Frequency and Temperature Dependence of the Sound Velocity in Amorphous Materials at Low Temperatures

G. Bellessa
Laboratoire de Physique des Solides, associé au Centre National de la Recherche Scientifique, Université Paris-Sud, 91405 Orsay, France

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Measurements of the sound velocity in vitreous silica and in amorphous metallic Pd-Si between 0.4 and 20 K and in the frequency range 10–450 MHz are reported. Above 4 K, the sound velocity varies linearly versus temperature and logarithmically versus frequency. From the analysis of the experimental results it is shown that the linear temperature dependence of the sound velocity cannot be explained with relaxation processes.

Sound velocity in amorphous materials has been extensively studied at low temperatures. It has been shown that below a certain temperature (around 2 K) the sound velocity decreases logarithmically as the temperature decreases.[1,2] This law results from a resonant interaction between the acoustic wave and some low-energy excitations. Above 2 K the velocity decreases with increasing temperature. This variation is usually explained by taking into account relaxation processes.[1] Between 4 and 20 K, a linear temperature dependence of the velocity has been reported in amorphous metallic Ni-P[2] and in amorphous selenium.[3] In vitreous silica such a law seems also to have been observed.[4] In the present Letter, I report measurements of the sound velocity versus temperature and frequency in vitreous soda silica and in amorphous metallic Pd-Si. The temperature dependence is indeed linear between 4 and 20 K. Thus, this law seems to be universal in amorphous materials. It is very different from the usual T^4 law of the sound velocity in crystals.[5] Furthermore, I report a velocity variation versus frequency which obeys a logarithmic law. These properties cannot be explained with the existing theory of the ultrasonic dispersion in glasses.[6,7] From the analysis of the frequency and temperature measurements, I deduce that the linear temperature dependence of the sound velocity cannot be explained with relaxation processes.

The sound velocity has been measured between 0.4 and 20 K and, in the frequency range 10–450 MHz. The velocity measurements are made with a phase comparison method. Hence, only a relative variation of the sound velocity can be measured precisely. The temperature dependence of the sound velocity for different frequencies is reported in Fig. 1 for vitreous soda silica.[8] It appears that the velocity variation in this temperature range is much stronger than in crystals[5] and that above 4 K the velocity depends linearly on the temperature within a very good accuracy. In Pd-Si the same law is observed in the same temperature range.[8] The corresponding measurements are not plotted on a $V(T)$ diagram in order to limit the number of figures. Nevertheless, they appear implicitly in Fig. 2. In Fig. 1, I have taken for the velocity reference the value at 0.4 K. In order to compare the sound velocity values for different frequencies, I suppose that at 0.4 K the velocity is not frequency dependent. This hypothesis is important and I shall prove it in the next paragraph. It appears in Fig. 1 that above 4 K the velocity is frequency dependent and that at a fixed temperature, the velocity increas-

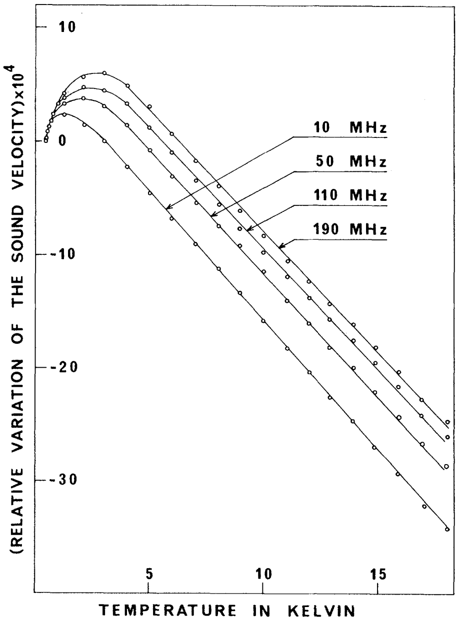

FIG. 1. Variation of the velocity of longitudinal sound waves as a function of temperature for different frequencies in vitreous soda silica. The velocity variation is relative to the value at 0.4 K. The longitudinal wave velocity is 5.8×10^5 cm/sec.

