

New mechanism for nonlinear sound absorption in metallic glasses

Yu. M. Gal'perin, V. L. Gurevich, and D. A. Parshin

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

(Submitted 5 October 1982)

Pis'ma Zh. Eksp. Teor. Fiz. **36**, No. 11, 386–389 (5 December 1982)

An explanation is offered for the frequency and temperature dependences observed for the nonlinear sound absorption in a metallic glass in experiments reported elsewhere. With increasing sound intensity the resonant absorption gives way to a relaxation absorption. The latter becomes nonlinear when the characteristic strain-induced change in the energy of two-level systems becomes greater than kT .

PACS numbers: 62.65. + k, 43.25.Ba

The nonlinear resonant absorption of sound is one of the basic effects confirming the existence of two-level systems in disordered materials (see Ref. 1, for example). The reason for the nonlinearity is that intense sound at a frequency ω equalizes the populations of the levels in a two-level system with a level spacing $E = \hbar\omega$. In metallic glasses (PdSiCu, PdSi, PdNiP, for example), however, the typical resonant absorption²⁻⁵ is also accompanied by a behavior of the nonlinear absorption which has no analog in insulating glasses and which is not explained by the existing theories. In this paper we wish to propose an explanation for these experiments and thereby offer a theory for the new nonlinear absorption mechanism in metallic glasses; this new mechanism is neither a resonant mechanism nor a relaxation mechanism.

The resonant sound absorption coefficient is¹

$$\Gamma^{(\text{res})} = (\pi n M^2 / \rho v^2) \omega \text{th}(\hbar\omega / 2kT) (1 + J/J_c)^{-1/2}. \quad (1)$$

Here n is the state density of the two-level system, assumed constant; M is the off-diagonal matrix element of the interaction of the states with the sound; ρ is the density; v is the sound velocity; J is the sound intensity; J_c is the critical value of this intensity (more on this below); and T is the temperature.

A nonlinear absorption was observed in the experiments of Refs. 3 and 4 with the same J dependence as in (1), but the coefficient of $(1 + J/J_c)^{-1/2}$ depended linearly on ω and was almost independent of T , in contrast with (1), where the dependence at $\hbar\omega \ll kT$ is $\Gamma^{(\text{res})}(0) \propto \omega^2/T$. The frequency and temperature intervals in Ref. 2 partially overlap those in Refs. 3 and 4.

It was recently shown⁵ that the results of Ref. 2 were actually obtained at different sound intensities J than in Refs. 3 and 4. With increasing J , the first region (region 1) on the $\Gamma(J)$ curve is a nonlinear region which is described well by Eq. (1); then comes a nonlinear region (region 2), in which the absorption behaves as in Refs. 3 and 4. It turns out that the critical intensities J_{c1} and J_{c2} differ by about an order of magnitude.

We believe that at $J \gg J_{c1}$ the resonant contribution to the absorption dies out almost completely, and only the relaxation contribution ($\Gamma^{(\text{rel})}$) remains. The mechanism for the relaxation absorption is as follows: In the field of the sound wave, the

distance between the levels of the two-level system changes periodically at the frequency ω . As a result, the distribution function of the upper level, F , is not an equilibrium function, since the equilibrium is reached only after a finite time $\tau(E)$, determined by the interaction of the two-level system with electrons. The result is a dissipation of acoustic energy. All the two-level systems with $E \lesssim kT$ contribute to the absorption, since these are the systems with which the electrons interact; in the course of this interaction, the electrons switch them from the ground state to the excited state.

In the field of a sound wave the distance between levels, $E(t)$, is a periodic function of the time t , which we write $E(t) = |\Delta + d \cos \omega t|$, where Δ is the distance between the levels in the absence of a strain, $d = D/(J/\rho v^3)^{1/2}$, and D is the "diagonal" strain energy. In the expression for $E(t)$ we have ignored the tunneling matrix element, but this simplification should not affect our order-of-magnitude estimates. We assume that the frequency ω satisfies the inequality $\omega \tau_{\min}(kT) \ll 1$, in accordance with the experimental situation in Refs. 3-5. Here τ_{\min} is the minimum value of the relaxation time of the two-level system (the minimum value for the given energy argument).

At intensities J satisfying the condition $d \ll kT$, the coefficient $\Gamma^{(\text{rel})}$ does not depend on J . In the region $\omega \tau_{\min}(kT) \ll 1$, this coefficient is given by the Jäckle formula,⁶ which holds in our case, although derived for insulating glasses:

$$\Gamma^{(\text{rel})} \simeq nD^2 \omega / \rho v^3. \quad (2)$$

The coefficient $\Gamma^{(\text{rel})}$ is thus proportional to ω and independent of T . This result is found from a relaxation formula of the Mandel'shtam-Leontovich type if we assume that for a given Δ there is a broad spectrum of two-level systems with a wide variety of relaxation times τ . The absorption is dominated by the systems with $\omega \simeq 1$ and $\Delta \simeq kT$.

At high intensities, with $d \gg kT$, the two-level systems with $\Delta \lesssim d$ have an energy $E(t) \gg kT$ over a large part of their period; only for a short time interval $\Delta t \simeq kT/d\omega \ll 1/\omega$ is the distance between levels less than or on the order of kT . It is at such times that the absorption occurs; the only significant absorption is by those two-level systems for which the condition $\tau \lesssim \Delta t$ holds (the other systems do not have time to relax). These two circumstances are responsible for the nonlinear behavior of $\Gamma^{(\text{rel})}$ at $d \gg kT$.

