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Acoustic propagation at low temperature in an amorphous rare-earth alloy : single-ion activation process

F. Lerbet and G. Bellessa

Laboratoire de Physique des Solides (*), Bât. 510, Université de Paris-Sud, 91405 Orsay Cedex, France

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Résumé. — L'atténuation et la vitesse d'ondes acoustiques dans un verre d'aluminosilicate à l'holmium sont étudiées. Les mesures sont faites jusqu'à 30 mK, dans le domaine de fréquences 10-500 MHz et dans un champ magnétique compris entre 0 et 60 kOe. Un pic d'atténuation suivant une loi d'Arrhenius et dépendant du champ magnétique est observé. Il est expliqué à l'aide d'un processus d'activation à un seul ion. Nous proposons pour ce processus, un modèle de renversement des ions de terre rare sur leur axe d'anisotropie.

Abstract. — The attenuation and the velocity of acoustic waves in a holmium aluminosilicate glass are studied. The measurements are performed down to 30 mK, in the frequency range 10-500 MHz and in a magnetic field up to 60 kOe. An attenuation peak following an Arrhenius law and depending on the magnetic field is reported. It is explained with a single-ion activation process. We propose for this process, a model of reversing of the rare-earth ions on their anisotropy axis.

1. Introduction.

There is a lot of magnetic measurements in spin glasses but there are rather few acoustic experiments in these materials. Hawkins *et al.* [1] and Huang [2] have performed ultrasonic measurements in the spin glass CuMn. They have found a small dip in the sound velocity near the freezing temperature T_g . To our knowledge, there is no acoustic-attenuation measurement reported in the literature. An effect of the magnetic field on the thermal conductivity of spin glasses has been observed [3, 4]. The authors proposed that this effect should arise from thermal activation of magnetic clusters. In this Letter, we report an acoustic study of the amorphous holmium aluminosilicate $(\text{Ho}_2\text{O}_3)_{0.194}(\text{Al}_2\text{O}_3)_{0.227}(\text{SiO}_2)_{0.579}$. This composition corresponds to a concentration of the holmium ions of 10 % at. The low temperature magnetic study of Chappert *et al.* has shown that this insulating material is a spin glass [5]. The holmium glasses are attractive for acoustic studies on account of the large spin-orbit coupling of the Ho ions. Consequently, the vibrations of the lattice are strongly felt by the magnetic moments. This is not the case for CuMn. We report in this Letter measurements of the attenuation and the velocity of acoustic waves down to 30 mK, in the frequency range 10-500 MHz and in a magnetic field up to 60 kOe. An attenuation peak following an Arrhenius law is reported. It can be explained with a single-ion activation process

(*) Laboratoire associé au C.N.R.S.

using the random anisotropy model of the rare-earth amorphous alloys [6, 7]. Other behaviours which cannot be explained with activation processes, are reported. At very low temperature, a logarithmic temperature variation of the sound velocity is observed. This effect, which is typical of the elastic two-level systems, is very small as compared with the magnetic effects.

2. Experimental results.

We have measured the attenuation and the velocity of longitudinal acoustic waves in the holmium aluminosilicate glass $(\text{Ho}_2\text{O}_3)_{0.194}(\text{Al}_2\text{O}_3)_{0.227}(\text{SiO}_2)_{0.579}$. The measurements have been performed between 30 mK and 10 K, in the frequency range 10-500 MHz and in the magnetic-field range 0-60 kOe. The temperature dependence of the acoustic attenuation at 10 and 110 MHz for two different values of the magnetic field is shown in figure 1. There is an attenuation peak and its location temperature depends on the magnetic field. We have studied the temperature dependence of the peak location as a function of the acoustic frequency in zero magnetic field (Fig. 2). This dependence agrees with the theoretical prediction given by the basic equation for the acoustic attenuation :

$$l^{-1} = \frac{\varepsilon}{2c} \frac{\omega^2 \tau}{1 + \omega^2 \tau^2} \quad (1)$$

where c is the sound velocity, ω is the acoustic angular frequency ε is the relaxation strength and τ is the relaxation time given by the Arrhenius law :

$$\tau = \tau_0 \exp\left(\frac{V_0}{k_B T}\right) \quad (2)$$

where V_0 is an activation energy and T is the temperature. Equations (1) and (2) lead to a logarithmic frequency dependence of the inverse peak temperature. The experimental points are well fitted to this law (Fig. 2). The best fit gives for V_0/k_B and τ_0 , 5.4 K and 5×10^{-12} s respectively. The activation energy above must be considered as a mean value, since the experimental peak is larger than the theoretical one. In order to give an account of the experimental peak width, it is

Fig. 1. — Relative attenuation of longitudinal acoustic waves for two frequencies and two magnetic fields. The left vertical scale is not the same as the right one.

