Direct Coupling of Magnetic Fields to Tunneling Systems in Glasses

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We report on investigations of spontaneous polarization echoes in the nonmagnetic multicomponent glass BaO-Al_2O_3-SiO_2 in static magnetic fields. While the echo decay is only marginally influenced, the echo amplitude depends strongly on magnetic fields. It seems that the intrinsic magnetic moment of tunneling systems causes dephasing effects which are detected in our echo experiments. In addition we find a strong increase of the echo amplitude with magnetic fields. This result shows that the coupling of the tunneling systems to magnetic fields is surprisingly strong and cannot be understood on the basis of current theories.

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Glasses at low temperatures exhibit properties which differ considerably from those of their crystalline counterparts [1,2]. They are caused by tunneling systems (TSs) stemming from the tunneling motion of small atomic entities in double-well potentials. The energy splitting E of these TSs is given by $E^2 = \Delta^2 + \Delta_0^2$, where Δ stands for the asymmetry energy due to the difference in the depth of the two wells and Δ_0 stands for the tunnel splitting originating from the overlap of the wave functions. Because of the randomness of the glassy structure these two quantities are widely distributed. The phenomenological tunneling model successfully describes the thermal, elastic, and dielectric properties of glasses at low temperatures [3,4]. However, experiments below 100 mK have revealed that deviations from the predictions of the tunneling model exist which cannot be explained by a modification of the distribution functions (for a recent overview, see [5]). Although a comprehensive theory is still missing, it seems that it is the interaction *between* the TSs which gives rise to these deviations [6–8].

A new and completely unexpected phenomenon was found recently: in nonmagnetic glasses, magnetic fields give rise to pronounced changes of their dielectric properties [9–11]. Before this discovery it was the general belief that glasses devoid of magnetic impurities are hardly sensitive to magnetic fields. Magnetic field effects were first observed in the multicomponent glass BaO-Al_2O_3-SiO_2 (“BAS”) which we have also used in our experiments. At 1.85 mK a field variation of only 10 μT led to clearly visible changes of the dielectric constant [9]. In addition at 5.84 mK an indication for a continuous phase transition was observed which has been interpreted as a transition from the uncorrelated tunneling motion of independent TSs to the collective motion of a large number of TSs [9]. Detailed measurements at higher temperatures and higher fields have shown that the dielectric constant varies nonmonotonically with magnetic fields [10–13].

Before reporting on our new results let us touch on the question how TSs can couple to magnetic fields. First, it is conceivable that interaction is mediated by nuclear magnetic moments. This mechanism can be ruled out because of the very long spin-lattice relaxation times of the order of 10^3 s measured in NMR studies on glasses down to 1.2 K [14]. Second, magnetic fields could interact with TSs if they were decorated by free electronic spins. Therefore, investigations were carried out on glasses with different concentrations of magnetic impurities [11]. It turned out that the magnitude of the magnetic effects is uncorrelated with the impurity concentration leading to the conclusion that magnetic impurities are not responsible for the observed effects. Third, the tunneling particles themselves could give rise to intrinsic magnetic moments and thus couple directly to magnetic fields. This is conceivable if tunneling from one potential minimum to the other does not occur along a single but along several paths. In a recent publication a hat shaped potential was considered with two potential minima in the azimuthal direction along the rim [15]. Tunneling of a charged particle along the two equivalent paths gives rise to currents in the opposite direction which cancel each other. The presence of a magnetic field breaks the symmetry thus leading to a net current and a change of the eigenvalues of the TSs. For isolated TSs noticeable changes would occur only at extremely high magnetic fields. Therefore, it has been suggested that a strong enhancement is brought about by the coupling between the TSs resulting in a large number of tunneling particles moving in a correlated manner and giving rise to effective current loops of mesoscopic scale.

Now we turn to our microwave experiments which shed new light onto the magnetic field effects. At low enough temperatures the characteristic relaxation times of the TSs become so long that the coherent motion of an ensemble of TSs becomes observable. In particular, dielectric polarization echoes can be generated which are analogous to magnetic spin echoes well known from NMR [16]. While in the later experiments the nuclear moments are driven by the rf-magnetic field, the TSs in our measurements couple to an electric field via their electrical dipole moment. In our experiments we used disk-shaped BAS samples, 0.5 mm thick and 8 mm in diameter which were placed in the uniform electric field region of a reentrant cavity with a resonance frequency $\omega/2\pi$ of about 1 GHz [17]. For cooling down to 10 mK the cavity was attached to the mixing chamber of a dilution refrigerator and magnetic fields up to $B = 230$ mT were applied. The inset in Fig. 1 shows the typical pulse sequence used to generate a spontaneous polarization echo which is also named "two-pulse echo." The signal is caused by TSs with an energy splitting $E \approx \hbar(\omega \pm \delta\omega)$, where ω is the angular frequency of the microwave pulse and $\delta\omega$ is its width given by the pulse spectrum.

