Ultrasonic attenuation and velocity studies of amorphous PdSiCu at low temperatures

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Measurements of ultrasonic attenuation and velocity changes were carried out on the metallic glass $Pd_{0.775}Si_{0.165}Cu_{0.06}$ as a function of amplitude, in the frequency range 10 to 90 MHz, for $0.3 \leq T \leq 10$ K. A comparison of the experimental results with the predictions of the current two-level-system (TLS) tunneling theory is presented. The amplitude-dependent attentuation changes observed in these studies are larger by a factor of 100 to 1000 than the values obtained from current TLS theory, with the use of parameters determined experimentally at higher frequencies by other workers. Below ~1 K these attenuation changes have a linear (rather than quadratic) dependence on frequency and very weak temperature dependence (rather than $1/T$). The saturated (amplitude-independent) attenuation was found to have a very weak temperature dependence, rather than T^3 dependence. A significant amplitude dependence of the slope of the sound velocity change as a function of $\ln T$ was also observed.

I. INTRODUCTION

The success of the two-level-system (TLS) tunneling theory[1-3] in explaining the thermal and ultrasonic properties of dielectric glasses[4] initiated numerous further investigations on metallic glasses. Similarly to their dielectric counterparts, at low-temperatures metallic glasses were found to have linear temperature dependence of the specific heat and quadratic temperature dependence of thermal conductivity.[5,6] Analysis of these results by applying the TLS theory gave, for both the density of states and the deformation potential of TLS, similar values to those for dielectric glasses, thereby suggesting that in this regard no significant difference exists in very different classes of disordered solids. Some other indications of the validity of the theory in metallic glasses were also found in ultrasonic properties; namely, the $\ln T$ dependence of the sound velocity change at low temperatures[7-9] and the saturation of the attenuation.[10,11] However, there have been reported[12] several discrepancies with the predictions of the theory and some of these remain unsettled.

New results on ultrasonic properties of a metallic glass PdSiCu are presented in this paper. A prominent saturation in attenuation was found in frequency and temperature regions where no measurable effect is expected from current TLS theory. Besides the magnitude, the temperature and frequency dependences of this saturable part of the attenuation were found to be quite different from the predictions. However, some characteristic aspects of the TLS model remain.[13]

In Sec. II we briefly review the pertinent aspects of the TLS theory, Sec. III covers the experimental procedure; we present and discuss our results in Sec. IV and Sec. V is devoted to summary and conclusions.

II. TLS THEORY

In this section, the main features of the TLS model pertaining to ultrasonic propagation are briefly reviewed.[4] This model assumes the existence of two level systems of unknown origin. The state of a TLS is conventionally described by a pseudospin of $S = \frac{1}{2}$ which interacts with the strain fields due to phonons. Phenomenologically we introduce a longitudinal and a transverse relaxation time of the pseudospin, τ_1 and τ_2, respectively. Then the equations of motion of the pseudospin become similar to those of the Bloch equations for NMR. The calculated attenuation coefficient α consists of two parts, namely, resonance attenuation and relaxation attenuation, which are, respectively, amplitude dependent and independent.

$$\alpha = \alpha_A + \alpha_I \ . \tag{1}$$

The amplitude dependence of α_A is described as

$$\alpha_A = \frac{\alpha_{A0}}{(1 + J/J_c)^{1/2}} \ , \tag{2}$$

where J is the acoustic intensity and J_c is the critical intensity

$$J_c = \frac{\hbar^2 \rho v^3}{2M^2 \tau_1 \tau_2} \ . \tag{3}$$

Here M is the deformation potential, ρ is the mass density, and v is the sound velocity. It is noted that the amplitude dependence, at high amplitudes, which is proportional to $1/\sqrt{J}$ rather than $1/J$, is characteristic of this model: this dependence originates from the fact that the distribution of the energy splitting of TLS is larger than the energy uncertainty due to power broadening. The frequency and temperature dependence of α_{A0} in Eq. (2) is

$$\alpha_{A0}=\frac{\pi n M^2}{\rho v^3}\omega\tanh\frac{\hbar\omega}{2kT} \quad , \tag{4}$$

where n is the density of states of the TLS. In the present experimental conditions $\hbar\omega << 2kT$ is always satisfied so that the factor $\tanh(\hbar\omega/2kT)$, which comes from the population difference between two levels, can be approximated by $\hbar\omega/2kT$; i.e.,

$$\alpha_{A0}\propto\omega^2/T \quad \text{for } \hbar\omega << 2kT \quad . \tag{5}$$

