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Résumé. — Une explication est proposée pour les effets non-linéaires récemment observés sur l'atténuation ultrasonore de PdSiCu amorphe et quelques autres métaux amorphes. Ces effets sont attribués à la saturation de l'atténuation de relaxation lorsque le flux acoustique est suffisamment intense pour égaliser la population des systèmes à deux niveaux dont les énergies s'étendent jusqu'à kT.

Abstract. — Strong non-linear effects recently observed in the acoustic absorption in amorphous PdSiCu and other amorphous metals are explained. We propose that saturation of the relaxation absorption due to tunneling sites might occur, when the acoustic power becomes so large that the whole ensemble of two level systems (TLS) is saturated up to energy splittings of the order of kT.

Amorphous metals and insulators contain low-energy excitations which determine to a large extent their low-temperature behaviour, in particular specific heat, thermal conductivity, acoustic absorption and dispersion [1]. These low-energy excitations are ascribed to tunneling sites, also known as two level systems (TLS) inherent in the amorphous structure [2,3]. Their distinct feature is an almost constant density of states over a wide energy range, at least up to a few K . The acoustic behaviour of these materials is mainly determined by two processes due to TLS [4]. In the resonant absorption, only TLS are participating whose energy splittings E match the energy of the ultrasonic phonon within the bandwidth $\hbar\Gamma_2 = \hbar/T_2$. Here, T_2 is the transverse relaxation time of the TLS. This absorption becomes power-dependent when the acoustic flux exceeds a certain critical value ϕ_c at which the decay rate $\Gamma_1 = 1/T_1$ (T_1 is the longitudinal relaxation time) of the TLS taking part becomes smaller than the absorption rate. In contrast, in the relaxational absorption process all TLS take part which are thermally excited and hence all TLS with energy splittings E up to a few kT. The time varying acoustic strains modulates E and absorption arises because the TLS return to equilibrium out of phase with the acoustic wave [4]. Of course both absorption mechanisms also contribute to dispersion, and in fact, are the only important dispersion mechanisms in amorphous materials at low temperatures. In amorphous metals, the TLS not only couple to phonons, but also to electrons thus dominating Γ_1 [5]. This in turn leads to a drastic increase of the critical intensity ϕ_c by four orders of magnitude compared to amorphous insulators.

Recently, a number of experiments have been reported [6,7] which indicated an hitherto unknown power-dependent absorption mechanism due to TLS. It can be described by the following empirical formula :

$$\alpha' = \alpha'_0 (1 + \phi/\phi'_c)^{-\frac{1}{2}} \tag{1}$$

Here, ϕ is the acoustic flux, and ϕ'_c is the acoustic flux above which this effect

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becomes noticeable. α_0' is very weakly temperature dependent. Strinkingly, the power
dependence resembles the one for the resonant absorption :

$$\alpha_{res} = \alpha_0 (1 + \phi/\phi_c)^{-1/2} \quad ; \quad \alpha_0 = \frac{\pi \bar{P} \gamma^2 \omega}{\rho v^3} \tanh\left(\frac{\hbar\omega}{2kT}\right) \tag{2}$$

Here, $\omega/2\pi$ is the sound frequency, ρ is the density, v is the sound velocity, $\bar{P}$ is
the density of states, and γ is the deformation potential TLS - phonons.
$\phi_c = \hbar^2 \rho v^3/(2\gamma^2 T_1 T_2)$ is the critical flux for saturation of the resonant absorption
[1]. If we would try to identify the observed nonlinear absorption with a resonant
absorption process using equation (2), this would yield a far too large $\bar{P}\gamma^2$. On the
other hand, α_0' corresponds in magnitude to the relaxational absorption. Experimental-
ly, this situation has been partially clarified very recently [8] by showing that two
power-dependent absorption mechanisms do exist in a-PdSiCu. One corresponds to the
already known saturable resonant absorption and is fully described by equation (2).
Since $\alpha_0' \sim \alpha_{rel}$, and because of the observed power dependence of α', one is tempted
to devise a mechanism which could lead to saturation of the relaxation absorption
hitherto unknown. We shall show that such a mechanism can exist and, in fact,
explains the experimental findings quite well.

As already mentioned above, resonant absorption occurs over a bandwidth of the
order of Γ_2. However, with increasing acoustic power the spectrum of TLS which couple
to the oscillating acoustic strain increases. This power dependent bandwidth can be
expressed as [9] :

$$\Gamma_2' = \Gamma_2 (1 + \phi/\phi_c)^{1/2} \tag{3}$$

We might expect saturation of the relaxational absorption when an appreciable number
of TLS taking part in this process are affected by the sound wave. Since all thermal-
ly excited TLS are participating in the relaxation process, Γ_2' should then be of the
order of $kT/\hbar$. From equation (3) and with $T \sim 1$ K (at which the experiments were
indeed carried out) we estimate $\phi/\phi_c \approx 10^2$ assuming $T_2 = 2 T_1 = 10^{-10}$ sec . Further-
more, since $\phi = \frac{1}{2}\rho v^3 S^2$ an acoustic strain S of the order of 10^{-4} is obtained. Such
high strain values, however, were indeed applied in the experiments reported recently
[6,7,8], and thus encourage this approach in order to further investigate its conse-
quences.

