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Magnetic-field effect on the phonon echoes in a rare-earth-doped glass

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Résumé.- Nous avons observé des échos de phonons dans un verre dopé avec de l'holmium, jusqu'à 10 mK, 60 kOe et dans le domaine de fréquences 450-800 MHz. L'amplitude des échos augmente d'un facteur 3 quand le champ magnétique croît de 0 à 60 kOe. Le champ magnétique agit aussi sur la décroissance des échos de phonons. Une expérience de désaturation est décrite. Le temps de relaxation état tunnel-réseau augmente d'un facteur 6 quand le champ magnétique croît de 0 à 60 kOe. Nous considérons la relaxation des états tunnel par les ions de terre rare.

Abstract.- We have observed phonon echoes in a holmium-doped aluminosilica glass, down to 10 mK, up to 60 kOe, in the acoustic frequency range 450-800 MHz. The echo amplitude increases by a factor 3 as the magnetic field increases from 0 to 60 kOe. The magnetic field acts also on the phonon-echo decays. A saturation recovery experiment is reported. The tunnelling-state-lattice relaxation time increases by a factor 6 as the magnetic field increases from 0 to 60 kOe. Relaxation of the tunnelling states by the rare-earth ions is considered.

Photon echoes in a rare-earth-doped glass have shown that the optical dephasing rates are enhanced in comparison to rates in crystalline materials [1,2]. This enhancement is due to the effect of the atomic tunnelling systems on the rare-earth ions [3–6]. In this letter, we report on an effect of the rare-earth ions on the relaxation rates of the tunnelling systems, also called two-level systems (TLS). Phonon echoes are a powerful tool to study the relaxation rates of TLS in glasses [7,8]. We have performed such an experiment in a holmium-doped aluminosilica glass. The holmium glasses are attractive for acoustic studies on account of the large spin-orbit coupling of the Ho ions [9]. We have observed an effect of the magnetic field on the echo amplitude and on the relaxation rates of the TLS. This new effect is different from the above-mentioned one, even though it implies the same coupling. Here, we observe directly the TLS and the rare-earth ions are perturbed by the magnetic field, whereas the photon echoes arise from optical transitions between the ground and excited levels. In other respects, it is known that magnetism acts on the coupling parameters between the TLS and the acoustic waves [10], result which has been confirmed recently in spin glasses [9,11]. Nevertheless, an external magnetic field does not change these parameters [9]. We shall see that it seems not possible to explain our effects by taking into account only the phonon-TLS coupling. So, it is tempting to consider that the magnetic moments relax the TLS in magnetic glasses as the conduction electrons do it in metallic glasses [12].

We have observed phonon echoes in an aluminosilica glass doped with holmium ions (1.5 % at.). The properties of these echoes have been studied down to 10 mK in a magnetic field up to 60 kOe, in the acoustic frequency range 450-800 MHz. Figure 1a shows the amplitude variation of the two-pulse echo at 10 mK and 450 MHz as a function of the incident acoustic intensity for 0 and 60 kOe. The pulse duration is \( \tau = 60 \) ns and the separation between the two excitation pulses is \( \tau_2 = 300 \) ns. It can be seen that the maximum of the echo amplitude happens at slightly different powers by about 2 dB. (Obviously, we have verified that the saturated acoustic attenuation in the sample does not change with magnetic field between 0 and 60 kOe). Figure 1b shows the variation of this maximum as a function of magnetic field. It increases continuously from 0 to 30 kOe and then saturates between 30 and 60 kOe.

