Effect of Nuclear Quadrupole Interaction on the Relaxation in Amorphous Solids

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Abstract

Recently it has been experimentally demonstrated that certain glasses display an unexpected magnetic field dependence of the dielectric constant. In particular, the echo technique experiments have shown that the echo amplitude depends on the magnetic field. The analysis of these experiments results in the conclusion that the effect seems to be related to the nuclear degrees of freedom of tunneling systems. The interactions of a nuclear quadrupole electrical moment with the crystal field and of a nuclear magnetic moment with magnetic field transform the two-level tunneling systems inherent in amorphous dielectrics into many-level tunneling systems. The fact that these features show up at temperatures $T < 100\,mK$, where the properties of amorphous materials are governed by the long-range $R^{-3}$ interaction between tunneling systems, suggests that this interaction is responsible for the magnetic field dependent relaxation. We have developed a theory of many-body relaxation in an ensemble of interacting many-level tunneling systems and show that the relaxation rate is controlled by the magnetic field. The results obtained correlate with the available experimental data. Our approach strongly supports the idea that the nuclear quadrupole interaction is just the key for understanding the unusual behavior of glasses in a magnetic field.

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I. INTRODUCTION

In 1998 Strehlow et al. observed a pronounced but unforeseen magnetic field dependence of the dielectric constant of certain multicomponent glasses at ultralow temperatures $T$, i.e., below $10mK$, for fields as small as $10\mu T$. The effect was found to be by several orders of magnitude larger than expected for an insulator in the absence of magnetic impurities. This result was especially astonishing since earlier experiments did not indicate noticeable magnetic field effects. Yet, careful measurements in magnetic fields ranging up to $25T$ and temperatures below $100mK$ revealed that the magnetic field causes drastic changes in the dielectric response. These experiments have caused further investigations. Following the original discovery, a number of different properties of the glasses were investigated in magnetic fields at these temperatures, e.g., by using the dielectric dipole echo technique.

Many low-temperature properties of glasses have been successfully described in the past by a standard tunneling model. To a good approximation a Tunneling System (TS) can be treated as a particle moving in a double-well potential (DWP). Because of the randomness of the glassy structure, the energy difference between the two wells as well as the tunneling matrix element have a broad distribution. This distribution is practically universal for all known dielectric glasses and results in an agreement between theory and experiment above $100mK$. Yet, below this temperature it is necessary to extend the model of isolated TS’s by taking into account the long-range interaction between them in order to interpret numerous experiments. The concept of resonant pairs (RP) plays an important role here. It captures the important physics as long as the interaction between TS is not so strong that the tunneling picture looses its meaning.

After the discovery of the anomalous glass behavior in a magnetic field, several extensions of the standard TM have been suggested. The dielectric properties of glasses at low temperatures are known to be due to the TS. It is reasonable to suppose that this is also the case for glasses in magnetic fields. Therefore, the principal question is how the magnetic field is acting on the TS’s. In our opinion, the model of Würger, Fleischmann and Enss deserves special mentioning and attention. In the framework of the model a direct coupling between the nuclear spin of a TS and the magnetic field takes place, though initially it was alleged that this possibility should be ruled out. However, the echo experiments convincingly evidence the influence of the nuclear moments on tunneling. Recently, Nagel et al. investigated isotope effects in polarization echo experiments in glasses. They observed magnetic field effects on amorphous glycerol when hydrogen, which has no electric quadrupole moment, was substituted by deuterium, that possesses nonzero quadrupole moment. Thereupon, one can conclude that the quadrupole electric moment of the TS’s is the key feature responsible for the effect.

A generalization of the standard tunneling model can be done as follows. Consider a tunneling particle with a nuclear spin. The energy levels of the system are degenerate with respect to the nuclear spin projection. This degeneracy is split if the particle has a quadrupole electric moment $Q$ which interacts with an electric field gradient (EFG), i.e., $q_{ij} = \frac{\partial^2 V}{\partial x_i \partial x_j}$, where $V(\vec{r})$ is the crystal-field potential. For the sake of simplicity, Würger et. al. supposed that the EFG possesses axial symmetry. In this case, the energy of the tunneling particle depends on the orientation of the quadrupole quantization axis. In general, in glasses the axes $u_R$ and $u_L$ differ in the two wells of the potential (see Fig.1). For
this reason, the quadrupole energy changes when the particle tunnels through the barrier. The magnetic field influences the energy spectrum since the particle acquires a Zeeman energy depending on the nuclear spin projection. Within that generalized tunneling model the previously mentioned echo experiment could be described\textsuperscript{24}.

The unusual dielectric response of glasses to an applied magnetic field occurs in the temperature region $T < 50mK$ where the standard tunneling model of non-interacting TLS cannot explain many experiments\textsuperscript{17}. Just within this temperature region RP are responsible for the relaxation in glasses. For this reason it is interesting to investigate whether or not the application of the magnetic field influences the dynamics of resonant pairs.

The principal goal of the paper is to show how the quadrupole interaction, if any, shows up in different characteristics of glasses. For this purpose we discuss first the explanation of the anomalous magnetic-field induced behavior of dielectric properties of glasses at ultra-low temperature based on the model\textsuperscript{24}. Then, we clarify how these features are reflected in the relaxation phenomena governed by the interaction between tunneling systems. We shall extend in the paper the concept of RP so that it includes the quadrupole interaction as well as the interaction with the magnetic field. We will also investigate how these interactions govern the relaxation rate of tunneling systems. A comparison of some of the results obtained with experiments will be made.