From many experiments it is known that the echo decay in glasses is governed by spectral diffusion, i.e., by the interaction of the resonating TSs with nonresonant systems in the neighborhood. In Fig. 1 we show the decay of the integrated echo amplitude in BAS at $T = 10$ mK as a function of the pulse separation time t_{12} for $B = 230$ mT and for zero field. In this, as in all the other measurements reported here, the length τ of the two microwave pulses was $\tau_1 = 0.1$ μs, $\tau_2 = 0.2$ μs. In the experiment shown here an ac electric field of strength $F_0 = 500$ V/m was applied. If at all, magnetic fields have only a minute influence on the echo decay. This means that the nature of the TSs contributing to the echo is not changed by the magnetic field.

Keeping the pulse separation t_{12} constant, the echo amplitude of a uniformly excited homogeneously broadened line is given by $A = A_0 \sin\theta_1 \sin^2(\theta_2/2)$ with $A_0 \propto \Delta N(p\Delta_0/E)$. Here p stands for the dipole moment of the TSs and $\Delta N = N \tanh(E/2k_BT)$ for the difference in the population of the two levels of those TSs which are in resonance with the applied microwave. The pulse area θ is given by $\theta = (\Delta_0/E)(\tau/\hbar)\mathbf{p} \cdot \mathbf{F}_0$.

Since the ratio $\theta_1/\theta_2 = \tau_1/\tau_2 = 1{:}2$ was kept constant in all our experiments, the echo amplitude simplifies to $A = A_0 \sin^3\theta$, with $\theta \equiv \theta_1$, or $\tau \equiv \tau_1$. Because of the wide distribution of the parameters of the TSs the assumption of a homogeneously broadened line is by no means valid. Nevertheless, this relation holds approximately for $\theta < 2\pi/3$. For $\theta > \pi$ the echo splits into two components with 180° phase difference [18].

Figure 2 shows the integrated echo amplitude as a function of the applied electric field for different magnetic fields. Let us first consider the zero field result. As expected, the echo amplitude A rises rapidly with electric field amplitude F_0 or, to be precise, with the pulse area θ. A maximum occurs at $\theta = \pi/2$ allowing one to determine the dipole moment of the TSs. Within the accuracy of our data—the absolute value of electric field strength in the cavity can be only roughly estimated—the maximum is found at about 500 V/m resulting in a mean dipole moment of 9×10^{-30} Asm [19]. A look at the other curves shows that the position of the maximum is hardly influenced by the applied magnetic field meaning that the dipole moment of the TSs remains unchanged within the accuracy of our data, in agreement with expectation.

As can be seen in Fig. 2, magnetic fields led to remarkable changes of the *echo amplitude*. Depending on the magnetic field an increase or a decrease of the amplitude is observed. In addition, the magnitude of this effect also depends on the applied electric field strength. This is shown in more detail in Fig. 3 for 11 and 230 mT. The echo amplitude $A(B)$ normalized to $A(B = 0)$ at zero magnetic field is plotted as a function of the electric field amplitude F_0. Clearly, the magnetic field effect stays constant at small electric fields and increases steadily with

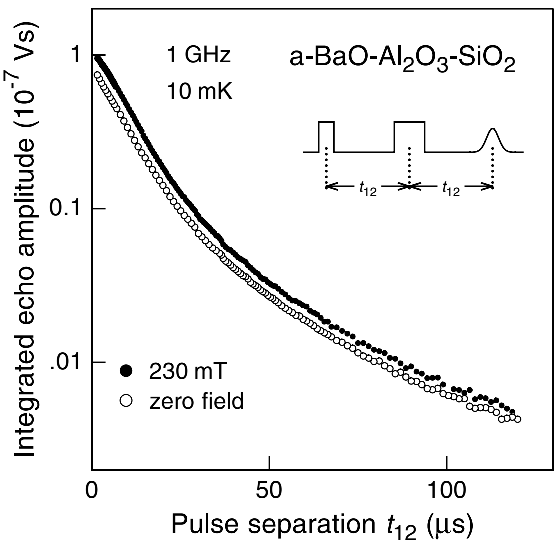

FIG. 1. Decay of the two-pulse echo amplitude at 10 mK in BAS as a function of the pulse separation time t_{12} in zero field and at $B = 230$ mT.

