Dynamic processes in dielectric glasses at low temperatures

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The behavior of dielectric glasses at low temperatures when acted on by electric and acoustic fields is discussed. The anomalous low-temperature behavior of the heat conductivity and the heat capacity is described. A phenomenological model of two-level systems in glasses arising from tunneling transitions is presented. Results are given of the experimental and theoretical studies of the phenomena of saturation, induced transparency of the medium by application of acoustic and electric pulses, and "hole-burning" in an inhomogeneously broadened line. Considerable attention is paid to discussing the experimental results on generation of coherent responses of echo signals. In order to describe the formation of phonon-echo signals in glasses, the concept is employed of spectral diffusion, which consists of a dynamic alteration of the resonance frequency. We stress both the aspects of the properties that are common with the phenomena of magnetic resonance, and the differences that arise owing to the broad spectrum of the two-level systems.

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CONTENTS

1. INTRODUCTION

Heightened interest has been seen recently in studying disordered systems such as spin glasses, amorphous metals, superionic conductors, etc. Besides these systems, considerable attention has been paid to such a broad class of amorphous compounds as the dielectric glasses. These objects have attracted little attention from theoreticians and experimentalists, owing to the complexity of the calculations of models of disordered systems and the lack of long-range order parameters. A breakthrough occurred after the studies of Zeller and Pohl,[1] who first turned their attention to the anomalous behavior of the heat capacity and the heat conductivity of various glasses at low temperatures and the study of Heinicke *et al.*,[2] who discovered the anomalous absorption of sound in analogous objects. Further studies have shown that the low-temperature anomalies are characteristic of all types of dielectric glasses, independently of their composition.[3,4]

Several models have been proposed to explain the experiments. The most successful model has proved to be that of Anderson, Halperin, Varma, and Phillips, which postulates the existence in glasses of tunneling transitions that lead to a broad energy spectrum (10^8–10^{12} Hz) of localized two-level systems. This subsequently led to a phenomenological description of a new spectrum of elementary excitations using the "pseudospin" $\hat{S} = 1/2$. Here a remarkable analogy arose between the phenomena in spin resonance and the low-temperature properties of glasses. A number of effects was found, such as saturation and an associated anomalous variation in the velocity of sound with temperature, induced transparency of the medium caused by acoustic and electric pulses, "hole-burning" in an inhomogeneously-broadened line, and the discovery of the electric and acoustic echo.[5,6] Here we must mention that the fundamental difference from radiospectroscopy is the extremely broad spectrum of two-level systems. Moreover, it is assumed that the number of states per unit energy interval is a constant or a slowly varying function. One of the consequences of this postulate is the deduction of a logarithmic dependence of the heat capacity on the time taken to perform the experiment.

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We note that there is as yet no consistent microscopic theory to substantiate the existence of localized two-level systems in dielectric glasses at low temperatures. More than that, the very possibility of existence of a low-energy spectrum of elementary excitations in glasses aroused some doubt until recently. Therefore the discovery and study of coherent responses in amorphous specimens upon pulsed excitation with electric and acoustic fields was the most convincing confirmation of the hypothesis of the existence of two-level systems arising from tunneling transitions. The description of the processes of formation of the phonon echo in glasses by a relatively narrow band of frequencies of exciting pulses, as compared with the broad spectrum of two-level systems that we have already mentioned, required the concept of spectral diffusion to explain it.[7] In turn, this led to a deeper understanding of the phenomenon of acoustic saturation. In particular, the conclusion was most interesting that the width of the spectrum of saturable frequencies depends on the duration of the saturating pulse. This was subsequently confirmed experimentally. We see from what we have said that up to now a large number of results has accumulated on the dynamic processes in dielectric glasses at low temperatures. However, the literature contains no sufficiently complete review reflecting the current state of this problem, apart from the excellent article of Hunklinger and Arnold,[8] which was concerned exclusively with acoustic studies in glasses up to 1975. We have set ourselves the problem of reflecting the principal theoretical and experimental results on dynamic processes in dielectric glasses in electric and acoustic fields. In presenting the material, we have deliberately restricted the treatment to a phenomenological description employing the concept of, the "pseudospin" $\hat{S}=1/2$. This has permitted us to put the major stress on the physics of the phenomena and to describe qualitatively the entire set of numerous experimental facts. Throughout the review, we stress both what the properties have in common with the phenomena of magnetic resonance and the differences that arise owing to the extremely broad spectrum of two-level systems. In spite of the breadth of problems discussed, the review is not complete. Thus, for example, we have hardly treated the anomalous variation of the velocity of sound with the temperature, which has been presented rather fully in Ref. 8. We should note that the fundamental views on this problem have not suffered substantial changes in the past five years.

2. LOW-TEMPERATURE ANOMALIES IN DIELECTRIC GLASSES

a) Microscopic structure of glasses

The current view on the microscopic structure of glasses is based on the hypothesis of Zachariasen,[9] who proposed that one can consider a glass to be a disordered network of atoms having the same interatomic distances as in the corresponding crystal structure. The simplest and best studied is the silica glass SiO_2. X-ray diffraction studies have shown that the mean distances between two nearest silicon and oxygen atoms are 1.62 Å, while those between nearest silicon atoms

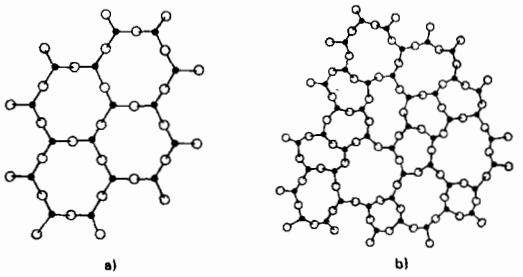

FIG. 1. a) Two-dimensional diagram of a crystal; b) the disordered structure of glasses.

are 3 Å. This practically matches the interatomic distances in the crystalline modifications of SiO_2. However, the glasses exhibit a large spread of Si-O-Si angles from 120° to 180° (Fig. 1). The experiments show that order is conserved in silica glasses at distances of 10–12 Å. In the more complex cases the irregular network of multicomponent glasses consists of both plane units of the type of SiO_2 and SiO_4 tetrahedra linked at random.

Kittel[10] first paid attention to the difference in behavior of the thermal properties of dielectric crystals and glasses in the temperature range from 100 to 300 K. In contrast to χ_{cryst}, the heat conductivity χ_{gl} of glasses declined with decreasing temperature and depended weakly on their composition. Here χ_{gl} was considerably lower than χ_{cryst} (Fig. 2). Kittel proposed that the mean free path of phonons is close in order of magnitude to the dimensions of the SiO_4 tetrahedra. This gave rise to the small spread in values of the heat conductivity of different glasses as compared with crystals. The decrease in the heat conductivity with decreasing temperature was explained by the difference between plane waves and the normal modes. They coincide in a regular lattice. In glasses an original plane wave becomes distorted and one can treat it as a set of normal modes having different eigenvalues. With decreasing temperature the mean free path of phonons begins to increase and becomes greater than the dimensions of the tetrahedral cell. Actually, Berman[11] discovered a plateau in the temperature-dependence of the heat conductivity at

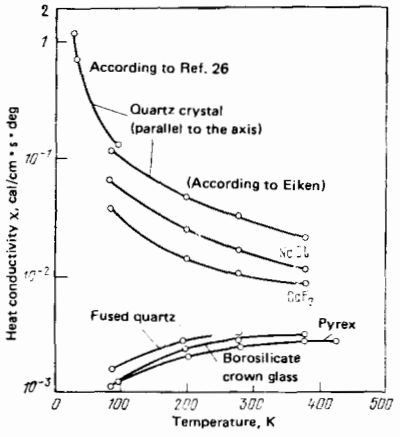

FIG. 2. Temperature-dependence of the heat conductivity of crystals and glasses.

