Spectral diffusion in glasses at low temperatures. The "burned-out hole" phenomenon and nonlinear resonant microwave and sound absorption

Yu. M. Gal'perin, V. L. Gurevich, and D. A. Parshin
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR

(Submitted 22 November 1986)
Pis'ma Zh. Eksp. Teor. Fiz. **45**, No. 2, 85–88 (25 January 1987)

The basic principles of the quantitative theory of spectral diffusion in glasses at low temperatures are discussed. In this diffusion the two-level systems are responsible for the resonant absorption of sound and for the microwaves. Expressions are obtained for the shape of the "burned-out hole" and for the correction to the resonant-absorption factor, which is proportional to the intensity of sound or microwave radiation.

Sound and microwave absorption in glasses at low temperatures is known to be caused by the interaction with two-level systems. We will analyze here the nonlinear effects induced by resonance absorption. In this category we can include the dependence of the absorption factor on the intensity of the external signal and the "burned-out hole" phenomenon: If against the background of absorption of a relatively strong signal of frequency ω is observed the absorption of a weak signal of a slightly different frequency ω_1, a decrease in the absorption of the latter signal can be recorded in a certain frequency interval $\nu = \omega - \omega_1$, $|\nu| \ll \omega$ (the "burned-out hole").

Let us briefly discuss the quantitative theory of these effects in the first approximation of the intensity of the signal for those cases in which the spectral diffusion is

 0021-3640/87/020107-04$01.00

dominant.[1] This phenomenon can be summarized as follows. The external signal of frequency ω interacts resonantly with the two-level systems with a level spacing $e = \hbar\omega$. Each resonant two-level system of this sort is, however, in close proximity to a certain number of thermal two-level systems, i.e., sytems with a level spacing on the order of T. The latter interact with the resonant system in different ways, depending on whether they are in the ground state or excited state. The change in the interaction energy resulting from their transitions is on the order[3] A/r^3. Here r is the distance between the two-level systems, and A is on the order of $D^2/\rho w^2$ (D is the strain energy of the two-level systems, ρ is the glass density, and w is the average sound velocity).

The transitions ("jumps") of the thermal two-level systems cause the level spacing of the resonant system to change. As a result, the resonant system sometimes deviates from resonance and sometimes returns to it. In the presence of spectral diffusion, there are thus many more two-level systems in the resonant state than in its absence.

The equations for the diagonal part, n, of the density matrix of the resonant two-level system and for the nondiagonal part, if $\exp(i\omega t)$, are

$$\frac{\partial n}{\partial t} = -\gamma(n - n_0) - F\mathrm{Re}f, \tag{1}$$

$$\frac{\partial f}{\partial t} + i[\omega - e/\hbar - \Delta\omega(t)]f + \frac{\gamma}{2}f = \frac{F}{2}(2n-1). \tag{2}$$

Here $\hbar F/2$ is the matrix element for the transition between the levels, which characterizes the interaction of the signal with the resonant two-level system, γ is the damping of the resonant two-level system, and $n_0 = [\exp(e/T) + 1]^{-1}$ $\Delta\omega(t) = \Sigma_l J_l \xi_l(t)$ is the random variation of the natural frequency of the resonant two-level system; the summation is carried out over all thermal two-level systems. We now introduce $J_l = A/\hbar r_l^3$, where r_l is the distance from the l-th thermal two-level system to the given resonant system. The function $\xi_l(t)$ alternatively takes on the values of 1 and -1 at random times; the aveage frequency of such jumps is Γ_l. The different functions ξ_l are assumed to be uncorrelated functions.

The change ΔQ in the absorption of a weak test signal of frequency ω_1 in the presence of a strong signal, which causes a change in the equilibrium population, Δn, is

$$\Delta Q = -\pi\omega_1 F_1^2 P \int_0^\infty de \langle\langle \Delta n(t)\delta[\hbar\omega_1 - e - \hbar\Delta\omega(t)]\rangle_\xi\rangle_c . \tag{3}$$

Here F_1 is the amplitude of a weak signal, and P is a constant which characterizes the state density of a two-level system (on the order of 10^{33} erg·cm^{-3}); one pair of angle brackets denotes averaging over all realizations of the random process $\xi(t)$ and the other pair denotes averaging over the thermal two-level systems.

Solving system (1),(2) by the method of iterations over F, we find the following expression in the lowest-order approximation in F^2:

$$\Delta Q = -B\int_0^\infty d\tau' e^{-\gamma\tau'} \int_0^\infty d\tau\, e^{-\gamma\tau/2} \cos\nu\tau \langle L(\tau, \tau')\rangle_c , \tag{4}$$

where $B = \pi\omega_1 F_1^2 F^2 P[1/2 - n_0(\hbar\omega)]$, and $L(\tau,\tau')$ is the product of independent means, each of which corresponds to a two-level system of some sort and is given by

$$\varphi(\tau,\tau') = \langle \exp[iJ\tau\xi(0) - iJ\int_{\tau'}^{\tau+\tau'} \xi(t)dt]\rangle_{\xi,\,\xi(0)} \qquad (5)$$

The averaging is carried out over all realizations of the random process $\xi(t)$ with the given initial values $\xi(0)$ at an arbitrary time $t=0$ and over all $\xi(0)$. Taking an average by using a method described in Ref. 6, we find the result

$$\varphi(\tau,\tau') = \cos J\tau\, e^{-\Gamma\tau}\left[\cosh\sqrt{\tau^2 - J^2}\,\tau + \frac{\Gamma}{\sqrt{\Gamma^2 - J^2}}\sinh\sqrt{\Gamma^2 - J^2}\,\tau\right]$$

$$+ \sin J\tau\, e^{-\Gamma(\tau+2\tau')}\frac{J}{\sqrt{\Gamma^2 - J^2}}\sinh\sqrt{\Gamma^2 - J^2}\,\tau \qquad (6)$$