The amplitude independent part of the attenuation coefficient, α_l, is expressed as

$$\alpha_l=\frac{nD^2}{8\rho v^3 kT}\int\operatorname{sech}^2\left(\frac{E}{2kT}\right)\frac{\omega^2\tau_1}{1+\omega^2\tau_1^2}\,dE \quad , \tag{6}$$

where D is the diagonal coupling constant between the pseudospin and phonons. If the dominant relaxation mechanism is via phonons, then

$$1/\tau_1\propto E^3\coth(E/2kT) \tag{7}$$

therefore, for the low-temperature limit, and $\omega\tau_1 >> 1$, we obtain

$$\alpha_l\propto\omega^0 T^3 \quad . \tag{8}$$

With regard to velocity changes, the theory gives for the resonance contribution

$$\frac{\Delta v}{v}=\frac{nM^2}{\rho v^2}\left\{\ln\left(\frac{T}{T_0}\right)+O\left[\left(\frac{\hbar\omega}{kT}\right)^2\right]\right\} \quad , \tag{9}$$

where T_0 is an arbitrary reference temperature. There is neither amplitude dependence nor frequency dependence in $\Delta v/v$, the latter as long as $\hbar\omega << 2kT$. The relaxation mechanism gives a constant contribution at low temperatures and negative temperature dependence at high temperatures, provided that Eq. (7) holds.

III. EXPERIMENTAL PROCEDURE

Amorphous $Pd_{0.775}Si_{0.165}Cu_{0.06}$ rods, 3 mm in diameter, were prepared by quenching the melt contained in thin-walled silica tubes, into water at 0 °C. In order to prevent crystallization the rods were carefully cut by a string saw into several pieces, some of which were used for calorimetry to check the state of the material. X-ray diffraction was also done on all samples. Pieces 10 to 15 mm in length were lapped and

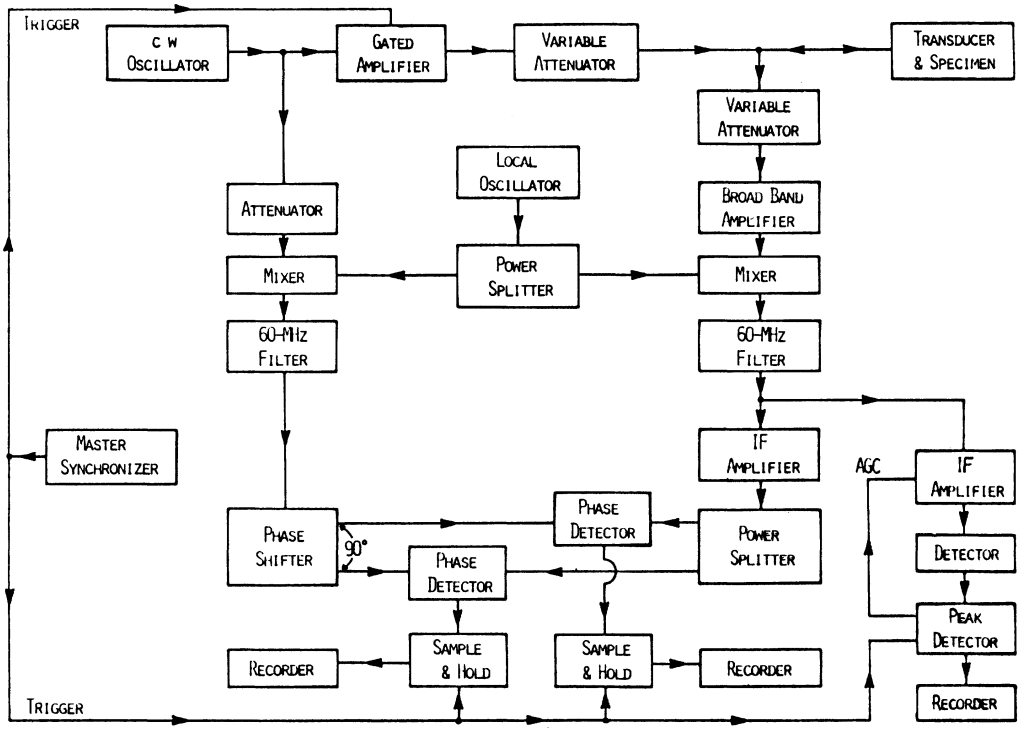

FIG. 1. Block diagram of equipment for the simultaneous measurement of the attenuation coefficient and the velocity change.

polished on the ends and used as specimens. Nonparallelism of the end faces was less than 10^{-4}.

