will show below. Therefore, when the temperature becomes comparable or larger than energy splitting between ground and excited states, TLS susceptibility becomes negligible. Thus, a structure of the ground state is only important for the resonant susceptibility of the system.

Next, we consider the effect of the quadrupole splitting on the ground state of the tunneling system. In the case of vanishing asymmetry energy $\Delta = 0$ the Hamiltonian of the tunneling system can be represented in the form

$$H = \frac{\hat{H}_r + \hat{H}_l}{2} + s^z(\hat{H}_r - \hat{H}_l) - \Delta_0 s^x. \qquad (34)$$

The total wave function of a tunneling particle is a product of the coordinate (pseudospin) wave function and the nuclear one.

If the tunneling amplitude Δ_0 is large, the ground state is described by the wave function $|g_{hyb}\rangle$ whose coordinate part is symmetric and the tunneling particles are shared equally between the two wells. In this case one has $\langle s^x \rangle \approx 1/2$, $\langle s^z \rangle \approx 0$ and the energy of the nuclear quadrupoles is given by the "mean" Hamiltonian

$$\frac{\hat{H}_r + \hat{H}_l}{2}. \qquad (35)$$

This regime is called the hybridized one.

In the opposite limit of the small tunneling amplitude, i.e., in the localized regime, one can neglect the tunneling term s^x in Eq. (34) and the energy minimum corresponds to either the ground state of the Hamiltonian $\hat{H}_l$ or to the ground state of the Hamiltonian $\hat{H}_r$. Then, the influence of the tunneling s^x term on the ground state is insignificant due to the strong reduction in the effective tunneling amplitude [see Eq. (32)] because of the small factor η^n. Let us find the crossover between the hybridized and localized regimes.

In the hybridized regime the ground-state energy is

$$-\frac{\Delta_0}{2} + \frac{1}{2}\langle g_{hyb}|\hat{H}_r + \hat{H}_l|g_{hyb}\rangle, \qquad (36)$$

while in the localized regime it is

$$\langle g_r|\hat{H}_r|g_r\rangle = \langle g_l|\hat{H}_l|g_l\rangle. \qquad (37)$$

Let us introduce a parameter λ_* describing the ground-state quadrupole energy difference between the two limiting regimes. It is the reorganization energy corresponding to the transition from the hybridized state to the localized one

$$\lambda_* = -\langle g_l|\hat{H}_l|g_l\rangle + \left\langle g_{hyb}\left|\frac{\hat{H}_r + \hat{H}_l}{2}\right|g_{hyb}\right\rangle. \qquad (38)$$

Comparing Eqs. (36) and (37), one finds that the hybridization regime is realized if

$$\Delta_0 > 2\lambda_*, \qquad (39)$$

while in the localized regime this inequality changes its sign, i.e.,

$$\Delta_0 < 2\lambda_*. \qquad (40)$$

Let n be the number of atoms of a TLS experiencing nuclear quadrupole interaction. The parameter

$$b_* = \lambda_*/n \qquad (41)$$

represents the reorganization energy per a tunneling atom. Let us estimate the parameter b_* for the case when the quadrupole interaction is described by Eq. (5). One has

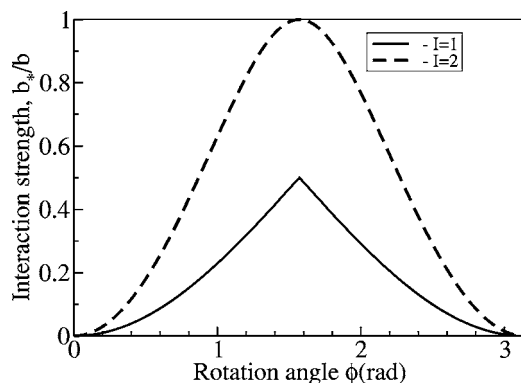$$\hat{H}_l = b[I_x^2 - I(I+1)/3], \qquad (42)$$

FIG. 3. Effective interaction energy b_* of nuclear spins vs angle between the nuclear quantization axis in two wells.

$$\hat{H}_r = b[(I_x \cos\phi + I_y \sin\phi)^2 - I(I+1)/3]. \qquad (43)$$

Then one can calculate the parameter b_* by using Eqs. (38), (42), and (43). To be more specific, consider the case $b > 0$ and integer spin $I = 1, 2$. The results of the numerical calculation of a single atom parameter b_* in the model Eqs. (42) and (43) for different rotation (mismatch) angles are represented in Fig. 3. One can see that the reorganization energy is always comparable with the nuclear quadrupole interaction energy.

In the hybridization regime (39) the quadrupole interaction is a weak perturbation. Then, the multiplet structure proves to be insignificant when one calculates the contributions of these tunneling systems to the susceptibility. In other words, the latter can be calculated within the standard TLS approach.

However, in the localized regime Eq. (40) a multiplet structure is the decisive feature. In this regime a renormalization of the tunneling amplitude (32) becomes important. If a tunneling amplitude Δ_0 is small enough, the ground state will be a superposition of the ground states in the isolated left and right wells (see Fig. 2). Let us estimate the upper limit of Δ_0 below which the two-level approximation for the ground state is still valid.

The nuclear-spin ground state in each well can be treated as nondegenerate (see Sec. II B). One can approximately construct the ground state and the lowest excited state of the tunneling system as a superposition of the unperturbed ground states in the two wells. The contribution of higher excited states are neglected. This is justified by calculating their contribution to a lowest-order perturbation theory. We estimate the correction factor c for the ground-state amplitude as

$$c = 1 - \sum_{i \neq gr} \frac{\Delta_0^2|\langle gl|i\rangle|^2}{(E_i - E_{gl})^2}, \qquad (44)$$

where i labels all states of the right well.

We determine the parameters of the regime for which the second term can be neglected as follows. The lowest excited states in each well are separated from the ground state by some characteristic energy $b \sim \hbar\omega_0$, where ω_0 is a frequency of the nuclear quadrupole resonance. The next group of

states is separated by the energy gap $\sim 2b$. The vast majority of states have energies exceeding that for the ground state by the energy $\lambda \simeq nb$. Because of the large statistical weight, these states provide the main contribution to Eq. (44). Therefore we may replace the denominator in Eq. (44) by λ. Then, the sum of the overlap factors $|\langle gl|i\rangle|^2$ in the numerator can be well approximated by unity. The requirement that the second term in Eq. (44) is small results in the condition

$$\Delta_0 < 2\lambda. \tag{45}$$

This condition is weaker than Eq. (40). Therefore, $c \approx 1$ is justified when Eq. (40) is fulfilled and one can ignore the excited states in both wells.