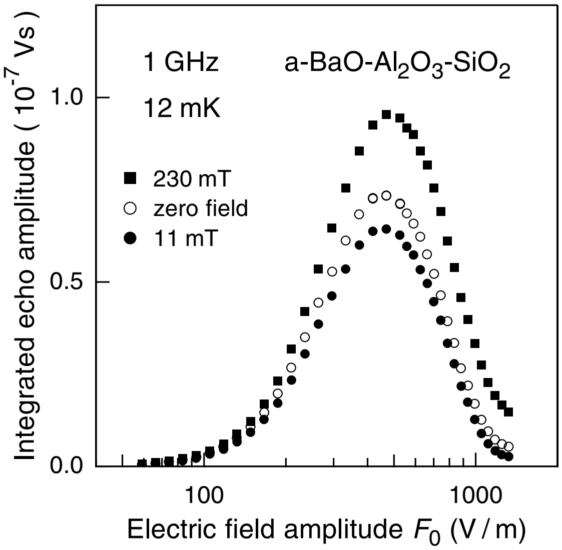

FIG. 2. Electric field dependence of the amplitude of the spontaneous polarization echo in BAS at zero field, 11 mT, and 230 mT.

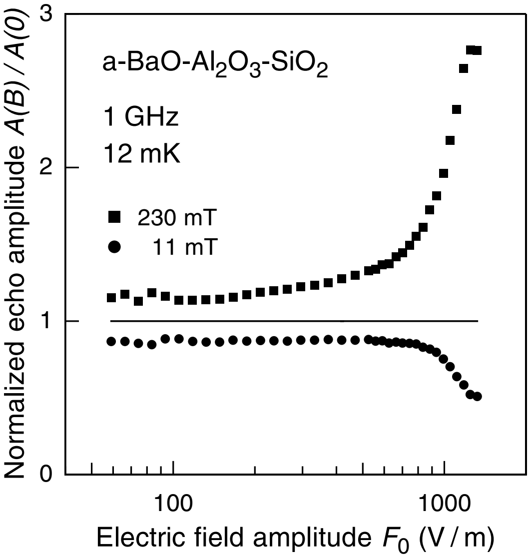

FIG. 3. Echo amplitude in BAS normalized to the zero field value versus applied electric field strength at two different magnetic fields.

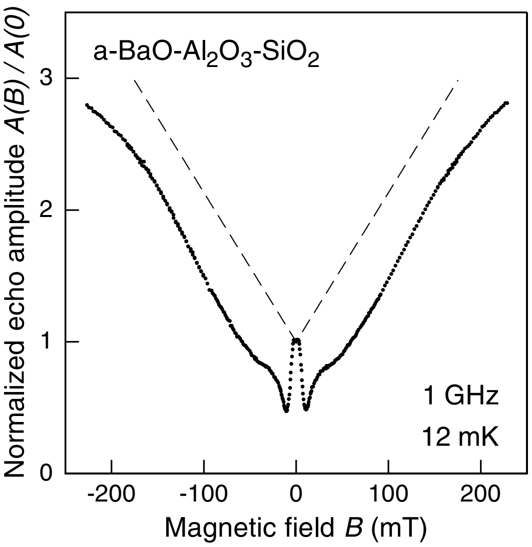

FIG. 4. Magnetic field dependence of the amplitude of the spontaneous polarization echo in BAS. The measurement was carried out at a frequency of 1 GHz and a temperature of $T = 12$ mK.

increasing field strength. At the highest fields this increase seems to level off or even shows the onset of an oscillatory behavior. Because of experimental uncertainties due to heating effects at high fields we cannot make an unambiguous statement. An electric field dependence has also been observed in low frequency dielectric measurements [11–13]. It is tempting to associate this effect with the electric flux acting on the quantum-mechanical state of the TSs thus linking magnetic and electric field effects, but no worked out explanation exists until now.

To study the magnetic effect in more detail we have investigated the echo amplitude as a function of the magnetic field keeping all other experimental parameters ($F_0 = 1.1$ kV/m, $\tau = 0.1$ μs, $t_{12} = 2$ μs, and $T = 12$ mK) constant. As shown in Fig. 4 a strong and non-monotonic variation of the echo amplitude was found: It first decreases with increasing field strength by about a factor of 2, goes through a minimum, and rises again with a slight oscillation. At the highest magnetic field of 230 mT the echo amplitude is roughly a factor of 3 higher than at zero field. From the data on the echo decay (see Fig. 1) it follows immediately that the magnetic field dependence of the amplitude cannot be due to changes of the relaxation times because this effect is in any case much too small. It is tempting to assume that the shape of the curve reflects two competing effects. One mechanism is thought to cause an initial decrease of the echo amplitude resembling a heavily damped cosine-shaped oscillation. We propose that dephasing effects give rise to these changes. The other mechanism is believed to lead to a more or less linear increase with rising field as indicated by the dashed line. We have to admit that for this surprising effect we have no explanation at all. With increasing temperature the magnetic field effects vanish rapidly and could not be observed anymore above 40 mK.