10 K, and associated its appearance with an increased mean free path in the low-temperature region. Subsequently this theory has been refined by a number of authors and some quantitative estimates have been offered.[12] It seemed that the thermal properties of glasses had been explained satisfactorily, at least qualitatively. However, subsequent experiments required a cardinal reexamination of the existing views on the mechanisms of heat conduction and heat capacity in glasses in order to interpret them.

b) Low-temperature anomalies of heat conductivity and heat capacity in dielectric glasses

In order to understand what was expected to be found in glasses at low temperatures, we recall how the heat conductivity and the heat capacity behave in dielectric crystals. According to the well-known Debye theory, heat is conducted in dielectric crystals by phonons, while the mean free path l determines the rate of energy exchange between the phonons belonging to different modes. The heat conductivity is determined by the expression

$$\chi = \tfrac{1}{3} Cvl. \qquad (1)$$

Here v is the mean velocity of the phonons and C is the heat conductivity of the lattice. At low temperatures l becomes comparable with the dimensions of the crystalline specimen, and the variation of the heat conductivity is determined by the temperature-dependence of the heat capacity according to the law T^3. Temperatures below 1 K correspond to thermal phonons with a wavelength greater then 1000 Å. This exceeds by a factor of hundreds the dimensions of the interatomic distances and those of the elementary tetrahedra of the basic structure of the glasses. Therefore, at low temperatures the phonons will propagate in glasses as in a homogeneous crystalline medium, and there should be no fundamental difference in the temperature behavior of the heat conductivity and the heat capacity of amorphous dielectrics and crystals. Zeller and Pohl[1] first studied the heat conductivity of various glasses at low temperatures. They showed that the temperature-dependence of the heat conductivity of all the studied glasses in the range from 0.1 to 1 K is determined by the expression $\chi = AT^{\sigma}$, where $\sigma \approx 1.8$ (Fig. 3). Moreover, the temperature-dependence of the mean free path of phonons was determined. Figure 4 indicates that the mean free path of phonons increases with decreasing temperature. This causes the values of the heat conductivity of quartz glass and crystalline quartz to approach each other at $T < 10$ K (Fig. 3). Figure 4 implies that we have $l \propto \omega^{-2}$ at temperatures from 0.1 to 1 K. In addition to these facts, Zeller and Pohl discovered an anomalous behavior of the temperature-dependence of the heat capacity

$$C = AT + BT^3. \qquad (2)$$

Here the linear term in the heat capacity varies little for different specimens. These experimental results

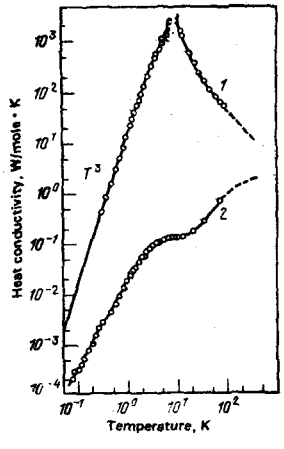

FIG. 3. Temperature-dependence of the heat conductivity.[1] 1—crystalline quartz, 2—quartz glass.

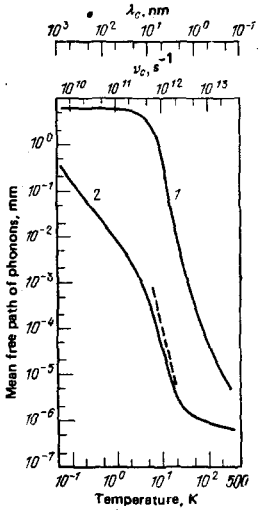

FIG. 4. Temperature-dependence of the mean free path.[1] 1—crystalline quartz, 2—quartz glass.

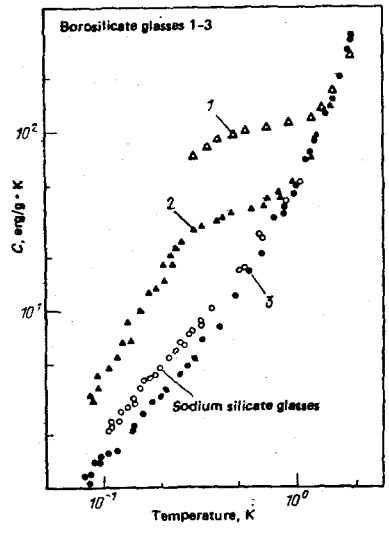

FIG. 5. Temperature-dependence of the heat capacity for glasses having differing contents of magnetic impurities.[3] 1—Pyrex No. 7740, Fe concentration 100 ppm; 2—Pyrex No. 9700, Fe concentration 12 ppm; 3—Pyrex No. 9700 in a magnetic field $H = 33$ kG.

are not explained by models based on various modifications of the Debye law. Then Stephens[3] performed detailed studies of a large number of specimens. He found that the heat conductivity $\chi \sim T^2$ practically did not depend on the impurities. The heat capacity was described by the expression (2), with the value of B about three times larger than the value obtained on the basis of the Debye theory, while the linear term predominates below 0.2 K. While the heat conductivity did not depend on the impurities, in contrast, an increase in the concentration of iron increased the heat capacity. Upon applying the magnetic field $H = 33$ kG, it became possible to eliminate the effect of magnetic impurities and the temperature-dependence of the heat capacity practically coincided with the analogous dependence for glasses without the magnetic impurities (Fig. 5). Several theories have been proposed to explain these phenomena,[13] but we shall take up in detail the theory of Anderson, Halperin, Varma, and Phillips,[14,15] which has not only explained the results that were obtained, but also has predicted a number of new effects, which have subsequently been confirmed experimentally.

3. METHODS OF THEORETICAL DESCRIPTION OF THE ANOMALOUS BEHAVIOR OF DIELECTRIC GLASSES

a) Model of localized low-energy excitations

This model is based on the hypothesis that atoms or groups of atoms exist in all dielectric glasses that can lie with almost equal probability in two equivalent positions. This situation is conveniently described by a double asymmetric well potential (Fig. 6). The most important parameters are: V–the height of the potential barrier, ε–the energy asymmetry of the two local minima, and d–the distance between the two minima. Tunneling leads to the binding energy $\Delta = \hbar\omega_0 \exp(-\lambda)$, where ω_0 is the frequency of the zero-point oscillations, $\lambda = \hbar^{-1} d\sqrt{2mV}$ is a parameter that allows for the overlap of the wave functions, and m is the mass of the tunneling particle or group of atoms. Tunneling leads to removal of degeneracy, so that the energy splitting of the two lower levels is

$$E = \sqrt{\varepsilon^2 + \Delta^2}. \tag{3}$$

At low temperatures one can restrict the treatment to the two lower levels, which have the splitting E and the population difference described by the Boltzmann factor $\exp(-E/2kT)$. The quantities ε and Δ vary randomly, owing to such factors as a different environment of the atoms around the two potential minima, local stresses,

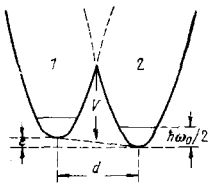

FIG. 6. Double asymmetric potential well. V—height of barrier, ε—asymmetry parameter, d—distance between the energy minima, $\hbar\omega_0/2$—energy of the ground state.

etc. In turn, this gives rise to a random splitting of the energy levels. Upon introducing the number $n(E)$ of two-level systems per unit energy interval, we can easily calculate the heat capacity of the set of such localized states:

$$C = k \int_0^\infty n(E) \left\{ \left(\frac{E}{2kT}\right)^2 \frac{\exp(-E/2kT)}{[1+\exp(-E/2kT)]^2} \right\} dE. \tag{4}$$

The important hypothesis has been advanced[14,15] that $n(E)$ is a slowly varying continuous function of E, with $n(0) \neq 0$. This leads directly to a linear temperature-dependence of the heat capacity:

$$C \propto \frac{\pi^2}{6} k^2 T n(0). \tag{5}$$

This model also allows one to describe the interaction of the two-level systems with electric and acoustic fields. Coupling between the localized two-level systems and phonons arises from the deformation of the double potential well by the elastic waves. We can write the Hamiltonian of the original two-level system in the form

$$\mathcal{H}_0 = \frac{1}{2}\begin{pmatrix} \varepsilon & \Delta \\ \Delta & -\varepsilon \end{pmatrix}. \tag{6}$$

Upon assuming for simplicity that the strain of a longitudinal acoustic wave mainly alters the asymmetry E, we can represent the interaction Hamiltonian as

$$\mathcal{H}_1 = -\gamma e \begin{pmatrix} 1 & \\ & -1 \end{pmatrix}. \tag{7}$$

Here $\gamma = \partial\varepsilon/\partial e$ is the deformation potential. Upon expressing the elastic strains in terms of the operators for creation and annihilation of phonons,[14] we can write the matrix element between the states 1 and 2 of the Hamiltonian of (6):

$$\langle k2 \mid 1\rangle = \sqrt{\frac{k}{2\rho v_l}} \gamma \frac{\Delta}{E}. \tag{8}$$

Here ρ is the density, and v_l is the velocity of a longitudinal acoustic wave having the wave vector $\mathbf{k}$. By using the matrix elements of (8), we can easily calculate[8,16] the time T_1 that it takes the two-level system to go over to the lower state upon emitting a phonon and the time τ_k for resonance absorption of a phonon by the localized mode:

$$T_1^{-1} = \left(\frac{1}{v_l^5} + \frac{2}{v_t^5}\right) \frac{\gamma^2 \Delta^2 E}{2\pi\rho\hbar^3} \coth\frac{E}{2kT}, \tag{9}$$

$$\tau_{k,\alpha}^{-1} = \frac{\pi\gamma^2 P\omega}{\rho v_\alpha^2} \tanh\frac{E}{2kT}. \tag{10}$$