A new method was developed[14] for precise measurements of ultrasonic attenuation and velocity changes. Its block diagram is shown in Fig. 1. To avoid any spurious amplitude dependences which come from the fact that the resonance frequencies and/or amplitude of a pulsed oscillator may be affected by a slight change in load, a gated amplifier was used for the ultrasonic pulse source. The received echo signals were converted to i.f. of 60 MHz, amplified, detected, and peak detected. The resolution of amplitude detection is determined by the signal-to-noise (S/N) ratio of the i.f. amplifier (Matec 6600) and peak detector (Matec 2470), and is better than 0.004 dB.

Use of the gated amplifier made it possible to obtain the phase information of the echo signals. The echo signal and the cw of the original oscillator are converted to i.f. (the conversion does not affect the phase information) in order to obtain high S/N ratios. The converted cw signal is split into two parts which have equal amplitude, but differ in phase by $\frac{1}{2}\pi$. Each of these cw signals is fed into a phase detector together with the converted echo signal to obtain the relative phase change in the specimen. (A sample-and-hold circuit follows each detector to obtain readable signals.) Resolution of the velocity change measurement is determined by the stability of the original oscillator (Wavetek 3001) and amplifier of the sample-and-hold circuit (homemade) and is better than 10^{-6}.

All the experiments were performed with a repetition rate of 10 or 20 pulses per second and a pulse width of $\sim$1.0 μs. By changing the repetition rate from 1 to 40 pulses per second, and the pulse width from 0.5 to 2.0 μs, it was found that no measurable heating effect due to the ultrasonic pulses occurred, even at the lowest temperatures and highest amplitudes used.

Lithium niobate crystals were used as ultrasonic transducers. They were bonded to the specimens by Dow Corning silicone oil No. 200, diluted with trichloroethylene.

IV. EXPERIMENTAL RESULTS AND DISCUSSIONS

A. Observations

Several frequencies between 10 and 90 MHz were chosen for measurements. Typical temperature dependence of attenuation is shown in Figs. 2(a) and 2(b) for 43- and 25-MHz transverse waves and in Fig. 3 for 27-MHz longitudinal waves, for different ultrasonic amplitudes whose origin is fixed arbitrarily for each data set. As is seen in these figures, below about 4 K a marked negative amplitude dependence is observed for transverse waves, whereas longitudi-

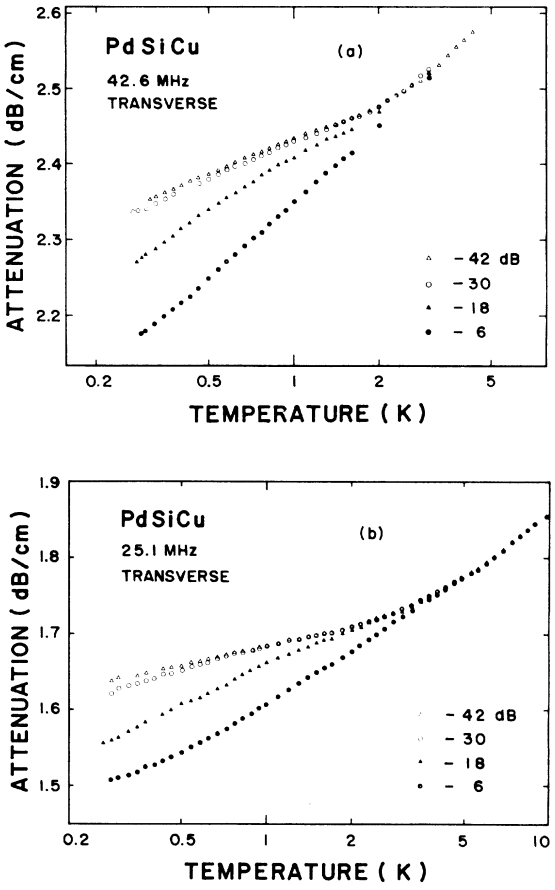

FIG. 2. Temperature dependence of the attenuation coefficient of (a) 42.6- and (b) 25.1-MHz transverse waves for four different amplitudes. Typical uncertainties in the results are about the size of the symbols for 25.1 MHz and about twice that size for 42.6 MHz.

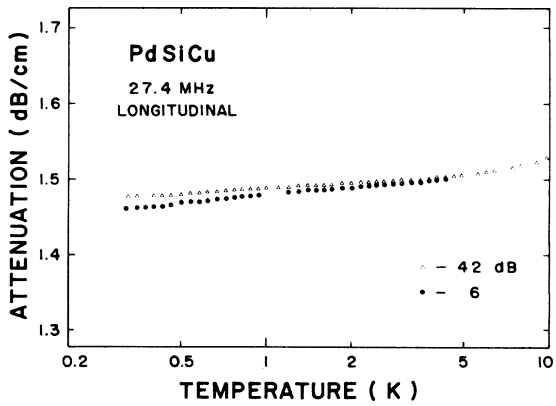

FIG. 3. Temperature dependence of the attenuation coefficient of 27.4-MHz longitudinal waves for two amplitudes.