II. GENERALIZATION OF THE STANDARD TUNNELING MODEL

Consider a double-well potential (DWP) characterized by the asymmetry energy $\Delta$. At low enough temperatures only two energy levels corresponding to the ground state in each well of the DWP are significant. These levels are connected by the tunneling amplitude $\Delta_0$. According to\textsuperscript{13,14,15}, the parameters $\Delta, \Delta_0$ obey the universal distribution

$$P(\Delta, \Delta_0) = \frac{P}{\Delta_0}$$

where $P$ as a constant. An isolated tunneling particle is usually described by the standard two-level pseudospin $1/2$ Hamiltonian

$$h = -\frac{\Delta}{2} \sigma^z - \frac{\Delta_0}{2} \sigma^x.$$
Suppose that the tunneling particle possesses nonzero spin \( \hat{I}^2 = I(I+1) \). Below we will identify it with the nuclear spin. Then the states of the particle are characterized by the sign of the pseudospin projection and by the particle spin projection onto a proper quantization axis \( n = -I, \ldots, I \). Thus, the dimension of the Hilbert space for the tunneling particle is \( 2(2I+1) \). Transitions of the particle between the wells of the DWP occur with conservation of the spin projection. Introducing the spin projection operator \( |n\rangle \langle n| \), one can define generalized pseudospin operators \( \sigma^z [n] \) and \( \sigma^x [n] \) so that \( \sigma^i [n] = \sigma^i \otimes |n\rangle \langle n| \).

As long as the spin of the particle does not interact with the environment, the energy levels are degenerate with respect to the spin projection and the tunneling Hamiltonian reads

\[
H = -\sum_n \left( \frac{\Delta^2}{2} \sigma^z [n] + \frac{\Delta_0}{2} \sigma^x [n] \right).
\]

(3)

In this case, the spin degrees of freedom do not exert an influence on the tunneling properties of the particle, no matter which Hamiltonian, either (2) or (3), is used to describe the particle motion.

Suppose that an uniform magnetic field \( B \) directed along the \( z \)-axes is applied. Then, the tunneling particle gains extra Zeeman energy dependent on the spin projection \( I_z \)

\[
E_{\text{int}} = g\beta BI_z,
\]

(4)

and the degeneracy of the energy levels is lifted. However, this splitting is irrelevant. Indeed, in both wells of the DWP the magnetic field has the same magnitude. For this reason, the Zeeman contribution depends only on the spin projection and does not depend on the pseudospin projection. For the case \( I = 1 \), the energy structure of the tunneling particle before and after application of the magnetic field is presented in Fig. (2). The states with fixed spin projection are the eigenstates. One should pay attention to the fact that the tunneling between the two sites \( L \) and \( R \) can happen only between eigenstates that have equal spin projection (like in the absence of a magnetic field). So, the magnetic field does not influence the overlap integral between the wave function of the left and the right well. This means that the application of a magnetic field alone does not influence the properties of the TS under consideration.

Let us now consider the case when the spin of the tunneling particle is \( I \geq 1 \). Then the tunneling particle possesses a quadrupole electric moment. It interacts with the crystal
field which is characterized by the electric field gradient (EFG) \( q_{ij} \). The Hamiltonian of the particle interacting with the crystal field reads\(^{26}\)

\[
H_Q = -\frac{eQ}{2I(2I-1)} \left[ q_{11} I_1^2 + q_{22} I_2^2 + q_{33} I_3^2 \right]
\]

(5)

where \( H_Q \) is written in the basis \( e_1, e_2, e_3 \) in which the tensor \( q_{ij} \) has a diagonal form. The EFG satisfies Laplace's equation \( q_{11} + q_{22} + q_{33} = 0 \). Introducing the asymmetry parameter

\[
\kappa = \frac{q_{22} - q_{33}}{q_{11}},
\]

(6)

one can rewrite Eq. (5) in the form

\[
H_Q = -b \left( 3I_1^2 + \kappa (I_2^2 - I_3^2) - I^2 \right).
\]

(7)

Here the parameter \( b = \frac{eQ q_{11}}{4I(2I-1)} \) designates the quadrupole interaction constant. We assume that the \( e_1, e_2, e_3 \) axes are chosen so that \( q_{33} \leq q_{22} \leq q_{11} \), since then \( 0 \leq \kappa \leq 1 \). If \( \kappa = 0 \), the EFG possesses axial symmetry. In this case the quadrupole energy is completely defined by the spin projection \( I_1 \) and the quadrupole quantization axis is directed along \( e_1 \).

