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INTENSITY-DEPENDENT ULTRASONIC ATTENUATION IN A METALLIC GLASS AT LOW TEMPERATURES

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Résumé. — L'atténuation d'ondes ultrasonores transverses à la fréquence de 720 MHz a été étudiée dans le métal amorphe $\text{Pd}_{0.775}\text{Si}_{0.165}\text{Cu}_{0.06}$ à basse température. A 0,062 K, l'atténuation dépend de l'intensité acoustique, comportement déjà observé dans les verres isolants et expliqué par l'existence d'une large distribution de systèmes à deux niveaux. La variation en température de l'atténuation a été mesurée entre 0,062 K et 1,5 K, à faible et forte intensités acoustiques. Elle est décrite par une interaction résonnante avec les systèmes à deux niveaux, plus une contribution de relaxation de la forme $A\omega T + B\omega^2 \log \frac{T}{T_0}$.

Abstract. — The attenuation of 720 MHz transverse acoustic waves has been studied in the metallic glass $\text{Pd}_{0.775}\text{Si}_{0.165}\text{Cu}_{0.06}$ at low temperatures. At 0.062 K the attenuation is found to be intensity-dependent, a behaviour already observed in amorphous insulators and explained by the existence of a broad distribution of two-level systems. The temperature variation of the attenuation has been measured between 0.062 K and 1.5 K at low and high acoustic intensities. It is described by a resonant interaction with two-level systems plus a relaxation contribution of the form $A\omega T + B\omega^2 \log \frac{T}{T_0}$.

Introduction. — During recent years the low temperature properties of amorphous solids have been intensively studied. Particularly anomalous behaviour has been observed on the specific heat, thermal conductivity [1] and acoustic propagation [2] of the amorphous insulators. These properties have been explained in the framework of the tunnelling model by the existence of low energy excitations in the form of two-level systems (T.L.S.) [3]. Owing to the fact that these T.L.S. are believed to be characteristic of the amorphous structure, independently of the chemical composition of the material, several experiments have been performed on metallic glasses in order to check their existence in these compounds.

Anomalous thermal conductivity (roughly varying as the square of the temperature) has been measured in metallic glassy alloys (PdSiCu , NiP , ...) [4]. Moreover a logarithmic temperature dependence of the sound velocity has been observed in various amor-

phous alloys including NiP [5], PdSiCu [6, 7] and CoP [8]. The intensity-dependent acoustic attenuation is one of the stronger arguments in favour of the T.L.S. model. Up to date, it has not been detected in glassy metals [9, 7]. We present in this letter the first experimental evidence of the saturation of the acoustic attenuation in an amorphous metallic alloy. In comparison with previous experiments we used lower temperatures and transverse acoustic waves. With these conditions larger effects are expected and the critical acoustic flux, which separates the saturated and unsaturated regimes, is pushed into an accessible range.

Experimental method and results. — The sample is a $\text{Pd}_{0.775}\text{Si}_{0.165}\text{Cu}_{0.06}$ alloy obtained by a rapid quenching from the liquid phase. It is derived from the same piece as those used in previous experiments [6, 10]. It has the shape of a cylindrical rod of diameter 2 mm and 3.74 mm long. The two end-faces are polished for acoustical experiments.

Ultrasonic waves were generated and detected by

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a AC-cut resonant quartz working on a high harmonic. The quartz resonator was bound directly to the amorphous metal with Dow Corning grease. The diameter of the acoustic beam is 1 mm. The attenuation was measured by using a standard pulse-echo technique. The pulse width ($\Delta t = 0.5 \mu s$) and the repetition rate (20 Hz) were low enough to prevent any heating effects.

The sample was mounted in a copper block which was fixed on the mixing chamber of a dilution refrigerator. The thermal contacts between the various elements were obtained using a silicone grease. The temperature was measured with a chip from a Speer carbon resistance.

We made two different experiments. First we recorded the amplitude of the ultrasonic signal as a function of the incident power at the lowest temperature ($T = 62 \text{ mK}$). In figure 1, we have plotted the attenuation versus the acoustic flux. Clearly saturation effects are observed : the attenuation is low at high

FIG. 1. — Relative attenuation of a transverse ultrasonic wave propagating in glassy PdSiCu as a function of the acoustic flux in the sample. The zero on the vertical scale does not take into account the non intensity dependent part of the attenuation. At high power the absolute attenuation is $16 \pm 3 \text{ dB cm}^{-1}$. The solid line is a calculated curve with the equation :

$$\alpha_R = \alpha_R^{\text{uns}} \left(1 + \frac{\Phi}{\Phi_c} \right)^{-1/2}$$

FIG. 2. — Attenuation change of a transverse ultrasonic wave propagating in glassy PdSiCu as a function of the temperature for low (upper curve) and high (lower curve) acoustic flux. The two sets of experimental points are shifted arbitrarily. The solid lines are calculated curves (see text).

power and increases with decreasing power. This result is similar to those observed in insulating glasses [11, 12]. In the second experiment we measured the ultrasonic attenuation α as a function of the temperature T in the range 0.06 to 1.5 K for two different values of the acoustic flux. In figure 2 the $\alpha(T)$ curves are given for low (unsaturated regime) and high (saturated regime) powers. In this case also the attenuation is clearly power-dependent.