Here v_t is the velocity of a transverse sound wave, $\omega = kv$, and $\alpha = l, t$. In the high-temperature approximation $h\omega < 2kT$, Eq. (10) implies that the absorption $l^{-1} = \tau v$ behaves as follows:

$$l^{-1} \propto \frac{\omega^2}{T}. \tag{11}$$

The expression (11) explains the observed temperature-dependence of the mean free path of phonons[1] (Fig. 4). Substitution of Eqs. (11) and (5) into (1) yields $\chi \propto T^2$. Thus the theory developed in Refs. 14 and 15 explains all the fundamental results on the anomalous behavior of glasses at low temperatures. Along with this, a set of conclusions stems from the obtained results that can be

directly tested experimentally. First there is the relationship $l^{-1} \propto \omega^2$. Further, the effect of diminished sound absorption, which is associated with the equalization of the populations, is taken into account with the factor $\coth(E/2kT)$. Such effects, which are due to saturation, are well known in magnetic resonance.[18,19] For a symmetric potential well ($\varepsilon = 0, E = \Delta$), Eq. (6) fully coincides with the expression for the time T_1 employed in calculations of one-phonon processes of spin-lattice relaxation.[20] However, we must note that the two-level systems possess the essential feature of the continuous distribution of splittings. And in some cases a simple analogy with magnetic resonance can give rise to false conclusions.

Another peculiar property of glasses is the slowly varying density of states $n(E)$ that we have already mentioned above. In line with this, it is interesting to trace what restrictions this feature imposes on the tunneling parameter λ.[14] To do this, let us replace the density of states $n(E)$ in Eq. (4) by

$$P(\varepsilon, \lambda)\, d\varepsilon d\lambda \approx P(0, \lambda)\, d\varepsilon d\lambda \equiv P(\lambda)\, d\varepsilon d\lambda. \tag{12}$$

Here $P(\varepsilon, \lambda)$ is the number of two-level systems having the asymmetry parameter ε and the tunneling parameter Δ per unit volume. For a given value, of E, the parameter λ cannot be smaller than $\lambda_{\min}$, which is defined by the condition that the corrections to the eigenvalues of the Hamiltonian of (7) due to the nondiagonal elements are small:

$$\lambda > \lambda_{\min} \equiv \ln \frac{\hbar\omega_0}{E}. \tag{13}$$

While employing Eq. (13) and the definition of Δ, let us represent Eq. (9) in the more convenient form:

$$T_1^{-1} = \left(\frac{1}{v_l^5} + \frac{2}{v_t^5}\right) \frac{\gamma^2 E^3 \exp[-2(\lambda - \lambda_{\min})]}{2\pi\hbar^4\rho} \coth\frac{E}{2kT}. \tag{14}$$

The energy splitting varies continuously from $\lambda_{\min}$ to some cutoff value $\lambda_{\max}$. We can conveniently introduce the parameter $\eta = \lambda_{\max} - \lambda_{\min} + \ln 2$, The parameter η denotes the width within which we have $\bar{P}(\lambda) = \bar{P}$. The number of two-level systems that contribute to the heat capacity is $n(E) = \eta P$. We see from Eq. (14) that the relaxation time varies by several orders of magnitude for small changes in η. The magnitude of η is unknown, and it has been taken to be from one to ten. On the other hand, one can find the limit $\lambda_{\max}$, which is defined by the shortest relaxation time $T_{1,\min}$ for the two-level systems having the energy splitting $E = 2kT$:

$$\lambda < \lambda_{\max} \equiv \frac{1}{2} \ln(\Gamma t), \tag{15}$$

here we have $\Gamma = T_{1,\min}^{-1}$. If the time t for measuring the heat capacity becomes comparable with $T_{1,\min}$, a logarithmic dependence of the heat capacity on the time for performing the experiment arises: $C \sim \ln(t/T_{1,\min})$.

No experimental confirmation of this behavior of the heat capacity has been obtained. Moreover, results have appeared that contradict these conclusions.[21,22] On the other hand, recent experiments to study the temperature profiles have detected an anomalous behavior for thermal pulses of duration 0.1 ms.[23]

b) Description of the low-temperature properties of glasses using the "pseudospin" $\hat{S} = 1/2$

A merit of the models developed in Refs. 14 and 15 also consists of the fact that the coupling of the two-level systems with the acoustic deformations can be represented in a form analogous to the interaction of spins $\hat{S} = 1/2$ with external magnetic fields.[19,24] Actually, upon diagonalizing the Hamiltonian of (6), we obtain

$$\hat{\mathscr{H}}_0 = \sum_i E^i \hat{S}_z^i. \tag{16}$$

Here E^i is defined by Eq. (3), and the superscript i means that the quantity E^i pertains to the concrete ith two-level system. However, the transformation that diagonalizes $\mathscr{H}_0$ simultaneously converts the interaction Hamiltonian of (7) into the form

$$\hat{\mathscr{H}}_1 = -\sum_i (B^i e^i \hat{S}_x^i + D^i e^i \hat{S}_z^i), \tag{17}$$

here we have

$$B^i = \frac{2\gamma\Delta^i}{E^i}, \quad D^i = \frac{2\gamma\varepsilon^i}{E^i}. \tag{18}$$

Since the two-level systems can have an electric dipole moment, then, by writing the Hamiltonian for interaction with electric fields in a form analogous to (7) and diagonalizing, we obtain the following instead of Eq. (17):

$$\hat{\mathscr{H}}_1 = -\sum_i (\bar{\mu}^i F^i \hat{S}_x^i + \mu^i F^i \hat{S}_z^i). \tag{19}$$

Here $\bar{\mu}^i$ and μ^i are the nondiagonal and diagonal dipole moments, and F^i is the electric field.

Thus we see that the tunneling model possesses an analogy with a spin $\hat{S} = 1/2$ in the "constant magnetic field" E^i. Here this field varies from node to node within broad limits. The interaction Hamiltonians in (17) and (19) give rise to both diagonal and nondiagonal transitions. Bearing this analogy in mind, we shall employ everywhere below the terms "spin", "spin-spin interaction", and "spin-phonon interactions" instead of "pseudospin-phonon interactions", etc. This allows us to transfer to the two-level systems, a number of conclusions well known in magnetic resonance, e.g., the Bloch equations. We need not reevaluate Eqs. (16)–(19). In contrast to the usual magnetic interactions,[25] the Hamiltonian in (7) is symmetry-invariant with respect to the time inversion $t \to -t$.

4. EFFECTS OF SATURATION BY ACOUSTIC AND ELECTRIC PULSES

a) The discovery of acoustic transparency in glasses

Simultaneously with the first results of Zeller and Pohl,[1] a disagreement was discovered between the mean free path l of thermal phonons as determined from the heat conductivity and the data on propagation of longitudinal acoustic waves of frequency 24 GHz.[2] Thermal phonons of this frequency make the major contribution to the heat capacity at 0.4 K, and their mean free path proves to be 0.005 cm. However, experiment[2] has shown that the mean free path is $l \sim 0.2$ cm for longitudinal acoustic waves, even at $T = 3.5$ K. This disagree-

ment has been explained in subsequent experiments to study the damping of acoustic pulses in glasses as a function of their power.[26,27] The absorption of acoustic pulses was measured in fused quartz and borosilicate glass from 0.1 to 2.5 K at frequencies 0.4–2 GHz. The energy of the acoustic pulses was varied from 10^{-7} to 10^{-2} ergs. A sharp alteration in the damping of the acoustic pulses was found as their energy was increased above 10^{-3} ergs. Figure 7 shows oscillograms of multiply reflected acoustic pulses of frequency 725 MHz propagating in a glass for two values of the introduced power. We see that the pulses of higher power are damped considerably more weakly. The dependence of the first reflected pulse on the energy introduced into the specimen was also studied. When $E > 10^{-3}$ erg, the medium becomes linear with a damping $l^{-1} = \ln(E/E_1)/2L$, where L is the length of the specimen. We note that, when one performs the same experiments on crystalline quartz, the acoustic absorption does not depend on the energy of the applied pulses. The quartz crystal was a linear medium throughout the energy range.

