Low-temperature acoustic properties of metallic glasses

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The dynamics of two-level systems (TLS) are substantially different in metallic glasses than in insulators because the TLS are coupled to conduction electrons in addition to phonons. This coupling does not merely cause a broadening of the energy eigenstates of a TLS; it can alter the fundamental nature of the tunneling. These effects are reflected in the low-temperature acoustic-attenuation and velocity changes of these materials. We compare theoretical calculations of ultrasonic properties of a distribution of TLS in a metal to previouly unexplained 1-GHz acoustic measurements of the sound velocity and attenuation of the metallic glass $Pd_{0.775}Si_{0.165}Cu_{0.06}$ below 1 K.

I. INTRODUCTION

The theory of two-level systems[1] (TLS) has had great success in explaining the low-temperature properties of glasses.[2–4] For insulating glasses the TLS theory provides a phenomenological model for the observed specific heat, thermal conductivity, and ultrasonic properties. The formal connection with $S=\frac{1}{2}$ dynamics is the basis for understanding acoustic resonance experiments such as phonon echoes.[5]

There is also substantial evidence that TLS's are present in metallic glasses.[6–10] However, at least some properties of metallic glasses are significantly different from those of insulating glasses. For example, in normal metals the sound intensity needed to observe saturation of ultrasonic attenuation is several orders of magnitude larger than in insulating glasses, and phonon echoes have not been observed. These observations have been ascribed to the significantly shorter relaxation times caused by the coupling of the TLS to conduction electrons, which has generally been assumed to lead to Korringa-like broadening of the energy eigenstates of a TLS. The greatly enhanced relaxation rates thus have been attributed to the quantitative issue that at low temperatures T the number of electronic excitations is proportional to T rather than T^3, as is the case for phonons. Therefore, the difference between insulating and metallic glasses has mostly been assumed to be due to the quantitatively shorter relaxation time rather than different qualitative behavior of the TLS.

However, some acoustic properties of metallic glasses are *qualitatively* different from those of insulating glasses. Here we focus on two features of the data of Ref. 9 on $Pd_{0.775}Si_{0.165}Cu_{0.06}$ that are not consistent with standard TLS theory. The first feature is that the saturable attenuation of sound of frequency $\omega/2\pi=0.96$ GHz at low temperatures (~ 10 mK) is much smaller than the TLS theory predicts based on velocity measurements. The second feature is that, given a value for the attenuation at temperatures T low enough that the sound frequency ω satisfies $\hbar\omega/k_BT \gg 1$ (which is expected to be resonant), standard TLS theory yields a prediction for the high-temperature relaxational attenuation which is much smaller than the experimentally measured nonsaturable attenuation for $T>0.1$ K. In Ref. 9, it was postulated that, for reasons unknown at the time, the resonant attenuation was partly unsaturable. A modified TLS distribution function with more asymmetric TLS's than the standard distribution function (and hence larger relaxation attenuation) was used to fit the data. However, both these modifications are completely *ad hoc* and one would like a more satisfying understanding of the experimental observations.

This work is motivated by recent measurements of dissipative tunneling of a single defect in a mesoscopic metal,[11] where it is demonstrated that the temperature dependence of the tunneling rate of a two-state defect in a mesoscopic metal can be *qualitatively* inconsistent with the predictions of standard TLS theory. The coupling of TLS to conduction electrons as well as phonons leads to new effects that cannot be described merely in terms of a short relaxation time T_1. The increased spectral density of electronic excitations compared to phonons leads to the destruction of coherent tunneling of the TLS when the temperature is substantially larger than the tunnel splitting.[12,13] This theory has been applied successfully to measurements of muon diffusion in metals[14] and of tunneling of hydrogen interstitials in niobium.[15] The relevance of this destruction of quantum coherence to the TLS in metallic glasses has been noted elsewhere in the literature;[16,17] in this paper we show that accounting for this leads to substantially improved agreement between theory and measurements of the sound attenuation and velocity over a broad temperature range.

The coupling between electrons and the TLS has nontrivial effects on the acoustic properties of all temperatures. At high temperatures the tunneling of a TLS is incoherent, and the temperature dependence of the tunnel-

ing rate is qualitatively different from that predicted by the usual TLS perturbation theory, where it is assumed that the effects of the electron bath on a coherently tunneling TLS are weak.[18] At low temperatures, the dominant "resonant" contribution can also be modified substantially by the coupling to electrons. One example of this is a (formally) resonant contribution to the nonsaturable attenuation that occurs for metallic and not insulating glasses that is important at very low temperatures. This contribution arises because at zero temperature the coupling of a TLS of energy E to electrons causes the resonance linewidth to broaden by an amount ΔE that is proportional to E. The integral over the TLS distribution yields a contribution to the attenuation that diverges logarithmically with the upper cutoff of the TLS distribution function. Since this contribution to the attenuation arises from TLS's that are very far from resonance, one expects it to represent a nonsaturable piece of attenuation whose amplitude has the $\tanh(\beta\hbar\omega/2)$ temperature dependence that is usually characteristic of resonant attenuation, where $\beta=(k_B T)^{-1}$. More generally, we find that the saturation behavior can be complex and sensitively dependent both on temperature (even when $\beta\hbar\omega/2 \gg 1$) and on the electron-TLS coupling.

We compare this theory to previous measurements of the sound velocity and attenuation of $Pd_{0.775}Si_{0.165}Cu_{0.06}$.[9] We show that the theory explains naturally some previously mysterious ultrasonic properties, although agreement between theory and experiment is not yet quantitative.

This paper is organized as follows. In Sec. II A we review standard TLS theory and show why it is not consistent with the data on $Pd_{0.775}Si_{0.165}Cu_{0.06}$. Section II B discusses the general framework of the calculations in this paper, with particular stress on the relationship between the acoustic properties of interest here and the quantities usually calculated in dissipative tunneling theory. In Sec. II C we discuss the linear response of a TLS in the incoherent regime (temperature larger than the renormalized tunneling matrix element) and in Sec. II D we discuss the low-temperature coherent regime. Section III is a comparison between this theory and experiment, and the results are discussed in Sec. IV.

II. THEORY

In this paper we assume that the low-temperature acoustic properties of a glass are determined by the effects on a sound wave of frequency ω when it couples to the distribution of the TLS in the material. An isolated TLS with asymmetry ϵ and tunneling matrix element Δ_0 has a Hamiltonian

$$H_0=\epsilon\sigma_z-\hbar\Delta_0\sigma_x \ , \tag{2.1}$$

where

$$\sigma_z=\frac{1}{2}\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix}$$

and

$$\sigma_x=\frac{1}{2}\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix} .$$

The isolated TLS has two energy eigenstates with energies $\pm E/2$, where $E=\pm[(\hbar\Delta_0)^2+\epsilon^2]^{1/2}$. The relaxation of a TLS is dominated by its coupling to conduction electrons, described by the Hamiltonian[19]

$$H=H_0+\sigma_z\sum_{kk'\eta}V_{kk'}c^{\dagger}_{k\eta}c_{k'\eta}+H_e \ , \tag{2.2}$$

where $V_{kk'}$ describes the scattering potential, and $c^{\dagger}_{k\eta}$ creates a fermion of wave vector k, energy ξ_k, and spin η. The bath itself is described by H_e; for noninteracting fermions, $H_e=\sum_{k\eta}\xi_k c^{\dagger}_{k\eta}c_{k\eta}$. The coupling of the defect to the bath is characterized quite generally in terms of a parameter K which describes the coupling strength as well as the spectral density of bath excitations.[13] For an s-wave potential ($V_{kk'}=V$) and a bath of free fermions, K is determined solely by the scattering phase shift for electrons at the Fermi surface;[20] in the limit of weak scattering, $K=\frac{1}{2}(n_0V)^2$, where n_0 is the density of electron states at the Fermi level.[21] In this paper we do not consider the relaxation of the TLS by phonons, which we do not expect to affect the ultrasonic properties below 1 K.

In Sec. II A we review the standard TLS treatment of this problem and the procedure used to fit the data of Ref. 9, and show why the results are unsatisfactory. In the succeeding sections we will show that dissipative tunneling theory can be used to calculate linear response functions that yield a much more complete understanding of the experimentally observed acoustic properties.

