Effect of a Magnetic Field on the Dipole Echo in Glasses with Nuclear Quadrupole Moments.

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The effect of a magnetic field on the dipole echo amplitude in glasses at temperatures of about 10 mK caused by nonspherical nuclei with electric quadrupole moments has been studied theoretically. It has been shown that in this case, the two-level systems (TLS’s) that determine the glass properties at low temperatures are transformed into more complicated multilevel systems. These systems have new properties as compared to usual TLS’s and, in particular, exhibit oscillations of electric dipole echo amplitude in magnetic field. A general formula that describes the echo amplitude in an arbitrary split TLS has been derived with perturbation theory. Detailed analytic and numerical analysis of the formula has been performed. The theory agrees qualitatively and quantitatively well with experimental data.

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I. INTRODUCTION

Glasses at temperature below 1 K display a number of universal properties which are fundamentally different from the properties of similar crystals. These properties are almost independent of the chemical composition and are therefore due to the structure of amorphous solid; more exactly due to absence of long-range order in the glass [1]. One of such universal phenomena is a two-pulse dipole echo.

The essence of the effect is the following. When a glass is subjected to two short electromagnetic pulses at a frequency 1 GHz separated by a time interval \( \tau \), a response at the same frequency may be detected at time \( \tau \) after the second pulse.

According to the standard theory by Anderson, Halperin, Varma, and Phillips [1], all these properties are associated with the existence of so-called two-level systems (TLS’s) in the glass, which are the groups of atoms tunneling between two minima of a double-well potential. However, the microscopic nature of the TLS’s in the majority of glasses remains unknown.

An interesting phenomenon was discovered in 2002 [2]. The amplitude of the dipole echo in some nonmagnetic glasses exhibited a strong nonmonotonic (oscillating) dependence on the magnetic field even in weak fields (about 10 mT). Wurger, Fleischmann, and Enss [3] and by one of us [6]. However, a detailed comparison of the theoretical results with experiments was not carried out in those works. This work is aimed at the detailed comparison of the theoretical results with the available experimental data. We perform a theoretical analysis of the derived general formula in various limiting cases and show that the theory agrees with the experimental data reasonably well, both qualitatively and, in some cases, quantitatively.

II. INTERACTION OF TWO-LEVEL SYSTEMS WITH NUCLEAR QUADRUPOLE MOMENTS

Let us briefly recall how the presence of atoms with nuclear quadrupole moments affects the properties of two-level systems and, in particular, the amplitude of the dipole echo in glasses [6]. Let one of the atoms that are displaced under the tunneling of the TLS have a nuclear quadrupole moment. The energy of its quadrupole interaction with the microscopic electric field and Zee- man interaction with the external magnetic field leads to TLS level splitting, so that the TLS becomes a system with two identical level sets each containing \( 2J + 1 \) levels. These sets are separated by the energy \( E \) of the original TLS, which is much greater than the energy splitting within the set.

The Hamiltonian of such a system may be written in

\( (J = 1 \) and a nonzero quadrupole moment) results in a more than an order of magnitude enhancement of the magnetic-field dependence of the echo amplitude.

The magnetic-field dependence of the echo amplitude was theoretically studied by Wurger, Fleischmann, and Enss [3, 5] and by one of us [6]. However, a detailed comparison of the theoretical results with experiments was not carried out in those works. This work is aimed at the detailed comparison of the theoretical results with the available experimental data. We perform a theoretical analysis of the derived general formula in various limiting cases and show that the theory agrees with the experimental data reasonably well, both qualitatively and, in some cases, quantitatively.
the form \[ (\mathbf{F} \cdot \mathbf{m}) \frac{1}{E} \left( \begin{array}{cc} \Delta & \Delta_0 \\ \Delta_0 & -\Delta \end{array} \right) \otimes \widehat{I}_Q + \right. 