Assume that in the left well the basis is \( e_1, e_2, e_3 \) while in the right well it is \( e_1', e_2', e_3' \). In general, these bases are different. Let us introduce a basis \( e_x, e_y, e_z \) common for both wells. We assume that \( e_1 \) and \( e_1' \) lie in the \( e_x, e_y \) plane. Suppose that \( e_1 \) coincides with \( e_x \) while \( e_1' \) forms an angle \( \theta \) with the \( e_x \) axis. Then, for the case \( I = 1 \) considered here, one can represent \( H_Q \) in the right well in the basis of the eigenfunctions of the operator \( I_z \), \(|-1\rangle, |0\rangle, |1\rangle \), as follows\(^{30}\)

\[
H_Q(\theta) = b \begin{pmatrix}
-\frac{1}{2} (1 + \kappa) & 0 & \frac{3}{2} (1 - \frac{\kappa}{3}) e^{-i\theta} \\
0 & \kappa + 1 & 0 \\
\frac{3}{2} (1 - \frac{\kappa}{3}) e^{i\theta} & 0 & -\frac{1}{2} (1 + \kappa)
\end{pmatrix}.
\]

(8)

\( H_Q(\theta) \) has the following eigenstates and eigenvalues

\[
|\beta^0\rangle = \begin{cases}
0 & \\
1 & 0
\end{cases} \leftrightarrow \varepsilon_0 = b (\kappa + 1);
|\beta^1\rangle = \begin{cases}
- e^{-i\theta} & 0 \\
0 & 1
\end{cases} \leftrightarrow \varepsilon_{\beta^1} = -2b;
|\beta^2\rangle = \begin{cases}
 e^{-i\theta} & 0 \\
0 & 1
\end{cases} \leftrightarrow \varepsilon_{\beta^2} = b (1 - \kappa)
\]

(9)

For the left well the eigenvalues are the same as for the right well while the eigenvectors are \(|\alpha^0\rangle, |\alpha^1\rangle, |\alpha^2\rangle\), resulting from Eq.(9) for \( \theta = 0 \). It follows directly from (9) that the spin state \(|\alpha^0\rangle = |\beta^0\rangle = |0\rangle\) is orthogonal to all the others, but because of the fact that the quadrupole quantization axes differ in the two wells, \( \langle \alpha^i | \beta^j \rangle \neq 0, \ i \neq j \). Thus, the state with spinor \(|0\rangle\) in the left well can couple only to the state with spinor \(|0\rangle\) in the right well,
while the remaining states in the left and the right well couple to each other. The energy level structure and possible transitions between the left and the right well states are shown in Fig. (3).

Let us investigate how the application of the external magnetic field changes the energy spectrum of the tunneling particle. The magnetic field shows up in two ways. First, it produces a Zeeman splitting. Then for different TS’s the planes formed by the vectors $e_1, e'_1$ are randomly oriented. For this reason the magnetic field is in general not orthogonal to this plane. Therefore it mixes the states with spinor $|0\rangle$ with states with spinors $|\alpha^i\rangle$ and $|\beta^i\rangle$ ($i = 1, 2$) of the two wells. In a new basis $|\alpha^i\rangle$ and $|\beta^i\rangle$ which is dependent on the direction of the magnetic field all $\langle \alpha^i | \beta^j \rangle \neq 0$, $i, j = 0, 1, 2$. Thus, additional transitions between the two wells become possible (see Fig. (4)).
It should be stressed that with increasing magnetic field, the Zeeman splitting will eventually exceed the quadrupolar one. Therefore, we discuss first the case when the TS’s are affected only by the magnetic field (see Fig. 2).

To simplify the further analysis, suppose that the magnetic field is orthogonal to the plane \( e_1, e'_1 \) and that the quadrupole splitting constant \( b \) is the same in both wells. Then the Hamiltonian of the TS in the presence of the quadrupole and Zeeman splitting reads

\[
\begin{pmatrix}
H_L & \Delta_0 \cdot I \\
\Delta_0 \cdot I & H_R
\end{pmatrix},
\]

where \( I \) is the rank 3 unit matrix and

\[
H_L = \begin{pmatrix}
-\frac{\Delta}{2} - m - \frac{b}{2} (1 + \kappa) & \frac{3}{2} (1 - \frac{\kappa}{3}) & 0 \\
\frac{3}{2} (1 - \frac{\kappa}{3}) & -\frac{\Delta}{2} + b (\kappa + 1) & 0 \\
0 & 0 & -\frac{\Delta}{2} + m - \frac{b}{2} (1 + \kappa)
\end{pmatrix},
\]

\[
H_R = \begin{pmatrix}
\frac{\Delta}{2} - m - \frac{b}{2} (1 + \kappa) & \frac{3}{2} (1 - \frac{\kappa}{3}) e^{-i\vartheta} & 0 \\
0 & \frac{3}{2} (1 - \frac{\kappa}{3}) e^{i\vartheta} & \frac{3}{2} (1 - \frac{\kappa}{3}) e^{i\vartheta} \\
\frac{3}{2} (1 - \frac{\kappa}{3}) & 0 & -\frac{\Delta}{2} + m - \frac{b}{2} (1 + \kappa)
\end{pmatrix}.
\]

III. PERMITTIVITY OF A TUNNELING SYSTEM WITH QUADRUPOLE AND ZEEMAN SPLITTING

Let us investigate how the changes of the energy spectrum induced by the quadrupole and Zeeman splitting described by Hamiltonian \( \text{(10)} \) influence the properties of a TS. First we examine the influence of these changes on the dielectric permittivity.