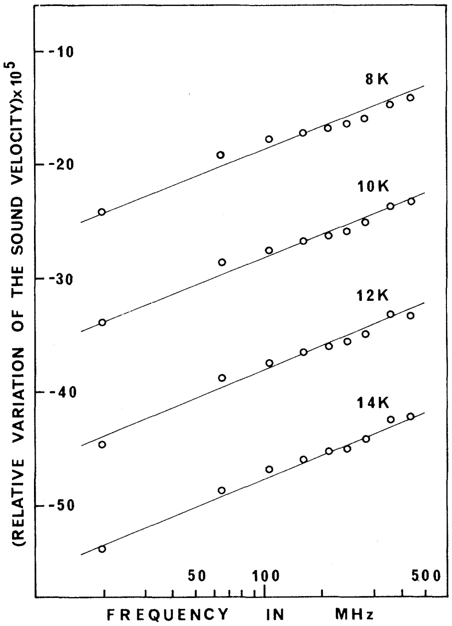

FIG. 2. Variation of the velocity of transverse sound waves as a function of frequency for different temperatures in amorphous metallic Pd-Si. The velocity variation is relative to the value at 0.4 K. The transverse wave velocity is 1.8×10^5 cm/sec.

es with increasing frequency. Measurements of these variations are reported in Figs. 2 and 3 for soda silica and Pd-Si, respectively. For the two samples, the velocity variation versus frequency obeys a logarithmic law. An important property which appears in Figs. 2 and 3 is that the straight lines fitting the experimental points are nearly parallel. There is indeed a slight departure from the parallelism but it can be neglected in a first analysis. The slopes of the straight lines are 2.8×10^{-4} and 3.5×10^{-5} for soda silica and Pd-Si, respectively. These values are almost the same as those obtained for the logarithmic temperature variation of the sound velocity below 2 K and which are 3.6×10^{-4} and 5.9×10^{-5} for soda silica and Pd-Si, respectively. The latter have been

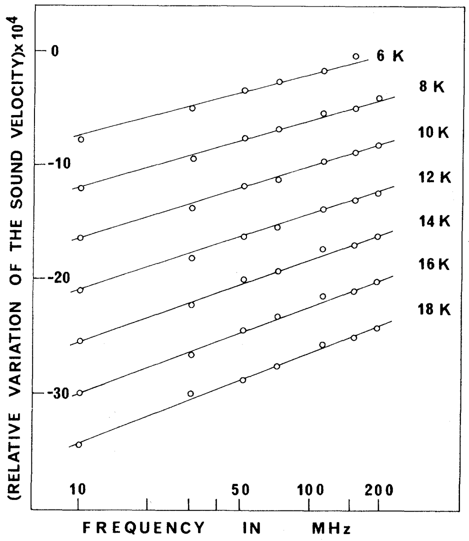

FIG. 3. Variation of the velocity of longitudinal sound waves as a function of frequency for different temperatures in vitreous soda silica. The velocity variation is relative to the value at 0.4 K.

obtained from velocity measurements between 0.4 and 2 K (Fig. 1) and are in agreement with the results already published on vitreous silica[1] and on Pd-Si.[9]