Thus we can specify two domains of system parameters. One is given by large tunneling amplitudes $\Delta_0 > 2\lambda_*$, where the nuclear quadrupole interaction can be ignored. The other is given by small tunneling amplitudes $\Delta_0 < 2\lambda_*$. In the latter case we can restrict our consideration to the pair formed by the two lowest-energy states. These eigenstates are superpositions of the ground states in the left and right potential wells. A nonperturbative approach developed in the next section, where nuclear spins are replaced with oscillators, leads to the similar results only with some minor deviations.

At this point we turn to the analysis of the dielectric constant using the structure of TLS ground states described above. For an improved analysis, it is convenient to write the resonant dielectric constant Eq. (22) in the following form:

$$\chi = \frac{\mu_0^2}{3}\sum_a \langle P_a(\Delta_0, \Delta; T)\chi_a(\Delta_0 = 0, \Delta)\rangle + \frac{\mu_0^2}{3}\sum_a \langle [P_a(\Delta_0, \Delta; T)$$
$$- P_a(\Delta_0, \Delta; 0)] \times [\chi_a(\Delta_0, \Delta) - \chi_a(\Delta_0 = 0, \Delta)]\rangle$$
$$+ \frac{\mu_0^2}{3}\sum_a \langle P_a(\Delta_0, \Delta; 0)[\chi_a(\Delta_0, \Delta) - \chi_a(\Delta_0 = 0, \Delta)]\rangle. \tag{46}$$

The logarithmic temperature dependence, often serving as an evidence for the TLS effects,[3] comes entirely from the first term. This is due to logarithmically uniform distribution of the tunneling amplitudes (2). The logarithmic divergence of the first term is suppressed for TLS's with the energies of the order of the thermal energy due to the compensation of a positive contribution from the ground state by negative contributions of excited states (see below). The third contribution is a temperature-independent constant. We will ignore it as a background correction to the susceptibility which does not affect its temperature dependence.

In the absence of the nuclear quadrupole interaction the important first and second terms can be evaluated. One can reexpress the first "logarithmic" term as

$$\chi_{\text{log}} = \frac{P\mu_0^2}{3}\int_0^W \frac{d\Delta_0}{\Delta_0}\tanh[\Delta_0/(2T)] \tag{47}$$

and the second "thermal" contribution as

$$\chi_T = \frac{P\mu_0^2}{3}\int_0^\infty \frac{d\Delta_0}{\Delta_0}\int_0^\infty d\Delta \left[\tanh\left(\frac{\sqrt{\Delta^2 + \Delta_0^2}}{2T}\right) - 1\right]$$
$$\times \left(\frac{\Delta_0^2}{2(\Delta^2 + \Delta_0^2)^{3/2}} - \delta(\Delta)\right). \tag{48}$$

In the second integral the upper cutoff for the tunneling amplitude is replaced by ∞ because the integrand decreases exponentially at large energies.

The first contribution can be written as

$$\chi_{\text{log}} = \frac{P\mu_0^2}{3}\left[\ln\left(\frac{W}{2T}\right) - I_1\right],$$

$$I_1 \approx \int_0^\infty \frac{\ln(x)dx}{\cosh^2(x)} \approx -0.82. \tag{49}$$

The "thermal" contribution is given by the dimensionless integral

$$\chi_T = \frac{P\mu_0^2}{3}\int_0^{+\infty} \frac{dx}{x}\int_{-\infty}^{+\infty} dy[\tanh(\sqrt{x^2 + y^2} - 1)]$$
$$\times \left(\frac{1}{2}\frac{x^2}{(x^2 + y^2)^{3/2}} - \delta(y)\right). \tag{50}$$

It vanishes because the upper limit of the integration over x has been replaced by ∞. This can be demonstrated by using, for instance, the trigonometric substitution $x = \alpha \cos(\beta), y = \alpha \sin(\beta)$ and evaluating the integral over β first.[25]

Below we calculate the susceptibility of tunneling systems for different temperature regimes. According to the above analysis [see Eqs. (39) and (40)] it is convenient to divide all tunneling systems into two parts depending on either $\Delta_0 > 2\lambda_*$ or $\Delta_0 < 2\lambda_*$.

In the first case $\Delta_0 > nb_*$ the effect of the nuclear quadrupole interaction is negligible and one can use the standard two-level approximation in order to estimate the contribution from these TLS's into the susceptibility as

$$I_1 = \int_{2\lambda_*}^W \frac{d\Delta_0}{\Delta_0}\tanh\left(\frac{E(\Delta_0, 0)}{2T}\right). \tag{51}$$

Consider temperatures $T > 2\lambda_*$. With $E(\Delta_0, 0) = \Delta_0$ one can estimate this integral as follows [see Eq. (49)]

$$I_1 \approx \ln\frac{W}{2T} - \frac{\lambda_*}{T} + 0.82. \tag{52}$$

The second term on the right-hand side is small in comparison with the first one and can be omitted. If the temperature is low $T < 2\lambda_*$, the tangent in Eq. (51) equals unity and we have

$$I_1 = \ln\frac{W}{nb_*}. \tag{53}$$

Now consider the second group of tunneling systems for which $\Delta_0 < \lambda_*$. For them the multiplet structure becomes important and we can confine ourselves to ground state levels

of the multiplet in each well. These levels are coupled by the tunneling amplitude $\Delta_0 \eta^n$. Then one has

$$I_2 = \int_0^{2\lambda_*} \frac{d\Delta_0}{\Delta_0} \tanh\left(\frac{\Delta_0 \eta^n}{2T}\right). \qquad (54)$$

Assume that $T < 2\lambda_* \eta^n$. Then

$$I_2 \approx \ln \frac{n b_*}{T/\eta^n}. \qquad (55)$$

This expression should be added to the term I_1 given by Eq. (53), also giving a contribution the temperature region considered. Thus, the total integral is given by

$$I_1 + I_2 = \ln \frac{W \eta^n}{T}. \qquad (56)$$

In the intermediate temperature range $2\lambda_* > T > 2\lambda_* \eta^n$ Eq. (54) is inapplicable strictly speaking. However, we expect that the contribution to the dielectric permittivity within the range $2\lambda_* > \Delta_0 > 2\lambda_* \eta^n$ is small because this range corresponds to the gap in the distribution of effective tunneling amplitudes [Eq. (33)]. In addition, the lower limit of the low-temperature dielectric constant (56) coincides with the upper one of the dielectric constant in the high-temperature range [Eq. (52)]. Thus, in the whole temperature region one has

$$\varepsilon_{\text{res}} \approx (P\mu^2/3)\ln(W/T), \quad 2\lambda_* < T,$$

$$\varepsilon_{\text{res}} \approx (P\mu^2/3)\ln(W/2\lambda_*), \quad 2\lambda_* \eta^n < T < 2\lambda_*,$$

$$\varepsilon_{\text{res}} \approx (P\mu^2/3)\ln(W\eta^n/T), \quad T < 2\lambda_* \eta^n. \qquad (57)$$

Our perturbation theory analysis is a phenomenological one and cannot be exact because we do not know the nuclear spin Hamiltonian. However, it is expected that the main qualitative features of the systems behavior are reproduced. For a more quantitative and non-perturbative analysis given in the next section, we replace nuclear spins with oscillators and investigate the tunneling amplitude behavior in such a toy model.