\left. \frac{1}{E} \left( \begin{array}{cc} \Delta & \Delta_0 \\ \Delta_0 & -\Delta \end{array} \right) \otimes \widehat{V}_Q. \right) \] (1)

Here, \( \widehat{I}_Q \) and \( \widehat{I}_\sigma \) are the 2 \( \times \) 2 and \( N \times N \) (\( N = 2J + 1 \)) unity matrices in the spaces of the TLS and the projections of the nuclear spin, respectively. \( \Delta \) is the difference between the double-well potential minima. \( \Delta_0 \) is the tunneling amplitude of the initial (unsplit) TLS, 
\( E = \sqrt{\Delta^2 + \Delta_0^2} \) is the total energy of the TLS disregarding the fine structure effects. \( \mathbf{F} \) is the ac electric field, and \( \mathbf{m} \) is the electric dipole moment of the TLS. 
\( \widehat{W}_Q = \mathbf{M} \cdot \mathbf{H} + \widehat{Q}_{\alpha\beta} \varphi_{\alpha\beta}(0) \), where \( \mathbf{M} \) is the nuclear magnetic moment, \( \mathbf{H} \) is the external magnetic field, \( \widehat{Q}_{\alpha\beta} \) is the operator of the nuclear quadrupole moment \( \overline{\mathbf{Q}}_{\alpha\beta} \) (expressed in terms of the nuclear spin operator), and \( \varphi_{\alpha\beta}(0) \) is the second-derivative tensor of the electric potential taken at the double-well potential maximum.

The second term in Eq. (1), which contains \( \widehat{W}_Q \), causes the fine splitting of the TLS. We assume that the basis nuclear spin wavefunctions are eigenfunctions of the operator \( \widehat{W}_Q \) and its matrix is diagonal in this basis.

The last term in the Hamiltonian (1) includes the operator 
\( \widehat{V}_Q = \widehat{Q}_{\alpha\beta} \varphi'_{\alpha\beta}(0)|x_{\text{min}}|/x_0 \), where \( \varphi'_{\alpha\beta}(0) \) — is the derivative of the tensor \( \varphi_{\alpha\beta}(x) \) with respect to the generalized coordinate \( x \) of the TLS at \( x = 0 \) and \( |x_{\text{min}}|/x_0 \) is the ratio of the atom displacement under the tunneling of the TLS to the characteristic interatomic distance \( x_0 \).

The first term in Eq. (1) determines the two-level system, the second one is responsible for the fine splitting of the TLS due to the magnetic and quadrupole moments of the nucleus, and the third term determines the matrix elements of the transitions between various levels in the microwave field. Finally, the fourth term couples the split levels of the system, which leads to allowed transitions between different fine structure levels and, ultimately, to the oscillations of the dipole echo in the magnetic field. Since \( |x_{\text{min}}|/x_0 \ll 1 \), the last term is small and may be taken into account perturbatively.

Performing the calculations in the lowest (second) order of perturbation theory, we come (similar to [4]) to the following expression for the echo amplitude:

\[ P_{\text{echo}} \propto -iV_1 V_2^2 \left( \frac{\Delta \omega}{E} \right)^4 \times \right. 

\left. \left[ 1 - \frac{64}{N} \sum_{n,m>n} \left( \frac{\Delta}{E} \right)^2 \left| (\overline{V}_Q)_{nm} \right|^2 \frac{\sin^4 (\varepsilon_{nm} \tau/2h)}{\varepsilon_{nm}^2} \right] \right]. \] (2)

Here, \( \tau \) is the time interval between two excitation pulses, \( \varepsilon_{nm} = (\overline{W}_Q)_{nn} - (\overline{W}_Q)_{nm} \) is the energy differences between the fine structure levels of the TLS. Equation (2) differs from similar Eq. (44) in [6] basically due to of the absolute-value square \( \left| (\overline{V}_Q)_{nm} \right|^2 \), which implies that our consideration is not limited by only the real-valued matrix elements \((\overline{V}_Q)_{nm}\).

The magnetic field enters Eq. (2) first via the energy difference \( \varepsilon_{nm} \) and, second, implicitly via the matrix elements \((\overline{V}_Q)_{nm}\), owing to the fact that they were calculated in the basis in which \( \widehat{W}_Q \) is diagonal.

According to our numerical calculations, the latter dependence is usually insignificant for finding the echo amplitude for the nuclei with integer spin \( J \). However, when the spin is a half-integer and the levels \( n \) and \( m \) are degenerate according to the Kramers theorem, the matrix element \((\overline{V}_Q)_{nm}\) cannot remove degeneracy and, consequently, must vanish in a zero magnetic field.

Formula (2) allows a numerical calculation of the echo amplitude for given parameters of the system. However, a direct application of this formula is complicated by the necessity of solving the algebraic equation of degree \( 2J + 1 \) with subsequent averaging of the results (which cannot be done in the general form even for \( J = 1 \)). Thus, Eq. (2) must be investigated in various limiting cases and analyzed numerically.