If we now average over all possible positions of the thermal two-level systems (see Ref. 7) and over their tunneling transmittances, on which the transition frequencies Γ depend, we find

$$\langle L(\tau,\tau')\rangle_c = \exp\left[-\frac{V(\tau,\tau')}{\tau_d}\right] \qquad (7)$$

where

$$V(\tau,\tau') = \int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}\int_0^{\infty}\frac{dJ}{J^2}[1 - \varphi(\tau,\tau')] \qquad (8)$$

Here Γ_0 is a typical frequency of the transitions of the thermal two-level systems due to the interaction with the phonons. In order of magnitude this transition frequency is $\Gamma_0 \simeq D^2T^3/\rho\hbar^4 w^5$, and $\hbar/\tau_d = A/\bar{r}^3 = APT$ is the scale energy of the interaction of thermal two-level systems at an average distance between them, given by $\bar{r} = (PT)^{-1/3}$.

For small values of $\tilde{F}$ the nonlinear absorption coefficient α can be written in the form $\alpha = \alpha_0(1 - F^2/F_c^2)$, where α_0 is the linear value. The critical amplitude of the signal F_c can be calculated by using the same method.

We present below the results of a calculation for the cases in which the spectral diffusion is appreciable. These results depend on the relationship between the temperature T and the characteristic temperature T_D, which is determined from the condition $\Gamma_0 = 1/\tau_d$ and which in order of magnitude is $(P\hbar^3 w^3)^{1/2} \simeq 1$ K.

1) At $T \gg T_D$ and $\gamma \ll 1/\tau_d$ we have

$$F_c^2 = \frac{\gamma}{\tau_d}L; \qquad L = \frac{\pi}{2}\ln(1/\gamma\tau_d). \qquad (9)$$

A large logarithmic factor sets this expression apart from the estimate in Ref. 3. The burned-out hole has a Lorentzian shape

$$\Delta Q = -\frac{B}{\gamma}\frac{L/\tau_d}{\nu^2 + (L/\tau_d)^2} \tag{10}$$

2) At $T \ll T_D$ and $\Gamma_0 \ll \gamma \ll \sqrt{\Gamma_0/\tau_d}$

$$F_c^2 = \frac{2\pi\Gamma_0}{\tau_d \ln(\Gamma_0/\gamma^2\tau_d)} \tag{11}$$

and the burned-out hole has essentially Lorentzian shape:

$$\Delta Q = -B\int_0^\infty d\tau \cos\nu\tau \frac{\exp(-\pi\Gamma_0\tau^2/2\tau_d)}{\gamma + (\pi\Gamma_0\tau/\tau_d)} \tag{12}$$

3) Finally, at $T \ll T_D$ and $\gamma \ll \Gamma_0 \ll \sqrt{\Gamma_0/\tau_d}$ we have

$$F_c^2 = \frac{\gamma}{\tau_d}L_1 \;; \qquad L_1 = \frac{\pi}{2}\ln\frac{\Gamma_0}{\gamma} \tag{13}$$

and the burned-out hole, which again has a Lorentzian shape, is given by Eq. (10), where L is replaced by L_1.

Although expressions (9) and (13) are the same, within the logarithmic factors, as the estimate of Ref. 3, their physics is entirely different. This is evident from the fact that these expressions correspond to various (large) logarithmic factors and various shapes of the burned-out hole. Estimate (11) was obtained, within a large logarithmic factor, in Ref. 5, where a nonstationary case was analyzed. Here, on the other hand, we are considering a stationary case. This means that the signal pulse length is assumed to be greater than γ^{-1} in cases 1) and 3) and greater than Γ_0^{-1} in case 2). Furthermore, it is essential for us to assume that the phonon distribution function is in the equilibrium state.

It would be of considerable interest to study experimentally the nonlinear resonant absorption and the shape of the burned-out hole in different cases. A detailed comparison with the results of the theory presented above would make it possible to understand the interaction of the two-level systems in glasses.

[1)]The concept of spectral diffusion was introduced by Klauder and Anderson[1] in the magnetic resonance theory. The importance of spectral diffusion in low-temperature physics of glasses was indicated in Refs. 2–5.

[1]J. R. Klauder and P. W. Anderson, Phys. Rev. **125**, 912 (1962).
[2]J. Joffrin *et al.*, J. Phys. (Paris) **36**, 811 (1975).
[3]S. Hunklinger and W. Arnold, in Physical Acoustics, ed. by W. P. Mason and R. N. Thurston, Academic Press, New York, 1976, Vol. 12, p. 155.
[4]J. L. Black and B. I. Halperin, Phys. Rev. B **16**, 2879 (1977).
[5]B. D. Laikhtman, Phys. Rev. B **33**, 2781 (1986).
[6]V. I. Klyatskin, Stokhasticheskie uravneniya i volny v sluchaĭno-neodnorodhykh sredakh (Stochastic Equations and Waves in Randomly Inhomogeneous Media), Nauka, Moscow, 1980,
[7]S. Chandrasekhar, Rev. Mod. Phys. **13**, 1 (1943).

Translated by S. J. Amoretty