The dependence of the absorption of acoustic pulses on their intensity J has been determined[28] at three fixed frequencies (Fig. 8). Within the limits of experimental error, the absorption varied as ω^2, in line with the conclusions of Eq. (11). For relatively large intensities the intensity varied as $l \sim J^{-1/2}$. One can explain these results qualitatively with the concept of localized two-level systems. Actually, starting at a certain intensity ($\sim 10^{-7}$ W/cm^2), the populations become equalized, i.e., saturation sets in, with a consequent decrease in sound absorption.

b) Application of the Bloch equations to saturation processes

For an analytic description of the effects of saturation we shall employ the Bloch equations, which have enjoyed widespread use in magnetic resonance[19]:

$$\left.\begin{aligned}
\frac{dM_x(\omega')}{dt} &= -\omega' M_y(\omega') - T_2^{-1} M_x(\omega'),\\
\frac{dM_y(\omega')}{dt} &= \omega' M_x(\omega') + \frac{Be}{\hbar} M_z(\omega') - T_2^{-1} M_y(\omega'),\\
\frac{dM_z(\omega')}{dt} &= -\frac{Be}{\hbar} M_y(\omega') - T_1^{-1}[M_z(\omega') - M_z^0(\omega')].
\end{aligned}\right\} \qquad (20)$$

Here we have $M(\omega') \equiv \langle S^i \rangle$ with energies $E^i = \hbar\omega'$. T_1 and T_2 are the longitudinal and transverse relaxation times, $Be_0/\hbar$ is the nutation frequency in accordance with Eqs. (17), $M_z(\omega') = (1/2)\tanh(\hbar\omega'/2kT)$, and the deformation e varies according to the law $e(t) = e_0 \cos(\omega t)$. One uses

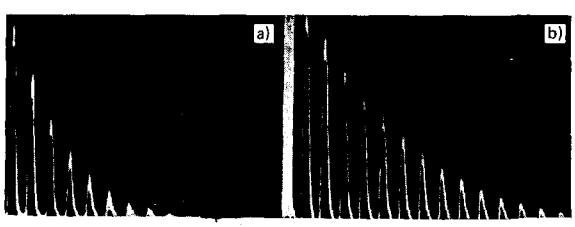

FIG. 7. Damping of pulses of longitudinal acoustic waves in borosilicate glass at $T = 0.35$ K.[8] Power introduced into the specimen: a) 5 μW/cm^2; b) 1 mW/cm^2.

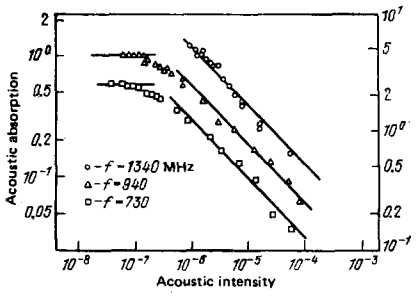

FIG. 8. Dependence of the acoustic absorption in borosilicate glass for longitudinal waves on their intensity for three fixed frequency values.[28]

the standard method of magnetic resonance to seek a stationary solution of Eqs. (20). The latter implies that saturation sets in when the condition is satisfied that $(Be_3/2\hbar)^2 T_1 T_2 > 1$.

In the saturation regime the resonance curve becomes broader in the ratio $[1 + (Be_0/2\hbar)^2 T_1 T_2]^{1/2}$. When we allow for the broadening, the formula (10) for the absorption takes on the form

$$l^{-1} = \frac{\pi B^2 \bar{p}}{\rho v_\alpha^3} \frac{\omega}{\sqrt{1 + (B^2 l_0^2 T_1 T_2/\hbar^2)}} \tanh\frac{\hbar\omega'}{2kT}. \qquad (21)$$

Let us introduce the value of the critical intensity I_c, which is

$$I_c = \frac{\hbar^2 \rho v_\alpha^3}{2B^2 T_1 T_2}. \qquad (22)$$

Taking Eq. (22) into account, we write the absorption in the form

$$l^{-1} = l_0^{-1} \frac{\tanh(\hbar\omega/2kT)}{\sqrt{1 + (J/J_c)}}. \qquad (23)$$

Here we have $l_0^{-1} = \pi \bar{p} B^2 \omega/\rho v_\alpha^3$, and $J = \rho v_\alpha^3 e_0^2/2$. When $J/J_c \ll 1$, which corresponds to resonance absorption in the absence of saturation, Eq. (23) goes over into Eq. (10). In the presence of saturation ($J/J_c \gg 1$), we have

$$l^{-1} \propto \frac{\omega^2}{T}\sqrt{\frac{J_c}{J}}. \qquad (24)$$

The results of the experiments[26-28] agree well with the conclusions of (24). Equations (21) and (24) yield a quadratic dependence of the absorption on the frequency, both at low powers and in a saturation regime.

c) Interaction with electric fields

As mentioned above, the "spins" are coupled not only with acoustic fields, but also with the electric component of an uhf field. Actually, an anomalous behavior of the dielectric constant with the temperature and with saturation by the electric field has been found in studies on dielectric absorption.[29,30] Figure 9 shows the relationship of the dielectric absorption at the temperature of 0.4 K and frequency of 10 GHz to the intensity of the electric component of the uhf field.

At intensities $I > 10^{-3}$ W/cm^2, a saturation process begins that leads to a decrease in the absorption proportional to $I^{-1/2}$. One can obtain an analytic expression for the dielectric absorption by replacing the acoustic quantities with the corresponding electric parameters[30]:

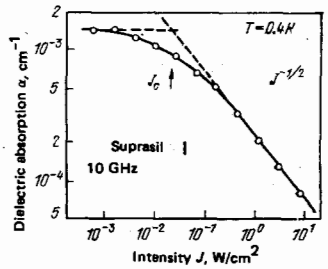

FIG. 9. Dependence of the dielectric absorption on the intensity of the electric component of the uhf field.[30]

$$\alpha = \frac{4\pi^2\omega n\mu^2}{3c\sqrt{\varepsilon_0}}\left(1+\frac{J}{J_c}\right)^{-1/2}\tanh\frac{\hbar\omega}{2kT}. \qquad (25)$$

Here c is the velocity of light and ε_0 is the dielectric constant. The natural question arises: are the same two-level systems responsible for the elastic and the electric properties? Recent experiments confirm this hypothesis. The acoustic absorption was measured in fused quartz and borosilicate glass while the specimen was simultaneously treated with an electromagnetic and an acoustic field at nearby frequencies. The measurements were performed at frequencies of 1 GHz at 0.5 K,[30] and 9 GHz at 1.5 K.[31,32] A strong dependence of the ultrasonic absorption on the power of the electromagnetic field was found (Fig. 10). The effect increases as the frequencies of the electromagnetic and ultrasonic pulses approach. Analogous results have been obtained upon employing a sensitive method of measurement in which the action of the ultrasonic pulse was detected from the variation in the electric impedance of a resonance cavity containing the specimen. These experiments resemble in methodology the studies on acoustic nuclear magnetic resonance and acoustic paramagnetic resonance.[33] Here the oscillating magnetic field is replaced with an electric field. These results have convincingly demonstrated that the two-level systems simultaneously possess both elastic and electric properties.

d) Temperature-dependence of the velocity of sound and the dielectric constant

By employing the Kramers-Kronig relationships, we can express the variation of the velocity of sound in terms of the absorption

$$\frac{\Delta v}{v} = \frac{v(T)-v(T_0)}{v(T_0)} = \int_0^\infty \frac{v}{\pi}\frac{l^{-1}(\omega', T)-l^{-1}(\omega', T_0)}{\omega^2-\omega'^2}\,d\omega'. \qquad (26)$$

Here the crossed integral sign denotes an integral in the sense of its principal value.

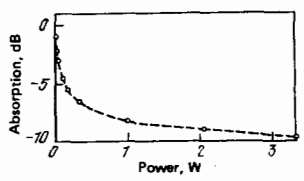

FIG. 10. Dependence of ultrasonic absorption in borosilicate glass on the power of the electromagnetic pulses.[32]

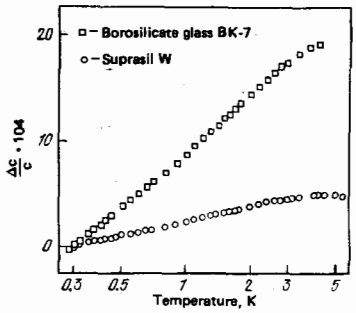

FIG. 11. Relative variation of the speed of light $\Delta c/c \cdot 10^4$ as a function of the temperature.[29]

In principle the study of the velocity of sound as a function of the temperature yields the same information as the absorption of sound. It depends less on the acoustic power, since both resonance and nonresonance two-level systems contribute. In the high-temperature approximation with $\hbar\omega < 2kT$, we obtain the following by using Eq. (26)[8]:

$$\frac{\Delta v}{v} = k^2 \ln\frac{T}{T_0}, \qquad (27)$$

here we have

$$k^2 = n_0 B^2/4\rho v^2. \qquad (28)$$

Similarly we can describe also the behavior of the dielectric constant in glasses:

$$\frac{\Delta C}{C} = -\frac{1}{2}\frac{\Delta\varepsilon_0}{\varepsilon_0} = \frac{p^2}{\varepsilon_0} n \ln\frac{T}{T_0}. \qquad (29)$$

Figure 11 shows the temperature-dependence of $\Delta c/c$.