Discussion. — The existence, in amorphous metals, of a broad distribution of T.L.S. highly coupled to acoustic waves, similar to those invoked in insulating glasses, can explain our results. An off-diagonal coupling between T.L.S. and the sound wave leads to resonant transitions and the corresponding acoustic attenuation has been calculated [3, 13] to be

$$\alpha_R (\text{cm}^{-1}) = \frac{n_0 G_{x,\sigma}^2 \pi \omega}{4 \rho v_\sigma^3 \sqrt{1 + \frac{\Phi}{\Phi_{c,\sigma}}}} \times \tanh \left[\frac{\hbar \omega}{2 k T} \right] = \frac{\alpha_R^{\text{uns}}}{\sqrt{1 + \frac{\Phi}{\Phi_{c,\sigma}}}} \quad (1)$$

where n_0 is the constant density of states of the T.L.S., $G_{x,\sigma}$ the coupling energy between T.L.S. and acoustic wave of frequency $\omega/2\pi$, propagating with the velocity v_σ in the material of specific mass ρ . $\sigma = l$ or t refers to the polarization of the wave. Φ is the acoustic flux in the material. $\Phi_{c,\sigma}$ is the critical flux separating the saturated from the unsaturated regime. Its value is given in the c.w. regime by :

$$\Phi_{c,\sigma} = \frac{2 \rho v_\sigma^3 \hbar^2}{G_{x,\sigma}^2 T_1 T_2} \quad (2)$$

where T_1 and T_2 are the longitudinal and transverse relaxation times of the T.L.S. The factor $\left(1 + \frac{\Phi}{\Phi_c} \right)^{-1/2}$ describes the saturation of the attenuation.

The working frequency and temperature are $\omega/2\pi = 720 \text{ MHz}$ and $T = 62 \text{ mK}$, respectively.

We describe our experimental results by equation (1), the unsaturated attenuation α_R^{uns} and the critical Φ_c flux being adjustable parameters. The best fit is obtained for

$$\alpha_R^{\text{uns}} = 5.6 \text{ dB} \cdot \text{cm}^{-1}$$

$$\Phi_c = 7.5 \times 10^{-5} \text{ W} \cdot \text{cm}^{-2}$$

with $v_t = 1.8 \times 10^5 \text{ cm s}^{-1}$ and $\rho = 10.5 \text{ g cm}^{-3}$, from the value of α_R^{uns} we deduce

$$\frac{n_0 G_{x,t}^2}{4} = (2.0 \pm 0.2) \times 10^7 \text{ erg cm}^{-3}$$

This value agrees well with that deduced from the velocity measurements in the same compound

$$(n_0 G_{x,t}^2/4 = 1.9 \times 10^7 \text{ erg cm}^{-3}) \quad [6].$$

The attenuation changes are measured at ± 0.1 dB. As a consequence the accuracy on α_R^{uns} (and on $n_0 G_{x,t}^2$) is better than 10 %. However the absolute acoustic flux is known only within a factor 4.

In previous experiments in amorphous metals NiP and PdSiCu at low temperatures, no saturation effect has been detected in the power range investigated here [7, 9]. We believe this is due to the use of longitudinal waves. If we compare the critical flux for transverse and longitudinal waves with the help of equation (2), we find :

$$\frac{\Phi_{c,t}(\omega, T)}{\Phi_{c,l}(\omega, T)} = \frac{v_l^3 G_{x,l}^2}{v_l^3 G_{x,t}^2} = \frac{\alpha_{R,l}^{\text{uns}}(\omega, T)}{\alpha_{R,t}^{\text{uns}}(\omega, T)}.$$

With $G_{x,l}^2/G_{x,t}^2 = 1.7[6]$, v_l as above and

$$v_l = 4.6 \times 10^5 \text{ cm s}^{-1},$$

we obtain :

$$\Phi_{c,t} \simeq 0.1 \Phi_{c,l}.$$

Therefore, with longitudinal waves the saturation effects would be observable at higher acoustic power (about 10 dB) and in any case the attenuation would be $\alpha_{R,l}^{\text{uns}}(\omega, T) \simeq 0.1 \alpha_{R,t}^{\text{uns}}(\omega, T) \simeq 0.5 \text{ dB cm}^{-1}$ in our conditions.