III. ANALYSIS OF LIMITING CASES

We begin the analysis of Eq. (2) with three independent energy scales appearing in it. The first two are the Zeeman energy \( E_H \) and the energy \( E_Q \) of the quadrupole interaction of the nucleus with the microscopic electric field. The third scale \( E_\tau = \hbar/\tau \) is determined by the time interval \( \tau \) between the pulses.

When the magnetic field is so high that \( E_H \) is greater than the other two scales, we can replace \( \sin^4 (\varepsilon_{nm} \tau/2h) \) by its average value and neglect the quadrupole interaction energy in the calculation of \( \widehat{W}_Q \) (and consequently \( \varepsilon_{nm} \)). In this case, the echo amplitude turns out to be

\[ P_{\text{echo}} \propto 1 - C/H^2, \] (3)

where the coefficient \( C \) is independent of the magnetic field.

Thus, the echo amplitude asymptotically (as \( 1/H^2 \)) approaches a constant value with an increase in the magnetic field, which agrees with the experimental data for high fields [4,8]. A less trivial magnetic-field dependence of the echo amplitude takes place when the Zeeman energy \( E_H \) is comparable with (or less than) at least one of the other energy scales \( E_Q \) or \( E_\tau \).
A. Small quadrupole splitting

Consider now the case where the quadrupole interaction energy is $E_Q \ll \mu H, E_r$. In this case, $E_H$ and $E_r$ may be comparable. Then, we can neglect the quadrupole energy in the calculation of $\tilde{W}_Q$ (but we still have to retain it in $\tilde{V}_Q$). Correspondingly, the set of the eigenfunctions of the projections of spin $J$ on the direction of the magnetic field can be used as the basis. In this case, the fine structure levels are $E_n = \mu H (J - n - 1)/J$, where $\mu$ is the nuclear magnetic moment and $n = 1, 2, ..., 2J + 1$.

In this case, since glass is isotropic and the trace of the tensor $\varphi'_{\alpha\beta}(0)$ is zero, the average squares of the absolute values of all matrix elements $|\tilde{V}_Q|_{nm}$ can be expressed in terms of a single constant. Then, the dipole echo amplitude (as a function of the magnetic field) is determined by the formula

$$P_{\text{echo}} \propto -i V_1 V_2^2 \left( \Delta u \over E \right)^4 \times$$

$$\left\{ 1 - C \frac{\Delta^2}{E^2} \left[ \sin^4(\mu H \tau / 2J) + \sin^4(\mu H \tau / J) \right] \right\}$$

(4)

Here, $C$ is the coefficient independent of the magnetic field (and different from $C$ in Eq. (3)).

The magnetic field enters into Eq. (4) as the product $\mu H \tau$. This implies that the variation of the dipole echo amplitude with $H$ should scale as $1/\tau$ along the horizontal axis.

Let us compare Eq. (4) with the measurement of the echo amplitude in the BK7 glass as a function of the magnetic field (Fig. 1). Obviously, the experimental dependence of the echo amplitude on $H$ has two components, one of which keeps its scale with a change in $\tau$ and the other one changes the horizontal scale as $1/\tau$.

The second component is well described by Eq. (4) with $\mu$ equal to the nuclear magnetic moment of the $^{10}$B atom (which is present in the BK7 glass and has a nuclear spin $J = 3$). We may suggest that the nonscalable component of the dependence is determined by the contribution of other atoms (e.g., Na or $^{11}$B) with the nuclear quadrupole moment, for which the condition $E_r \gg E_Q$ is not fulfilled.

B. Magnetic field dependence of echo amplitude in weak fields

Consider now another limiting case where $E_Q \simeq E_r$ but the Zeeman energy is low, $E_H \ll E_Q, E_r$. This will give us an idea of the behavior of an echo amplitude in small fields.

At $H = 0$, the energy differences $\varepsilon_{nm}$ between the fine structure levels of the TLS are fully determined by the energy of the quadrupole interaction of the nucleus with the internal field and enter the final formula for the echo amplitude through the function

$$\sin^4 y/y^2, \quad y = \varepsilon_{nm} \tau / 2\hbar.$$

(5)

The plot of this function (see Fig. 2 left pane) is a set of peaks with the $1/y^2$ envelope. The character of the magnetic-field dependence of the dipole echo amplitude in weak fields is determined by the fact that the values of $y_0$ corresponding to the zero-field splitting may fall to a minimum or maximum of dependence (5).