In order to justify the hypothesis made in the preceding paragraph, I undertake now to show that the sound velocity does not depend on frequency around 0.4 K. It is well known that there are in amorphous materials two-level systems with a broad distribution of their energy splitting.[10, 11] At low temperatures, there is a resonant interaction between these two-level systems and the sound wave. This interaction leads to an increase of the sound velocity with increasing temperature according to a logarithmic law.[1] It has been already pointed out that this law does not depend on frequency.[1] This does not mean that the velocity is itself frequency independent. To prove this point, it is necessary to return to the general formula giving the sound velocity variation due to the resonant interaction[1]

$$\frac{V(\omega)-V(\omega_0)}{V}=\frac{n_0M^2}{\rho V^2}\,\mathrm{P}\int_0^{\omega_M}d\omega'\omega'\left(\frac{1}{\omega^2-\omega'^2}-\frac{1}{\omega_0^{\,2}-\omega'^2}\right)\tanh\left(\frac{\hbar\omega'}{2k_BT}\right), \tag{1}$$

where ω and ω_0 are two ultrasonic frequencies, ω_M is a cutoff frequency, M is the coupling constant between the two-level systems and the ultrasonic wave, and n_0 is the two-level system density of states which is assumed constant up to ω_M. The integral of Eq. (1) can be performed on the interval $[0,\infty)$ analytically[12] so that

$$\frac{V(\omega)-V(\omega_0)}{V}=\frac{n_0M^2}{\rho V^2}\mathrm{Re}\left[\psi\left(\frac{1}{2}+\frac{\hbar\omega}{2i\pi k_BT}\right)-\psi\left(\frac{1}{2}+\frac{\hbar\omega_0}{2i\pi k_BT}\right)\right], \tag{2}$$

where ψ is the logarithmic derivative of the γ function. At low temperatures ($\hbar\omega \gg k_B T$), $\operatorname{Re}\psi(z)$ can be approximated with $\ln|z|$. Then Eq. (2) becomes

$$\frac{\Delta V}{V}=\frac{n_0 M^2}{\rho V^2}\ln\frac{\omega}{\omega_0}. \tag{3}$$

Hence, the slope is the same as that of the logarithmic temperature variation.[1] At high temperature ($\hbar\omega \ll k_B T$), Eq. (2) can be written

$$\frac{\Delta V}{V}\simeq\frac{n_0 M^2}{\rho V^2}\psi''\left(\frac{1}{2}\right)\left[\left(\frac{\hbar\omega}{2\pi k_B T}\right)^2-\left(\frac{\hbar\omega_0}{2\pi k_B T}\right)^2\right]. \tag{4}$$

This variation is very small (as compared with the velocity variations measured in the preceding paragraph) and can be neglected. These results are consistent with those of Golding, Graebner, and Kane who have evaluated numerically the variation of the velocity versus temperature.[13] Equation (4) means that there is no frequency dependence of the sound velocity in the temperature range where the logarithmic temperature law holds ($\hbar\omega \ll k_B T$). I have experimentally verified that around 0.4 K the sound velocity increases logarithmically with increasing temperature for all the frequencies used. Hence the hypothesis made in the preceding paragraph is justified. However it must be mentioned that a deviation from the logarithmic temperature dependence has been reported elsewhere.[13] Nevertheless, this deviation is very small and appears at higher frequencies than those considered here.

My results show a frequency and temperature dependence of the sound velocity above 4 K. So, it is natural to attempt to explain it with the existing theory of structural relaxation.[6,7] In this theory, a complete description of the effect of the relaxation of two-state structural defects on the elastic properties of glasses at low temperatures is given. The sound velocity variation due to this relaxation is given by[7]

$$\frac{\Delta V}{V}=-\int_0^{\hbar\omega_M} dE\, 2\,\frac{n_0 D^2}{\rho V^2}\frac{1}{k_B T}\coth^{-2}\left(\frac{E}{2k_B T}\right)\frac{1}{1+\omega^2\tau^2(E)}, \tag{5}$$

where D is the deformation potential of the two-level systems and τ is a relaxation time. This formula leads in the limit $\omega\tau \gg 1$ to a sound velocity variation which is strongly temperature dependent (proportional to T^6) and inversely proportional to the square of the frequency.[14] These laws are not experimentally observed between 4 and 20 K. This does not mean that the relaxation of the two-level systems does not exist. It has been observed in sound attenuation experiment below 2 K.[7] However, above 4 K it can be supposed that the relaxation has reached its maximum (in the limit $\omega\tau \ll 1$). Hence it would no longer change the sound velocity.