A. Standard TLS theory

Here we review the standard TLS theory used in the previous fits and discuss the difficulties that arise when this theory is compared to experiment. To calculate the effects of the TLS on a sound wave of angular frequency ω,[3,4,22,23] one divides the attenuation into resonant and relaxational parts. The resonant attenuation arises from absorption by the TLS of energy splitting $E\approx\hbar\omega$. The relaxational attenuation comes from relaxation processes induced by the change in energy asymmetry caused by the sound wave. These two contributions are both calculated using the TLS theory. The two parameters that enter are K, the electron-TLS coupling constant, and $\bar{P}\gamma^2$, which is the product of the density of the TLS $\bar{P}$, and the square of the phonon-TLS coupling constant γ. Here we do not vary the distribution function of TLS $P(\epsilon,\Delta_0)$; it is assumed to be of the usual form[1]

$$P(\epsilon,\Delta_0)=\frac{\bar{P}}{\Delta_0} \ , \tag{2.3a}$$

or, equivalently,

$$P(E,r)=\frac{\bar{P}}{2r\sqrt{1-r}} \ , \tag{2.3b}$$

with $E=[(\hbar\Delta_0)^2+\epsilon^2]^{1/2}$ and $r=(\hbar\Delta_0/E)^2$. The resonant attenuation $\alpha_{\rm res}$ is given by[1]

$$\frac{\alpha_{\rm res}}{\omega}=\pi\left[\frac{\bar{P}\gamma^2}{\rho v^3}\right]\tanh\left[\frac{\beta\hbar\omega}{2}\right] , \tag{2.4}$$

where ρ is the mass density and v is the sound velocity. [For $Pd_{0.775}Si_{0.165}Cu_{0.06}$, $\rho = 10.52$ g/cm^3 and $v = 1.8 \times 10^5$ cm/sec (Ref. 24).]

The resonant contribution to the sound velocity is obtained by a Kramers-Kronig transform of the attenuation;[25] it has the form[23]

$$\frac{\Delta v_{\rm res}}{v} \equiv \frac{v(T) - v(T=0)}{v} = \frac{\bar{P}\gamma^2}{\rho v^2}\left[\Psi\left(\tfrac{1}{2} + \beta\hbar\omega\right) - \ln(\beta\hbar\omega)\right] , \quad (2.5)$$

where Ψ is the digamma function. At high temperatures where $\beta\hbar\omega \ll 1$,

$$\frac{\Delta v(T)}{v} \rightarrow \frac{\bar{P}\gamma^2}{\rho v^2} \ln\left[\frac{T}{T_0}\right] ,$$

where T_0 is a reference temperature.

The relaxational attenuation and velocity shift depend both on $\bar{P}\gamma^2$ and K; they are given by[9,22]

$$\frac{\alpha_{\rm relax}}{\omega} = \int_0^\infty dE \, \mathrm{sech}^2\left[\frac{\beta E}{2}\right] \times \int_0^1 dr \frac{\bar{P}\gamma^2}{\rho v^3} \frac{\sqrt{1-r}}{2r} \frac{\beta\omega T_1}{(\omega T_1)^2 + 1} , \quad (2.6a)$$

$$\frac{\Delta v_{\rm relax}}{v} = -\int_0^\infty dE \, \mathrm{sech}^2\left[\frac{\beta E}{2}\right] \times \int_0^1 dr \frac{\bar{P}\gamma^2}{\rho v^2} \frac{\sqrt{1-r}}{4r} \frac{\beta}{(\omega T_1)^2 + 1} . \quad (2.6b)$$

Here, T_1^{-1} is the relaxation rate of the TLS induced by the conduction electrons.

In the standard TLS picture, one assumes that the coupling to the conduction electrons is sufficiently weak that their effects can be accounted for using second-order time-dependent perturbation theory on the eigenstates of an isolated TLS. Thus, one takes the Hamiltonian (2.2), diagonalizes the first two terms, and then accounts for the third term using lowest- (second-) order perturbation theory. One obtains a relaxation rate T_1^{-1}:

$$T_1^{-1} = \left[\frac{\pi}{\hbar}\right] KE \coth\left[\frac{\beta E}{2}\right] . \quad (2.7)$$

The resonant contribution in this picture is expected to be independent of the relaxation rate T_1^{-1} and hence the TLS-electron coupling parameter K;[26] increasing T_1^{-1} lowers the height of the resonance peak but causes the number of TLS's contributing to the absorption to increase proportionality. [Of course, the *nonlinear* (saturation) properties of the attenuation do depend on T_1 and hence K.]

Now we compare this theory to experimental measurements of sound velocity and attenuation at fixed frequency $\omega/2\pi = 0.96$ GHz as a function of temperature for $0.01 < T < 1$ K.[9] We first consider the velocity measurements. The two parameters that enter, K and $\bar{P}\gamma^2$, have correlated uncertainties, so we fit the data to yield values for $\bar{P}\gamma^2$ for different values of K. We then compare the low-temperature attenuation calculated using these values of $\bar{P}\gamma^2$ and K to the measured attenuation. Since it is difficult to measure absolute attenuation, we assume that the attenuation is determined experimentally only up to an additive, temperature-independent constant.

We find that satisfactory fits to the velocity measurements over the temperature range 20 mK–1 K can be obtained for a broad range of K (~ 0.02–0.2), through for $K > 0.15$ or so the quality of the fit for $T < \hbar\omega/k_B$ degrades. Figure 1 shows fits for $K = 0.02$ and 0.2; fits for K between these two values are of higher quality than the ones shown. Table I shows the values of $C \equiv \bar{P}\gamma^2/\rho v^2$ extracted from these fits for different values of K. We estimate the uncertainty in the values of C extracted to be a few percent for a given value of K.

The description between theory and experiment arise when one attempts to fit the experimentally measured attenuation. One problem is that the observed intensity dependence of the low-temperature attenuation is qualitatively inconsistent with theoretical expectations.

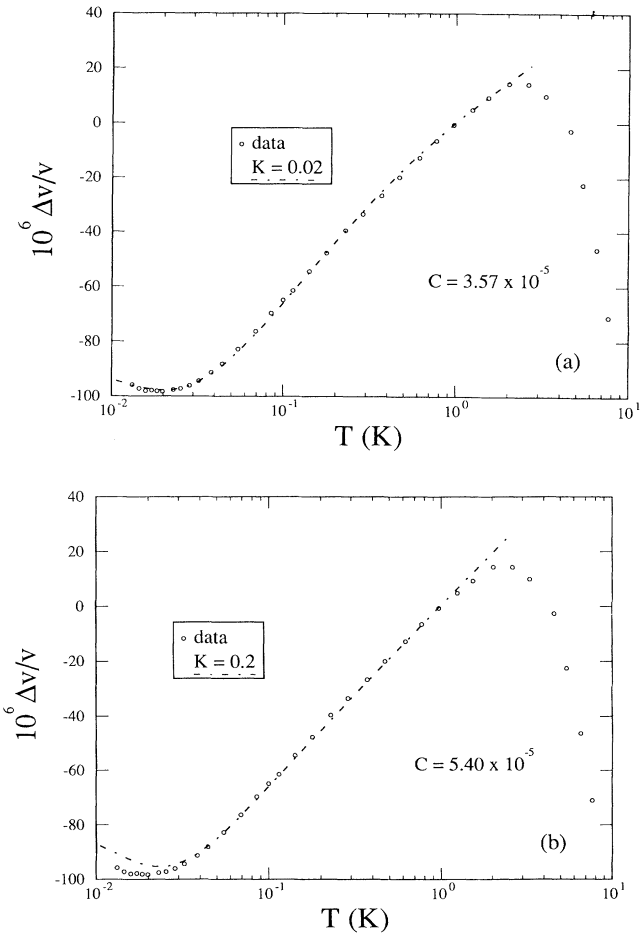

FIG. 1. Fit of standard TLS theory to data of Ref. 9; relative velocity shift $\Delta v/v$ vs temperature T (on a log scale). (a) Fit using $K = 0.02$; (b) fit using $K = 0.2$.

TABLE I. Table of values of $C \equiv \bar{P}\gamma^2/\rho v^2$ obtained for different values of K using the standard TLS calculation described by Golding *et al.* (Ref. 9) and Black (Ref. 18). The uncertainties in the values of C are a few percent.