The initial decrease of the echo amplitude resembles the free induction decay in NMR which is caused by the dephasing of resonating spins after switching off the driving field. We believe that both phenomena have a common origin. In our attempt to explain the decrease of the amplitude we assume that TSs carry a magnetic moment. This is plausible if the tunneling motion takes place in a potential offering two different paths for tunneling as in the hat shaped potential mentioned above. In this case a small magnetic moment μ of the order of $\mu = e\hbar/2m$ could be related with this motion where m is the mass of the tunneling particle. A magnetic field changes the eigenfrequency $\omega = E/\hbar$ by the amount $\Delta\omega = \mu B/\hbar$ leading to a phase change $\Delta\varphi = (\mu B/\hbar)t$ in comparison to motion in zero field. The inversion of the phase of the wave function by the second microwave pulse does not necessarily leave the magnetic moment unchanged, because the TSs are in general not spherical and neither the ground nor the excited state is an eigenstate of the angular momentum operator. Therefore the "effective" magnetic moment of a TS will be different for each eigenstate. This means that the phase changes accumulated during the time t_{12} before the inverting pulse and afterwards do not cancel exactly when the echo appears. Consequently, the amplitude of the polarization echo will be reduced. In other words, the magnetic field breaks the time reversal symmetry in this experiment. If we assume that the minimum observed in our experiment corresponds to a phase change of $\Delta\varphi \approx \pi/2$, we obtain a magnetic dipole moment of $\mu \approx 4 \times 10^{-27}$ A m^2 which is much smaller than Bohr's magneton but of the order of a nuclear magnetic moment. Thus we can exclude once more the possibility that the effects observed in our experiment are caused by impurity spins. Having in mind that tunneling particles are expected to be small clusters of atoms, the value deduced above is very large. We have no explanation for this large value but believe that for a more sophisticated estimate the interaction between the TSs has to be taken into account. It should be pointed out that the distribution of the magnetic moments will probably be centered at $\mu = 0$ and the value estimated from our experiment thus reflects the width of their distribution function.

We are not able to offer even a vague explanation for the second strange effect, namely, for the increase of the echo amplitude at higher fields. The echo amplitude A_0 is proportional to the product $\Delta N p$, i.e., proportional to the number of TSs oscillating in phase with the driving field. As discussed above p is practically constant; therefore, the effective number of TSs must be increased by the magnetic field. The magnitude of this effect suggests that not only a small fraction but most of the TSs react to magnetic fields. Again we believe that this phenomenon has to do with the interaction between the TSs, but no theoretical approach exists until now.

It should be mentioned that a magnetic field dependence of the amplitude of coherent echoes has also been reported by other authors. However, in these experiments the glasses were doped typically with 1% of magnetic ions. For example, an increase of the amplitude with the magnetic field was observed in an acoustic echo experiment at 11.5 mK and 450 MHz on an aluminosilicate glass doped with 1.5% holmium [20]. In a series of experiments polarization echoes were generated in different glasses between 1.8 and 4.2 K with 5 kW microwave pulses at 10 GHz [21]. However, in this case the echoes are definitely not caused by TSs because their relaxation is too fast at such high temperatures. The authors reported a decrease of the echo amplitude at small magnetic fields, and an increase at higher magnetic fields. Although attempts have been made to explain these data it seems that not only the magnetic effect but also the nature of the echo itself is not fully understood [22].

In summary, we have observed pronounced changes of the amplitude of dielectric polarization echoes with moderate magnetic fields. The magnitude of the effect indicates that not only a small subset of TSs responds to magnetic field but more or less all TSs participate in the phenomenon. Since the echo decay itself is hardly influenced by magnetic fields, other effects must be responsible which are not yet understood. It seems that TSs carry a magnetic moment leading to dephasing effects. The surprising increase of the echo amplitude at higher magnetic fields beyond its original value at zero field can probably be understood only by taking into account the interaction between the TSs leading to a considerable modification of the dynamics of TSs.

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Note added.—Recent work by Würger [23] indicates a strong influence of magnetic fields on the properties of pairs of TSs.

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