The following expression can be derived¹⁾ for the dissipation function TS of a two-level system with a level spacing E :

$$T\dot{S} = - (kT/4) \text{ch}^{-2}(E/2kT) (\delta F \partial F / \partial t), \quad (3)$$

where δF is the deviation of the distribution function F from the equilibrium value F_0 . The function F is determined from the equation

$$\partial F / \partial t = - \delta F / \tau(E), \quad (4)$$

with $\tau(E)$ replaced by $\tau(kT)$. Under the condition $\omega \tau d / kT \ll 1$ ($\tau \ll \Delta t$) Eq. (4) can be solved by iterations, through a series expansion of δF in this small parameter. We wish to emphasize that this condition is compatible with the inequality $d \gg kT$ only if $\omega \tau(kT) \ll 1$. In this case we have

$$T\dot{S} = (4kT)^{-1} \text{ch}^{-2}(E/2kT) \tau (\partial E / \partial t)^2. \quad (5)$$

Expression (5) can also be used for estimates at $\tau \simeq \Delta t$.

Calculating the energy absorbed over a period (i.e., actually over a time $\simeq \Delta t$), and

dividing it by $2\pi/\omega$, we find the average rate of energy absorption by one two-level system. The result must be summed over all the two-level systems which contribute to the absorption (for which $\Delta \lesssim d$). Of these systems, those with $\tau \approx \Delta t$ make the greatest contribution. Because of the exponential dependence of τ on the overlap integral of the two levels (which is a random quantity with a smooth distribution function¹⁾), the number of such systems decreases only logarithmically with increasing d . Dividing the resulting absorbed-energy density by the acoustic energy flux density, we therefore find the sound absorption coefficient, at logarithmic accuracy:

$$\Gamma^{(\text{rel})}(J) \approx \Gamma^{(\text{rel})}(0)(J_{c2}/J)^{1/2} \propto J^{-1/2}. \quad (6)$$

The critical intensity²⁾ $J_{c2} \approx \rho v^3 (kT/D)^2$ is proportional to T^2 , in accordance with the experimental results of Ref. 4 (there may be a small numerical coefficient in this estimate).

Expression (6) holds up to the intensity J_m which satisfies the condition $d(J_m)\omega\tau_{\min}/kT \approx 1$. Using³⁾ $\hbar/\tau_{\min}(V_{\perp}/V_0)^2 kT$ for metallic glasses, where $1/V_0$ is the electron state density per atom at the Fermi level, and V_{\perp} is the transition matrix element (≈ 1 eV), we find

$$J_m \approx \rho v^3 (kT/D)^2 (kT/\hbar\omega)^2 (V_{\perp}/V_0)^2 \approx J_{c2} (\omega\tau_{\min})^{-2} \gg J_{c2}. \quad (7)$$

The necessary condition for the existence of region 2 is therefore the inequality³⁾ $\omega\tau_{\min} \ll 1$. In metallic glasses, this condition is satisfied by sound frequencies $f = \omega/2\pi$ below 1 GHz. In insulating glasses, τ_{\min} is three or four orders of magnitude longer at 0.1–1 K. Accordingly, nonlinear region 2 can occur there only at $f \approx 1$ MHz, at which absorption measurements are difficult.

The arguments outlined above agree with the behavior observed in the experiments of Refs. 3–5, although these arguments do not determine the numerical coefficients. The order-of-magnitude estimates are consistent with the data of Refs. 3–5, however. Finally, we note that this theory can also explain the nonlinear behavior observed for the sound velocity in Ref. 4.

¹⁾It can be shown that under the inequality $\hbar\omega \ll T, d$ the nonequilibrium density matrix of the two-level system is diagonal in the representation which diagonalizes the Hamiltonian in the given sound-wave field. We can therefore use the concept of a distribution function. A systematic derivation has been offered by one of the present authors (Yu. M. G.) and also by B. D. Laikhtman, whom we thank for furnishing his results.

²⁾It follows that the nonlinear effects should be more clearly expressed for transverse sound than for longitudinal sound, as was observed in Ref. 4.

³⁾At $\omega\tau_{\min} \gg 1$, a third nonlinear region with $\Gamma \propto J^{-1}$ can arise (region 3). Evidence for a broad spectrum of relaxation times τ at a given E comes from the fact that it is possible to observe a resonant nonlinear absorption in addition to the nonlinearity discussed in the present letter, at the same frequency but at different sound intensities. This result means that at $\omega\tau_{\min} \ll 1$ there is still a significant fraction of two-level systems with $\omega\tau > 1$ which are involved in the resonant absorption.

¹⁾S. Hunklinger and W. Arnold, in: *Physical Acoustics*, Vol. 12, New York, 1976, p. 155.

²⁾P. Doussineau, *J. Phys. Lett. (Paris)* **42**, L83 (1981).

³⁾H. Araki *et al.*, *Phys. Rev.* **B 21**, 4470 (1980).

⁴G. Park *et al.*, *Phys. Rev. B* **24**, 7389 (1981).

⁵A. Hikata *et al.*, *J. Low Temp. Phys.* **49**, 339 (1982).

⁶J. Jäckle, *Z. Phys.* **257**, 212 (1972).

⁷J. L. Black, in *Glassy Metals I*, Springer-Verlag, New York, 1981, p. 167.

Translated by Dave Parsons

Edited by S. J. Amoretty