memory terms which have been neglected so far in Akcasu's approach. In view of the simplifications made in the calculations of Dubois-Violette and de Gennes, it is not surprising to also find some discrepancies at long times. While the preaveraging of the Oseen tensor has only a minor influence on the linewidth[12] and probably also on the line shape in the q region considered, the neglect of the excluded-volume interaction seems to be the most severe simplification. An experimental check of this statement will be carried out by future measurements under θ conditions.

At higher concentrations, Fig. 3 clearly shows strong deviations from the q^3 behavior in going to smaller q. This can be taken as an experimental indication for the crossover from a q^3 to a q^2 behavior, as predicted by de Gennes[4] for entangled systems. (This crossover is not to be confused with the expected q^2 behavior at very low q due to the diffusion of the polymer as a whole.) However, the small number of data points prevents an accurate determination of the exponent in the low-q range. For this purpose, more experiments will be necessary. Within the de Gennes theory the position of the crossover point q^* in momentum space allows a dynamical estimate of the screening length ξ, although in order to give a numerical value, one has to know the proportionality factor. The values given on Fig. 3 were obtained assuming $q^*\xi = 1$, and their order of magnitude agrees quite well with those found in static experiments on polystyrene in benzene.[2] Using $q^*\xi = \sqrt{6}$ as given by Akcasu[12] would increase ξ. To prove the predictions concerning the concentration dependence of ξ or D_c again more experiments will be necessary. For the two concentrations we have $D_c(0.3\ \mathrm{g/cm^3})/D_c(0.15\ \mathrm{g/cm^3}) = 1.54 \pm 0.09$ and for the monomer concentrations $(0.26/0.15)^{3/4} = 1.51$, which is in agreement with the proposed behavior.

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Relaxation of Tunneling Systems by Conduction Electrons in a Metallic Glass

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Acoustic experiments on the metallic glass $Pd_{0.775}Si_{0.165}Cu_{0.06}$ are presented which show a tunneling-system decay rate at 0.01 K enhanced by a factor of 10^4 over insulating glass. A theory is presented for decay based upon the coupling of tunneling systems to conduction electrons, and the parameters of the theory are obtained from experiment. Relaxational absorption and dispersion in this amorphous metal are interpreted in terms of a tunneling model with a broad spectrum of decay rates.

In this Letter we show that a strong interaction exists between the conduction electrons and the intrinsic two-level tunneling systems of an amorphous metal. This coupling is inferred by observation in acoustic experiments of a greatly enhanced tunneling-system decay rate T_1^{-1} in the metallic glass $Pd_{0.775}Si_{0.165}Cu_{0.06}$ in comparison with amorphous insulators such as SiO_2. A theory

of conduction-electron–tunneling-system relaxation is presented which provides a description of the energy and temperature dependence of the decay rates. Finally, we argue that a distribution of decay rates, as would arise from an atomic tunneling process, is required to yield reasonable agreement with these experiments.

Like insulating glasses below 1 K, metallic glasses exhibit a nearly linear temperature-dependent contribution to their specific heats,[1] a nearly T^2 phonon thermal conductivity,[1,2] and also show resonant phonon absorption[3] and dispersion.[3,4] These phenomena can thus be interpreted in terms of a general model which describes amorphous solids as supporting a broad distribution of two-level atomic tunneling systems which arise as an intrinsic feature of disorder.[5] In spite of the similarities among disordered solids, recent experiments[3] on PdSiCu revealed the following unusual features: (i) The acoustic saturation intensity I_c was inferred to be anomalously large, so that the usual tunneling-system relaxation process involving one-phonon decay seemed unlikely. (ii) A nonsaturable contribution to acoustic absorption, varying approximately as $\log T$, was observed between 0.1 and 4 K. (iii) Negative deviations from $\log T$ behavior in the resonant sound velocity were observed at temperatures as low as 0.2 K. We shall show that all of these anomalies occur as a result of tunneling-system–conduction-electron interactions.

In the present experiments, the acoustic absorption and velocities have been measured for PdSiCu at a frequency $\omega/2\pi \approx 1$ GHz, as a function of acoustic intensity, for temperatures between 10 mK and 10 K for longitudinal and transverse phonon polarizations; see Fig. 1.