Consider a particle that can occupy \( (2I + 1) \) levels in a DWP. We introduce the dipole moment operator in the form

\[
\hat{p} = \frac{p_0}{2} \begin{pmatrix} -I & 0 \\ 0 & I \end{pmatrix},
\]

where \( I \) is the unit matrix of rank \( (2I + 1) \) and \( p_0 \) defines the value of the dipole moment. (For \( I = 0 \) this expression transforms into the well known dipole moment operator for a two-level-system). The interaction of the TS with the external electrical field \( F \) reads

\[
\hat{V} = -F \hat{p}.
\]

In second-order perturbation theory the correction to the energy of a TS induced by this interaction is given by

\[
\delta \varepsilon = F^2 Z^{-1} \sum_{b \neq a} e^{-\frac{E_b}{T}} |\langle a | \hat{p} | b \rangle|^2 \frac{E_a - E_b}{E_a - E_b},
\]

where \( T \) is the temperature, \( Z = \sum_a e^{-\frac{E_a}{T}} \) is the partition function and \( E_a \) denotes the eigenvalues of the TS in the presence of a magnetic field.
On the other hand, as the electric field changes from zero to a certain final value $F$, the energy increases by $\chi F^2/2$, where $\chi$ is a permittivity of the system. Comparing this value to $\delta \varepsilon$ one obtains

$$\chi = 2Z^{-1} \sum_{b \neq a} \frac{e^{-E_b/T} |\langle a | \hat{p} | b \rangle|^2}{E_b - E_a} = Z^{-1} \sum_{b \neq a} \frac{\left(e^{-E_b/T} - e^{-E_a/T}\right) |\langle a | \hat{p} | b \rangle|^2}{E_b - E_a}$$

(16)

Let us apply this relation for the TS described by Hamiltonian (10).

The permittivity of a tunneling system is a function of the parameters $\Delta, \Delta_0, b, \theta, m$. (For a preliminary analysis in this part of the paper we confine ourselves to the case of an axial symmetric EFG by setting the parameter $\nu = 0$). Keeping in mind a possible fit of the available experimental data, we choose the following values for the parameters $\Delta_{\text{max}} = \Delta_{0\text{max}} = 10K, b = 10mK$. The temperature varies between $10mK$ and $100mK$. The Zeeman energy of the nuclear spin ranges from zero to $30mK$. With the Landé factor $g = 3$, this corresponds to a maximal magnetic field of about $15T$.

The permittivity has been estimated numerically. First, we calculated the eigenvalues and eigenfunctions for the matrix (10) with values of the TS parameters in the above mentioned range. Then, we estimated the permittivity $\chi (\Delta, \Delta_0, b, \theta, m, T)$ by using Eq. (16). Finally, the result was averaged over the parameters $\Delta, \Delta_0, \theta$ by using Eq. (11) and assuming a uniform distribution of the angles formed by the quadrupole quantization axes in the wells. As an example, we present here the result of our calculations for the temperature $T = 40mK$

Figure 5: Magnetic field dependence of the permittivity $\varepsilon'$ for typical quadrupole splitting $b = 10mK$ at $T = 40mK$

The permittivity exhibits a pronounced peak at an energy approximately corresponding to the magnetic field $B \approx 15T$. This result corresponds well to Ref. 3 where a pronounced peak in the permittivity was observed for a similar value of the magnetic field and a temperature of $T = 64mK$.

As mentioned before we have assumed $\nu = 0$ in calculating the permittivity shown in Fig. 5. When we introduce $\nu$ a second energy scale appears when $\nu$ is small. This scale is relevant for weak magnetic fields. It might explain another peak observed in Ref. 3 in the
low field regime. Also a second atom of the tunneling entity with a different quadrupole moment may result in a second energy scale. We have modelled a low energy scale by simply redoing the calculations for \( b = 0.3 \text{mK} \) and plotting the results for the permittivity in a form which can directly compared with the experimental findings of Ref. 3. Figure 6 shows an agreement with experiments that the permittivity at fixed value of magnetic field is the higher the lower the temperature is.

Figure 6: Magnetic field variation \( \Delta \varepsilon \) of the permittivity.

IV. RELATION BETWEEN THE PERMITTIVITY AND MANY-BODY RELAXATION

As mentioned in the introduction, the temperature region \( T < 100 \text{mK} \) where certain glasses show an unusual response to an applied magnetic field coincides with the region where the relaxation of tunneling system is due to the \( R^{-3} \) interaction between TLS rather then single phonon processes. In the previous section we have seen that a magnetic field affects the permittivity provided the quadrupole effect is taken into account. Therefore, it is of interest to investigate whether a magnetic field influences the relaxation induced by this interaction.

The relaxation induced by the long range \( R^{-3} \) interaction is strongly connected with the concentration of resonant pairs (RP). Resonant pairs of TS’s are the main concept for this relaxation mechanism. First we recall briefly the main idea of this approach.

Consider a pair of two-level-systems. In general, it possesses four different configurations. Only two of them (configurations A and B ) shown in Fig. 7 are important. The special feature of these configuration is that one TLS is in the ground state while the other is in the excited state.