K	$C \times 10^5$
0.02	3.57
0.03	3.77
0.04	3.92
0.10	4.74
0.15	5.15
0.18	5.33
0.20	5.40

By this we mean that theory predicts that there should be a saturable attenuation of magnitude $\pi C(\omega/v)\tanh(\beta\hbar\omega/2)$. The smallest value of C consistent with the velocity measurements occurs for small K; when one uses this value of C to calculate the resonant attenuation (which is independent of K), one obtains the result shown in Fig. 2(a). It is clear that the saturable attenuation measured experimentally is much smaller than the theoretical prediction. Increasing K only exacerbates this problem because it increases C. In addition, the temperature dependence of the saturable attenuation is not well described by the $\tanh(\beta\hbar\omega/2)$ function predicted by Eq. (2.4); the attenuation appears to be all unsaturable for $T > 0.08$ K, whereas $\tanh(\beta\hbar\omega/2) = 0.28$ at 0.08 K.

If we argue that C is indeed fixed by the low-temperature velocity and for some reason the resonant attenuation is not completely saturated, we can then adjust the zero of attenuation so that the low-temperature part fits the data, as is done in Fig. 2(a). However, if we do this, no matter what K we choose, the temperature dependence of the calculated total attenuation is qualitatively inconsistent with experiment because the theory predicts a relaxational contribution that is too small.

The problem can be illustrated simply by noting that the expression for the attenuation (2.6) using the expression (2.7) for T_1^{-1} is bounded above by

$$\frac{\alpha_{\text{relax}}}{\omega} \leq \frac{1}{2} C \left[\frac{\omega}{v} \right] \int_0^\infty d(\beta E) \operatorname{sech}^2 \left[\frac{\beta E}{2} \right] \times \int_0^1 dr \frac{\omega/g(E)}{r^2 + \omega^2/g^2(E)} , \tag{2.8}$$

where $g(E) \equiv (\pi/\hbar) KE \coth(\beta\hbar\omega)$, so that

$$\frac{\alpha_{\text{relax}}}{\omega} \leq \frac{\pi}{2} C \left[\frac{\omega}{v} \right] \quad \text{for all } K . \tag{2.9}$$

This bound holds also in the presence of a parallel relaxation of the TLS by phonons. Since the resonant attenuation as $T \to 0$ tends to $\pi C(\omega/v)$, this theory predicts that the relaxational attenuational should be substantially smaller than the resonant attenuation. The data are grossly inconsistent with this.

The comparison of standard TLS predictions for the attenuation with experiment is shown in Figs. 2 and 3. The data shown[9] are taken at different power levels; the low-intensity curve reflects the linear response of the system (resonant plus relaxational) whereas the high-intensity curve is expected to correspond to relaxational attenuation only. Figure 2 shows the division into resonant and relaxational parts for $K = 0.02$ and 0.2. In Fig. 3, we compare the theoretical prediction for the total linear attenuation for $K = 0.02$, 0.1, and 0.2 with the low-power attenuation measurements. (K was chosen to be between 0.02 and 0.2 on the basis of the velocity fits.) It is clear that neither the temperature dependence of the linear response nor the saturation behavior are well described by any of the theoretical curves. Thus, we have two main questions—why the saturable attenuation is smaller than the resonant contribution predicted by the TLS theory, and why the unsaturable attenuation is larger.

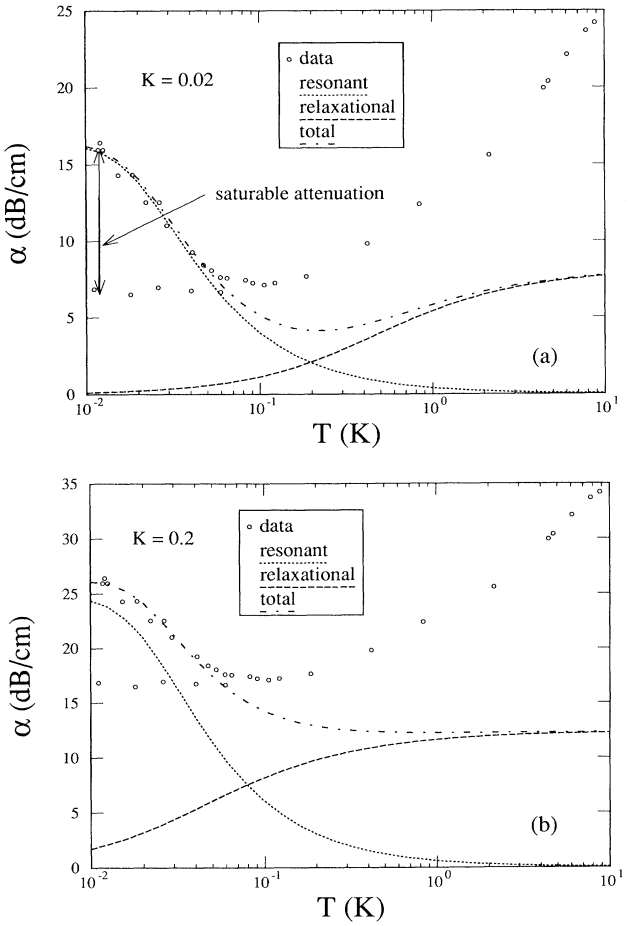

FIG. 2. Fit of standard TLS theory to data of Ref. 9: attenuation α vs temperature T. Parameters for the theoretical curves are obtained from the velocity fit. The two sets of data at low temperature correspond to low- and high-intensity sound; the difference is the saturable attenuation. Both resonant and relaxational attenuation are shown; resonant attenuation should be compared to the experimentally measured saturable attenuation. The experimental attenuation zero is unknown and has been adjusted so that the low-temperature attenuation agrees with theory. (a) $K = 0.02$, (b) $K = 0.2$. TLS theory clearly fails to explain the data.

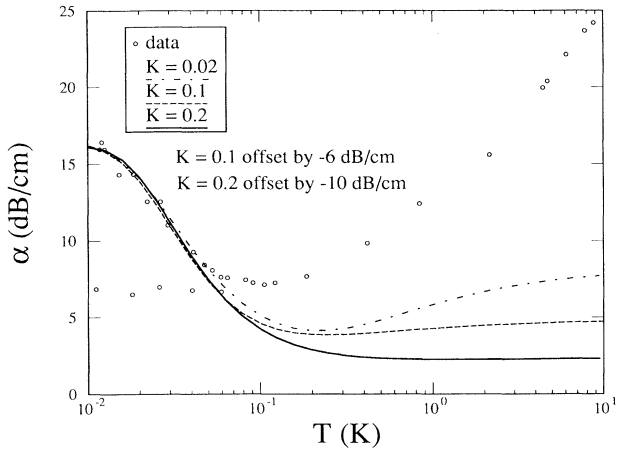

FIG. 3. Linear response attenuation α vs temperature T for $K=0.02$, 0.1, and 0.2 compared to low-intensity attenuation data of Ref. 9. Parameters for theoretical curves are obtained from the velocity fit. The $K=0.1$ curve has been offset by -6 dB/cm and the $K=0.2$ curve by -10 dB/cm. The experimental attenuation zero has been adjusted to agree with the $K=0.02$ curve at low temperature.

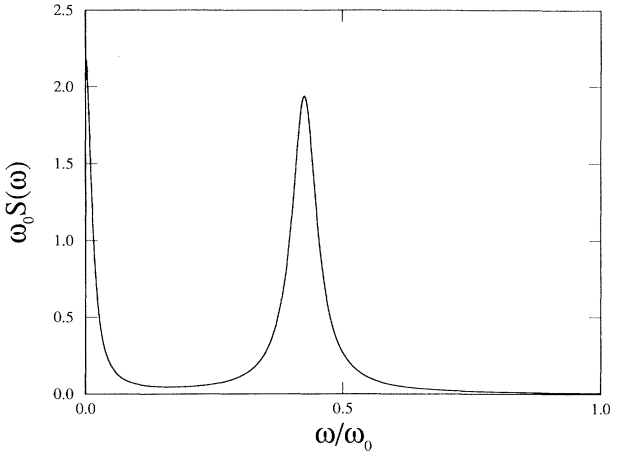

FIG. 4. Structure factor $S(\omega)$ of a single TLS vs frequency ω for parameter values $K=0.05$, $\epsilon=0.4$ K, $\hbar\Delta_r=0.2$ K, and $T=0.09$ K, from Gorlich *et al.* (Ref. 34). The reference frequency ω_0 corresponds to a temperature of 1 K. This structure factor is qualitatively similar to the result of standard TLS theory; it consists of Lorentzian peaks of width T_1^{-1} [from Eq. (2.7)] centered about $\omega=0$ and $E/\hbar$.