Saturation phenomena play an important rôle in spin resonance as well as optical
resonance. There exists a wealth of theoretical approaches to describe saturation of
an ensemble of two state systems, supported by a large number of experiments. But
indeed a complete and rigorous treatment of all relevant nonlinear effects do not
exist. Therefore, we are reduced to assume a reasonable hypothesis: because of
saturation, the longitudinal susceptibility which is the fundamental quantity is
multiplied by a factor w:

$$w = \frac{\Gamma_2^2 + \omega_0^2}{\omega_0^2 + \Gamma_2^2 (1 + \phi/\phi_c)} \tag{4}$$

where the energy splitting E of the TLS is $\hbar\omega_o$. Using Eq.(4) we then obtain for
the relaxation attenuation:

$$\alpha_{rel} = \frac{2\omega C}{v} \int d\omega_0 \frac{\hbar}{4kT} \operatorname{sech}^2\left(\frac{\hbar\omega_0}{2kT}\right) \int dr \frac{\sqrt{1-r}}{r} \frac{\omega T_1}{1 + \omega^2 T_1^2} \frac{\omega_0^2 + \Gamma_2^2}{\omega_0^2 + \Gamma_2^2(1 + \phi/\phi_c)} \tag{5}$$

It should be noted that equation (5) contains the distribution of relaxation
strength according to the tunneling model, and in order to be consistent, this dis-
tribution also has to be invoked in w (equation (4)). We now calculate α_{rel} using
$T_2 = 2 T_1$, and adding the resonant absorption according to equation (2). We then
obtain the solid curve in figure 1. As can be seen, there is quite good agreement
with the experiment (open circles). Also the other main features of the experiment
can be explained, and we mention only two :

a) The weak temperature dependence of α_0' : keeping in mind that an upper limit ϕ_u
of acoustic power is applied in the experiment, we write
$\alpha' = \alpha_{rel}(T, \phi \to 0) - \alpha_{rel}(T, \phi_u)$. This procedure is an oversimplified schema for the
analysis in [6,7,8] which used an extrapolation out of the experimentally explored
domain of acoustic power. However, in spite of the crudeness of our procedure we
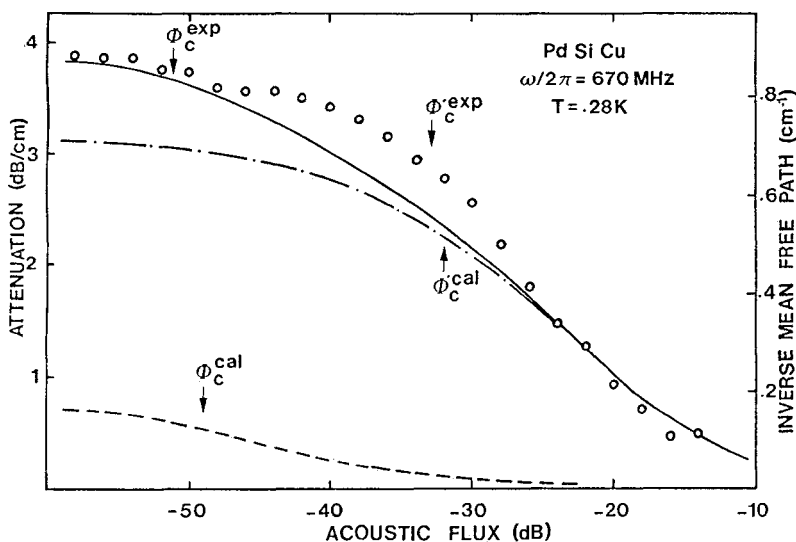
Figure 1. Power-dependent absorption in the amorphous metal PdSiCu for a frequency of 670 MHz and a temperature of 0.28 K. Theoretical results are represented by the solid line, and experimental points by the open circles [8]. The theoretical calculation is the sum of the resonant absorption (dashed line) and the power-dependent relaxation absorption (dashed-dotted line). ϕ_c^{exp} and $\phi_c^{'exp}$ are experimental values from reference [8]. ϕ_c^{cal} and $\phi_c^{'cal}$ are the values calculated as explained in the text.

achieve quite good agreement with the results given in [8].

b) The effect was not observed in fused silica at 0.3 K [6]. This follows straight-forwardly from the fact that in amorphous insulators α_{rel} has then vanished almost completely ($\alpha_{rel} \sim 0.06$ dB/cm at T=0.3 K). In contrast, in a-metal α_{rel} decreases less rapidly because it is mostly governed by electrons and not by phonons. However, also in a-metals α_{rel} should tend to zero at mK temperatures. Finally, we should like to mention that according to our model the sound velocity change caused by relaxation also becomes saturable, but we shall present corresponding calculations in a more detailed paper.

In summary, we have proposed an explanation for the non linear relaxation absorption observed in amorphous metals. Our model is based on saturation of the TLS ensemble over a spectral width of the order of kT/ℏ.

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