Fig. 2. — Location temperature of the activated peak as a function of frequency. In the inset the peak amplitude as a function of frequency.

sufficient to take an activation energy distribution between 3 and 7 K. Equation (1) leads to a peak height proportional to the acoustic frequency. It is indeed what is observed, as it appears in the inset of figure 2. From the proportionality factor, the relaxation strength can be deduced and is found to be 2.7×10^{-4} .

Until now, we have described the behaviour of the peak in zero magnetic field. However, as it appears in figure 1, the location temperature of the peak changes as a magnetic field is set up. For a fixed value between 0 and 10 kOe, the frequency dependence of the peak temperature behaves as expected from (1) and (2). From the fit of the experimental points to the theoretical law, we obtain for τ_0 the same value as in zero magnetic field and for the mean activation energy a value proportional to the magnetic field H according to (Fig. 3).

$$V = V_0 + \alpha H \tag{3}$$

where $\alpha = 0.46 \text{ K/kOe}$. Above 10 kOe and up to 30 kOe the attenuation peak is still observable but it is so broadened that its location temperature becomes meaningless.

3. Interpretation.

We have described the behaviour of an internal friction peak in an amorphous rare-earth alloy. However, it is well known that such peaks exist often in glasses [8]. The property which is not very common is the peak shifting with the magnetic field. This is an experimental evidence for the thermal activation of some magnetic objects. Hence, it is interesting to compare our results with the magnetic study on the same sample of Chappert *et al.* [5]. These authors have found a peak in the ac susceptibility as a function of temperature in the frequency range 10-200 Hz. This spin-glass behaviour is consistent with the model of thermal activation of magnetic clusters in a double-well potential [9-12]. Using this model, Chappert *et al.* have found for the mean activation energy and

Fig. 3. — Activation energy of the peak (as deduced from Eqs. (1) and (2)) as a function of magnetic field.

τ_0 , 9.8 K and 5.4×10^{-9} s respectively [5]. These results as compared with ours, show that the activation processes which appear in the two experiments are different. The so short characteristic time τ_0 that we observe (three orders of magnitude smaller than the value above) is an experimental evidence that it is rather atoms than large clusters which are thermally activated. Moreover, the fact that τ_0 does not change with the magnetic field means that it is always the same objects which are thermally activated. Then, the question is what are these magnetic objects ? In the framework of the random anisotropy model of the rare-earth alloys [6, 7], it is possible to describe simply the rare-earth ions as magnetic moments in a double-well potential. In this model Harris *et al.* [6] assume that each rare-earth spin i is subjected to a local anisotropy field of random orientation z_i and the lowest states are $|J_{z_i}\rangle = |\pm J\rangle$. Fert and Campbell [7], considering the electric quadrupole plus magnetic dipole ion, have shown there is an energy minimum for one orientation of the quadrupole. When J is large, they found that the ground state is practically a doublet $|J_{z_i}\rangle = |\pm J\rangle$. Hence, alloys with Ho^{3+} ($J=8$) are well described with this model. What we add to this description (in fact we give an explicit form to a property which is implicit in the model) is to assert that the magnetic moment at each site has to overcome an energy barrier, in order to reverse its direction on its anisotropy axis. This assumption is reasonable, since in order to reverse the magnetic moment, it is necessary to draw out of its minimum-energy orientation the electric quadrupole. Hence, we have defined at each rare-earth site a phenomenological double-well potential. Moreover, these objects will be strongly coupled to the acoustic waves due to the associated modulation of the crystal field. In figure 4a such a double-well potential is shown. The x -coordinate is an angular one. We have chosen a symmetrical potential for simplicity. We shall show later that an asymmetry independent of H does not change the results. When a magnetic field H is set up, the two states of the doublet have no longer the same energy. The corresponding double well becomes asymmetrical (Fig. 4b). The asymmetry is $2\Delta = 2g_J J\mu_B H_{z_i}$ where g_J is the Landé factor ($= 1.25$), μ_B is the Bohr magneton and H_{z_i} is the component of H on the local anisotropy axis. We point out that we are not dealing with two-level systems in the sense used for glasses. In the present case, we do not consider tunnelling between the two wells but only activation

Fig. 4. — Energy of the magnetic moment as a function of its orientation. The x-coordinate is an angular one, (a) and (b) are without and with magnetic field, respectively.

above the barrier. The relaxation time of the rare-earth moments in the double wells of figure 4 is given by :

$$\tau = \tau_0 \exp\left(\frac{V + \Delta}{k_B T}\right) \operatorname{sech}\left(\frac{\Delta}{k_B T}\right). \quad (4)$$

The last factor of (4) which is sometimes omitted cannot be neglected here. Obviously, equation (4) is identical to equation (2) in zero magnetic field. In the presence of a magnetic field H , the asymmetry $\Delta = 10 \mu_B H$ is 7 K at 10 kOe and this value is larger than T in the temperature range of the experimental peak. Hence, the factor $\operatorname{sech}(\Delta/k_B T)$ can be replaced by $\exp(-\Delta/k_B T)$. As a result the asymmetry disappears in (4). This shows that it is not possible to explain the peak displacement with the magnetic field if V is treated as field independent. However the experimental results agree with equation (4), if V is replaced by the expression given in (3) (τ_0 is field independent as proved by experiment). This last relation is indeed an experimental proof that the mean barrier height V as defined in figure 4 varies linearly with the magnetic field. If the factor α in (3) is written in the form $\alpha = K g_J J \mu_B$, then K is of the order of unity. This so simple result is not easy to explain since a theoretical model for the barrier is lacking.