V. TOY MODEL

Since the nuclear spin interaction in the right and left wells is unknown, we consider instead a toy model in which nuclear spins are replaced by classical oscillators. This "bosonization" approach to nuclear spins is justified when one deals with the low energy states of many spins. In particular, it has been used by Prokofev and Stamp to investigate the nuclear spin interaction effect on the large spin tunneling.[26] We consider a symmetric TLS ($\Delta = 0$) characterized by a coherent tunneling amplitude Δ_0 and coupled to n oscillators representing the nuclear spins of n tunneling atoms forming the TLS concerned. All oscillators have the frequency Ω and the mass M. These n oscillators are linearly coupled to the TLS and have the shifted equilibrium positions $x_i = \pm x_0/2$, $i = 1, \ldots, n$ when the TLS occupies the right or left wells, respectively. Then the Hamiltonian of the system can be expressed as

$$\hat{H} = -\Delta_0 s^x + \sum_{i=1}^{n}\left(\frac{p_i^2}{2M} + \frac{M\Omega^2 x_i^2}{2}\right) - s^z M\Omega^2 x_0 \sum_{i=1}^{n} x_i. \qquad (58)$$

The spin values $s^z = \pm 1/2$ stand for the TLS residing in the right or left wells, respectively. The models similar to Eq. (58) have extensively been studied within the polaron theory (see e.g., Refs. 27 and 28, and references therein). Following the standard approach one can determine an approximate ground state of the problem by minimizing the "classical" part of Eq. (58). This excludes the kinetic energy term of the oscillators. The "classical" energy of the system can be expressed as

$$E_{\text{cl}} = -\frac{1}{2}\sqrt{\Delta_0^2 + \left(M\Omega^2 x_0 \sum_{i=1}^{n} x_i\right)^2} + \sum_{i=1}^{n}\left(\frac{M\Omega^2 x_i^2}{2}\right), \qquad (59)$$

where the spin-Hamiltonian has been replaced by its ground-state energy, thereby using the relationship $E_{\text{ground}} = -\frac{1}{2}\sqrt{\Delta^2 + \Delta_0^2}$ for the Hamiltonian $-\Delta s^z - \Delta_0 s^x$. Oscillators are treated classically. This is justified when their number is large, i.e., $n \gg 1$. We suppose that this regime is applicable here.

The total energy can be minimized with respect to the oscillator coordinates x_i. Then, one has for the derivatives of the classical energy (59) with respect to all coordinates

$$0 = \frac{\partial E_{\text{cl}}}{\partial x_i} = -\frac{(M\Omega^2 x_0)^2 X}{2\sqrt{\Delta_0^2 + (M\Omega^2 x_0 X)^2}} + M\Omega^2 x_i, \quad X = \sum_{i=1}^{n} x_i. \qquad (60)$$

These equations can be solved analytically by taking into account that at the energy minimum all equilibrium coordinates x_i are identical, that is $x_i = X/n$. The following relations are found:

$$\Delta_0 > 2\lambda_* : X = 0,$$

$$\Delta_0 < 2\lambda_* : X = \pm \frac{n x_0}{2}\sqrt{1 - \left(\frac{\Delta_0}{2\lambda_*}\right)^2},$$

$$\lambda_* = n b_*, \quad b_* = M\Omega_0^2 x_0^2/2. \qquad (61)$$

The quantity b_* has been introduced to describe the change in the oscillator energy induced by its interaction with TLS. For spins, this energy is equivalent to the quadrupole splitting energy b_* [see Eq. (41)].

Thus, depending on the relationship between the oscillator energy λ_* and the tunneling amplitude Δ_0, a TLS ground state can have different structures. When tunneling is stronger than the TLS interaction with oscillators, i.e., when $\Delta_0 > 2\lambda_*$, the tunneling atoms are distributed equally between the two wells and all oscillators have their energy minimum at $x_i = 0$. This state is energetically most favorable because an identical internal structure for the left and right states minimizes the energy of the system. In other words, the tunneling of TLS is so fast that the oscillators see it in its average

position in both wells simultaneously. This state is equivalent to the hybridized state considered previously in Sec. IV.

In the opposite case of weak tunneling $\Delta_0 < 2\lambda_*$ the symmetry between the right and left wells is broken because of a strong displacement of oscillators. In this case the system has two energy minima depending on the sign in the definition of the displacement X in Eq. (61). For the nuclear quadrupole interactions, these two states are represented by the nuclear spin configurations minimizing the energy of TLS's localized either in the right or left wells.

Both the energy minima are still coupled by tunneling, but the tunneling amplitude is much weaker then in the case of vanishing oscillator displacements. Consider the non-adiabatic tunneling regime applicable to the nuclear spins.[26] In this case we may express the effective tunneling amplitude Δ_{0*} as the product of the coherent tunneling amplitude Δ_0 and the overlap integral $\langle l | r \rangle$ of the left and right states of the environment, i.e.,

$$\Delta_{0*} = \Delta_0 \langle l | r \rangle. \tag{62}$$

In order to estimate the overlap integral, one can use the harmonic approach. We will use it for the wave functions in the left and right wells and consider the case of zero temperature, reasonable if $T < 2\lambda_*$. The domain $T \approx 2\lambda_*$, where excited states are important, is relatively narrow at the large number of oscillators n and can be approximately ignored due to a logarithmically weak dependence of the dielectric constant on T.