Let \( V_{12} = U/R^3 \) denote the weak interaction between these TLS’s where \( U \) is the interaction constant. Such a pair is in resonance when

\[
| E_1 - E_2 | < V_{12}
\]  \hspace{1cm} (17)
Here $E_i = \sqrt{\Delta_{i0}^2 + \Delta_i^2}$, $\Delta_{0i}, \Delta_i, i = 1, 2$ are the tunneling parameters of two TLS constituting a pair. We are interested in the case $E_1 \approx E_2 \approx T$ and $V_{12} \ll T$ which means a weak interaction. Due to the constraint (17), the states (A) and (B) are coupled with each other and are well separated from the states (C) and (D) by an energy gap of the order of $T$. For this reason, they can be excluded from the consideration. The two states of the TLS pair (A) and (B) are referred to as a flip-flop configuration and the transition between them is called a flip-flop one. When such two TLS’s form a resonant pair (RP) they can be considered as a new type of the two-level system with the asymmetry $|E_1 - E_2|$. Resonant pairs are responsible for the many-body relaxation induced by the $R^3$ interaction. The transition amplitude between the levels (A) and (B) is described by the following expression\textsuperscript{16,18}

$$U_{12} = V_{12} \sigma_1^{x} \sigma_2^{x} \frac{\Delta_{01} \Delta_{02}}{E_1 E_2}.$$  \hspace{1cm} (18)

Next we write down the expression for the concentration of pairs with tunneling parameters $\Delta_p, \Delta_{0p}$\textsuperscript{16,18}:

$$P^{(2)}(\Delta_p, \Delta_{0p}) = \frac{P^2}{4} \int \frac{d\Delta_{01}d\Delta_{02}}{\Delta_{01} \Delta_{02}} \cdot \int \frac{d\Delta_{01}d\Delta_{02}}{\Delta_{01} \Delta_{02}} \left[ 1 + e^{-\frac{E_1}{T}} \right]^{-1} \left[ 1 + e^{-\frac{E_2}{T}} \right]^{-1} \cdot \int d^3R \delta \left( \Delta_{0p} - \frac{\Delta_{01} \Delta_{02} U}{E_1 E_2 R^3} \right) \delta(\Delta_p - (E_1 - E_2)).$$  \hspace{1cm} (19)

The integration over $d^3R$ gives

$$\int d^3R \delta \left( \Delta_{0p} - \frac{\Delta_{01} \Delta_{02} U}{E_1 E_2 R^3} \right) = \frac{\Delta_{01} \Delta_{02} U}{E_1 E_2 \Delta_{0p}^2}. \hspace{1cm} (20)$$

For resonant pairs one has $\Delta_p \leq \Delta_{0p} \ll E_1 \approx E_2 \approx T$ and, therefore, one can omit $\Delta_p$ in the argument of the $\delta-$ function in (19). The concentration of resonant pairs $P_r^{(2)}(\Delta_{0p})$ is...
obtained by integration of $P^{(2)}(\Delta_p, \Delta_{0p})$ over the interval $0 < \Delta_p < \Delta_{0p}$

$$P^{(2)}(\Delta_{0p}) = P^2 \frac{U_0}{\Delta_{0p}} \int d\Delta_{01} d\Delta_1 \frac{d\Delta_{02}}{\Delta_{01}} \int d^2 \frac{1}{\Delta_{02}} \left( \Delta_{01} \Delta_{02} \right) \delta (E_1 - E_2)$$

(21)

On the other hand, the resonance permittivity of a TLS is given by the well known expression (see e.g. 14)

$$\chi = P \int_0^B d\Delta \int_0^{\Delta_0} \frac{d\Delta_0}{\Delta_0} \frac{1}{E} \left( \frac{\Delta_0}{E} \right)^2 \tanh \frac{E}{2T}$$

(22)

Let us calculate the derivative

$$\frac{\partial \chi}{\partial \ln T} = -\frac{P}{2T} \int_0^B d\Delta \int_0^{\Delta_0} \frac{d\Delta_0}{\Delta_0} \frac{1}{ch^2 \frac{E}{2T}} \left( \frac{\Delta_0}{E} \right)^2$$

(23)

Comparing expressions (21) and (23), we can conclude that $P^{(2)}(\Delta_{0p})$ and the square of $\frac{\partial \chi}{\partial \ln T}$ are approximately proportional to each other. Therefore we expect similar features in the behavior of the permittivity and in the rate of the interaction induced relaxation which is proportional to the RP concentration. This means that the multilevel systems can again be considered as effective two level systems. When the Zeeman splitting exceeds the quadrupole one, the multilevel tunneling systems behaves exactly as two-level-systems.

In Figure 8 we present in the framework of the model described above the results of the calculation of the parameter $(\frac{\partial \chi}{\partial \ln T})^2$ for different temperatures as a function of the applied magnetic field. One notices that for magnetic fields causing a Zeeman splitting smaller or of the order of the quadrupole splitting, i.e., $b \lesssim 10 mK$ it holds that the larger the temperature the larger is $\frac{\partial \chi}{\partial \ln T}$. This can be understood as follows: The relaxation rate induced by the interaction between TS’s is proportional to the concentration of RP, which in turn is proportional to the temperature. This conclusion agrees with the sequence of different curves in Fig. 8. However, some curves show a monotonous magnetic field dependence, while others display a non-monotone behavior. This feature is explained in the following section.

V. RELAXATION OF THE INTERACTING MANY-LEVEL TUNNELING SYSTEMS

The main goal of the current section is a qualitative treatment of the results obtained in Sec. IV. In particular, we want to explain the temperature dependencies shown by Fig. 8.

In our previous papers[16,18,19,20,21] we have demonstrated that the many-body $R^{-3}$ interaction between TLS’s results in a new relaxation mechanism responsible for the low temperature relaxation. On the other hand, it was shown above that if the tunneling particle possesses a nuclear spin $I \geq 1$, the energy spectrum of a tunneling system consists of several lines, as distinct from one line in the case of a TLS. Below we investigate how these changes in the energy spectrum of the tunneling system caused by the interaction of the tunneling particle with the electric field gradient and the magnetic field influence the relaxation rate produced by the $R^{-3}$ interaction.