4. Other experimental results.

In the same sample we have observed other behaviours which cannot be explained, at least simply, in the framework of activation processes. These behaviours are reported in figures 5 and 6. In figure 5 it appears that in addition to the activation peak which is displaced toward the high temperatures as the magnetic field is increased, there is a small attenuation peak which appears and then increases. The location of this peak is at 0.37 K and is independent on the acoustic frequency and the magnetic field. Its height is roughly proportional to $\omega^{3/2}$. The behaviour of the sound velocity is shown in figure 6. The vertical shifts of the curves with respect to each other are not arbitrary. Therefore, the velocity variation as a function of magnetic field at a given temperature can be measured in figure 6. In zero magnetic field, the velocity behaves roughly as usual when an activation process is present and the variation agrees qualitatively with the attenuation peak of figure 1. As the magnetic field is increased, the activation process and the corresponding sound velocity variation are shifted toward the higher temperatures. Simultaneously, one can see an

Fig. 5. — Relative attenuation as a function of temperature for different magnetic fields. The peak which appears as the magnetic field increases is located at a temperature independent on the acoustic frequency and the magnetic field.

Fig. 6. — Relative velocity variation of the acoustic waves as a function of temperature for different magnetic fields. The vertical shifts of the curve with respect to each other are not arbitrary. Hence, the velocity variation as a function of magnetic field at a given temperature can be measured on this figure.

increase of the velocity with increasing temperature up to 3 K (Fig. 6). Such a variation could be explained by considering a resonant interaction of the acoustic wave with a spin system distribution [13] or with a two-level system distribution [14]. These systems could arise from the tunnelling of the rare-earth ions in double-well potentials as described in figure 4. The kink around 0.3 K on the velocity variation would mean that the splitting distribution does not spread down to 0 but rather down to 0.5 K [15]. Correlatively, the attenuation peak, which is located at a temperature independent on the acoustic frequency could be also explained by the relaxation of such a distribution of two-level systems [14]. This would imply that the relaxation times τ of these systems are such that $\omega\tau \ll 1$ in all the temperature range. Here, it must be pointed out that the peak height should be proportional to ω^2 rather than to $\omega^{3/2}$. Moreover, so short τ would require unphysical

deformation potentials for the direct one-phonon process and it would be necessary to consider other effects as for example the Orbach process [16]. However, we think that there are too many unknown parameters (4 or 5 at least) and any fit to the experimental results would not be very convincing. Lastly, one must remember that the freezing temperature of our sample is around 0.4 K [5] but there is no physical argument to connect the attenuation peak to this temperature.

5. Elastic two-level systems.

Up to now, we have never considered the effect on the acoustic wave of the two-level systems which always exist in the amorphous matter [17, 18]. They give rise to a logarithmic temperature dependence of the sound velocity at low temperature [14]. This behaviour appears in zero magnetic field in figure 6. As the magnetic field is increased, there is another effect which appears and which hides the first. From the slope of the logarithmic temperature variation of the velocity, the coupling parameter nM^2 can be deduced [14]. We obtain $nM^2 = 4 \times 10^7/\text{erg. cm}^3$. This value is 5 times smaller than the one in fused silica [14]. The reduction of this parameter as magnetism is present in the sample has been already pointed out [19].

6. Conclusion.

The study of the acoustic-wave propagation in an amorphous rare-earth alloy has given many results. Some of them are not understood. However, there is an attenuation peak which is well explained with an activation process of some light magnetic objects. We have proposed for this process a model of reversing of the rare-earth ions on their anisotropy axis. It is interesting to point out that this process must lead to a remanent magnetization. We know that in a similar sample, a remanent magnetization has been already observed by Chappert *et al.* [5] but it is not the same as the one we propose. These authors have reported a time constant of about 10^2 s at 0.34 K. For the process that we propose, the sample put in a magnetic field at 0.34 K and then removed from the field, will be demagnetized with a time constant determined with equation (4). Its value is 4×10^{-5} s which is too short to be observed. However, by choosing a more suitable temperature like 0.2 K, equation (4) gives a time constant of 3 s which is easily observable. In order to avoid the cluster effects at this temperature it would be then necessary to study a more dilute sample.

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