5. DIRECT INTERACTIONS BETWEEN TWO-LEVEL SYSTEMS

In the Bloch equations (20), one introduces phenomenologically two relaxation times T_1 and T_2. The time T_1 is governed by direct single-phonon processes and can be calculated by Eq. (9). The hypothesis was advanced in Ref. 34 of the existence of a "spin-spin interaction" in glasses. Here, by analogy with the spin-spin interactions in magnetic resonance, it has been possible to define a time

$$T_2 \ll T_1. \qquad (30)$$

Further studies have shown that the width arising from the static "spin-spin" interactions in glasses are often masked by dynamic processes, e.g., spectral diffusion. Owing to the importance of the "spin-spin" interactions in interpreting a number of the experimental results, let us take up in greater detail the physical nature of the source of the coupling between the two-level systems.

Let us write the Hamiltonian of the interaction of the two-level systems with elastic fields, taking into account the fact that the deformations cause not only a change in the asymmetry ε, but also in the parameter Δ. Then we obtain the following instead of Eq. (7):

$$\mathcal{H}_1 = \sum_{\alpha,\beta}\left(e_{\alpha\beta}\frac{\partial\varepsilon}{\partial e_{\alpha\beta}} + e_{\alpha\beta}\frac{\partial\Delta}{\partial e_{\alpha\beta}}\right)\frac{1}{E}\begin{vmatrix} \varepsilon & \Delta \\ \Delta & -\varepsilon \end{vmatrix}, \qquad (31)$$

here we have

$$e_{\alpha\beta} = \partial_\alpha u_\beta + \partial_\beta u_\alpha. \tag{32}$$

Again, upon diagonalizing the expression (31) and employing the spin operators, we obtain instead of Eq. (16) the interaction Hamiltonian in the form

$$\hat{\mathcal{H}}_i = \sum_{i,\alpha,\beta} (e^i_{\alpha\beta} B^i_{\alpha\beta} \hat{S}^i_x + e^i_{\alpha\beta} D^i_{\alpha\beta} \hat{S}^i_z), \tag{33}$$

$$B^i_{\alpha\beta} = 2\left(-\frac{\partial \varepsilon^i}{\partial e_{\alpha\beta}} \frac{\varepsilon^i}{E^i} + \frac{\partial \Delta^i}{\partial e_{\alpha\beta}} \frac{\Delta^i}{E^i}\right), \tag{34}$$

$$D^i_{\alpha\beta} = 2\left(\frac{\partial \varepsilon^i}{\partial e_{\alpha\beta}} \frac{\varepsilon^i}{E^i} + \frac{\partial \Delta^i}{\partial e_{\alpha\beta}} \frac{\Delta^i}{E^i}\right). \tag{35}$$

Taking into account the interaction via the phonon field field analogously to the calculations in EPR,[35,36] we obtain the Hamiltonian of the "spin-spin" interactions

$$\hat{\mathcal{H}}_{s\text{-}s} = \sum_{i,j} J^{xx}_{ij} (\hat{S}^i_+ \hat{S}^j_- + \hat{S}^i_- \hat{S}^j_+) + \sum_{i,j} J^{zz}_{ij} \hat{S}^i_z \hat{S}^j_z. \tag{36}$$

The quantities J^{xx}_{ij} and J^{zz}_{ij} depend in an essential manner on the relationship between the wave vector q and the distance r from the defect i to the defect j. When $r_{ij} \ll q^{-1}$, we have

$$J^{xx}_{ij} = \frac{B^i_{\alpha\beta} B^j_{\alpha\beta}}{A(r_{ij})^3}, \qquad J^{zz}_{ij} = \frac{D^i_{\alpha\beta} D^j_{\alpha\beta}}{A(r_{ij})^3}. \tag{37}$$

Here we have $A = 64\pi\rho^2$, and $B^i_{\alpha\beta}$ and $D^i_{\alpha\beta}$ are deformation potentials of the order of 1 eV.[34] Thus Eq. (36) resembles in structure the dipole-dipole interaction Hamiltonian well known in magnetic resonance. The quantities J^{xx}_{ij} couple only the "spins" having identical energy splittings, whereas the J^{zz}_{ij} couple all the two-level systems. The continuous distribution of two-level systems fundamentally distinguishes the situation in glasses from the case of magnetic resonance, where the unlike spins are, e.g., the electronic and nuclear spins. In glasses all the "spins" are in some sense unlike. We can consider the like two-level systems to be those that satisfy the condition[37] $|E_i - E_j| < J^{xx}_{ij}$. One can determine the concentration of like "spins" for which we must take into account both J^{xx}_{ij} and J^{zz}_{ij}. It turns out that[37] $c \sim 10^{-2}$. Therefore, below in treating the phonon echo, we shall restrict the treatment to the terms containing J^{zz}_{ij}.

The treatment of the interaction via the phonon field assumes a regular lattice. This description is ill-suited to such disordered systems as glasses. Therefore the derivation of the spin-spin interactions from the equations of propagation of sound in glasses as a continuous medium is of undoubted interest.[7] Here $D^i_{\alpha\beta} S^i_z$ plays the role of an "elastic dipole moment" and can be treated as the source of the elastic field, just as the magnetic moment is the source of the magnetic field. Therefore the term $e^i D^i S^i_z$ represents the external stress localized at the node i, and according to Hooke's law[38] we have

$$\rho v_t^2 \nabla^2 u_\alpha + \rho (v_e^2 - v_t^2) \partial_\alpha (\partial_\gamma u_\gamma) = (D_{\alpha\beta} S_z) \partial_\beta \delta(r). \tag{38}$$

Solution of Eq. (38) yields the displacement $u_\alpha(r_{ij})$, from which we can derive the elastic deformation tensor by employing Eq. (32) for isotropic coupling $\gamma_{\alpha\beta} = \gamma\delta_{\alpha\beta}$:

$$e_{\alpha\beta} = \frac{-2\gamma (\Delta/E) S_z}{4\pi\rho v_t^2 r^3} \left(\delta_{\alpha\beta} - \frac{3 r_\alpha r_\beta}{r^3}\right). \tag{39}$$

This expression recalls the dipole field well known from electrostatics. Upon substituting (39) into (33), we again obtain the Hamiltonian of the "spin-spin" interactions $\mathcal{H}_{s\text{-}s} = \sum_{ij} J^{zz}_{ij} S^i_z S^j_z$. However, for the isotropic case we have[7,34]

$$J^{zz}_{ij} = \sum_{\alpha\beta} 2\gamma^i \frac{\Delta^i}{E^i} e_{\alpha\beta} = 0.$$

We can derive an expression for $J^{zz}_{ij} \neq 0$ by employing the general solution of Eq. (38).[7] If we know the Hamiltonian of the spin-spin interactions (36), we can determine the characteristic time T_2 and also study the dynamics of pulsed excitations of amorphous systems.

6. GENERATION OF COHERENT ELECTRIC AND ACOUSTIC RESPONSES IN GLASSES

a) Fundamental concepts and methods of observation of echo signals

The detection of electric and phonon echo signals has been one of the most convincing confirmations of the existence of two-level systems arising from tunneling transitions. It has turned out that the echo in glasses has much in common with other echo phenomena, such as spin, photon, ferromagnetic echos, etc. Therefore we shall take up briefly the fundamental concepts of echo phenomena. Usually one excites echo signals with two pulses of an electromagnetic or acoustic field, or by a combination of them. The duration of the pulses must be shorter than all the relaxation times in the system being studied. The echo signals appear after a time equal to twice the interval between the exciting pulses. The very generation of such a response involves the presence in the system of an inhomogeneous width $1/T_2^*$,[39] in addition to the irreversible relaxation times T_1 and T_2. The term "inhomogeneous broadening" essentially reflects the existence in the system of interactions that give rise to a scatter in the effective resonance frequencies. Therefore only a fraction of the spins participates in the resonance excitation. Usually the inhomogeneous width $1/T_2^*$ can be treated as a superposition of a large number of Lorentzian lines, each of which has the homogeneous width $1/T_2$. Here, as a rule, we have $T_2^* < T_2 < T_1$. After the resonance excitation of the spins with a uhf pulse, a dephasing process begins, which is due to the inhomogeneous width $1/T_2^*$. This dephasing can be compensated by applying a second uhf pulse after the time τ, so that at the instant 2τ the system of spins itself generates a coherent response, which has been termed the spin echo signal.[40] For the simplest case $\hat{S} = 1/2$, the amplitude of the spin echo signal is described for the formula[40,41]

$$E(2\tau) \sim \sin(\omega_1 \tau_1) \sin^2\left(\frac{1}{2}\omega_1 \tau_2\right). \tag{40}$$

Here τ_1 and τ_2 are the durations of the first and second pulses. The amplitude of the echo is maximal when $\omega_1\tau_1 = \pi/2$ and $\omega_1\tau_2 = \pi$. When the durations of the exciting pulses are the same, the echo signal is maximal when $\omega_1\tau_1 = \omega_1\tau_2 = 2\pi/3$. With increasing interval between the pulses, the intensity of the signal declines as $\exp(-t/T_2)$.