In the harmonic approach one may expand the energy (59) near the local minima given by (61) up to the second order in a coordinate displacement. This expansion can be written as

$$E_{\mathrm{cl}}(X) \approx -\lambda_* + \frac{\lambda_*}{2}\left[1 - \left(\frac{\Delta_0}{2\lambda_*}\right)^2 \right]$$

$$+ \frac{1}{2}\sum_{i,j} A_{ij}(x_i - X/n)(x_j - X/n);$$

$$A_{ij} = \frac{\partial^2 E_{\mathrm{cl}}}{\partial x_i \partial x_k} = \frac{4b_*}{x_0^2}\left[\delta_{ij} - \frac{1}{n}\left(\frac{\Delta_0}{2\lambda_*}\right)^2 \right], \tag{63}$$

where δ_{ij} is the Kronecker symbol. The expansion is written near the potential minimum at the right well, while for the left well the sign of X should be changed to the opposite.

The harmonic part of the Hamiltonian (63) can be represented by n independent modes including the symmetric mode

$$u = \frac{1}{\sqrt{n}}\sum_{i=1}^{n} x_i, \quad \Omega_s = \Omega_0 \sqrt{1 - \frac{\Delta_0^2}{\lambda_*}}, \tag{64}$$

and $n-1$ asymmetric, degenerate modes α

$$u_\alpha = \sum_{i=1}^{n} c_i^\alpha x_i, \quad \Omega_a = \Omega_0, \quad \sum_i c_i^\alpha = 0, \quad \sum_i |c_i^\alpha|^2 = 1. \tag{65}$$

A factor $1/\sqrt{n}$ is introduced in Eq. (64) to conserve the commutation rules between the coordinate and momentum opera-

tors. Only the symmetric mode interacts with the tunneling motion between the right and left minima, while asymmetric modes remain unchanged and can therefore be ignored. Therefore the overlap integral in Eq. (62) is given by the overlap between two ground-state wave functions of the symmetric harmonic modes with the mass M and the frequency Ω_s, and the equilibrium positions shifted by $\pm |X|/\sqrt{n}$ [Eq. (61)] from the origin. Using the Gaussian wave functions for oscillator ground states, one can express this integral in the form

$$\langle l | r \rangle = \exp\left(-\frac{M\Omega X^2}{n\hbar} \right) = \exp\left\{ -\frac{nM\Omega x_0^2}{\hbar}\left[1 - \left(\frac{\Delta_0}{\lambda_*}\right)^2 \right]^{3/2} \right\}. \tag{66}$$

This result can be used to characterize approximately the tunneling of a TLS consisting of n atoms coupled to the nuclear spins. The overlap integral for a single oscillator i is

$$\eta = \langle li | ri \rangle = \exp\left(-\frac{M\Omega x_0^2}{\hbar} \right) \tag{67}$$

in analogy to the single-spin overlap integral introduced before. The parameter λ_* can be represented by the single atom quadrupole splitting energy multiplied by the number of atoms in a TLS, i.e., by

$$\lambda_* \approx nb_*. \tag{68}$$

Thus we may use the following approximate relationship between the initial (coherent) tunneling amplitude Δ_0 and the effective tunneling amplitude Δ_{0*}:

$$\Delta_{0*} \approx \Delta_0 \exp\left\{ -n \ln(1/\eta)\left[1 - \left(\frac{\Delta_0}{\lambda_*}\right)^2 \right]^{3/2} \right\}. \tag{69}$$

In the following we will use this relationship in order to describe the dielectric response of TLS.

The tunneling amplitude distribution modified by Eq. (69) has a dip (pseudogap) due to sharp decrease in Δ_{0*} when the coupling strength Δ_0 becomes smaller than the effective rearrangement energy $\lambda_* = nb_*$. We can define the modified distribution of TLS's over Δ_{0*} for $\Delta_{0*} < \lambda_*$ by using

$$P(\Delta_{0*}) \propto \int_0^\infty \frac{d\Delta_0}{\Delta_0} \times \delta\left(\Delta_{0*} - \Delta_0 \right.$$

$$\left. \times \exp\left\{ -n \ln(\eta)\left[1 - \left(\frac{\Delta_0}{\lambda_*}\right)^2 \right]^{3/2} \right\} \right) \tag{70}$$

$$= \frac{1}{\Delta_{0*}}\frac{1}{1 + 3n\ln(1/\eta)\left(\dfrac{\tilde{\Delta}_0}{\lambda_*}\right)^2 \sqrt{1 - \left(\dfrac{\tilde{\Delta}_0}{\lambda_*}\right)^2}}, \tag{71}$$

where the amplitude $\tilde{\Delta}_0$ is an implicit function of Δ_{0*} defined by Eq. (69). In the case of a large tunneling amplitude $\Delta_{0*} > \lambda_*$ the distribution is logarithmically uniform because $\Delta_0 = \Delta_{0*}$ in that regime.

For the proof of Eq. (71) we employ

$$\delta[a - \phi(x)] = \frac{1}{\phi'(\tilde{x})} \delta(x - \tilde{x}), \qquad (72)$$

where $\tilde{x}$ is the solution of equation $a - \phi(x) = 0$. We identify

$$a = \Delta_{0*}, \quad x = \Delta_0, \qquad (73)$$

$$\phi(x) = x \exp\left\{- n \ln(\eta)\left[1 - \left(\frac{x}{\lambda_*}\right)^2\right]^{3/2}\right\} \qquad (74)$$

and derive

$$\phi'(\Delta_0) = \exp\left\{- n \ln(\eta)\left[1 - \left(\frac{\Delta_0}{\lambda_*}\right)^2\right]^{3/2}\right\}$$
$$\times \left\{1 + 3n \ln(\eta)\left[1 - \left(\frac{\Delta_0}{\lambda_*}\right)^2\right]^{1/2}\left(\frac{\Delta_0}{\lambda_*}\right)^2\right\}. \qquad (75)$$

By substituting Eq. (69) into (75), we obtain

$$\phi'(\Delta_0) = \frac{\Delta_{0*}}{\Delta_0}\left\{1 + 3n \ln(\eta)\left[1 - \left(\frac{\Delta_0}{\lambda_*}\right)^2\right]^{1/2}\left(\frac{\Delta_0}{\lambda_*}\right)^2\right\}. \qquad (76)$$

When this expression is put into Eq. (72) and then into Eq. (70), we obtain

$$\int_0^\infty \frac{d\Delta_0}{\Delta_0} \frac{1}{\frac{\Delta_{0*}}{\Delta_0}\left\{1 + 3n \ln(\eta)\left[1 - \left(\frac{\Delta_0}{\lambda_*}\right)^2\right]^{1/2}\left(\frac{\Delta_0}{\lambda_*}\right)^2\right\}_{\Delta_0 = \tilde{\Delta}_0}} \delta(\Delta_0 - \tilde{\Delta}_0)$$

which coincides with Eq. (71). The modified distribution of tunneling amplitudes $W(\ln(\Delta_{0*})) = P(\Delta_{0*})\Delta_{0*}$ is shown in Figs. 4 and 5.