The ability to observe a large saturation effect for the transverse mode at 10 mK has allowed us to perform two-pulse saturation-recovery experiments. Two 100-nsec–duration acoustic pulses are applied to the glass. The first, with large amplitude, is capable of nearly saturating the two-level systems; the second, much weaker, probes the recovery of the resonant energy levels at the pulse separation time τ_{12}. At 10 mK, we observe no effect on the second pulse by the first when τ_{12} is as small as 100 nsec. Since our sensitivity is sufficient to discern an effect after approximately 4 time constants, *we place an upper limit on T_1 of 25 nsec at 0.96 GHz and 10 mK.* For comparison, in fused silica glass under similar conditions, it has been observed[6] that $T_1 \approx 200$ μsec, a decay time longer by four orders of magnitude.

In glassy insulators below 1 K, it is well established that decay of tunneling systems is governed by a one-phonon process.[6] If such a process were to explain the anomalously rapid decay time in PdSiCu it would require a phonon–tunneling-system coupling more than an order of magnitude larger than existing in SiO_2.[3,6] Since this possibility seems unreasonable, we propose that a *parallel* decay of energy through the conduction-electron system of a metallic glass is possible. To make this idea quantitative the following Hamiltonian is introduced[7]:

$$H = H_0 + (2M_\nu S_x + D_\nu S_z)e_\nu + N^{-1}(v_\perp S_x + v_\parallel S_z)\sum_{kk'} c_k^\dagger c_{k'}, \quad (1)$$

where $H_0 = ES_z + H_{\rm ph} + H_{\rm el}$ represents, respectively, the noninteracting pseudo-spin-$\frac{1}{2}$ tunneling systems of energy E, the phonons, and the conduction electrons; and N is number of atoms in the glass. The coupling of a tunneling system to a strain e_ν of polarization ν is given by off-diagonal M_ν and diagonal D_ν elements, whereas coupling of tunneling systems to the conduction-electron density is given by off-diagonal and diagonal elements $v_\perp$ and $v_\parallel$, respectively. We describe by the latter term a process in which conduction electrons with density of states $n(E_F)$ per atom at the Fermi energy are inelastically scattered off the tunneling system with energy change $\pm E$, with a resulting tunneling-system decay rate

$$R_{\rm el} = (\pi/2\hbar)[n(\epsilon_F)v_\perp]^2 E \coth(\tfrac{1}{2}\beta E). \quad (2)$$

This process is formally analogous to the Korringa relaxation[8] of nuclear spins due to conduction electrons in metals.[9,10] With the previously stated limit on $T_1 < 25$ nsec, we find that $n(E_F)v_\perp > 0.04$.

The effect of this fast energy decay on other acoustic processes is profound. For example, the diagonal strain coupling term in Eq. (1) gives rise to an intensity-independent interaction known as "relaxational" absorption[11] (and dispersion) which depends on the size of the product ωT_1 with respect to unity. Here T_1 is the decay time of thermally excited tunneling systems ($E \approx 2k_B T$). In glassy insulators, the condition $\omega T_1 \gg 1$ is generally obeyed in the temperature regime in which tunneling is believed to occur.[12] In PdSiCu, however, the fast electronic decay process permits relaxational effects in the regime $\omega T_1 \lesssim 1$ to be

observed for temperatures as low as 100 mK where the behavior of T_1 is well characterized. We suggest that the anomalous behavior discussed above (items ii and iii) can be explained by a "relaxational" process if a distribution of decay times is included.

We assume that the tunneling picture[5] applies to PdSiCu at temperatures $T \lesssim 10$ K. Thus we consider a distribution of tunneling entities residing in double-well potentials with tunneling matrix element $\Delta_0 = (\hbar\Omega)e^{-\lambda}$, where $\hbar\Omega$ is a zero-point energy, and with asymmetry Δ. The system's lowest two eigenvalues are then $E = \pm\frac{1}{2}(\Delta_0^2 + \Delta^2)^{1/2}$. The diagonalized coupling terms become $M = \gamma\Delta_0/E$, $D = 2\gamma\Delta/E$, and $v_\perp = K_\parallel\Delta_0/E$, where γ and $K_\parallel$ are phonon and electron deformation potentials. The relaxational contribution to the absorption $\alpha_{\rm rel}$ and velocity shift $\Delta V_{\rm rel}/V$ is given by