In addition to the two-pulse methodology that we have described, there is a three-pulse excitation of the signals. In this case there is a series of responses, the most interesting of which is the so-called stimulated echo, which decays as one varies the interval between the second and third pulses as $\exp(-t/T_1)$. Owing to these features, the echo phenomenon proves to be a relatively simple and practically sole method of measuring the relaxation times T_1 and T_2.

The spin and photon echo signals described above are characteristic of a system having a discrete spectrum. Echo signals have also been found in purely classical systems: the echo in a plasma,[42] the electroacoustic and polarization echo, in piezoelectric powders[43,44] and crystals,[45] etc. In spite of such a great variety of objects studies by the echo methods, the following general requirement must be satisfied for generation in them of coherent responses: a nonlinear mechanism must exist for the excitation or formation of the echo signals. For example, the spin echo signal arises from the nonlinear interaction of the spin system with the magnetic component of the uhf pulse. This nonlinear character of the interaction gives rise to the expression (40) for the amplitude of the spin echo signals. Gould[46] has shown that the echo in a plasma is characterized by a linear interaction of the oscillator systems with the field of the uhf pulse and a decay or dispersion that is nonlinear in amplitude of the free oscillations after the action of the exciting pulses. Interestingly, in contrast to the spin echo signals, these responses first increase as one increases the interval between the exciting pulses and only later begin to decline. Up to now all the known studies of the echo in glasses have shown no such features. This is one of the arguments for adopting the Hahn mechanism of formation of the echo signals in glasses by analogy with the spin or photon echo.

b) Experimental observations of echo signals in glasses

The first report of the experimental observation of an anomalous echo in glasses at the frequency 10 GHz at a temperature of 4.2 K was published in Ref. 5 (Fig. 12). The effect was observed at zero magnetic field and declined rapidly as the latter was increased. The physical nature of the echo signals in glasses remained unelucidated at that time. A phonon-echo effect was predicted in glasses,[47] while the experiment cited above was interpreted as a photon echo involving levels linked by tunneling transitions. Golding and Graebner[6] first performed an experiment on the phonon echo at the superlow temperature of 20 mK. The echo signals were observed in fused quartz at the frequency 0.68 GHz. Two acoustic pulses of the same intensity were generated with a film transducer made of zinc oxide and were propagated in the glass specimen. Signals of the two- and three-pulse echo were observed with the times $T_2 = 24$ μs and $T_1 = 200$ μs. The mean coupling coefficient γ_e between the longitudinal acoustic deformation and the two-level systems that contribute to the phonon-echo signal can be easily established from the pulse area θ. The Bloch equations (20) imply that the nutation frequency ω_1 corresponds in our case to the quantity $Be_0/\hbar = \gamma_e/\hbar$ for a symmetric double potential

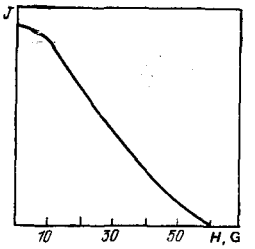

FIG. 12. Decline in intensity with increasing magnetic field.[5] Glass doped with $CeO_2 + TiO_2$.

well ($E = \Delta$). Therefore the pulse area is defined as

$$\theta = \frac{\gamma}{\hbar}\int_{-\infty}^{\infty} e\,dt = \frac{\gamma_e l_0 \tau}{\hbar}. \qquad (41)$$

This is true for pulses of rectangular shape. Upon expressing the area θ in terms of the measurable acoustic energy $E = (1/2)\rho v^2 l_0^2$, we can determine the mean parameter γ, which proves for fused quartz to be 1.6 eV. This agrees well with the results obtained in saturation experiments.[25,27] In experiments on the phonon echo, one measures not the amplitude of the signal as described by Eq. (40), but the intensity. Just as in the case of the photon echo,[48] the latter is determined by the expression

$$I = I_0 N^2 \sin^2(\theta_1)\sin^4\left(\frac{1}{2}\theta_2\right). \qquad (42)$$

Here I_0 is the emission intensity of one of the two-level systems, and N is their effective number per unit volume. In view of the extremely large inhomogeneous width, the acoustic pulse excites only those frequencies that correspond to its spectrum, with the width $\hbar\tau^{-1}$, where τ is the duration of the pulse. Hence the effective number of excited two-level systems is $N = n\hbar\tau^{-1}\tanh(\hbar\omega/2kT)$. It was of interest to verify the validity of Eq. (42) for glasses. For two equal pulses having a small pulse area ($\theta_1, \theta_2 \ll 1$), the intensity of the echo signal is proportional to

$$I \propto \frac{\theta^6}{\tau^2} \propto \tau^4. \qquad (43)$$

In the case of two identical pulses that excite the maxi-

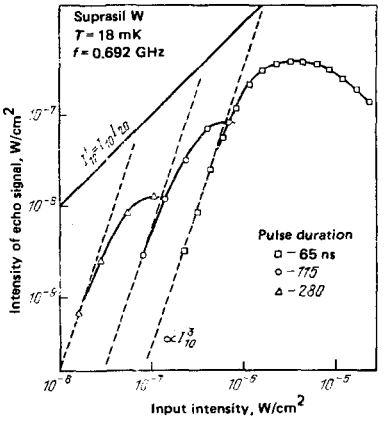

FIG. 13. Magnitude of the echo signals as a function of the intensity of the exciting acoustic pulses for three fixed values of their duration.[6]

mal response ($\theta = 2\pi/3$), the intensity of the echo signal is governed by a different dependence on the duration of the pulses:

$$I \propto \tau^{-2}. \tag{44}$$

The results of the experiments, which agree well with the theoretical relationships (42)–(44), are shown in Fig. 13. Actually we see from Fig. 13 that the maximal intensities of the echo signal vary as τ^{-2}. Thus they showed that the signal intensity is determined by the square of the number of two-level signals, as is characteristic of coherent emission.[49]

c) Model of spectral diffusion

As we have pointed out above, a spin echo signal can arise when a spread of resonance frequencies of the individual emitters exists in the system and gives rise to an inhomogeneous width. Of course, in the case of glasses the "spin-spin" interactions lead to a static shift in the splitting and a corresponding spread in the resonance frequencies. According to Eq. (36) we have

$$E^i_{\text{eff}} = E^i + \sum_{i \neq j} J^{zz}_{ij} S^j_z. \tag{45}$$

However, this shift proves extremely small in comparison with the already existing vast inhomogeneous width.