The model under investigation assumes that the particle can occupy $n = 2I + 1$ levels each in the left and in the right well of a TS. Let us generalize the concept of resonant pairs
Figure 8: The square of the logarithmic derivative of the permittivity as a function of the magnetic field.

Figure 9: Two tunneling systems: TS 1) is in one of its ground state multiplet. TS 2) is in the excited state multiplet. The occupied states are marked by solid line. The arrow lines show which levels are occupied after flip-flop process has happened.
to the case \( n > 1 \). Consider the tunneling system (1) in Fig. 9. There we are dealing with a ground-state multiplet the states of which are denoted by their spinor parts \( \alpha_1^i \) and an excited state multiplet denoted by \( \beta_2^i \). The corresponding energies are \( \delta_{\alpha_1^i} \) and \( \delta_{\beta_2^i} \). They are derived from the energies 0 and \( E_1 = \sqrt{\Delta_{10}^2 + \Delta_2^2} \) of the TS in the absence of the quadrupolar and Zeeman interaction. Therefore the various transition energies between the two multiplets are

\[
E_{1ij}^* = E_1 + (\delta_{\alpha_1^i} - \delta_{\beta_2^j}) , \quad i, j = 1, \ldots, n .
\]  

Like in the case of a TLS only those with \( E_{1ij}^* \approx T \) contribute significantly to relaxation processes. Below we will assume that also

\[
\delta_{\alpha_1^i}, \delta_{\beta_2^j} \ll T .
\]  

holds\textsuperscript{31}. Therefore only \( E_1 \) defines the Gibbs distribution and the states within a multiplet are occupied with nearly equal probability. A similar analysis holds for the TS(2) of Fig. 9. Thus, two multilevel TS’s give raise to flip-flop processes \( \alpha_1^i, \beta_2^j = \beta_2^k, \alpha_1^l \) when the corresponding energy differences between the two configurations differ in energy by less than a characteristic interaction matrix element. The latter and the various different processes are discussed in the following.

Let \( V [\alpha_1^i, \beta_2^j; \beta_2^k, \alpha_1^l] \) denote the transition amplitude of such a flip-flop transition. Then the condition for a resonant pair is

\[
|E_{1ij}^* - E_{2kl}| = |E_1 - E_2 + (\delta_{\alpha_1^i} - \delta_{\beta_2^j}) - (\delta_{\alpha_1^l} - \delta_{\beta_2^k})| < V [\alpha_1^i, \beta_2^j; \beta_2^k, \alpha_1^l] .
\]  

When all the energy splittings \( \delta = 0 \), i.e., for TLS’s the resonating pair concentration is given by\textsuperscript{16,18,19,20,21}

\[
N_* = (PT)(PU) .
\]  

Let us investigate how the concentration of resonating pairs is modified for multilevel systems. For simplicity we set all \( V [\alpha_1^i, \beta_2^j; \beta_2^k, \alpha_1^l] \equiv V \). We fix the energy \( E_1 \) and also the splitting energies \( \delta_{\alpha_1^i} \). Then the energy range of the parameter \( E_2 \) is defined by Eq. (20) and equals \( V \). For a given \( E_1 \) there are \( n^4 \) different state configurations \( \alpha_1^i, \beta_2^j; \alpha_2^l, \beta_2^k \) possible, i.e., the energy difference \( (\delta_{\alpha_1^i} - \delta_{\beta_2^j}) - (\delta_{\alpha_1^l} - \delta_{\beta_2^k}) \) takes \( n^4 \) different values. So there are also \( n^4 \) different energy ranges for \( E_2 \). Different values of \( E_2 \) relate to different tunneling systems. However, one must take into account that the probability of finding an initial configuration \( \alpha_1^i, \beta_2^j \) is \( 1/n^2 \). Hence the total probability of forming resonating pairs would seem to increase like \( n^4/n^2 = n^2 \) as compared with TLS’s. This would hold true if the transition amplitude \( V [\alpha_1^i, \beta_2^j; \beta_2^k, \alpha_1^l] \) would coincide with the previously discussed amplitude \( V_{12} \) of TLS’s. But this is not so as is easily seen as follows. The transition between two TS’s (1) and (2) can be described similarly as in Eq. (18) by

\[
\sum_{\alpha_1^i, \beta_2^j; \beta_2^k, \alpha_1^l} V [\alpha_1^i, \beta_2^j; \beta_2^k, \alpha_1^l] \sigma_1^+ \sigma_2^+ |\alpha_1^i\rangle |\beta_2^k\rangle \langle \alpha_1^i | \langle \beta_2^k | ,
\]  