The phase memory time \( T'_2 \) can be measured by
observing the decay of the phonon echo as a function of the time separation \( r_{12} \) between the two pulses (we use in this letter the terminology and the notations of Black and Halperin [13] and Golding and Graebner [7,8]. Figure 2a shows the variation of the echo amplitude as a function of \( 2r_{12} \) for different magnetic fields at 10 mK and 450 MHz. It can be seen that, for each magnetic field value, there are two different exponential decays with a cross-over time which varies continuously with magnetic field. The characteristic times of the two different decays do not change with magnetic field, except at 0 kOe where it seems that the initial decay is two times faster than the others. However, the time range of this decay is very small. If we choose for \( T_2 \) the characteristic time of the initial decay (which is very well defined in high magnetic field), we obtain \( T_2 = 3.4 \mu s \). This value is very short as compared with the one obtained by Graebner and Golding in fused silica [8] and which is \( T'_2 = 20 \mu s \) at 18 mK (thus, leading to the value \( T'_2 = 65 \mu s \) at 10 mK according to the \( T^{-2} \) dependence reported by these authors). We have also studied the variation of \( T'_2 \) as a function of temperature. \( T'_2 \) varies as \( T^{-1} \) in the temperature range 10-50 mK [14]. This dependence is different from the \( T^{-2} \) law reported by Graebner and Golding for phonon echoes in fused silica [8] but is the same as the temperature dependence for the electric dipolar echoes in a silica glass doped with OH\(^-\) [15].

We have observed three-pulse echoes in our sample. Figure 2b shows the echo amplitude as a function of \( r_{13} \) (the delay between the first and the third pulse) at 10 mK and 450 MHz for 0 and 60 kOe. The decay is not exponential. This behaviour is also observed in fused silica [8] and predicted by the theory [13]. If, following Black and Halperin [13], we measure \( T_1' \) (defined as the value of \( r_{13} - r_{12} \) at which the echo falls to e\(^{-1}\)) in figure 2b, we can see that \( T_1' \) is shorter at 0 kOe (15 \( \mu s \)) than at 60 kOe (40 \( \mu s \)). However, it must be pointed out that these values depend crucially on the initial value. Here again they are by one order of magnitude smaller than those reported in fused silica [8].

Measurements of the recovery time from saturation have been made. In a two-pulse saturation recovery experiment the first pulse saturates the resonant TLS and the attenuation change of the weak second pulse at a time \( r_{12} \) later, is measured as a function of \( r_{12} \) [16]. In principle, the recovery time is a direct measurement of \( T_1 \). Experimentally, there is a distribution of relaxation times and the decay is not exponential. Our results are reported in figure 3 for different magnetic field values at 10 mK and 450 MHz. It appears that the saturation recovery varies strongly with the magnetic field. If one chooses for \( T_1 \), the characteristic time of the initial decay (which corresponds to the shortest \( T_1 \)), one obtains \( T_1 = 300 \mu s \) at 60 kOe and \( T_1 = 50 \mu s \) at 0 kOe. The \( T_1 \) variation between 0 and 60 kOe is very similar to the curve in figure 1b. We have also studied the temperature dependence of \( T_1 \). We have found a roughly \( T^{-1} \) dependence between 10 and 50 mK [14]. Our \( T_1 \) value at 60 kOe and 50 mK (\( T_1 = 70 \mu s \)) is very near the one reported (\( T_1 \approx 60 \mu s \)) by Golding and Graebner.
in fused silica at the same temperature [16].

The first step, before considering the effect of the magnetic field on the TLS, is to try to explain self-consistently all the changes observed in the framework of the phenomenological TLS model [17]. The echo amplitude increase could be explained with a change of $P\gamma^2$ (where $P$ is the density of TLS and $\gamma$ the deformation potential). By another way, it is possible to measure $P\gamma^2$ through the logarithmic dependence of the sound velocity at low temperature [17]. By measuring the velocity variation of acoustic waves at 70 MHz down to 10 mK, we have observed that the slope of the logarithmic temperature dependence does not change with magnetic field between 0 and 60 kOe [14]. Hence, we have obtained $P\gamma^2 = 3 \times 10^8$ erg/cm$^3$ in our aluminosilica glass containing 1.5 % of Ho ions [14] and $P\gamma^2 = 0.7 \times 10^8$ erg/cm$^3$ in a same glass except for the Ho content which is 10 % [9]. Since the magnetic ions are coupled to the lattice, it can be understood that these parameters change due to the magnetic interactions between the ions. However, at the low temperatures where we are working, the expected effect of the magnetic field is rather static. In the framework of the random anisotropy model of the rare-earth alloys, each rare-earth ion is subject to a local anisotropy field of random orientation [19,20]. The application of a magnetic field changes the direction of the magnetic moments ("it closes the umbrella" [21]). Hence the rare earth ions in a static magnetic field induce static strains on the TLS due to the magneto-elastic coupling. An increase of Ho content between 1.5 % and 10 % leads to a decrease of $P\gamma^2$ but the effect of the magnetic field on the dynamics of the TLS is the same : $T_2$ and $T_1$ are roughly the same in all our samples of different Ho contents between 1.5 % and 10 % [14]. This is not surprising, since there are always many magnetic ions between 2 TLS of a given splitting, in our concentration range. Moreover, the sample containing 10 % Ho exhibits a spin glass transition around 400 mK [22], but this seems not to change anything on the effects reported here [14]. We have never considered the activation processes which have been observed in these glasses because their characteristic parameters are such that they must be quenched at very low temperature [9]. If it is easy to see that the magnetic field can induce static strains on the TLS, it is difficult to understand how any static strain can change the dynamics of the TLS, since there is always in glasses a large distribution of splitting with a constant density of states.