B. Linear response theory

We will now show that many of the puzzles raised in the previous section can be resolved if one accounts properly for the TLS-electron interaction. In this section we discuss how results obtained in the context of muon diffusion in metals[12] and quantum measurement theory[13] can be used to understand acoustic properties of metallic glasses.

It is useful to think in terms of linear response theory. For small intensities, the attenuation from a single TLS is determined solely by the phonon-TLS matrix element and the structure factor $S(\omega)$ of the TLS. These $S(\omega)$ are the Fourier transforms of the expectation values $\langle\sigma_z(0)\sigma_z(t)\rangle$, where $\sigma_z(t)$ is the z component of the pseudospin at time t. For an isolated TLS, $S(\omega)$ consists of two δ-function peaks at frequency $\Omega=\pm E/\hbar$, where $E=[(\hbar\Delta_0)^2+\epsilon^2]^{1/2}$. If the coupling to the electrons is weak, the finite frequency peaks develop a width T_1^{-1}, and in addition a Lorentzian central peak emerges, which also has width T_1^{-1}. This structure factor is shown in Fig. 4.

The attenuation $A(\omega)$ from a single TLS is simply related to the imaginary part of the response function $\chi''(\omega)$, which is related by the fluctuation-dissipation theorem to $S(\omega)$:

$$\frac{A(\omega)}{\omega}=\chi''(\omega)=\tanh\left[\frac{\beta\hbar\omega}{2}\right]S(\omega)\ . \tag{2.10}$$

The total attenuation $\alpha(\omega)$ is obtained by integrating over the distribution function of TLS.

The velocity change is obtained by a Kramers-Kronig transform of the attenuation:

$$\frac{\Delta v}{v}=\frac{1}{\pi v}\int_0^\infty d\omega'\frac{\alpha(\omega')}{\omega^2-\omega'^2}\ . \tag{2.11}$$

It is obvious that the resonant attenuation (2.4) is consistent with this formalism when $S(\omega)$ consists of a δ function; in addition it is straightforward to forward to show that in the limit $\beta\hbar\omega\ll 1$, the expressions (2.6a) and (2.6b) for the relaxational attenuation and velocity shift correspond to the linear response expressions (2.10) and (2.11) if $S(\omega)$ is a Lorentzian of width T_1^{-1} and amplitude $(\epsilon/E)^2\mathrm{sech}^2(\beta E/2)$.[23] For $\epsilon\approx E$ this Lorentzian structure factor describes a TLS which is incoherently tunneling between two states whose relative occupations are determined by detailed balance.[11] In addition, these linear response expressions are valid for those TLS's whose structure factor is qualitatively different from that shown in Fig. 4; we will see that sometimes $S(\omega)$ consists of a central peak plus power law edges, where the width of the central peak and the effective width of the finite frequency edges are not simply related. Such an $S(\omega)$ is shown in Fig. 5.

The experimentally relevant division of the attenuation is into saturable and nonsaturable attenuations. Although in insulators the saturable attenuation is resonant and the nonsaturable attenuation is relaxational, in metallic glasses the separation into resonant and relaxational pieces is misleading because nonsaturable attenuation also arises from processes that are formally resonant. We expect the only saturable attenuation to arise from a near-resonant coherently tunneling TLS, so that all other attenuations are nonsaturable.[27]

In the next section we discuss theoretical predictions for the structure factor and hence the linear response. There is substantial overlap between our discussion (which follows the presentation of Leggett *et al.*[13]) and that of Kagan and Prokofe'ev.[17] However, differences arise because our emphasis is on presenting the back-

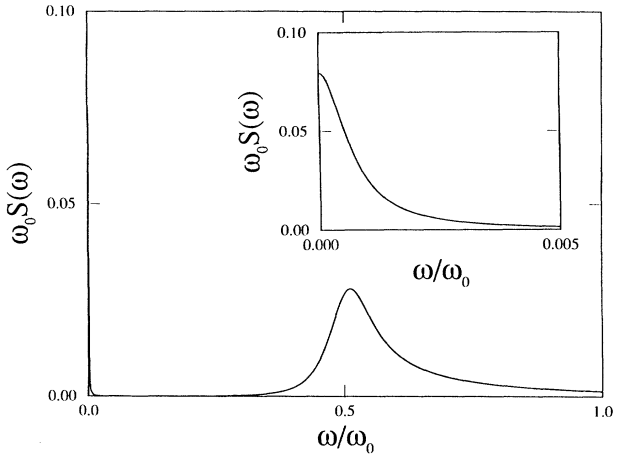

FIG. 5. Structure factor $S(\omega)$ vs frequency ω of a single TLS in the incoherent regime; parameter values are $T=0.042$ K, $K=0.2$, $\epsilon=0.5$ K, and $\hbar\Delta_r=0.01$ K. The reference frequency ω_0 corresponds to a temperature of 1 K. The structure factor consists of a Lorentzian central peak and a smeared power-law edge at frequencyy $\omega=\epsilon/\hbar$. The inset shows the Lorentzian central peak on an expanded scale. Note the width of the central peak and of the feature at $\omega\sim\epsilon/\hbar$ are substantially different.

ground needed to conduct a detailed comparison between the theory and the experimental data over a broad temperature range.

C. Dissipative tunneling

One obtains the expression of Golding *et al.*[9] for the relaxation rate (2.7) of a TLS by assuming that the coupling to the conduction electrons can be accounted for using lowest-order time-dependent perturbation theory. However, Kondo[12] has shown that the fourth-order term in this perturbation expansion depends logarithmically on the ratio of the electron bandwidth to the thermal energy. Therefore, the assumption that the fermionic bath is a weak perturbation is generally not valid at low temperatures.

The problem of the dynamics of a two-level system coupled to an ohmic heat bath has received substantial attention in the literature, and the basic phenomenology is well understood. If one performs a renormalization-group transformation successively integrating out fermionic degrees of freedom of high energy, then one can state the results for the TLS dynamics in terms of a renormalized tunneling matrix element Δ_r, where $\Delta_r=\Delta_0(\Delta_0/D)^{K/(1-K)}$ and D is the ultraviolet cutoff.[28] We shall only discuss $K<\frac{1}{2}$, which is the regime relevant to these experiments.[29] When k_BT is less than $\hbar\Delta_r/K$, then the standard result, Eq. (2.7), is qualitatively correct.[30] On the other hand, when $k_BT>\hbar\Delta_r/K$, then the interaction between the electron bath and the TLS is effectively very strong, so that the proper procedure is to diagonalize the terms involving σ_z in (2.1) and (2.2) and then to include the effects of the tunneling matrix element Δ_0 perturbatively. One finds an $S(\omega)$ that is qualitatively different from that obtained in Sec. II A. Thus, we will find it useful to divide the TLS into "coherent" and "incoherent" depending on whether $Kk_BT<\hbar\Delta_r$ or $Kk_BT>\hbar\Delta_r$, respectively.