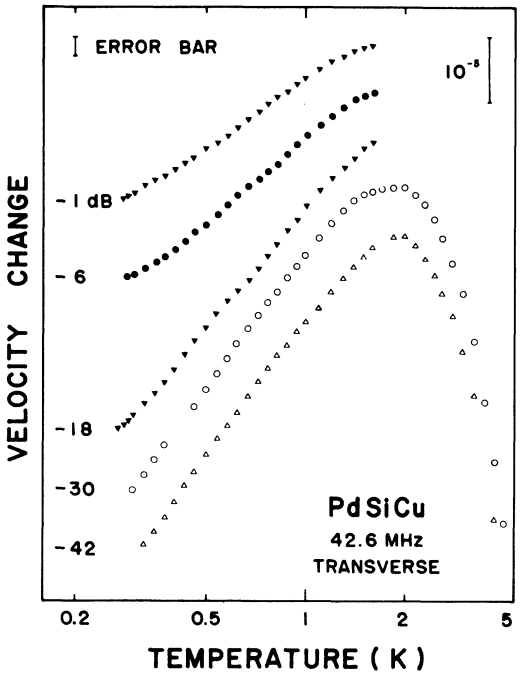

FIG. 4. Temperature dependence of the velocity changes of 42.6-MHz transverse waves for several amplitudes.

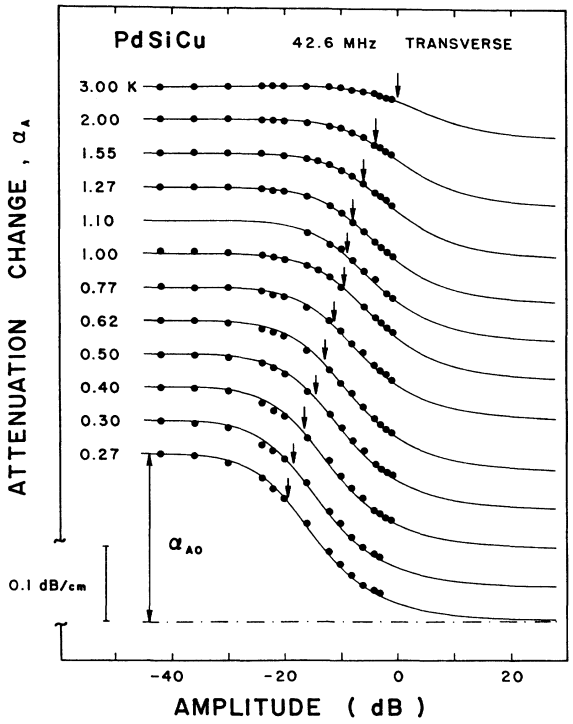

FIG. 5. Amplitude dependence of the attenuation coefficient of 42.6-MHz transverse waves for various temperatures (dots). Fitted curves, using Eq. (2), are also shown. The arrows mark values of J_c obtained from the fitting.

nal waves give a much smaller effect. The temperature dependence of velocity change was also measured simultaneously. The results for 43-MHz transverse waves are shown in Fig. 4. As the theory predicts, the velocity change, $\Delta v/v$, is proportional to $\ln T$ below a maximum around 2 K. Contrary to the theoretical prediction, however, an amplitude dependence of this slope was observed. Longitudinal waves also give $\ln T$ dependence, but with a slope about $\frac{1}{3}$ of that observed for transverse waves. Some amplitude dependence of the slope was also observed for longitudinal waves.

B. Amplitude-dependent part of attenuation

In order to examine the amplitude dependence of the amplitude-dependent attenuation, α_A, in more detail, we plotted α_A for transverse waves against ultrasonic amplitude, at various temperatures. (The changes of the attenuation of longitudinal waves were too small to be analyzed in this manner.) The observed amplitude dependences were found to fit very well the theoretical curves given by Eq. (2). An example of the fit is shown in Fig. 5 for 43 MHz. Other possibilities of amplitude dependence, such as $1/(1+J/J_c)$, were also tested, however, Eq. (2) was found to give the best fit, as indicated by the solid curves in Fig. 5. From this fitting we can obtain two quantities, α_{A0} and J_c (the latter is indicated by arrows in Fig. 5).