Let us consider now the possible effects of the magnetic field on the TLS. It is known that magnetism acts on the parameters of the elastic TLS. This property has been established in ferromagnetic glasses [10] and confirmed recently in insulating magnetic glasses [9,11]. Thus, $P\gamma^2 = 3 \times 10^8$ erg/cm$^3$ in our aluminosilica glass containing 1.5 % of Ho ions [14] and $P\gamma^2 = 0.7 \times 10^8$ erg/cm$^3$ in a same glass except for the Ho content which is 10 % [9]. Since the magnetic ions are coupled to the lattice, it can be understood that these parameters change due to the magnetic interactions between the ions. However, at the low temperatures where we are working, the expected effect of the magnetic field is rather static. In the framework of the random anisotropy model of the rare-earth alloys, each rare-earth ion is subject to a local anisotropy field of random orientation [19,20]. The application of a magnetic field changes the direction of the magnetic moments ("it closes the umbrella" [21]). Hence the rare earth ions in a static magnetic field induce static strains on the TLS due to the magneto-elastic coupling. An increase of Ho content between 1.5 % and 10 % leads to a decrease of $P\gamma^2$ but the effect of the magnetic field on the dynamics of the TLS is the same : $T_2$ and $T_1$ are roughly the same in all our samples of different Ho contents between 1.5 % and 10 % [14]. This is not surprising, since there are always many magnetic ions between 2 TLS of a given splitting, in our concentration range. Moreover, the sample containing 10 % Ho exhibits a spin glass transition around 400 mK [22], but this seems not to change anything on the effects reported here [14]. We have never considered the activation processes which have been observed in these glasses because their characteristic parameters are such that they must be quenched at very low temperature [9]. If it is easy to see that the magnetic field can induce static strains on the TLS, it is difficult to understand how any static strain can change the dynamics of the TLS, since there is always in glasses a large distribution of splitting with a constant density of states.

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\[ T_1^{-1} = \left( \frac{1}{C_1^0} + \frac{2}{C_1^0} \right) \frac{\gamma^2 E \Delta_0^3}{2 \pi \rho h^4} \coth \left( \frac{E}{2k_B T} \right) \] (1)
It has not been possible to explain the effects reported here by considering only the TLS-phonon coupling and the magneto elastic coupling of the rare-earth ions. So, it seems that there must be some magnetic excitations at very low temperature. Continentino has considered a coupling between TLS and magnons [23]. These excitations give rise to an additional relaxation mechanism. In our samples, the magnetic-ion concentration is low and these excitations do not exist. Nevertheless, if one suppose that some rare earth ions can flip on their anisotropy axis at very low temperature, then it is possible to explain the effect of the magnetic field on $T_1$. As the magnetic field is set up, the rare earth ions which can flip, are those perpendicular to the field (the others are quenched due to the magnetic energy). These spins can relax the TLS. As the field increases "the umbrella" progressively closes [21], thus removing the relaxing spins and increasing correlatively $T_1$. Obviously this possibility is hypothetical. Magnetization measurements at very low temperature would be very useful to know more about the rare-earth ion motions. In any case, they will be insufficient to explain conclusively our effects as long as the theory describing the dynamics of the TLS in the presence of dilute magnetic moments will be lacking. The present experiments show that there are some interesting physical effects and we hope that they will stimulate theoretical works in this field.

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