Consider for example the simplest case of the nucleus with $J = 3/2$. This nucleus has four fine structure levels and, respectively, three independent energy differences $\varepsilon_{nm}$. However, two of them ($\varepsilon_{12}$ and $\varepsilon_{34}$) vanish in zero magnetic field, according to the Kramers theorem. The corresponding matrix elements ($\tilde{V}_Q$)$_{nm}$ also vanish. As a result, the contribution of these two pairs of levels appears to be small compared to the contribution of the remaining $\varepsilon_{23}$ pair.

Thus, in this case, the behavior of an echo in weak fields is determined by the single energy difference $\varepsilon_{23}$. Suppose that the time interval $\tau$ between the pulses is such that $y_0 = \varepsilon_{23}(H = 0) \tau / 2\hbar = \pi k$ with integer $k$, which corresponds to a minimum of function (5). Then, in the presence of the magnetic field, $y$ must shift from the minimum position and, according to Eq. (2), the echo amplitude must decrease, irrespective of the properties of the system. Therefore, the magnetic-field dependence of the dipole echo should have a maximum at $H = 0$.

Figures 2a and 2b show the numerical results for the echo amplitude calculated with the use of Eq. (2) for the

![Figure 1: Magnetic-field dependence of the dipole echo amplitude in the BK7 glass (in arbitrary units) for $\tau$ = (from top to bottom) 0.75, 1.5, 2, 3, 6, and 12 $\mu$s. The points are the experimental data from 8 and solid lines are the theoretical curves for the nuclear magnetic moment of $^{10}$B calculated with the use of Eq. (4).](image-url)
case of $J = 3/2$ and $y_0 = \pi$ and $2\pi$, respectively (contributions of all levels were taken into account). Clearly, in agreement with the prediction, the echo amplitude has a maximum at zero magnetic field.

On the contrary, if $\tau$ is such that $y_0$ corresponds to a maximum of function (5), then the echo amplitude must increase in the presence of the magnetic field and the magnetic-field dependence of an echo should have a minimum at $H = 0$. The corresponding numerical results are shown in Figs. 2a and 2c.

For an arbitrary spin, this analysis is possible if the contribution of one energy splitting $\varepsilon_{nm}$ prevails over the contributions of the other pairs of levels. This occurs, e.g., if the other $\varepsilon_{nm}$ values are large and can be disregarded in Eq. (2) due to the term $1/\varepsilon_{nm}^2$.

Thus, it turns out that a zero-field maximum of the echo amplitude should change to a minimum and vice versa with a change in the time interval $\tau$ between the pulses.

A similar behavior was experimentally observed in glycerol with hydrogen replaced by deuterium [9, 10]. Deuterium has spin 1, which corresponds to three zero-field energy splittings $\varepsilon_{12} \propto 2\eta$, $\varepsilon_{23} \propto 3\eta$, and $\varepsilon_{13} \propto 3 + \eta$, where $0 < \eta < 1$ is the parameter of the microscopic field asymmetry. If $\eta \ll 1$, the first level splitting is much smaller than the other two and, in agreement with the above analysis, may play the main role in the magnetic-field dependence of the echo amplitude.

Figure 3 presents the experimental results [9, 10] and the positions of $y_0$ for different values of $\tau$. They were calculated with the fitted value $\varepsilon_{12}/h \approx 110$ kHz (which is comparable with an experimental value of 150 kHz [9]). Clearly, the echo amplitude as a function of the magnetic field has a minimum at $H = 0$ when $y_0$ appears near the maximum of function (5) and vice versa; i.e., the echo amplitude has a maximum at zero field when $y_0$ appears near the minimum of the function (5).

Summarizing, the elaborated theory of the dipole echo in the magnetic field with the inclusion of the nuclear quadrupole moments provides the explanation of all characteristic features observed in experiments and, in some cases, yield even a quantitative agreement between the experiments and analytic calculations. However, the comparison of the magnetic-field dependence of the dipole echo amplitude in an arbitrary glass with the theory is complicated by superimposed contributions from the atoms with different nuclear magnetic and quadrupole moments.

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