Leggett *et al.*[13] review how one uses the functional integral technique to integrate out the bath and reduce the problem to that of the time evolution of the two-state system where transitions have an effective nonlocal (in time) interaction between them. They calculate the Laplace transform $\tilde{P}_\epsilon(\lambda)$ of the time evolution $P_\epsilon(t)$ of a TLS with asymmetry ϵ started at time $t=0$ with $\sigma_z(t=0)=1$, which is related to $S(\omega)$ by[31]

$$S(\omega)=\frac{2}{e^{\beta\epsilon}+1}[\tilde{P}_\epsilon(\lambda=i\omega)+e^{\beta\epsilon}\tilde{P}_{-\epsilon}(\lambda=i\omega)] . \tag{2.12}$$

The functional integral that yields the time evolution of the TLS is not solvable in general, so we use the noninteracting blip approximation of Leggett *et al.*, which can be viewed as a self-consistent Born approximation.[32] As Leggett *et al.* discuss, this approximation is well controlled in an extreme incoherent regime $\hbar\Delta_r\ll k_BT$ and for a symmetric TLS at $T=0$ in small-K limit. We want to apply the theory for the entire distribution of the TLS, so this approximation must be viewed as uncontrolled. However, we do expect these calculations to yield accurately the qualitative behavior of the TLS. The limitations of the noninteracting blip approximation are most serious for the coherent TLS and are discussed further in Sec. II C. Expressions for the linear response valid for an asymmetric TLS at low temperatures in the small-K limit have also been obtained,[33,34] which we discuss in Sec. II C 2. However, there are no calculations of the structure factor that are expected to be quantitatively accurate in the regime of moderate K and intermediate temperature.

Within the noninteracting blip approximation one obtains an expression for $\tilde{P}_\epsilon(\lambda=i\omega)$ [which is related to the structure factor $S(\omega)$ by Eq. (2.12)]:

$$\tilde{P}_\epsilon(\lambda=i\omega)=\frac{g_1+h_1+(h_1g_2-g_1h_2)/\omega}{g_1^2+(\omega+g_2)^2} , \tag{2.13}$$

where

$$g_1(\epsilon,\omega)=\Delta_r^{2(1-K)}\frac{1}{4}\left[\frac{2\pi}{\beta\hbar}\right]^{2K-1}\frac{1}{\Gamma(2K)} \times\left\{\left|\Gamma\left[K+i\frac{\beta}{2\pi}(\hbar\omega-\epsilon)\right]\right|^2\cosh\frac{\beta}{2}(\hbar\omega-\epsilon) +(\epsilon\rightarrow-\epsilon)\right\} , \tag{2.14a}$$

$$g_2(\epsilon,\omega)=-\Delta_r^{2(1-K)}\frac{1}{4}\left[\frac{2\pi}{\beta\hbar}\right]^{2K-1}\frac{\cot(\pi K)}{\Gamma(2K)} \times\left\{\left|\Gamma\left[K+i\frac{\beta}{2\pi}(\hbar\omega-\epsilon)\right]\right|^2\sinh\frac{\beta}{2}(\hbar\omega-\epsilon) +(\epsilon\rightarrow-\epsilon)\right\} , \tag{2.14b}$$

$$h_1(\epsilon,\omega)=\Delta_r^{2(1-K)}\frac{1}{4}\left[\frac{2\pi}{\beta\hbar}\right]^{2K-1}\frac{1}{\Gamma(2K)}\times\left\{-\left|\Gamma\left[K+i\frac{\beta}{2\pi}(\hbar\omega-\epsilon)\right]\right|^2\times\sinh\frac{\beta}{2}(\hbar\omega-\epsilon)-(\epsilon\rightarrow-\epsilon)\right\}, \quad (2.14c)$$

$$h_2(\epsilon,\omega)=\Delta_r^{2(1-K)}\frac{1}{4}\left[\frac{2\pi}{\beta\hbar}\right]^{2K-1}\frac{\tan(\pi K)}{\Gamma(2K)}\times\left\{-\left|\Gamma\left[K+i\frac{\beta}{2\pi}(\hbar\omega-\epsilon)\right]\right|^2\times\cosh\frac{\beta}{2}(\hbar\omega-\epsilon)-(\epsilon\rightarrow-\epsilon)\right\}. \quad (2.14d)$$

Here, $\Gamma(z)$ is the complex γ function.

In practice, we will only use this expression in situations where either only the lowest-order terms in Δ_r need be considered or else $\epsilon=0$; in these situations $\tilde{P}$ reduces to

$$\tilde{P}(\lambda=i\omega)=\frac{g_1(\epsilon,\omega)+h_1(\epsilon,\omega)}{g_1^2(\epsilon,\omega)+(\omega+g_2)^2}. \quad (2.15)$$

Physical quantities are determined by the integral of the response over the distribution function of asymmetries and tunneling matrix elements. Whether one uses Δ_r or Δ_0 only affects a constant which can be absorbed into $\bar{P}$; We use Δ_r since it is the observable low-energy quantity. The attenuation is given by

$$\frac{\alpha(\omega)}{\omega}=\frac{C}{v}\tanh\left[\frac{\beta\hbar\omega}{2}\right]\int_0^\infty d\epsilon\int_0^\infty\frac{d\Delta_r}{\Delta_r}S(\omega,\epsilon,\Delta_r), \quad (2.16)$$

and the velocity change is given by the Kramers-Kronig transform of the attenuation, Eq. (2.11). In the next subsection we describe the results of the noninteracting blip approximation for the regimes of incoherent tunneling.

1. *Incoherent regime*

We first discuss the regime of incoherent tunneling, $\hbar\Delta_r<Kk_BT$. When this condition is satisfied, the energy eigenstates of the TLS are completely destroyed and one should view the system as incoherently tunneling between position eigenstates.

In this regime the structure factor has the form shown in Fig. 5; in addition to a Lorentzian central peak there is a feature at frequency $\epsilon/\hbar$, which reflects the possibility that transitions from the lower-energy well to the higher-energy one can be induced. The central peak is calculated by Grabert and Weiss[35] and Fisher and Dorsey;[36] the edge is calculated for $\Delta_r\ll\omega$ by Chakravarty and Kivelson.[37] Both contributions are contained in Eqs. (2.14), which are equivalent to the results of Dattagupta *et al.*[38]

At zero temperature the structure factor consists solely of power-law edges with threshold $\hbar\omega=\epsilon$. The absence of response for frequencies less than this value is reasonable because one needs to conserve energy when inducing transitions from a lower-energy to a higher-energy state. At nonzero temperature the edge broadens and, in addition, a Lorentzian central peak emerges.

By using (2.12)–(2.14), we have calculated the incoherent contribution to the quantity

$$\int_0^\infty d\epsilon\int_0^{K/\beta\hbar}\frac{d\Delta_r}{\Delta_r}\tanh\left[\frac{\beta\hbar\omega}{2}\right]S_{\epsilon,\Delta_r}(\omega) \quad (2.17)$$

for several values of K and $\omega/2\pi=0.96$ GHz. We have verified that the integral of the full structure factor over the TLS distribution function is the same as the one over just the central peak to within a few percent. Therefore, it is simpler to consider the central peak only and calculate this contribution to the attenuation using the analog of (2.6) [where the absence of a factor of $(\epsilon/E)^2$ can be attributed to the destruction of the energy eigenstates]:

$$\frac{\alpha_{\rm rel}}{\omega}=\frac{C}{v}\int_0^\infty d\epsilon\,\mathrm{sech}^2\left[\frac{\beta\epsilon}{2}\right]\times\int_0^{K/\beta\hbar}\frac{d\Delta_r}{\Delta_r}\frac{\beta\omega T_1}{(\omega T_1)^2+1}, \quad (2.18a)$$

$$\frac{\delta v_{\rm relax}}{v}=-\frac{C}{2}\int_0^\infty d\epsilon\,\mathrm{sech}^2\left[\frac{\beta\epsilon}{2}\right]\times\int_0^{K/\beta\hbar}\frac{d\Delta_r}{\Delta_r}\frac{\beta}{(\omega T_1)^2+1}, \quad (2.18b)$$

using the dissipative tunneling result for the relaxation rate T_1^{-1}:

$$T_1^{-1}=\tfrac{1}{2}\Delta_r^{2(1-K)}\left[\frac{2\pi}{\beta\hbar}\right]^{2K-1}\times\frac{\cosh(\beta\epsilon/2)}{\Gamma(2K)}\left|\Gamma\left[K+i\frac{\beta\epsilon}{2\pi}\right]\right|^2. \quad (2.19)$$

This result for the relaxation rate differs qualitatively from the standard result Eq. (2.7); as Fig. 6 demonstrates, for temperatures T obeying $Kk_BT>\epsilon$, T_1^{-1} increases as the temperature is decreased as $T_1^{-1}\propto T^{-(1-2K)}$. The temperature where the relaxation rate is largest occurs at $Kk_BT\sim0.2\epsilon$.