In Fig. 6 the temperature dependence of α_{A0},

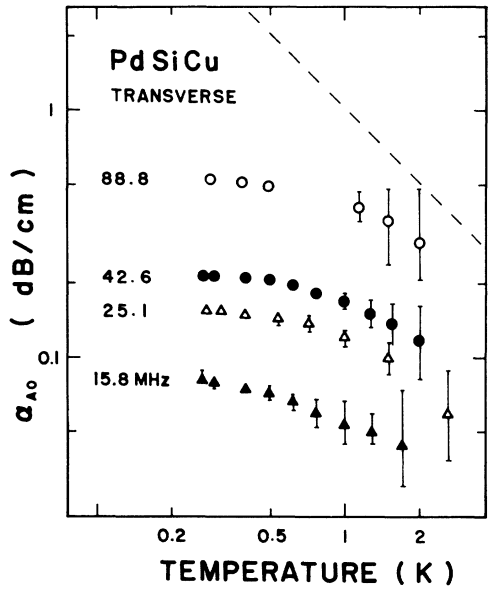

FIG. 6. Temperature dependence of α_{A0} of transverse waves at four different frequencies. The dashed line represents the slope of $1/T$ variation [see Eq. (4) for reference].

determined by the above procedure, is shown for several frequencies. It is obvious that the temperature dependence does not follow Eq. (5), i.e., $1/T$, at least below about 1 K, where α_{A0} tends to a constant value. Every data set follows a similar temperature dependence. In order to compare the experimental frequency dependence to that displayed by Eq. (5), i.e., ω^2, in Fig. 7 we show a plot of α_{A0} against the frequency used, at 0.3 K, where little temperature dependence is observed. As is seen in the figure, the frequency dependence is linear rather than quadratic. Moreover, the data by Doussineau *et al.*[10] taken on the same type of metallic glass at 0.062 K and 720 MHz falls on the linear frequency extrapolation of our data, despite the difference in temperature. This fact strongly suggests that the temperature-independent region can be extended down to 0.06 K.

The temperature dependences of the critical intensity J_c, obtained from fitting α_A to Eq. (2) for four different frequencies, are plotted in Fig. 8. This figure shows, within the temperature range covered, roughly quadratic temperature dependence of J_c for all frequencies. The frequency dependence of J_c appears to be very weak, although the absolute values of the acoustic intensity are very difficult to determine experimentally. Therefore, phenomenologically the following relation holds:

$$J_c \propto \omega^n T^2, \quad |n| < 1 \ . \tag{10}$$

We note here that relation (10) with $n=0$ holds experimentally for dielectric glasses.[4] As is seen above, the agreement of the functional form of Eq. (2) with the measured amplitude dependence of the attenuation suggests not only that the saturation occurs through the exhaustion of absorbing systems, but also that there exists a continuous distribution of such states. The critical intensity obtained here at 0.3 K is one order of magnitude larger than that deduced by Doussineau *et al.*[10] These results are not inconsistent with each other provided that relation (10) holds also for the frequency and temperature range these workers used. From the derived J_c we can estimate the relaxation time. Assuming $\tau_1 = \tau_2 = \tau$, $M = 0.1 \sim 1$ eV, $J_c \sim 10^{-3}$ W/cm^2, for 0.3 K we obtain (in ns)

$$\tau \simeq 1-10 \ . \tag{11}$$

This is, again, not inconsistent with the Golding *et al.*[11] upper limit for relaxation time ($\tau < 25$ ns at 10 mK) obtained from the double pulse measurement; provided that relation (10) holds, the upper limit of their relaxation time extrapolates to 750 ns at 0.3 K.

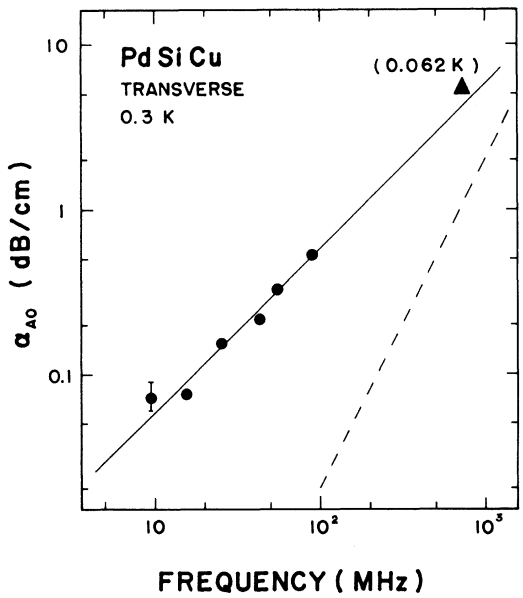

FIG. 7. Frequency dependence of α_{A0} at 0.3 K (dots). The solid triangle represents the data of Ref. 10 taken at 62 mK, and the dashed line shows α_{A0} extrapolated to 0.3 K from the data of Ref. 10, on the basis of Eq. (4).