Along with the static shifts in the splittings, interesting effects arise that involve dynamic changes in E^i_{eff}.[8] A treatment of this kind of the dynamic effects based on the concept of spectral diffusion[50] was first performed in Refs. 7 and 51. In the mechanism of spectral diffusion, all the spins are provisionally divided into two types: A-spins and B-spins. The splittings of the A-spins lie in the frequency band of the uhf pulse, while the B-spins are all the rest, outside the given frequency band, Then they considerably exceed in number the A-spins, owing to the broad spectrum of the tunneling splittings. We shall assume that flipping of the B-spins arises from spin-lattice relaxation. This process leads to the dynamic shift $\Delta\omega(t)$ for the A-spins[7]:

$$\Delta\omega(t) = \frac{2\pi^2}{3}\frac{C_{\text{av}}}{\hbar}\left\langle\left|\frac{\Delta}{E}\right|\right\rangle_A \left\langle\left|\frac{\Delta}{E}\right| n_B\right\rangle_{E,\lambda}. \tag{46}$$

Here $\langle \ldots \rangle_A$ denotes averaging over the A-spins, we have $C_{\text{av}} = \sqrt{\langle J^2_{ij}\rangle}$, and $\left\langle\left|\frac{\Delta}{E}\right| n_B\right\rangle$ pertains to the B-spins, whose flipping leads to the spectral diffusion. An important feature of the mechanism of spectral diffusion for small t is the monotonic dependence on the time. This involves the fact that the flipping probability of a B-spin is proportional to t. Therefore, for small times $t \ll T_1$ we have[7]:

$$(2\pi)^{-1}\Delta\omega(t) \propto T^4 t. \tag{47}$$

Let us examine this process as applied to the formation of the two-pulse phonon echo in glasses. The most substantial contribution of spectral diffusion to the process of echo formation consists in the loss of phase coherence after the first and second pulses. For a two-pulse sequence, according to Ref. 41, the echo signal at the instant of time $t = 2\tau$ is determined as follows:

$$E(2\tau) = \left\langle \exp\left(i\int_0^{\tau} \Delta\omega(t)\,dt - i\int_{\tau}^{2\tau} \Delta\omega(t)\,dt\right)\right\rangle. \tag{48}$$

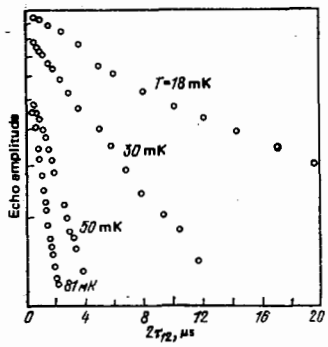

FIG. 14. Decline of the two-pulse echo for four different temperatures.[6]

Here $\langle \ldots \rangle$ denotes averaging over all the A-spins and all the flipped B-spins. In the limiting case in which $\tau \ll T_1$, Klauder and Anderson[50] have shown that

$$E(2\tau) = \exp(-m\tau^2), \tag{49}$$

here we have

$$m = \lim_{t\to 0} t^{-1}\Delta\omega(t). \tag{50}$$

If we estimate the relaxation time T_1[12] by Eq. (9), where $E = 2kT$, then it turns out that the condition $\tau \ll T_1$ is satisfied at temperatures 20–80 mK. Upon substituting the value $\Delta\omega(t, T)$ from Eqs. (47) and (50), we get

$$m \propto T^4. \tag{51}$$

Upon defining T_2' as the time of phase memory during which an echo at the instant 2τ declines by a factor of e, we find on the basis of Eqs. (49) and (51) that

$$T_2' \propto T^{-2}. \tag{52}$$

One of the fundamental features of the mechanism of spectral diffusion consists of the fact that the decay of the stimulated echo depends on the interval between the first and second pulses τ_{12}.[41] It was shown[7] that for times $\tau_{12}, \tau_{13} \ll T_1$ that we have

$$E(\tau_{12}, \tau_{13}) = \exp(-m\tau_{12}^2)\exp[-\tau_{12}m(\tau_{13}-\tau_{12})]. \tag{53}$$

In connection with the mechanism of spectral diffusion proposed in Refs. 7 and 51, experiments have been performed on the phonon echo that confirm a number of the theoretical conclusions.[6] The temperature-dependence of the decline of the two-pulse echo was studied. Figure 14 shows the results for four different temperatures. Analysis shows that the relationship $T_2' \propto T^{-2}$

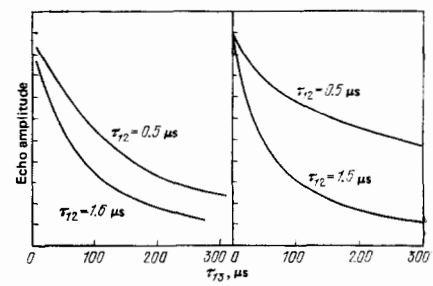

FIG. 15. Decline of the stimulated echo for two fixed intervals between the first and second pulses.[6,7]

holds, in agreement with Eq. (52). An analogous temperature-dependence has also been observed for the stimulated echo. The decline of the stimulated echo has been studied for two different intervals between the first and second pulses (Fig. 15). The right-hand side of the same diagram shows the theoretical curves as calculated by Eq. (53). Thus the results of the experiments on the two- and three-pulse echo show that the mechanism of spectral diffusion plays a substantial role in the formation of the phonon echo in glasses.

d) Other echo-type effects in glasses

In addition to the rather well studied phonon echo, there have been a number of experiments in glasses on excitation of coherent responses by the electric components of uhf pulses (Fig. 16). The observed signals[5,53-56] have a number of features that distinguish them from the well known phenomenon of the spin echo. Further studies have shown that the echo effect is manifested in a broad class of glasses of different compositions, both without special introduction of paramagnetic impurities, and with various paramagnetic dopants of rare-earth ions and ions of the iron group. A characteristic feature of all glasses containing paramagnetic dopants is the strong dependence of the intensity of the echo on the external magnetic field. It is also interesting to note the identical course of this variation for ions belonging to the same group. Figures 17 and 18 show typical curves for glasses doped with rare-earth ions and ions of the iron group. The suggestion naturally arose of whether the echo signal in glasses is a mixture of the electric and spin echos caused by inexact placement of the specimen at the maximum of the electric component of the uhf field, and hence, a partial effect on the specimen of the magnetic component. A control experiment was performed for this purpose. A plane parallel cylindrical X-cut specimen of quartz was placed in the cavity of a volume resonator, and the quartz rod was displaced until a series of reflected ultrasonic pulses of greatest amplitude had appeared. This corresponded to positioning the quartz at an antinode of the electric component of the uhf field. Thereupon the specimen under study was placed at the very same position in the resonator, which corresponded to echo signals of maximum amplitude. Of course, this arrangement of the specimen does not rule out a weakened action of the magnetic component of the uhf field, in particular, arising from distortion of the magnetic and electric

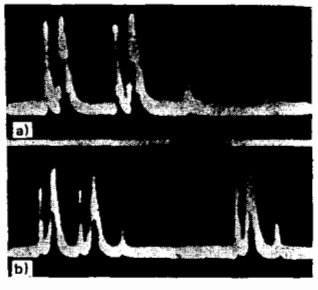

FIG. 16. a) Two-pulse echo in an undoped glass; b) two-pulse and three-pulse (stimulated) echo in a glass doped with Nd^{3+} ions.

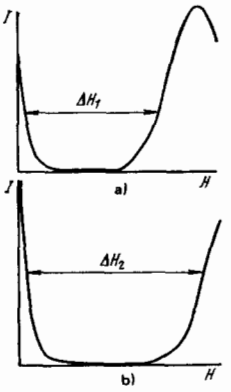

FIG. 17. a) Dependence of the intensity of the two-pulse echo signal in a glass containing Ce^{3+} on the magnetic field when the power of the exciting pulses is maximal and the interval between them minimal ($\Delta H_1 = 9.25$ kG); b) the corresponding dependence for minimal power and maximal interval ($\Delta H_2 = 12.9$ kG).[53]

fields around the specimen, which could give rise to a spin echo. Yet a number of factors contradict this: 1) All the specimens exhibit an echo signal for $H = 0$. It is improbable that the initial splittings for different ions coincide with the quantum energy at the frequency of measurement. 2) the EPR absorption lines do not coincide in field with the maxima of the echo signal, but sometimes are found at field values in which the echo signals are zero. 3) A shift and narrowing of the peak of maximum intensity of the echo signal as a function of the interval between the pulses and of the power of the uhf pulses is unusual for the spin echo. Figure 19 shows the dependence of the echo signal on the magnetic field in sodium aluminosilicate glass for three different values of the interval between the exciting pulses. We see from Fig. 19 that the width of the peak declines with increasing Δt, and its maximum shifts with respect to the field. Therefore the cited phenomenon is most likely due to tunneling transitions in the glasses, and it has been called the microwave tunnel echo.