We have chosen a representative value of $\phi = 1.23$ rad for the rotation angle [see Eq. (9)] in a field of axial symmetry bI_a^2 and $b > 0$. In accordance with Eq. (9) and Fig. 3 the effective interaction parameters are $\eta = 1/3$ and $b_* = 0.33$ mK for $I=1$, and $\eta = 1/3$ and $b_* = 0.85$ mK for $I=2$. The nuclear quadrupole effect shows up at an energy which is about three times larger for $I=2$ then for $I=1$.

One can see from Figs. 4 and 5 that the dip in the distribution of tunneling amplitudes appears in the domain $\{\lambda_* \exp[-n \ln(1/\eta)], \lambda_*\}$. This becomes sizable for $n=4$. A similar behavior is found for the other values of the overlap integral η. This dip causes a change of the temperature de-

pendence of the resonant susceptibility as discussed below.

The resonant dielectric susceptibility of the TLS ensemble can be estimated from the "logarithmic" integral

$$\delta\varepsilon \approx \int_0^W \frac{d\Delta_{0*}}{\Delta_{0*}} P(\Delta_{0*})\tanh\left(\frac{\Delta_{0*}}{2T}\right). \qquad (77)$$

By integrating the distribution with the dip we obtain the results shown in Figs. 6 and 7.

The quadrupole interaction parameter $b = 1$ mK and the rotation angle $\phi \sim 1.23$ rad are the same as before. One notices a plateau in the dielectric constant separating two logarithmic temperature dependences characterized by the same slope at sufficiently large number n of atoms per single TLS. In the experiment[15,16] a plateau is found for $T < 5$ mK, while
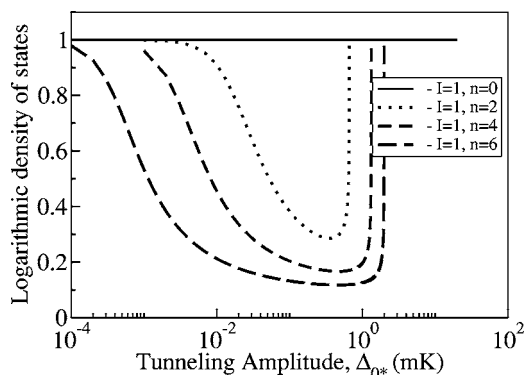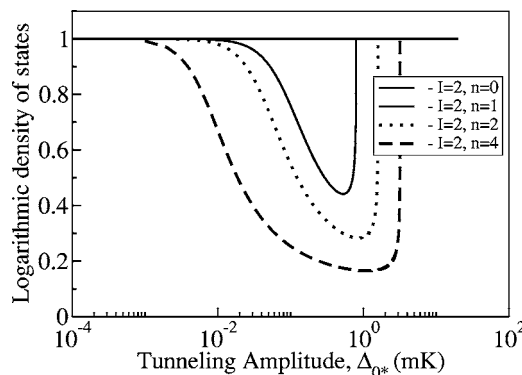

FIG. 4. The dip in the logarithmic TLS distribution over tunneling amplitudes due to the nuclear quadrupole interaction. The logarithmic density of states $P(\Delta_0^*)\Delta_0^*$ is shown for $I=1$ and different numbers n of tunneling atoms. The case $n=0$ corresponds to the lack of the interaction. The rotation angle is set to 1.23 rad.

FIG. 5. The dip in the logarithmic TLS distribution over tunneling amplitude due to the nuclear quadrupole interactions. The logarithmic density of states $P(\Delta_{0*})\Delta_{0*}$ is shown for $I=2$ and different numbers n of tunneling atoms per TLS.
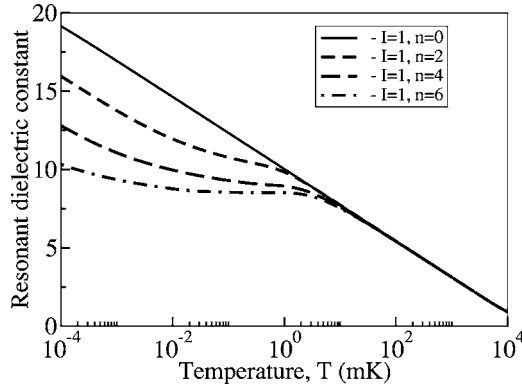
FIG. 6. Resonant dielectric constant affected by the nuclear quadrupole interaction in the case of the nuclear spin $I=1$.

the low-temperature edge of the plateau has not been observed because it requires much lower temperature than those reached experimentally. A temperature of 5 mK gives a reasonable estimate of the quadrupole interaction energy.

In all cases the plateau begins at the characteristic maximum temperature

$$T_{max} \simeq \lambda_*, \tag{78}$$

and extends down to the characteristic minimum temperature

$$T_{min} \simeq \lambda_* \eta^n. \tag{79}$$

For $T < T_{min}$ the behavior of the standard tunneling model is restored and the dielectric constant shows a logarithmic temperature dependence with the same slope as at high temperatures $T > T_{max}$. The logarithmic width of the plateau $\ln(T_{max}/T_{min})$ is thus directly proportional to the number of atoms per single TLS.

We expect that in the plateau regime the dielectric constant can be controlled by the external magnetic field. In fact, the overlap integral of the nuclear spin states in the left and right well can be affected by the magnetic field. This effect is discussed in the next section.
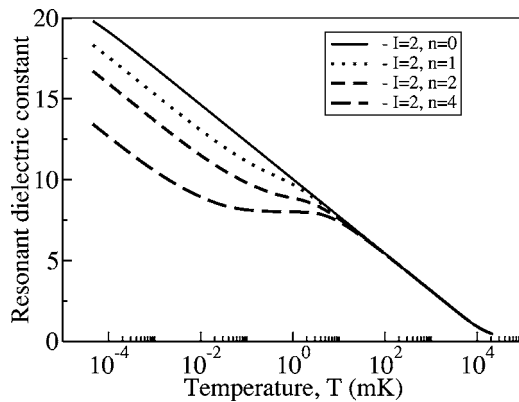

FIG. 7. Resonant dielectric constant affected by the nuclear quadrupole interaction in the case of the nuclear spin $I=2$.