There is in glasses another structural relaxation which is a thermally activated relaxation.[15] In this process there is no longer tunneling through a potential barrier but only thermal activation. In order to attempt to explain our results with this relaxation, it is necessary to know the distribution of relaxation times. Anderson has put forward a very simple hypothesis on this distribution in amorphous materials.[16] He considers a relaxation time

$$\tau_i=\tau_0\exp(V_i/k_B T), \tag{6}$$

with a distribution of activation energies $\rho(V_i)$ ~const. Using Eq. (6) it is then easy to solve the general equation giving the sound velocity variation due to relaxation processes[17]:

$$\frac{V(\omega)-V(\infty)}{V}=A\int_0^\infty\frac{\rho(\tau)d\tau}{1+\omega^2\tau^2}. \tag{7}$$

Eqs. (6) and (7) give immediately

$$\frac{\Delta V(\omega,T)}{V}=AT\ln(\omega\tau_{\min}), \tag{8}$$

where $\tau_{\min}$ is a normalization constant ($\omega\tau_{\min} < 1$). Equation (8) gives the frequency and temperature dependence consistent with our experimental results. Unfortunately, according to Eq. (8), the slopes of the curves in Figs. 2 and 3 should be proportional to the temperature and this is not the case. So the thermally activated relaxation model fails also.

The preceding analysis shows that our experimental results cannot be explained with a relaxation theory. My experimental law can be put with a good approximation in the form

$$\Delta V/V=AT+B\ln\omega, \tag{9}$$

where A and B are two constants independent of frequency and temperature, respectively. Thus, it is reasonable to assume that the two terms of Eq. (9) arise from different physical processes. As it has been already pointed out, the constant B is almost equal to the factor $n_0M^2/\rho V^2$ of Eq. (3) for soda silica and Pd-Si. So the second term of Eq. (9) may result from the resonant interaction between the ultrasonic wave and the two-level systems. The last point but not the least is that the first term of Eq. (9) does not seem to be a relaxation term. It gives a large variation of the sound velocity (in comparison with the variation of the sound velocity in crystals in the same temperature range), and may be due to the strong anharmonicity of the disordered lattice.

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Stimulated Phonon Emission

W. E. Bron and W. Grill
Indiana University, Bloomington, Indiana 47401

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Inversion, through direct absorption of far-infrared laser light, of a three-level electronic system of V^{4+} ions in dilute solution in Al_2O_3 leads to stimulated emission of 24.7-cm^{-1} phonons. Stimulated phonon emission manifests itself in the presence of ballistic flow of longitudinal phonons along the c axis of Al_2O_3, but this flow is not observed under spontaneous phonon emission.

We report direct observation of the stimulated emission of 24.7-cm^{-1} (0.74 THz) phonons. Stimulated phonon emission is achieved by inversion of a three-level electronic system of V^{4+} ions in Al_2O_3 and is detected through an analysis of the time-of-flight spectrum of the phonon propagation.

The lowest-lying three electronic states of the $3d^1$ configuration of Al_2O_3:V^{4+} have been known for some time,[1-3] as have the group-theoretic selection rules for electric-dipole transitions among these states.[4] As indicated in Fig. 1, the ground state (numbered 1 in the figure) transforms as the $E_{3/2}$ ($A_{3/2}$ in McClure's notation[4]) representation of the C_3 double group; the first excited state (numbered 2) appears at 28.1 cm^{-1} (0.84 THz) above the ground state with a half-width of ~2 cm^{-1} at liquid helium temperature and is known to transform as $E_{1/2}$, and the next excited state (numbered 3), which appears at 52.8 cm^{-1} (1.58 THz) above the ground state with half-width of ~3.5 cm^{-1}, also transforms as $E_{1/2}$. Electric-dipole absorptive transitions from the ground state