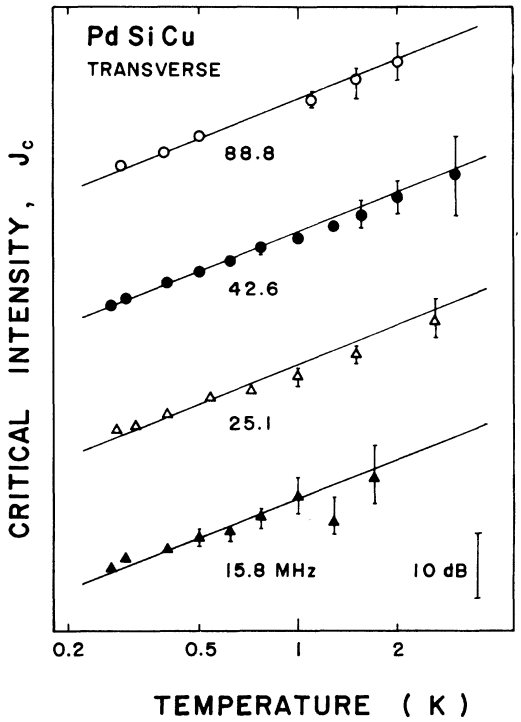

FIG. 8. Temperature dependence of the critical intensity, J_c, at four frequencies (see Fig. 5). Because of the uncertainties in the absolute intensities for each frequency, the origin of each data set is shifted arbitrarily.

Despite the agreement of the present results with the functional form of Eq. (2) and the consistency of the value for the relaxation time found here and those found by other workers, there remain serious discrepancies with current TLS theory. In particular, this theory predicts ω^2/T dependence of α_{A0}, which is not found here. Moreover, the value of nM^2 deduced by Doussineau *et al.*[10] on the basis of the theory [see Eq. (4)], i.e., $nM^2 \simeq 10^7$ erg/cm^3, is, respectively, two to three orders of magnitude smaller than the value obtained from the present results for 100 to 10 MHz. The dashed line in Fig. 7 represents the predicted α_{A0} [Eq. (4)] at 0.3 K, using $nM^2 = 10^7$ erg/cm^3. In order to check whether this large discrepancy is confined to metallic glasses, an experiment on a dielectric glass, i.e., fused silica Suprasil 1 (Ref. 15) was carried out in the same temperature range, using 30-MHz transverse waves. Over a dynamic range of about 60 dB of ultrasonic amplitude (the highest amplitude was similar to that for PdSiCu), no amplitude dependence was found, within the accuracy of our experiment. This means either that the amplitudes used are too high to observe the unsaturated region, or too low to observe the saturation, or that α_{A0} itself is too small to be observed. The former two possibilities are less likely than the last, because, if the frequency independence of J_c observed experimentally at high frequencies[4] can be extended to lower frequencies, then after taking into account the T^2 dependence, the expected J_c should fall in the range covered by our experiment. On the other hand, if ω^2/T dependence predicted by the TLS theory holds in this case, by extrapolating the data of Hunklinger *et al.*[16] on Suprasil W, at 750 MHz and 0.48 K, to 30 MHz and 0.3 K, we obtain (in dB/cm)

$$\alpha_{A0} \simeq 0.006 \ . \tag{12}$$

This is too small to be observed. However, if α_{A0} is linear in frequency and nearly independent of temperature, as for the metallic glass, we obtain

$$\alpha_{A0} \simeq 0.1 \ . \tag{13}$$

In this case α_{A0} should be observable. Therefore, we can conclude that the frequency dependence of α_{A0} for metallic glass is essentially different from that for dielectric glass, whose behavior the TLS theory explains very well.

The very small amplitude dependence observed for longitudinal waves prevents us from drawing any firm conclusions. However, it should be noted that the TLS theory predicts for longitudinal waves one order of magnitude smaller values for α_{A0} and one order of magnitude larger values for J_c than for transverse waves. Therefore, in this regard the experimental results are not inconsistent with the theory.