VI. EFFECT OF THE EXTERNAL MAGNETIC FIELD ON THE ANOMALOUS DIELECTRIC PROPERTIES AT ULTRALOW TEMPERATURES

The application of the external magnetic field affects the orientation of nuclear magnetic moments. When Zeeman splitting becomes comparable with the nuclear quadrupole interaction, the quantization axes in both potential wells of the given TLS are aligned with the direction of the magnetic field. Accordingly, the mismatch of nuclear quadrupole axes will be reduced by the field and the overlap integral of nuclear quadrupole states in different wells increases and approaches unity in the high-field limit. The effective interaction constant b_* vanishes in that case. Thus, high magnetic field reduces the influence of the nuclear quadrupole interaction and the dielectric constant behaves similar to the standard tunneling model. In this section we estimate the effect of the external field on the low-temperature resonant dielectric constant by using the simple oscillator and axially symmetric models for the nuclear quadrupole interaction formulated in previous sections.

Consider the effect of the external magnetic field on parameters characterizing the mismatch of the nuclear spins in the right and left wells. The set of parameters necessary to characterize the given TLS includes the overlap integral for single nuclear spin η, the number n of nuclear spins involved into the tunneling process, and the characteristic nuclear spin interaction energy b_* [see Eq. (69)]. The number n is field independent, while the overlap integral η and the single atom interaction constant b_* are subjected to the changes with the external field. We investigated numerically the change of the parameters η and b_* in an external field for the axially symmetric quadrupole interaction. The nuclear spins are $I=1$ and $I=2$ with the same angle $\cos(\phi) \approx 1/3$ and the nuclear quadrupole interaction constant $b=1$ mK as has been assumed in previous sections. The sign of the nuclear quadrupole interaction constant $b>0$ is chosen as previously. Therefore, the ground state has the zero spin projection onto the quantization axis. The zero-field dielectric constant for these regimes is shown in Figs. 6 and 7. We have calculated numerically the dependence on the external magnetic field B of the overlap integral between the nuclear spin ground states in the left and right wells. Also we have estimated numerically the dependence of the effective interaction constant on the magnetic field. The calculations have been made as follows. The quadrupole Hamiltonian is chosen in the form $b(I^a)^2$ with a the left-well axis $a=x$ and the right-well axis rotated by the angle $\phi=1.23$ rad in the x-y plane with respect to the x direction. Then the Zeeman term describing the interaction of the nuclear magnetic moment with magnetic field, is augmented to the Hamiltonian (5). The magnetic field with a fixed absolute value B has been generated in the random directions. The overlap integral of ground states η as well as the effective interaction constant b_* have been computed and then averaged over $\sim 10^4$ realizations of the random field. The results of calculations are shown in Figs. 8 and 9.

We express the magnetic field in units of the Zeeman energy splitting (mK). In fact, it is unclear which nuclear spins are most important in the experiment. A reasonable scale we use for estimates is about 1 mK spin splitting in a field of $B \sim 5$ T.
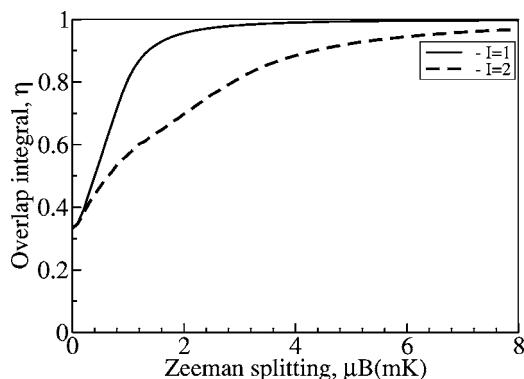
FIG. 8. The effect of the magnetic field on the overlap integral. The 1 mK Zeeman splitting approximately corresponds to field 5 T.

FIG. 10. Effect of a magnetic field on the dielectric constant in the case of spin $I=1$ and the number of atoms per TLS $n=4$.

As is clear from Figs. 8 and 9, the overlap integral increases monotonously with the field and approaches unity when $\mu B \gg 1$ mK. The effective interaction constant b_* vanishes in the same limit. Therefore, the application of magnetic field results in the disappearance of the plateau in the temperature dependence of the dielectric constant and the standard logarithmic dependence is restored. To examine this effect, we compute the temperature dependence of the TLS resonant dielectric constant at various fields. We have used Eq. (77) with Eq. (69) for the effective tunneling amplitude and input parameters η and b_* obtained from our field-dependent calculations (Figs. 8 and 9).

The field-dependent dielectric constant is shown in Figs. 10 and 11 for $n=4$ atoms per single TLS and nuclear spins $I=1$ and $I=2$, respectively. The field effect is similar in both cases. However, a stronger field is needed to suppress the nuclear quadrupole interaction in the case of larger spin $I=2$.

We can interpret the field dependence as follows. A relatively small field $\mu B \leq 0.2$ mK and $B \leq 1$ T affects the resonant dielectric constant only for low temperatures $T \leq 5$ mK. At higher temperatures the field effect cannot be viewed beeing a small correction. The further increase of the field leads to linear reduction in the width of the plateau from both the high- and low-temperature sides. When the field is sufficiently high $\mu B \geq 1$ mK, and $B \geq 5$ T, it competes with the nuclear quadrupole splitting and the plateau narrows by orders of magnitude and almost vanishes at $\mu B \sim 4$ mK.
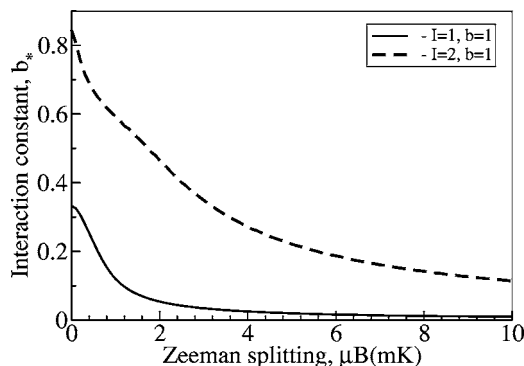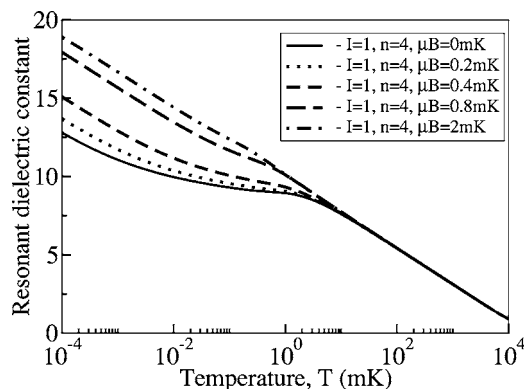
Then the standard tunneling model behavior is restored. It would be very interesting to perform measurements of the dielectric constant at very low temperatures, i.e., for $T \leq 5$ mK and a strong magnetic field around 10 T. In this case the plateau in the dielectric constant should disappear. We hope that our work will stimulate such measurements.