\[
V [\alpha_1^i, \beta_2^j; \beta_2^k, \alpha_1^l] = V_{12} \langle \beta_2^k | \alpha_1^i \rangle \langle \alpha_2^l | \beta_2^k \rangle .
\]
As before the notation $|\alpha^i_1\rangle$, $|\beta^i_2\rangle$ etc. refers to the spinor part only of the wavefunction of the particle. Here $|V_{12}| = \frac{\Delta \alpha \Delta \beta U}{E_1 R_2^3}$ and $R_{12}$ is the distance between the TS. Let us estimate the transition amplitude $V[\alpha^i_1, \beta^i_2; \beta^i_1, \alpha^i_2]$ in terms of $V_{12}$. By making use of Eq. (29) and the completeness condition in the form of $\sum_{\alpha^j_1} |\langle \beta^j_2 | \alpha^j_1 \rangle|^2 = 1$, we obtain
\[
\sum_{\alpha^i_1; \alpha^i_2} |V[\alpha^i_1, \beta^i_2; \beta^i_1, \alpha^i_2]|^2 = |V_{12}|^2 \sum_{\alpha^i_1} |\langle \beta^i_2 | \alpha^i_1 \rangle|^2 \sum_{\alpha^i_2} |\langle \beta^i_1 | \alpha^i_2 \rangle|^2 = |V_{12}|^2 \quad (30)
\]
For further estimation, we make the simplest assumption that all the matrix elements entering Eq. (30) are equal. The total number of terms in the left-hand sum is $n^2$. So
\[
|V[\alpha^i_1, \beta^i_2; \beta^i_1, \alpha^i_2]| = \frac{|V_{12}|}{n} \quad (31)
\]
Thus, the tunneling amplitude $V[\alpha^i_1, \beta^i_2; \beta^i_1, \alpha^i_2]$ entering Eq. (29) is $n$ times smaller as compared with the case of pairs formed by TLS. Therefore, the factor $n^2$ found above for increasing the probability of formation of resonating pairs is further reduced by a factor $n$. So, the total probability to form a resonating pair increases for multilevel systems $\xi = n$ times as compared with TLS’s.

A similar analysis can be carried out when some of the energy levels remain degenerate or when the transitions between some of them are forbidden. This case is investigated by using as an example the spectrum of TS with a nuclear spin $I = 1$ when only quadrupolar effects are taken into account (see Fig. 3). In order to find the matrix element of the flip-flop transition let us again make use of Eq. (29). Here we must differentiate between three cases.

In the first case, the transition in both TS’s of the pair occurs between levels with $I_2 = 0$. The corresponding states are denoted by their spinor parts $\alpha^0_1, \beta^0_1$ and $\alpha^0_2, \beta^0_2$. By taking into account that the spinor parts remain unchanged it follows that
\[
V[\alpha^0_1, \beta^0_2; \beta^0_1, \alpha^0_2] = V_{12} \langle \alpha^0_1 | \beta^0_1 \rangle \langle \beta^0_2 | \alpha^0_2 \rangle = V_{12} \quad (32)
\]
In the second case, the transition between $I_2 = 0$ states takes place only in one TS of the pair. In that case one finds
\[
V[\alpha^0_1, \beta^0_2; \beta^0_1, \alpha^0_2] = V_{12} \langle \alpha^0_1 | \beta^0_1 \rangle \langle \beta^0_2 | \alpha^0_2 \rangle \quad (33)
\]
The sum of the square of the matrix elements is
\[
\sum_{\alpha^0_2} |V[\alpha^0_1, \beta^0_2; \beta^0_1, \alpha^0_2]|^2 = |V_{12}|^2 \sum_{\alpha^0_2} |\langle \beta^0_2 | \alpha^0_2 \rangle|^2 = |V_{12}|^2 \quad (34)
\]
Hereby the condition $\sum_{\alpha^0_2} |\langle \beta^0_2 | \alpha^0_2 \rangle|^2 = 1$ has been used. Since the total number of terms in the sum (34) is two, one can estimate
\[
|V[\alpha^0_1, \beta^0_2; \beta^0_1, \alpha^0_2]| = \frac{|V_{12}|}{\sqrt{2}} \quad (35)
\]
In the third case, when the transitions are between levels with nonzero spin projection in both TS’s, one finds

\[
\sum_{\alpha_1^i, \alpha_2^j} |V[\beta_1^i, \beta_2^j, \beta_1^j, \alpha_2^i]|^2 = |V_{12}|^2 \sum_{\alpha_1^i} |\langle \beta_1^i | \alpha_1^i \rangle|^2 \sum_{\alpha_2^j} |\langle \beta_2^j | \alpha_2^j \rangle|^2 = |V_{12}|^2. \tag{36}
\]

The total number of terms in the left-hand sum in Eq. (36) is equal to \(2^2\) and, therefore,

\[
|V[\alpha_1^i, \beta_2^j, \beta_1^j, \alpha_2^i]| = \frac{|V_{12}|}{2}. \tag{37}
\]

Using Eqs. (26), (32), (35), (37) one estimates that the probability to form a resonating pair increases by a factor of \(\xi = 1 + \frac{8}{9\sqrt{2}} \approx 1.6\).

Thus, for the case of a nuclear spin \(I = 1\) the quadrupolar interactions result in an increase of the probability of finding resonating pairs by a factor \(\xi \approx 1.6\) as compared with TLS’s. When in an applied magnetic field the Zeeman splitting is of the order of the quadrupolar interaction, the probability of forming a resonating pair increases by a factor of \(\xi = 3\). Finally, when the Zeeman energy exceeds the quadrupolar splitting, the factor is \(\xi = 1\).