C. Amplitude-independent part of attenuation

Knowing the quantity α_{A0}, one can deduce the completely saturated attenuation, α_l, as a function of temperature. Figure 9 shows the dependence of α_l on temperature for various frequencies (each data set has an apparent attenuation due to geometric effects, etc., so that only its temperature dependence is relevant). The dashed and the dotted curves in the figure represent the theoretical prediction [Eq. (6)] for two extreme regimes, i.e., $\omega\tau_1 >> 1$ and $\omega\tau_1 << 1$, respectively. We used τ_1 due to phonons [Eq. (7)] to obtain $\alpha_l \propto \omega^0 T^3$ for $\omega\tau_1 >> 1$ and τ_1 due to electrons [see below: Eq. (14)] to obtain $\alpha_l \propto \omega^2 T^{-1}$. $M^2 = D^2 = 0.1$ (eV)^2 and $nM^2 = nD^2 = 10^7$ erg/cm^3 were chosen; in addition, for the dotted curve we used $n_e V_\perp = 0.1$ and $\omega/2\pi = 100$ MHz. It is obvious that the temperature dependence of α_l of our data is quite different from either T^3 or T^{-1}; it ranges from $\ln T$ to $T^{1/2}$. Moreover, as seen in Fig. 9 the temperature-dependent part of α_l increases as frequency increases, and the change is approximately linear in the frequency used.

As pointed out by Golding *et al.*,[11] this temperature and frequency dependence might be explained by a very short relaxation time. Assuming the relaxation due to electrons, through a Korringa-type mechanism, dominates at low temperatures over that due to phonons, one obtains for τ_1 (Ref. 11)

$$\tau_1 = \frac{2\hbar \tanh(E/2kT)}{\pi (n_e V_\perp)^2 E} \ , \tag{14}$$

where n_e is the density of states of electrons at the

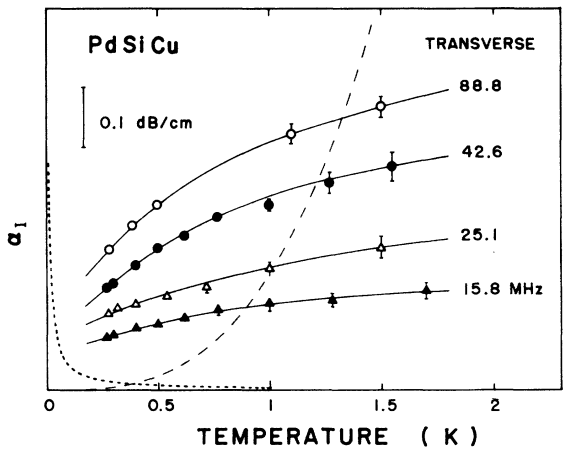

FIG. 9. Temperature dependence of the amplitude-independent attenuation coefficient, α_l, at four frequencies. Temperature-independent part of α_l was arbitrarily subtracted (see Ref. 17). The dashed curve shows T^3 theoretical dependence of α_l [Eq. (8)] for $\omega\tau_1 >> 1$ and the dotted curve represents T^{-1} dependence [Eq. (14)] for $\omega\tau_1 << 1$ (see text).

Fermi surface and $V_\perp$ is a coupling constant between a TLS and conduction electrons.

Using Eq. (14) together with Eq. (6), one obtains for the relaxation contribution to the attenuation, α_l,

$$\frac{\alpha_l(x)}{\omega} = \frac{nD^2}{4\rho v^3}\int_0^\infty \mathrm{sech}^2\xi \frac{\tanh\xi/2x\xi}{1+(\tanh\xi/2x\xi)^2}\,d\xi \ , \tag{15}$$

with

$$x = \tfrac{1}{2}\pi(n_e V_\perp)^2\frac{kT}{\hbar\omega}, \quad \xi = E/2kT \ . \tag{16}$$

Therefore, the logarithmic decrement, α_l/ω, taken at different frequencies, should fall on the same curve, when plotted against T/ω. A family of curves representing α_l/ω of Eq. (15) was computed, using nD^2 and $n_e V_\perp$ as parameters. A plot of the experimental decrement α_l/ω for four frequencies 16, 25, 43, and 89 MHz vs T/ω was then compared with these theoretical curves, and no satisfactory fit could be obtained.[17] The discrepancy between experimental and calculated values was particularly striking for the data obtained at 90 MHz. Finally, we note that a spread in the values of τ_1, at a given energy, does not account for the experimental results.