Our prediction concerning the value of the critical flux for longitudinal waves does not agree with the results of reference [7]. Some reasons for that discrepancy may be suggested : poor accuracies for absolute acoustic flux measurements, conditions for c.w. regime not evidently fulfilled, etc...

When the temperature was varied the acoustic wave is attenuated by the resonant interaction with T.L.S. (described by equation (1)) and by a non-resonant or relaxation mechanism. In glasses the relaxation of the T.L.S. due to thermal phonons leads to an $\omega^0 T^3$ attenuation dependence [14]. In metals this contribution has not been observed and previous experimental results have been fitted with an empirical law of the form [10] :

$$\alpha_{NR} = \alpha_0(\omega) + A\omega T + B\omega^2 \log \frac{T}{T_0} \quad (3)$$

where $\alpha_0(\omega)$ is the temperature independent part probably due to electrons, impurities, and defects... T_0 is an arbitrary temperature reference, and A and B are two constants to be determined by adjusting equation (3) to the experimental points.

Here, in the case of the curve (a) of figure 2, the acoustic power is not sufficiently low so that the completely unsaturated regime is not reached. We have $\Phi/\Phi_c \simeq 0.55$ at 62 mK and $\alpha_R \simeq 0.8 \alpha_R^{\text{uns}}$. But

another problem arises when the temperature increases, because Φ_c also increases. Due to lack of information on the relaxation of T.L.S., we do not know the temperature dependence of T_1 and T_2 and consequently the temperature dependence of Φ_c . Moreover we are not sure if we are in the c.w. regime where $T_1 \ll \Delta t$; if we are not, Δt must replace T_1 in equation (2). However if we suppose Φ_c increases as T^3 (which is the expected law for insulators) we can roughly take into account the temperature variation of $\left(1 + \frac{\Phi}{\Phi_c}\right)^{-1/2}$. We fit our experimental results at low acoustic power with an attenuation given by the sum of equations (1) and (3). With $n_0 G_{x,t}^2/4 = 2 \times 10^7 \text{ erg cm}^{-3}$ obtained above, the best fit is given by :

$$A = 9.5 \times 10^{-11} \text{ cm}^{-1} \text{ K}^{-1} (\text{rad/s})^{-1}$$

$$B = 5.4 \times 10^{-20} \text{ cm}^{-1} (\text{rad/s})^{-1}.$$

For the high power curve we have the same problem. At 62 mK, $\Phi/\Phi_c \simeq 30$ and $\alpha_R \simeq 0.2 \alpha_R^{\text{uns}}$. We operate as for the low power curve and we calculate the corresponding attenuation with equations (1) and (3) with the parameters A and B just determined (Fig. 2).

These values for A and B are in agreement with an independent determination from acoustic attenuation measurements on the same sample at higher temperatures ($1 < T < 6 \text{ K}$) and various frequencies between 185 MHz and 1 080 MHz [15].

The actual relaxation mechanism is still unknown. We have already discussed and discarded various possible contributions in a previous paper [10].

From our measurement of the critical acoustic flux we can obtain some further information on the values of the relaxation times T_1 and T_2 . Equation (2) can be written :

$$\frac{n_0}{T_1 T_2} = \frac{n_0 G_{x,\sigma}^2 \Phi_{c,\sigma}}{2 \hbar^2 \rho v_\sigma^3}.$$

In the right-hand side all quantities are known or have been determined in this paper and at 720 MHz and 62 mK we obtain :

$$\frac{n_0}{T_1 T_2} \simeq 5 \times 10^{47} \text{ erg}^{-1} \text{ cm}^{-3} \text{ s}^{-2}.$$

The density of states n_0 is not known in amorphous PdSiCu. Experiments have indicated an excess specific heat in PdSiCu [16], but no definite linear term. In insulators specific heat results give a value of $n_0 \simeq 2.5 \times 10^{32} \text{ erg}^{-1} \text{ cm}^{-3}$ at 700 MHz [17]. In the superconducting disordered metal $\text{Zr}_{0.7}\text{Pd}_{0.3}$ at very low temperatures, a linear T dependence of the specific heat has been recently observed. A value

$$n_0 \simeq 2.7 \times 10^{33} \text{ erg}^{-1} \text{ cm}^{-3}$$

has been inferred [18]. With $n_0 \simeq 10^{33} \text{ erg}^{-1} \text{ cm}^{-3}$ we deduce $T_1 T_2 \simeq 2 \times 10^{-15} \text{ s}^2$. It can be noticed that

this is the actual value of the product $T_1 T_2$ only if the condition for c.w. regime ($\Delta t \gg T_1$) is fulfilled. On the other hand it is the product $\Delta t T_2$ which is measured and we have an upper limit for $T_1 T_2$. In vitreous SiO_2 pseudo-spin echo experiments provided direct measurements of T_1 and T_2 [19]. At 60 mK and 680 MHz, $T_1 T_2 \simeq 10^{-11} \text{ s}^2$. As previously remarked the relaxation times T_1 and T_2 of the T.L.S. are much shorter in amorphous metals than in glasses. It is probably the reason why, as another group [7], we have not observed pseudo-spin echoes in PdSiCu.