The results obtained in this section are based on the fact that a tunneling system is a multilevel one. In other words, the energy levels of the TS should be well resolved. Yet, in an ensemble of interacting TS’s the energy levels fluctuate due to spectral diffusion. When the scale of spectral diffusion exceeds the quadrupole splitting the transition occurring from the different levels of the TS’s can not be considered as statistically independent. The scale of spectral diffusion is about \(\gamma T\), \(\gamma = \frac{P U}{27}\). So our approach is valid when the quadrupole splitting \(b\) is

\[
b > \gamma T. \tag{38}
\]

Due to a similar reason the Zeeman splitting should obey

\[
g \mu B > \gamma T. \tag{39}
\]

This condition establishes the minimal value of the \(B\) in order that magnetic field effects show up.

The results obtained in this section are based on the energy level classification described in Sec. II. This requires that the tunneling amplitude \(\Delta_0\) fulfills the relation \(\Delta_0 \ll \Delta, b\). For that reason, in the above analysis the factor \(\frac{\Delta_0}{E_1 E_2}\) entering Eq. (18) is a small parameter formed by strongly asymmetric tunneling systems.

VI. DISCUSSION AND CONCLUSIONS

We have shown that the strong magnetic field dependence of the electric susceptibility in ultracold glasses can be understood by taking into account the interactions of tunneling systems in the presence of nuclear quadrupolar moment. The essential point is the following: Even by a small applied magnetic field the number of different energy levels of a tunneling system is increased. This in turn, modifies the concentration of resonant tunneling pairs and leads this way to observable effects.
Two kinds of experiments can be related to the present investigation. The first kind deals with the experimental determination of the real part of the response of the system to an applied magnetic field. We find for the field dependence of the static electric susceptibility the right order of magnitude of the effect. That a too small effect was found in\[32\] may result from the use of perturbation theory. Another unexplained feature, namely a plateau in the temperature dependence of the electric susceptibility, sometimes called dielectric saturation is most probably due to a dependence of the tunneling matrix element $\Delta_0$ on the spinors of the right and left well.

The second group of experiments is related to measurements of the imaginary part of the response. In this case information on the relaxation rate of elementary excitations can be obtained. For example, measuring the echo amplitude allows to determine the behavior of the TS coherence time $\tau_2$ as function of field. The echo amplitude has been found to show a non-monotonic dependence on the magnetic field\[5,6,7,8,9,10,11\]. It is defined by the transverse relaxation rate $\tau_2^{-1}$. In Sec. 5 we have introduced the parameter $\xi$ depending on the magnetic field which controls the concentration of RP. If the relaxation in the system is due to the resonant pairs, the transverse relaxation rate is directly proportional to the concentration of RP. Without the quadrupolar effect, the relaxation rate $\tau_2^{-1} = \gamma^2 T^{18}$. If the quadrupolar effects are taken into account, the concentration of RP increases by the factor $\xi$. Therefore, the relaxation rate becomes

$$\tau_2^{-1} \approx \xi \gamma^2 T . \quad (40)$$

The non-monotonic behavior of the parameter $\xi$ on the magnetic field correlates with the behavior of the echo amplitude found in the experiments mentioned above. Note that in high magnetic fields $\xi$ reduces to $\xi = 1$, because in that limit the energy levels are equally spaced as in Fig. 2.

The standard scheme of interpreting the echo experiments is based on the assumption that TS’s are two-level ones. In that case they can be described in terms of $\tau_2^{-1}$. This is not the case when the quadrupolar effects are taken into account. Therefore it is no surprise that the calculated $\tau_2^{-1}$ can only qualitatively but not quantitatively describe the echo experiments. Instead the approach in\[24,34,35\] based on a multilevel description of TS’s instead of the standard Bloch equations seems very reasonable.

For this reason, another kind of low-temperature experiments should be made to clarify the role if the $R^{-3}$ interaction between the tunneling system. For example, measurements of the magnetic field dependence of dielectric loss or internal friction would allow to extract information on the lifetime $\tau_1$ of the excitations. We want to show that in this case the factor $\xi$ should show up in an even more important way.

The spectral diffusion reaches its maximal value $\gamma T$ at time $\tau_1$. Therefore, the rate of the spectral diffusion is $\gamma T/\tau_1$. On the other hand the spectral diffusion rate relates to $\tau_2^{-2} \quad 18,27$. Then

$$\tau_1^{-1} = \xi^2 T \gamma^3 . \quad (41)$$

Thus the relaxation rate $\tau_1^{-1}$ shows an even stronger quadrupole and magnetic field dependence than $\tau_2^{-1}$ and investigation of dielectric loss or internal friction in the presence of the magnetic field opens an attractive opportunity to investigate the role of the $R^{-3}$ interaction.

The above analysis is based on the assumption that the EFG has approximately axial symmetry, i.e., that the parameter $\varkappa$ is small. Generally this is not the case. Nevertheless, the magnetic field induced relaxation mechanism has a quite universal character. Indeed,
let the nuclear spin be half-integer. It follows from Kramers’ theorem that in zero magnetic field the energy spectrum is degenerate. So, after application of the magnetic field the total number of levels of the multilevel TS increases and an effect takes place.

The nature of the tunneling systems in amorphous solids remains puzzling despite of the large theoretical efforts probing various models. The main problem of the theory is the lack of experiments which test particular models versus the original phenomenological model\textsuperscript{13}, which simply employs the distribution $\mathcal{N}$. We expect that the magnetic field experiments allow to reveal which and how many atoms participate in the tunneling. This should shed new light on the microscopic nature of the tunneling systems in glasses.

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To obtain the quadrupole Hamiltonian for the right well one should put $\theta = 0$ in Eq. (8).

The opposite case will be examined in a separate paper.