D. Velocity change

As is seen from Fig. 4, the velocity change, $\Delta v/v$, below about 2 K is linear in $\ln T$, in agreement with the prediction of the TLS theory. The slopes of $\Delta v/v$ give values for nM^2, i.e., $(0.4-1.2)\times 10^7$ erg/cm^3, which are of the same order as other workers' results.[8,11,18] However, a significant amplitude dependence of the slope was found,[19] contrary to the predictions of the theory [Eq. (9)]. Figure 10 shows the dependence. There is also an increase found in the slope of $\Delta v/v$ with increasing frequency. For example, at the lowest amplitudes used, the slope increases from 2.2×10^{-5} to 3.5×10^{-5} as frequency increases from 10 to 90 MHz, with a monotonic change for intermediate frequencies. The theoretical expression, Eq. (9), has a correction term of order $(\hbar\omega/kT)^2$, which in this case is $\sim 10^{-5}$ and cannot, therefore, explain the observed frequency dependence.

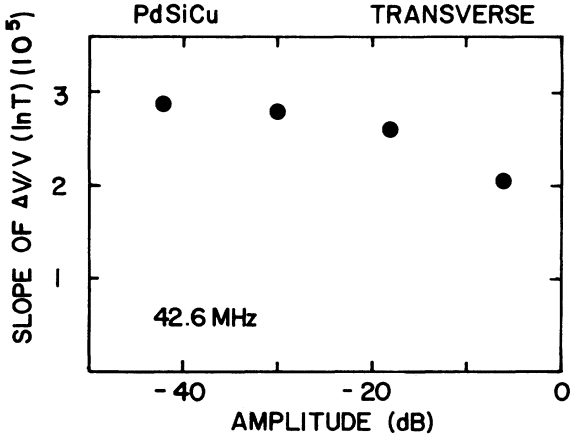

FIG. 10. Slopes of the velocity changes vs $\ln T$ at 42.6-MHz transverse waves for four different amplitudes.

V. SUMMARY AND CONCLUSIONS

As has been shown, some of our results agree very well with the current TLS theory, i.e., (a) the form of amplitude dependence of the attenuation, and (b) the temperature dependence of the velocity change. Some results are not inconsistent with the theory, i.e., (c) the fact that longitudinal waves have a smaller amplitude dependence of attenuation and smaller velocity change than transverse waves. Some of the parameters obtained from our results are consistent with those of other workers, i.e., (d) the relaxation time obtained from the critical intensity, J_c, and (e) nM^2 obtained from the velocity measurements. However, there are serious discrepancies with the theory, i.e., (f) the different temperature dependence of the saturable attenuation, α_{A0}, (g) the different frequency dependence of α_{A0}, (h) the different temperature and frequency dependencies of the amplitude-independent part of the attenuation, α_l and (i) the existence of amplitude dependence as well as of some frequency dependence of the low-temperature slope of $\Delta v/v$.

As pointed out by Black,[12] a very short relaxation time might be one of the clues to solving some of these discrepancies. However, as we have shown in Sec. IV C, a Korringa-type relaxation mechanism does not give results which are in agreement with our observations. In addition, the frequencies we used are so small that all our experiments were carried out in the regime $\omega\tau \lesssim 1$. This fact raises a conceptual difficulty concerning "resonance" attenuation. In other words, in this regime the amplitude dependent part of the attenuation should be negligibly small or, if any, according to current TLS theory it should be quadratic in frequency, as long as we assume a density of TLS states independent of energy.[20]

In view of the above, it appears that additional mechanisms, or an alternative theory will be required to explain our observations.

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[18]J. R. Matey and A. C. Anderson, Phys. Rev. B 17, 5029 (1978).
[19]For larger amplitudes the curve deviates from $\ln T$ at low temperatures. We verified that this is not due to heating, by performing experiments with different repetition rates.
[20]A linear dependence of α_{A0} can be obtained, of course, by selecting an energy-dependent density of TLS states, $n(E)$. An appropriate choice of constants in $n(E)$ may yield such a fit without introducing an inconsistency with the existing data for the specific heat and thermal conductivity, which for PdSiCu were taken only down to 1 and 0.1 K, respectively [B. Golding, B. G. Bagley, and F. S. L. Hsu, Phys. Rev. Lett. 29, 68 (1972); and Ref. 5]. Thus very much higher frequency phonons are involved in these properties than the ultrasonic ones used in the present study. Moreover, such an energy-dependent density of states may be helpful in accounting for the data of Matey and Anderson (Ref. 18), who found for PdSiCu values of nM^2 six times larger than those deduced from ultrasonic velocity (as a function of temperature) measurements. However, such an $n(E)$ does not affect the $1/T$ dependence of α_{A0} expected from current theory. Thus, items (f), (h), and (i) in the summary would still remain unresolved.