At high temperatures arguments similar to those used to derive Eq. (2.9) can be used to show that this contribution to the attenuation saturates at a value $\pi C(\omega/v)/[2(1-K)]$. We expect the incoherent contribution to the velocity to be also dominated by the central peak, whose contribution is calculated using (2.18). We have verified that the contribution from the edge is negligible when $\beta\hbar\omega\ll1$; when $\beta\hbar\omega\gg1$ we expect the coherently tunneling TLS to dominate the acoustic properties. Thus, the effects of the edge on both the attenuation and velocity can be ignored at all temperatures.

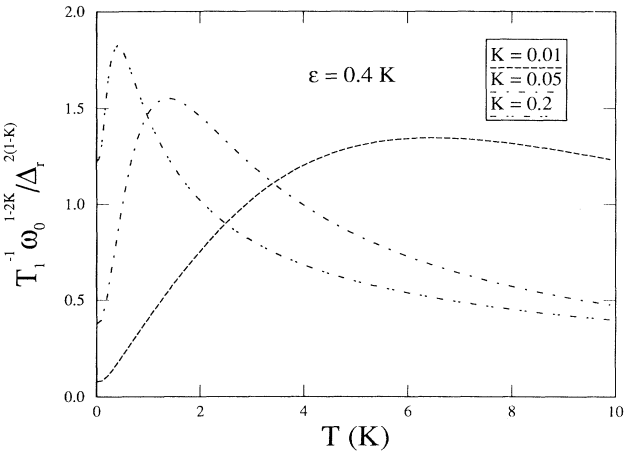

FIG. 6. Scaled relaxation rate $T_1^{-1}/\Delta_r^{2(1-K)}$ of a single TLS in the incoherent regime as a function of temperature T for $\epsilon=0.4$ K, and $K=0.01$, 0.05, and 0.2. The reference frequency ω_0 corresponds to a temperature of 1 K. For $T \gtrsim 0.2\epsilon/Kk_B$, T_1^{-1} increases as the temperature is decreased as $T_1^{-1} \propto T^{-(1-2K)}$; this result differs qualitatively from the standard TLS theory prediction.

2. *Coherent regime*

We now consider the attenuation induced by the TLS's which satisfy the condition $\hbar\Delta_r > Kk_BT$. Although to a large extent standard TLS theory can be applied in this regime, the attenuation from the coherent TLS is different from that calculated by Golding *et al.*[9] and Black.[18] The main differences arise from contributions to the nonsaturable attenuation that arise from formally resonant processes. There are two main reasons for this. The first is a contribution that arises even in the small-K limit because the width of the resonance is proportional to its frequency; the second is that when K becomes greater than 0.1 or so the structure factor becomes extremely temperature dependent; the resonant finite-frequency peaks become indistinct at quite low temperatures.

(a) Small K. We first discuss the limit of small K, where Gorlich *et al.*[34] and Weiss and Wollensak[33] have calculated the structure factor of a TLS of energy splitting E. Within a Lorentzian approximation for the line shapes, the structure factor has the form

$$S(\omega)=\frac{a}{2}\frac{\Gamma}{(\Omega-\omega)^2+\Gamma^2}+\frac{a}{2}\frac{\Gamma}{(\Omega+\omega)^2+\Gamma^2}+\frac{\epsilon^2/\Omega^2}{\cosh^2(\hbar\beta\Omega/2)}\frac{\Gamma_r}{\omega^2+\Gamma_r^2}\ , \tag{2.20}$$

where $a=1-\epsilon^2/\Omega^2$, $\Gamma_r=\pi K\hbar(\Delta_r^2/E)\coth(\beta E/2)$, $\Gamma=\Gamma_r/2+K\pi(\epsilon/E)^2/(\beta\hbar)$, and

$$\Omega^2=\epsilon^2+\Delta_r^2[1-2\,\mathrm{Re}\{\psi(1+i\beta E/2\pi\hbar)\}+\ln(2\pi/\beta\hbar\Delta_r)]\ ,$$

with ψ the digamma function. This form is valid only at low temperatures; when $E \lesssim Kk_BT$, this solution must break down because it predicts that Ω becomes negative.

In the limit $E \ll k_BT$, this structure factor corresponds exactly to that expected within the "old" TLS theory described in Sec. II A. When $k_BT \gtrsim E$, there are nontrivial corrections to the "resonant" part of $S(\omega)$, but there are two reasons why these changes do not affect the attenuation significantly. The main difference between Eq. (2.20) and the theory described in Sec. II A lies in the difference between Ω and E, which affects the "matrix element" a. However, a is unity when $\epsilon=0$ at all temperatures, and the symmetric TLS's dominate the resonant attenuation. In addition, the contribution of the TLS to the attenuation when $E \lesssim k_BT$ is small because of the $\tanh(\beta\hbar\omega/2)$ factor. Thus, we expect standard TLS theory to be adequate to calculate the attenuation contribution for the TLS with $\Delta_r > Kk_BT$.

Since the relaxational part is suppressed by a factor of $\mathrm{sech}^2(\beta\hbar\omega/2)$, at the lowest temperatures only the resonant attenuation is observed (e.g., only the finite-frequency peaks in the structure factor need be considered). However, here we show that even in the limit $T\to 0$ there is a substantial *unsaturable attenuation*, which arises because the width of the resonance peak is proportional to the resonance frequency.

First consider the case $T=0$. The averaged structure factor $\bar{S}(\omega)$ is obtained by averaging $S(\omega)$ for the single TLS over the distributions for Δ_r and ϵ. Equation (2.20) yields the scattering rate of a TLS of energy E of $r\eta E/\hbar$, where $r=(\hbar\Delta_r/E)^2$ and $\eta=\pi K/2$. The contribution to $\bar{S}(\omega)$ from the resonance at $\omega=E/\hbar$ is thus found to be

$$\bar{S}_+(\omega)=\int_0^1\frac{\bar{P}\,dr}{2(1-r)^{1/2}}\times\int_0^{E_{\max}}dE\frac{r\eta E/\hbar}{(E/\hbar-\omega)^2+(r\eta E/\hbar)^2}\ . \tag{2.21}$$

The integral over the energy E diverges logarithmically as the upper cutoff $E_{\max}$ tends to infinity. In the Appendix we show that this divergence does not depend on the assumption of a Lorentzian line shape. However, it is crucial that the width of the line be proportional to its peak frequency; the relevant integral $\int dE\{E^x/[(E-E_0)^2+E^{2x}]\}$ diverges only for $x=1$.

To proceed further, it is simplest to rewrite Eq. (2.21) as the sum of two terms $\bar{S}_{+\mathrm{res}}$ and $\bar{S}_{+\mathrm{anom}}$, where

$$\bar{S}_{+\mathrm{res}}(\omega)=\int_0^1\frac{\bar{P}\,dr}{2(1-r)^{1/2}}\frac{\eta\omega r}{1+\eta^2r^2}\times\int_0^{E_{\max}}dE\frac{1}{(E/\hbar-\omega)^2+(r\eta E/\hbar)^2} \tag{2.22}$$

and

$$\bar{S}_{+\mathrm{anom}}(\omega)=\int_0^1 \frac{\bar{P}\,dr}{4(1-r)^{1/2}}\frac{\hbar\eta r}{(1+\eta^2 r^{2)}}\int_0^{E_{\max}} dE\,\frac{d/dE[(E/\hbar-\omega)^2+(r\eta E/\hbar)^2]}{(E/\hbar-\omega)^2+(r\eta E/\hbar)^2}\ . \tag{2.23}$$

The first term, $\bar{S}_{+\mathrm{res}}$, corresponds to the usual resonant contribution to the attenuation, for $\eta r \ll 1$ the limits of integration over E can be set to $\pm\infty$, yielding

$$\bar{S}_{+\mathrm{res}}(\omega)=\int_0^1 \frac{dr\,\bar{P}}{2(1-r)^{1/2}}\frac{\hbar\pi}{1+\eta^2 r^2}\ . \tag{2.24}$$

The second term $\bar{S}_{+\mathrm{anom}}$, which comes from the TLS far from resonance, is

$$\bar{S}_{+\mathrm{anom}}(\omega)=\int_0^1 \frac{dr\,\bar{P}}{2(1-r)^{1/2}}\frac{\hbar\eta r}{1+\eta^2 r^2}\times\ln\left[\frac{E_{\max}}{\hbar\omega}\sqrt{1+r^2\eta^2}\right], \tag{2.25}$$

where we have assumed $E_{\max}/\hbar\omega \gg 1$. The resonance at $\omega=-E/\hbar$ contributes an equal amount to the anomalous piece; it does not affect the resonant piece.