VII. RELATIONSHIP OF THE MODEL TO REAL SYSTEMS

In this section we address two questions. First question is whether our choice of parameters for the nuclear quadrupole interaction is justified and these parameters can be used to fit the existing experimental data. The second question is related to glasses having no (or negligable) nuclear quadrupole interaction. These glasses (Mylar, SiO_2) should not show any anomalies in the dielectric constant temperature dependence. The experimental data related to these glasses are discussed below.

According to the previous consideration we expect that the plateau observed experimentally in the temperature dependence of the dielectric constant for $T \leq 5$ mK is due to the fact that the parameter nb [see Eq. (6)] amounts to a value of 5 mK. Experimentally, a qualitative change in the TLS resonant dielectric susceptibility is seen in the temperature range of $T \sim 5 - 10$ mK (see Refs. 13, 15, and 16). There is no problem to intepret the saturation behavior using our formal-

FIG. 9. The effect of magnetic field on the effective single-atom interaction constant b_*.
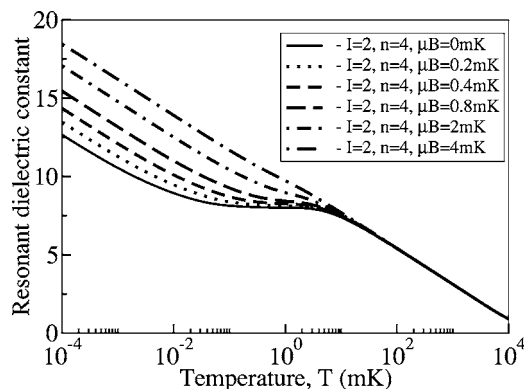

FIG. 11. The effect of magnetic field on the dielectric constant in the case of spin $I=1$ and the number of atoms per TLS $n=4$.

TABLE I. Nuclear spin, magnetic moment, quadrupole moment, and frequency of nuclear quadrupole resonance for different chemical elements, possibly participating in the tunneling.

Nucleus	I	μ_N	Q (barn)	$\hbar\omega_0$ (mK)
^{1}H	1/2	4.837	0	0
D=^{2}H	1	1.213	0.00286	0.7×10^{-2} Ref. 18
^{12}C	0	0	0	0
^{16}O	0	0	0	0
^{23}Na	3/2	2.863	0.104	0.14 (Na), 0.4 (NaF) Ref. 30
^{27}Al	5/2	4.309	0.147	0.9 in Al Ref. 31
^{29}Si	1/2	−0.962	0	0
^{29}K	3/2	0.505	0.0585	
^{135}Ba	3/2	1.82	0.160	0.86 (BaBiO$_3$) Ref. 32

TABLE II. Saturation temperature below which the dielectric constant ε and/or sound velocity v become temperature independent.

Glass	Nuclei	T_{sat} (mK)	Exp.	Refs.
Mylar (C$_{10}$H$_8$O$_4$)$_n$	No	<1 mK	ε	15
5% K-SiO$_2$	K	4 mK	ε	15
10% K-SiO$_2$	K	4 mK	ε	15
BK7 (Ref. 33)	Na	5 mK	ε	15
SiO$_x$	No	8 mK	ε	15
BaO-Al$_2$O$_3$-SiO$_2$	Al,Ba	~5 mK	ε	16
a-SiO$_2$	No	<2 mK	v	34 and 35
BK7	Na	<5 mK	v	36

ism within the experimental accuracy choosing properly the fitting parameters.[21] However, the question arises whether or not the nuclear quadrupole interaction is sufficiently strong to display itself in this temperature range. We also expect that glasses lacking the nuclear quadrupole interaction should not show any deviations from the standard model.

The nuclear quadrupole interaction constant b is defined by Eq. (6). The values of the electric field gradient are close to each other in different glasses, while the nuclear quadrupole moments Q differ strongly for different elements, leading to the broad distribution in the observed energies of the nuclear quadrupole resonance $\hbar\omega_0\sim b$ Ref. 29. The data for the chemical elements present in glasses for which the low temperature measurements have been made are summarized in Table I.

As one can see from Table I, the typical range of nuclear quadrupole interactions for Na, K, Al, Ba nuclei is around but slightly less than 1 mK. This interaction should be larger in glassy materials because the above-mentioned elements are bound with nonmetal atoms there. The covalent and ionic bonds are expected to be stronger than metallic bonds, in particular, due to the absence of screening. Therefore, their binding energy and, accordingly, the electric field gradient affecting the nuclear quadrupole interaction are expected to be larger in these glasses.

The experimental data indicating the strong changes in low-temperature dielectric and, probably, acoustic properties are summarized in Table II. The saturation in a temperature dependence of the dielectric constant below the temperature T_{sat} takes place in all materials containing Na, K, Al, or Ba which have relatively high quadrupole moments (see Table I). The high saturation temperature observed in BaO-Al$_2$O$_3$-SiO$_2$ is most likely due to large values of the Ba and Al quadrupole moments compared with the other elements in Table I.

The absence of the low-temperature saturation in the dielectric constant of Mylar is in the full agreement with the theory. Indeed, Mylar is an organic polymer composed of C, H, and O atoms, for which the most stable isotopes have vanishing nuclear quadrupole moments. Similarly according to the most recent experimental data[34,35] there is no saturation in the temperature dependence of the sound velocity in

a-SiO$_2$ having no nuclear quadrupole interaction. A saturation in the low-temperature dependence of the dielectric constant has also been observed in SiO$_x$ which poses a puzzle. One possible explanation is that unpaired electrons may be present in this material.[37] They act like nuclear quadrupole moments in that aspect.

The problem of interpretation of the acoustic experiment[36] is a knotty one. If the same two-level systems control the acoustic and dielectric behaviors of the glasses, the saturation should take place below the same T_{sat} both for the dielectric constant and for the sound velocity.[3] On one hand, there is no saturation in the logarithmic temperature dependence in α-SiO$_2$ having no nuclear quadrupoles in agreement with the theoretical expectations. It is difficult, however, to understand the absence of any deviations from the standard tunneling model for the sound velocity measurements in BK7 down to 5 mK Ref. 36. Although the major contribution to the dielectric constant and the sound velocity can be due to different subsets of TLS's possessing either larger dipole moments or stronger elastic coupling to lattice vibrations,[38] there must be some amount of TLS's containing quadrupole nuclei and contributing to both dielectric and acoustic properties. These TLS's should lead to an anomalous acoustic behavior. One possible explanation of the absence of any saturation effect is that the measurements have been made for $T>5$ mK, while $T_{\text{sat}}\approx5$ mK (see Table II). An extension of acoustic measurements in BK7 to lower temperatures should help to clarify the puzzling situation.