$$\begin{pmatrix} \alpha_{\rm rel}/\omega \\ -2\Delta V_{\rm rel}/V \end{pmatrix} = \int_0^\infty dE\, \mathrm{sech}^2(\tfrac{1}{2}\beta E) \int_{r_{\min}}^1 dr\, P(r) D^2(r) \frac{1}{(\omega/R)^2+1} \begin{pmatrix} \frac{\omega}{R}\frac{\beta}{4\rho v^3} \\ \frac{\beta}{4\rho v^2} \end{pmatrix}. \tag{3}$$

The inner integration is performed over the distribution function[11,13] $P(r) = \frac{1}{2}\bar{P}(1-r)^{-1/2} r^{-1-q}$, where $r \equiv \Delta_0^2/E^2$ and q is to be determined by experiment.[14] The rate R is the sum of two rates, $R = R_{\rm el} + R_{\rm ph}$, where[11] $R_{\rm ph} = \sum_\nu M_\nu^2 E^3 \coth(\frac{1}{2}\beta E)/2\pi\rho\hbar^4 V_\nu^5$ and $R_{\rm el}$ is given by Eq. (2). Because of its stronger E dependence, the one-phonon rate $R_{\rm ph}$ will dominate decay above a few K. With the above assumptions the resonant absorption and velocity contributions are $\alpha_{\rm res}/\omega = \frac{1}{2}\pi\,(\bar{P}\gamma^2/\rho V^3) \times B(1-q, \frac{1}{2})\tanh[\frac{1}{2}\beta\hbar\omega]$ and $\Delta V_{\rm res}/V = (\bar{P}\gamma^2/2\rho V^2) \times B(1-q, \frac{1}{2})\ln(T/T_0)$, where B is the β function and T_0 is a reference temperature. The total absorption and velocity change is the sum of resonant and relaxational contributions.

We have evaluated Eq. (3) numerically as a function of the parameters q, $mv_\perp$, γ, and $\bar{P}$ with the results shown in Fig. 1. The parameter values used here are $q = 0.4$, $mv_\perp = 0.16$, $\gamma_t = 0.4$ eV, and $\bar{P} = 2.2\times10^{31}$ erg^{-1} cm^{-3}. The electron-coupling parameter is very close to the lower limit, $mv_\perp > 0.04$, found by the saturation-recovery experiment. The value of $v_\perp$, which is the difference in the scattering pseudopotential seen by the electrons for the two tunneling configurations, is estimated at ~ 1 eV, taking $n(E_F) = 3N/2E_F$ and $E_F \approx 10$ eV. The phonon-coupling parameter is about a factor of 2 smaller than the value for SiO_2 found by phonon echo techniques, $\gamma_t(SiO_2) \approx 1.1$ eV, and $\bar{P}$ is the same[6] found for SiO_2.

It should be emphasized that we have attempted to describe here the acoustic properties of a glass in a temperature region varying by three orders of magnitude utilizing only four parameters and the tunneling model. Since there are four contributions involved (the resonant and the relaxational absorption and velocity) the problem is determined, and there is no assurance that four parameters can describe the experimental results. For example, it would be possible to describe the absorption data in Fig. 1 to arbitrary precision if the constraint on either velocity contribution were relaxed or, alternatively, if an