If one chooses $E_{\max}=300$ K, for sound of frequency 0.96 GHz, in the limit of small K, $\bar{S}_{\mathrm{anom}}/\bar{S}_{\mathrm{res}}\sim 5.85$ K. The K dependence that arises from the integration over r is small; for $K=0.1$ one finds $\bar{S}_{\mathrm{anom}}/\bar{S}_{\mathrm{res}}\approx 0.58$ and for $K=0.3$, $\bar{S}_{\mathrm{anom}}/\bar{S}_{\mathrm{res}}\approx 1.7$. However, as we discuss in the next section, when K becomes large the Lorentzian approximation for the line shape breaks down and this calculation can be viewed only as a rough estimate.

Since the TLS that contribute to the anomalous attenuation are all far from resonance, the anomalous contribution should be nonsaturable. The temperature dependence is roughly the same $\alpha\propto\tanh(\beta\hbar\omega/2)$ displayed by the resonant attenuation; the contribution of the TLS with $E<k_BT$ is suppressed because the $\coth(\beta E/2)$ factor in the scattering rate causes the width of the peak to be no longer proportional to E, but in this regime the $\tanh(\beta\hbar\omega/2)$ factor is already suppressing the attenuation. The relaxational attenuation from the coherently tunneling TLS is seen from Eq. (2.20) to be well described by the theory of Sec. II A when K is small.

(b) Large K. When K is large, the structure factor is no longer well described by Eq. (2.20). When $\epsilon=0$ the noninteracting blip approximation (NIBA) appears to give a reasonable description; in addition to working for small K, it is exact for $K=\frac{1}{2}$.[13] However, when $\epsilon\neq 0$ and $k_BT<E$, for some frequencies the noninteracting blip approximation yields a negative structure factor, which is clearly an unphysical result. This problem has been pointed out by Weiss and Wollensak[33] and Gorlich *et al.*,[34] who have derived forms for the structure factor which do not have this unphysical feature. However, their calculations are not expected to be quantitatively accurate in the regime of intermediate K.

Although there is no quantitatively accurate calculation of the structure factor for intermediate K, qualitative insight can be obtained by investigating the properties of the NIBA. It is reasonable to do this because the main qualitative changes occur in the resonant attenuation, which is dominated by the TLS's which have $\epsilon\sim 0$.

The structure factor $S(\omega)$ for a symmetric TLS within the NIBA has been calculated by Grabert *et al.*[39] We have calculated the evolution of the structure factor $S(\omega)$ within the NIBA as the temperature is increased for a symmetric TLS with $\Delta_r/2\pi=0.96$ GHz. Figure 7 shows $S(\omega)$ obtained within NIBA at five different temperatures, for $K=0.2$ and 0.05. Recall that in the limit of small K, there is no central peak in the structure factor for a TLS with $\epsilon=0$ at any temperature. However, for $K=0.2$ [Fig. (7a)] the resonant line shape of the structure factor washes out rapidly as the temperature is

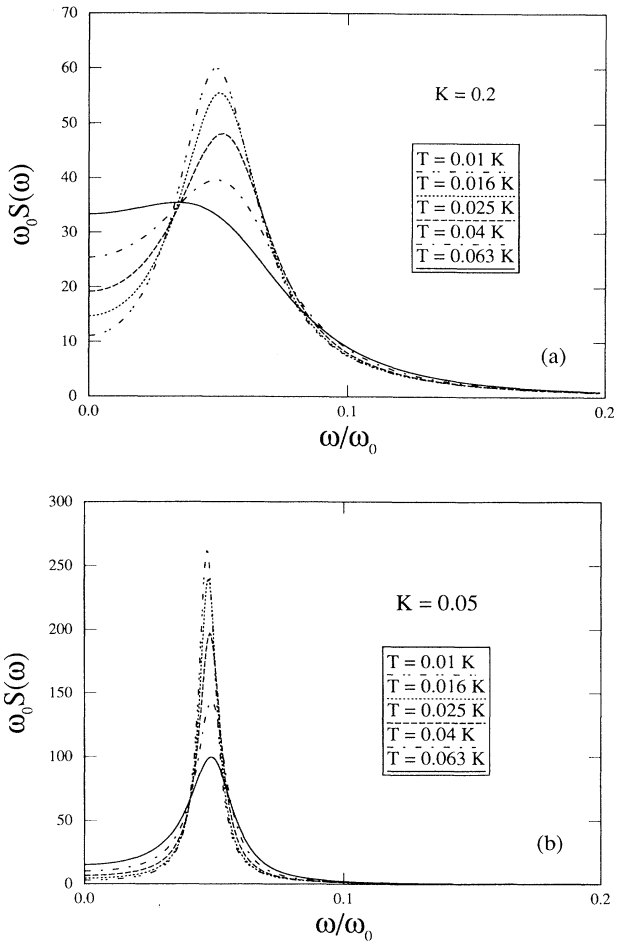

FIG. 7. Structure factor $S(\omega)$ calculated using NIBA as a function of frequency ω at five different temperatures T for parameter values $\epsilon=0$ and $\Delta_r/2\pi=0.96$ GHz. The reference frequency ω_0 corresponds to a temperature of 1 K. (a) $K=0.2$, (b) $K=0.05$. For large K one finds that small changes in temperature lead to large qualitative changes in the structure factor. This effect is significantly less marked when K is small.

raised. If we denote the peak value of $S(\omega)$ as $S_{\max}$, we find that the ratio $S(\omega=0)/S_{\max}$ increases from ~ 0.18 at $T=0.01$ K to ~ 0.94 at $T=0.063$ K. These changes are not simply a change in the linewidth because $S(\omega)$ is hardly affected as the temperature is raised from 0.01 to 0.063 K for $\omega > 1.5\Delta_r$. Figure 7(b) shows the evolution of $S(\omega)$ when $K=0.05$; it demonstrates that this effect is substantial only when K is reasonably large.

We expect that the disappearance of the finite-frequency resonance in the structure factor corresponds to the disappearance of saturable attenuation. Therefore, we expect the saturable attenuation to decrease much more quickly than usual TLS theory predicts as the temperature is raised. Unfortunately, the saturation behavior is not simply related to the linear response, so detailed comparison to the experimentally observed saturation behavior is not possible.

In addition to the attenuation from the TLS with $\Delta_r \sim \omega$, the TLS's far from resonance will contribute to the "anomalous" attenuation, which is also unsaturable. This contribution is discussed in the preceding subsection. However, when K is large, that calculation yields only an estimate for the structure factor; first, the approximation that the structure factor is a sum of Lorentzians becomes worse, and second, K dependence arises from the fact that extending the limits of integration to $\pm\infty$ becomes a worse approximation as K increases.

III. COMPARISON TO EXPERIMENT

In this section we compare the theory to experimental measurements of the acoustic properties of $Pd_{0.775}Si_{0.165}Cu_{0.06}$.[9] The method used here is completely analogous to that described in Sec. II A; we first fit the velocity curves to obtain values of C for various K, and then use these values to calculate the attenuation.

Comparison between theory and experiment is complicated by the fact that theoretically we would like to calculate the attenuation by integrating the structure factor over the TLS distribution function. However, use of the rather complicated functional forms for $S(\omega)$ discussed in the previous sections is computationally prohibitive because detailed fitting requires calculation of the velocity shift, which involves a triple integral (over Δ_r, ϵ, and frequency ω'); in any case we do not expect any structure factors obtained to date to be quantitatively accurate in the regime of intermediate temperature and K. Therefore, we employ approximations that enable us to calculate both the velocity and attenuation with no more than double integrals; these approximations are all well controlled in the small-K limit.

Three contributions to the attenuation need to be calculated. They can be viewed as (1) the "resonant" contribution (attenuation from finite-frequency peaks in the structure factors of the coherently tunneling TLS), (2) a relaxational contribution from the coherently tunneling TLS, and (3) a relaxational contribution from the incoherently tunneling TLS.