Conclusion. — We have given experimental evidence for intensity-dependence of the acoustic attenuation in the amorphous metal PdSiCu. Our results are a supplementary indication on the existence of a broad distribution of two-level systems in this material. Moreover the high value of the critical acoustic flux allows us to infer very short relaxation times for the T.L.S.

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Note. — After a remark by a referee, it seems to us worthwhile to clarify the meaning of the quantities n_0 and G_x^2 . We adopt the following definitions : n_0 is the total density of states of intrinsic two-level defects with a given energy splitting $2E$, as obtained from specific heat measurements on ideally pure glass sample ; G_x^2 is the mean square value of the coupling constant taken over all intrinsic defects (it would be better to write $\overline{G_x^2}$). Ultrasonic attenuation measurements provide numerical value for the product $n_0 \overline{G_x^2}$ as a whole, noted $(n_0 \overline{G_x^2})_{\text{us}}$.

Some authors [13] adopt other definitions : G_x^2 is relative to symmetric defects with energy splitting $2E$ which are the more strongly coupled to strains (it could be written $(G_x^{\text{max}})^2$) ; $\tilde{n}_0$ is the density of states of these symmetric T.L.S. One must evidently have $n_0 G_x^2 = \tilde{n}_0 (G_x^{\text{max}})^2$ and $\tilde{n}_0 < n_0$; typically $\tilde{n}_0 \sim 10^{-1}$ or $10^{-2} n_0$.

Both of these conventions may be chosen. But care must be taken when defining related quantities such as the relaxation time T_1 .

For instance, let us examine the critical acoustic flux Φ_c given by equation (2). Among all the defects,

consider first a subset of defects all with the same characteristics Δ and Δ_0 and therefore the same G_x (without average : $G_x \simeq 2b \frac{\Delta_0}{E}$). In a one-phonon process, T_1^{-1} for this subset is proportional to G_x^2 (we neglect the occurrence of two phonon polarizations) and therefore $G_x^2 T_1$ is independent of G_x . This result is valid for all the subsets of T.L.S. with energy $2E$ and Φ_c is actually independent of the off-diagonal coupling constant G_x . Then two consequences arise : first, Φ_c has a well-defined value in spite of the broad distribution of individual coupling constant values ; secondly if equation (2) is written with $(G_x^{\text{max}})^2$, we must take T_1 as the relaxation time T_1^{min} of the (short-living) symmetric defects and conversely if we put $\overline{G_x^2}$ (following our definitions), T_1 must be the mean relaxation time $\overline{T_1}$ with an average over all T.L.S. with energy splitting $2E$. $\overline{T_1}$ and T_1^{min} are related by : $n_0 T_1^{\text{min}} = \tilde{n}_0 \overline{T_1}$.

Every type of experiments give its own mean value for G_x . For instance pseudo-spin experiments in glasses with pulse widths $\Delta t_1 = \Delta t$ and $\Delta t_2 = 2 \Delta t$ give an echo amplitude [20].

$$A(\varepsilon) \propto \left| \sin \left(\frac{G_x \varepsilon \Delta t}{\hbar} \right) \right|^3$$

and the measurement of the strain which produces a maximum for the echo provides another (complicated) mean value $(\overline{G_x})_{\text{echo}}$. It is adventurous to compare these different values and probably one has $(\overline{G_x})_{\text{echo}} \leq (\overline{G_x^2})^{1/2}$ where $\overline{G_x^2}$ is obtained by dividing $(n_0 \overline{G_x^2})_{\text{us}}$ by $(n_0)_{\text{spec. heat}}$.

Evidently all these considerations fully apply to the dielectric properties of glasses, with the dipolar momentum p_x instead of G_x .

Two conclusions are drawn from the previous discussion :

1. we may take $n_0(2E)$ as the total density of states with energy splitting $2E$. In fact, n_0 is an experimentally well-defined quantity ; this is not true for $\tilde{n}_0$. As a consequence, the numerical value given in the present article concerns $\overline{T_1}$;

2. different mean values obtained from different measured quantities cannot be compared without a careful examination.

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