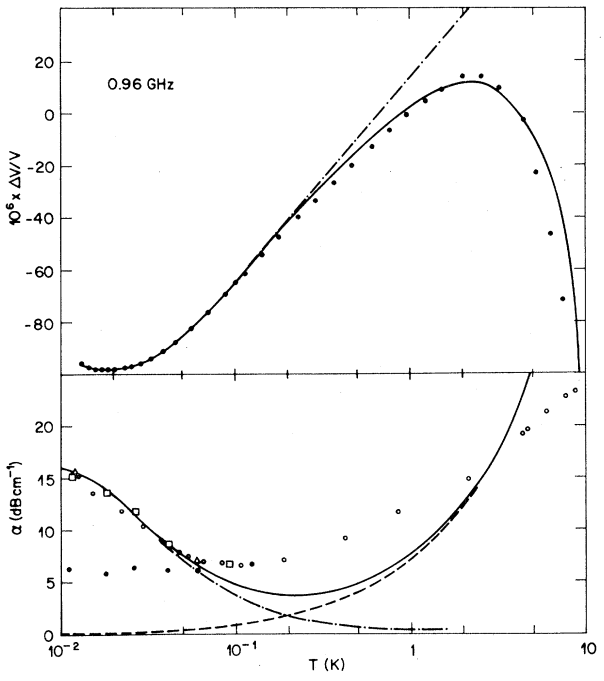

FIG. 1. Transverse acoustic velocity (upper panel) and absorption coefficient (lower panel) of $Pd_{0.775}Si_{0.165}Cu_{0.06}$ at low temperatures. The solid line is the theoretical calculation composed of two components: resonant, indicated by a dot-dashed line; and relaxational, indicated by a dashed line. The solid circular points in the absorption data refer to an acoustic intensity of 10^{-2} W cm^{-2}; the other points are for an intensity as low as 10^{-6} W cm^{-2}.

additional coupling parameter were introduced to describe the relaxational processes, an approach followed previously.[12] However, the tunneling model, taken strictly, allows only *one* coupling constant in the description of resonant and relaxational response.

In our model calculation, the relative sizes of resonant and relaxational behavior are governed by q, the position of the velocity maximum and the velocity temperature dependence above the maximum are determined by the phonon coupling γ and by q, and the deviation from $\ln T$ velocity behavior above 0.2 K is caused by relaxational dispersion. The deviation of theory from experiment at low temperatures arises[15] from the overly simple power-law form for the q-dependent distribution function $P(\gamma)$. At higher T, the deviations result from neglecting correlations between Δ_0 and Δ, i.e., from assuming that Δ is uniformly distributed.

A sensitive test of the coupling parameters can be made by calculating the critical intensity for saturation I_c, since saturation at very low temperatures occurs as a balance between phonon-induced transitions and electron-induced decay. At 10 mK since the pulse duration is much greater than T_1, a steady-state solution to the rate equations exists with $I_c = 2\rho V^3(\hbar/\gamma)^2 T_1^{-2}$, where we assume $T_1 \approx T_2$. For transverse phonons the calculation gives $I_c = 2\times 10^{-3}$ W cm^{-2}, whereas from measurement we find $I_c \approx 1\times 10^{-3}$ W cm^{-2}, well within experimental uncertainty in obtaining absolute[16] intensities.

The proper description of the relaxational acoustic behavior in PdSiCu requires a distribution of decay rates for two-level systems of a given energy.[17,18] If a single decay rate were assigned by taking $M = \gamma$ in Eq. (2) (and assuming a roughly uniform density of states), then a maximum in $\alpha_{\rm rel}$ would occur when ωT_1 ($E \approx 2k_B T$) ~ 1, i.e., just below 1 K, in clear disagreement with the data in Fig. 1. The extent of the distribution can be estimated by examining the maximum value of $\gamma_{\rm min}$ in Eq. (3) which is consistent with experiment. For systems of energy $E/k_B \approx 0.1$ K, for example, we find a *minimum* distribution in T_1 of four orders of magnitude. Thus the present results constitute strong evidence for a two-level *tunneling model* of intrinsic defects in glasses, as opposed to simply a general two-level model.

It has been claimed that tunneling systems can contribute to anomalies in the *electrical* resistivity of glassy metals.[19] The present description of the tunneling-system–conduction-electron interaction is fundamentally different[7] from this previous theory and, based upon our experimental value for $K_{\|}$, does not lead to readily observable effects in electronic transport.

In a recent paper, Bellessa[20] has claimed that the acoustic velocities of glasses in the region $4 < T < 20$ K cannot be explained by the tunneling model: Our calculation for the velocity exhibits a nearly linear T dependence and a logarithmic frequency behavior in this limited temperature region, and a precise fit to experiment can easily be obtained by minor modification of the distribution functions. Thus we do not believe that a different model must be invoked to explain these properties.

In conclusion, we have shown that the presence of conduction electrons is the origin of the unusual tunneling system dynamics in metallic glasses. The existence of greatly enhanced decay rates for these systems has provided a unique opportunity for examining acoustic processes in a region in which they are very sensitive to assumptions about the spectrum of relaxation times.

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