VIII. DISCUSSION AND CONCLUSIONS

In this work we have considered various aspects of the effect of the nuclear quadrupole interaction on the low-temperature properties of glasses. The significance of this interaction has been pointed out by Würger, Fleischmann, and Enss.[17] In the present paper this interaction has been employed to characterize the resonant dielectric susceptibility of amorphous solids at ultralow temperatures $T\leqslant5$ mK where major deviations from the predictions of the tunneling model are observed. The analysis of the typical interaction parameters shows that the nuclear quadrupole interaction is strong enough to explain these deviations. To our knowledge the materials having no nuclear quadrupole interaction and

no unpaired electrons, i.e., the organic polymer Mylar,[5] and α-SiO$_2$ Refs. 34 and 35 do not show the deviations from the logarithmic temperature dependence of a dielectric constant or a sound velocity down to the lowest temperatures accessible experimentally around $T \sim 1$ mK. These results support our theory.

Our theory uses the fact that the nuclear quadrupole interaction is generally different in the right and left wells of a two-well tunneling system. Therefore, it affects the coherent coupling between the wells. The overall quadrupole interaction effect is governed by the relative magnitude of two parameters, i.e., $\lambda_* = nb_*$ which is the characteristic nuclear quadrupole interaction strength of TLS's consisting of n atoms and the tunneling matrix element Δ_0 between the left and right wells. When $\Delta_0 > \lambda_*$, the nuclear quadrupole interaction can be neglected. At smaller tunneling amplitude $\Delta_0 < \lambda_*$ the effective tunneling coupling is reduced exponentially due to the small overlap between different nuclear spin eigenstates in different wells. This is similar to the polaron effect and we use this similarity in order to suggest a solvable toy model based on the replacement of the nuclear spins with oscillators.

The resonant dielectric constant is determined by the contribution of all resonant TLS's with a characteristic tunneling amplitude of order of energy, which varies from the thermal energy to some characteristic maximum value $T < \Delta_0 < W$. The deviations from the standard tunneling model are observed at temperatures comparable with the nuclear quadrupole interaction, i.e., for $T \simeq \lambda_*$. They show up as a temperature-independent plateau due to the breakdown of coherent tunneling in the energy range $T < \Delta_0 < \lambda_*$, where resonant TLS's do not exist. Therefore, TLS's with tunneling amplitudes belonging to this domain do not contribute to the resonant dielectric constant. Note that the hypothesis of the breakdown of coherent tunneling below $T \sim 10$ mK has been proposed by Enss and Hunklinger[39] assuming, however, that it is due to the interaction between TLS's. Our work suggests the valuable realization of their hypothesys.

The results agree qualitatively with a number of low-temperature dielectric measurements made by different groups. To obtain quantitative agreement for the temperature at which the plateau forms, we have to assume that the relevant two-level systems consist of, at least, four atoms ($n = 4$). This assumption agrees qualitatively with the TLS model based on the renormalization group theory[12] and is in line with molecular dynamics studies of glasses.[22]

At very low temperatures $T < T_{min}$ ($T_{min} < 0.1$ mK) the plateau in the resonant dielectric constant should go over into a logarithmic temperature dependence characterized by the same slope $d\varepsilon_{res}/d\ln(T)$ as found at high temperatures, i.e., for $T > T_{max} \sim \lambda_*$. Therefore, it is worth while attempting an experimental observation of this behavior. It is unclear whether it is possible to perform measurements at such low temperatures at present. A comparison of the measurements with the theory can be used to estimate the number n of tunneling atoms involved into single TLS because the logarithmic width of the plateau $\ln(T_{max}/T_{min})$ is approximately equal to n.

The nuclear quadrupole interaction should affect the sound velocity since it enters its resonant part in the same manner as to the resonant dielectric susceptibility. Although it is clear from available experiments that there is a difference in the nature of TLS's contributing to dielectric and acoustic properties, there are observed correlations between the two responses both in the hole burning and nonequilibrium dielectric measurements.[3,5,40] We propose to extend acoustic measurements of glasses possessing atoms with nuclear quadrupole moments to lower temperatures in order to search deviations in the temperature dependence of a sound velocity. The most promising candidates are those materials in which the plateau in the dielectric constant is seen, including BK7, 10% K-SiO$_2$, and BaO-Al$_2$O$_3$-SiO$_2$ glasses (see Table II).

We have also analyzed the dependence of the resonant dielectric constant on the external magnetic field. We show that a high magnetic field $B \sim 5 - 10$ T affects nuclear spins stronger than the nuclear quadrupole interaction and therefore will restore coherent tunneling. This is because the nuclear spin states in the right and left wells get aligned parallel to the field. We have performed the model calculations of the dielectric constant in a magnetic field by combining numerical simulations for the behavior of a single spin affected by both a quadrupole interaction and an external field. The experimental verification of our theory can be made measuring the dielectric constant in the strong external magnetic field, which should eliminate the temperature independent plateau and restore the logarithmic temperature dependence.

To our knowledge, most of experimental studies of the magnetic field effect on the dielectric constant have been performed at relatively high temperatures, i.e., for $T > 10$ mK.[16] These studies show certain similarities and also distinctions compared to our predictions. The major changes in the dielectric constant are observed at $T \simeq 5$ mK. The application of magnetic field leads to an increase in the dielectric constant in agreement with expectations. At a certain magnetic field the dielectric constant of the standard tunneling model is restored. However, the change in the dielectric constant does not show a monotonous dependence on the magnetic field strength. A relatively strong effect is observed at very small fields (for $T \simeq 30$ mK and $B \simeq 0.1$ T). So, the behavior of the system is more complicated in this temperature range. Perhaps, the relaxational contribution to the dielectric constant is still significant there. Then, both the phonon-stimulated relaxation[41] and the interaction-induced relaxation[42] of TLS's could be affected by an external magnetic field and our analysis would not be applicable. The experiments at lower temperatures $T \simeq 5$ mK can test the model proposed here and can be used to characterize the internal structure and nuclear quadrupole interaction for a single TLS.

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