The discovery of an electric-dipole echo in fused quartz[55,56] at superlow temperatures of 20 mK in the frequency range from 10^8 to 10^9 Hz is also of undoubted interest. As usual, the decline of two- and three-pulse echo signals was studied as a function of the interval between the applied pulses. The decline is nonexponential, just as in the phonon echo discussed above (see Fig. 13). The times for decline are proportional to Δt, both for the two-pulse echo and the stimulated echo, as

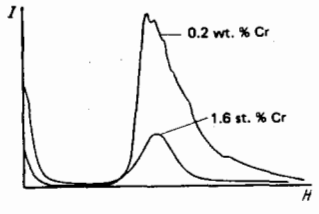

FIG. 18. Dependence of the intensity of the echo signal on the magnetic field for two specimens of lithium aluminosilicate glasses having different concentrations of Cr ions (weight %).[54]

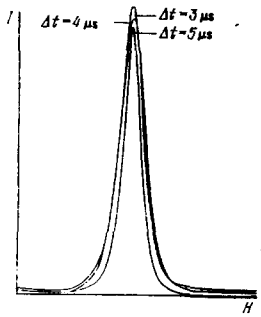

FIG. 19. Dependence of the intensity of echo signals on the magnetic field in sodium aluminosilicate glass containing Fe ions for three values of the interval Δt between the exciting pulses.

is characteristic of the mechanism of spectral diffusion. The effect of an electric field on the formation of echo signals has also been studied.[57,58] In order to do this, after the first exciting pulse, a static electric field was applied during the time Δt (Fig. 20), and the amplitude of the echo was studied as a function of the value of the electric field for a fixed interval between the two pulses. The application of the electric field accelerates the natural process of dephasing of the electric dipoles after the first pulse, and consequently the echo signal decreases.

7. "HOLE-BURNING" IN AN INHOMOGENEOUSLY BROADENED LINE

In optical spectroscopy and magnetic resonance, the effect is widely known of saturation of individual spin packets of an inhomogeneously broadened line, which has been termed "hole-burning".[39,59,60] Mathematically this stems from the nonlinear dependence of the absorption on the energy of the electromagnetic or acoustic field. Before we take up its features as applied to glasses, we shall briefly discuss its manifestations in spin resonance. Here the concept of an inhomogeneously broadened line that we have discussed above is of fundamental importance. As we know, the resonance curve of an inhomogeneously broadened line is the envelope of the ensemble of spin packets, each of which has a homogeneous width determined by the irreversible relaxation processes. Therefore, if a specimen having an inhomogeneously broadened line is placed in a saturating uhf field, the spin packets will be saturated whose intrinsic frequencies satisfy the condition

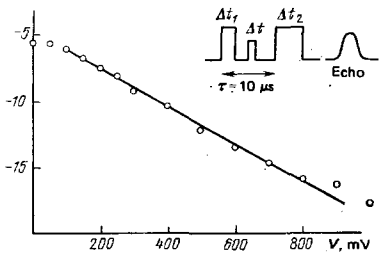

FIG. 20. Dependence of the intensity of the echo signal on the amplitude of the static electric field.[57] The static field acts between the first and second uhf pulses.

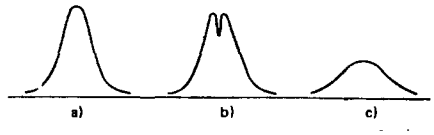

FIG. 21. a) Unsaturated line; b) "hole burning" in an inhomogeneously broadened line; c) saturation of a homogeneously broadened line.

$\nu = g\beta H/\hbar$. All the other spin packets will not be saturated, and a hole is formed in the inhomogeneously broadened line at the site of the resonating and saturated spin packets. Naturally, a homogeneously broadened line will be uniformly saturated, which will lead to diminished resonance absorption and to a broadening of the entire curve. Figure 21 shows an unsaturated line, an inhomogeneously broadened line with a "burned hole", and a saturated homogeneously broadened line.

The most substantial difference of glasses from magnetic resonance is the vast inhomogeneous width of the spectrum of two-level systems. Therefore a powerful acoustic pulse will saturate only those two-level systems that lie in the frequency band of the given pulse. Experiments of this type were first performed in Ref. 61. In view of the distinctive character of the experiment, let us take up the methodology of performing it in somewhat greater detail. A powerful saturating pulse J_1, whose carrier frequency could be varied was applied to one of the ends of a glass rod (Fig. 22) located in a broadband resonator. The other end of the rod was located in a narrow-band resonator and a pulse J_2 was applied to it with an intensity somewhat below saturation. The electric components of the pulses J_1 and J_2 excited in the glass rod acoustic pulses generated by cadmium sulfide transducers and propagating in opposite directions. The reflected acoustic pulses excited by J_2 were detected as a function of the frequency of the pulse J_1. Figure 23 shows the results of the experiment for two values of the intensities of the saturating pulse J_1. It turned out that the minimum absorption of the acoustic pulse excited in the narrow-band resonator is observed when the frequencies of the pulses J_1 and J_2 coincide. Very interestingly, the widths coincide for the resonance curves of sound absorption for two different intensities of the pulse J_1 differing by an order of magnitude. The dotted line shows the curve that would have existed for a homogeneously broadened line in agreement with Fig. 21, c.

The interesting question arises of what determines the width of the "burned hole" and how it will change shape after the saturating pulse ceases. When the pulse acts on a spin system, the width of the hole is deter-

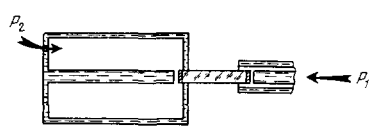

FIG. 22. Diagram of the experimental apparatus for studying the inhomogeneous width of two-level systems.[61]

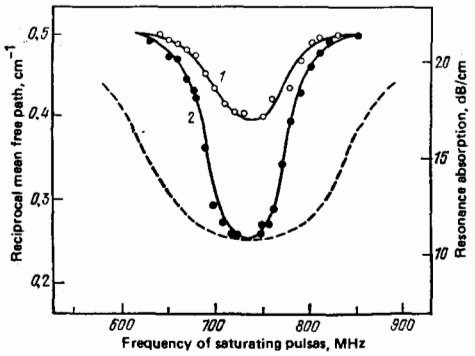

FIG. 23. Resonance absorption of the probe acoustic pulse as a function of the frequency of the pulse I_1 in borosilicate glass at 0.55 k.[61] The curve 1 corresponds to intensity $I_1 = 5\times 10^{-6}$ W/cm^2, 2—$I_1 = 5\times 10^{-7}$ W/cm^2.

mined by the lifetime of the quantum states between which the transitions occur. Theoretical and experimental studies of the echo in glasses[6,7] have shown that the mechanism of spectral diffusion contributes substantially to the width. Moreover, in a number of cases the width $\Delta\omega(t, T)$ is larger than T_1^{-1}. In line with this, we can neglect the latter. We have shown above [see Eq. (47)] that the width $\Delta\omega(t) \propto t$. In this case t is nothing other than the duration of the applied pulse. Therefore the width of the hole should depend on the duration of the saturating pulses, and this has recently been confirmed experimentally[62] (Fig. 24).

8. CONCLUSION

Recently the number of studies on dielectric glasses by physical methods has been growing rapidly with most of the results being rather clearly explained on the basis of the Anderson-Halperin-Varma-Phillips model. Nevertheless, in a number of aspects this theory offers no satisfactory explanation. For example, contradictory results exist on the problem of the dependence of the heat capacity on the duration of the experiment. We should note the difficulty of performing such experiments. The striking similarity in the acoustic properties of glasses of the most varied composition as yet remains mysterious. The question remains open of the role of OH impurities, which possess a dipole moment, and of their contribution to the electric echo in glasses.[63,64] In particular, studies[65] have appeared that advance the hypothesis of the existence of two types of centers that yield an electric-echo signal: OH impurities and two-level localized states. There are a number of studies that have tried to overcome the stated

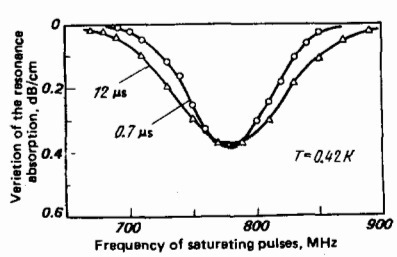

FIG. 24. Variation of the resonance absorption as a function of the frequency of saturating pulses of different durations.[62]

difficulties by partial alterations of the Anderson-Halperin-Varma-Phillips model or by constructing a microscopic theory of the low-temperature anomalies in glasses.[66-69] However, the results of these studies have not become widely disseminated up to now. For this reason we have restricted the treatment in this review to a detailed examination of the problems that have received an experimental confirmation.

Characteristically, in studying the properties of dielectric glasses we encounter the same features that arise in completely different types of amorphous compounds. Up to now a number of anomalies have been discovered in the heat capacity, heat conductivity, and the temperature-dependence of the velocity of sound and of the dielectric constant in superionic conductors, amorphous metals, and in a number of other amorphous systems.[70-77] We must note that the cited physical processes amazingly recall the behavior of dielectric glasses. Moreover, a number of hypotheses have been advanced that in a number of cases one can also apply the two-level model to these objects taking into account the specifics of the concrete systems, e.g., conduction electrons in amorphous metals. This indicates a number of universal properties inherent in disordered systems, to whose study much attention is currently being paid.

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