We first discuss the "resonant" contribution (which we again stress is partly unsaturable). To estimate this contribution, we use the fact that the resonant attenuation is dominated by the symmetric ($\epsilon=0$) TLS and estimate

$$\alpha_{\rm res} = C\left[\frac{\omega}{v}\right]\int_{K/\beta}^{E_{\max}} dE \tanh\left[\frac{\beta\hbar\omega}{2}\right] S(E, r=1, \omega) ,$$

where the structure factor is calculated using NIBA and we have used $E_{\max}=200$ K. This procedure is exact as $K\rightarrow 0$ and automatically includes the anomalous piece of Sec. II C 2. It does not include any relaxational component because only symmetric TLS's are considered.

Figure 8 shows $\alpha_{\rm res}/(C\omega/v)$ obtained in this fashion as a function of temperature for $\omega/2\pi=0.96$ GHz and several different values of K. If C is kept fixed, $\alpha_{\rm res}(T=0.01\ {\rm K})$ changes by less than 10% as K is increased up to 0.15, but then increases rapidly as K is increased further.

The relaxational attenuation is calculated in two parts, depending on the relative sizes of $\hbar\Delta_r$ and Kk_BT. The coherent relaxational attenuation comes from the asymmetric TLS's which satisfy $\hbar\Delta_r > Kk_BT$. The NIBA yields a negative $S(\omega)$ in this parameter regime and hence cannot be used to calculate this contribution. We calculate this piece using (2.6), but restricting the integral over the distribution function to the coherently tunneling TLS with $\hbar\Delta_r > Kk_BT$. We believe this is reasonable for the reasons outlined in Sec. II C 2. The contribution of the incoherently tunneling TLS is calculated using (2.18) and (2.19), restricting the integral to the incoherent regime $\hbar\Delta_r < Kk_BT$. The expressions for the relaxation rates in the two regimes match smoothly in the $K\rightarrow 0$ limit.

The total attenuation is obtained by adding together these three contributions. Figure 9 shows $\alpha/(C\omega/v)$ for different values of K; as K is increased both the low-temperature and high-temperature asymptotes increase. The increase of the high-temperature asymptote is expected because $\alpha/(C\omega/v)$ tends to $\pi/[2(1-K)]$ at high temperatures. The shape of the attenuation versus tem-

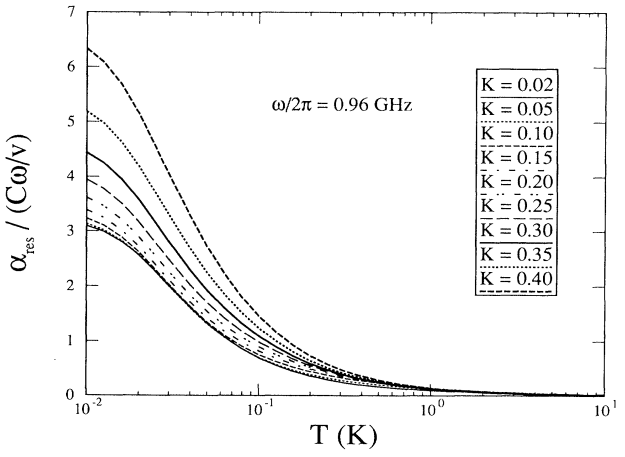

FIG. 8. Scaled resonant attenuation $\alpha_{\rm res}/(C\omega/v)$, estimated as described in the test, vs temperature T for $\omega/2\pi=0.96$ GHz and several values of K. For small K increasing K changes $\alpha_{\rm res}$ only slightly; when K is large $\alpha_{\rm res}$ increases substantially when K is increased.

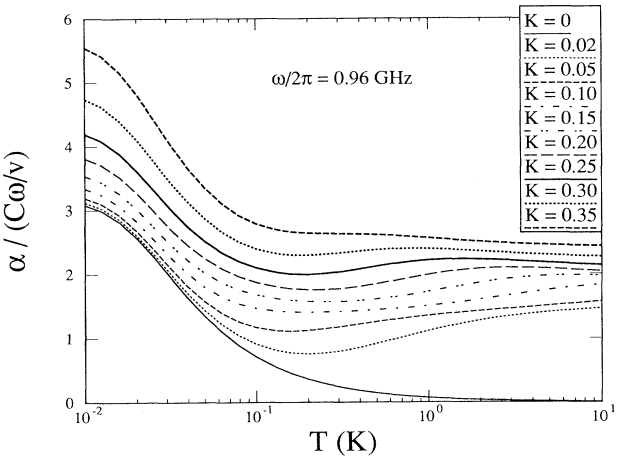

FIG. 9. Scaled total attenuation $\alpha/C(\omega/v)$ vs temperature T for different values of K; in contrast to the results of the theory in Sec. II A, there is a minimum in the attenuation as a function of temperature up to K of ~ 0.25.

perature curve remains roughly the same for K between 0.02 and 0.25; above 0.25 the minimum in the attenuation washes out, and for $K=0.35$ the total attenuation looks very similar to the resonant part only (e.g., $K=0$), but with a temperature-independent offset.

A. Velocity

The velocity shift is obtained by a Kramers-Kronig transform of the attenuation. Analytic information is available about the dependence of the velocity on the temperature for small K in the limit of high temperatures ($\beta\hbar\omega \ll 1$). In this regime $\Delta v/v$ is proportional to $\ln(T)$ because the response function $\alpha(\omega)/\omega$ is independent of frequency for both regimes $\beta\hbar\omega \gg 1$ and $\beta\hbar\omega \ll 1$. The coefficient of the logarithm is the difference in $\alpha/\pi\omega$ for $\beta\hbar\omega \gg 1$ and $\beta\hbar\omega \ll 1$. Thus, at high enough temperatures, for small K

$$\Delta v/v = C[1-1/(2-2K)]\ln(T/T_0) ,$$

where T_0 is a reference temperature. This temperature dependence is different than for phonon relaxation; for phonons, the high-temperature limit of $\pi/2$ occurs for frequency $\propto T^3$, leading to a slope of $C(1-3/2) = -C/2$.

When K is larger, α/ω is no longer independent of frequency for $\beta\hbar\omega \gg 1$; for $K=0.35$, $\alpha_{\rm res}/\omega$ drops by about a factor of 2 between $\omega = 1$ and 100 K. Therefore, one no longer expects $\Delta v/v$ to depend linearly on $\log T$.

The experiment reported in Ref. 9 measures attenuation and velocity over a broad range of temperatures, including the crossover region where asymptotic forms are inadequate and numerical fitting is necessary. Therefore, we have calculated the velocity shift numerically. Once again we calculate separately the contributions of the symmetric coherently tunneling TLS, the asymmetric coherent TLS, and the incoherently tunneling TLS. Since Fig. 8 demonstrates that the total resonant attenuation is only weakly K dependent for $K < \sim 0.15$, we have approximated the resonant contribution using the usual form Eq. (2.5).[40] For larger K the velocity can be obtained by numerically performing a Kramers-Kronig transform on the attenuation. The two parts of the relaxational contribution are calculated with the same approximations used for the attenuation.

First we look at the velocity and find, for different values of K, the values of C that fit the velocity curves. We find that satisfactory fits can be obtained for K in the range 0.02–0.15; the corresponding values of C are shown in Table II. For K's in this range the quality of the fit of the velocity is very similar to that of the "old" theory, shown in Sec. II A. Figure 10 shows the comparison of the two calculations for $K=0.15$.

There are several reasons why we believe K cannot be too large. First, the qualitative discrepancy between theory and experiment for the attenuation becomes worse as K is increased above 0.25. Second, when K exceeds 0.20, the relaxational velocity curve develops a marked kink below 1 K as the slope crosses over to its asymptotic high-temperature value; this kink leads to marked disagreement between theory and experiment for very large K. Figure 11 shows the comparison between the measured velocity shift and theory for $K=0.22$; there is qualitative disagreement between the theoretical curve and the data for $T \lesssim 0.02$ K as well as for $T \gtrsim 0.5$ K. In this curve we have used Eq. (2.5) to calculate the resonant velocity shift; accounting more accurately for the resonant contribution will not help this situation because the resonant contribution varies smoothly with temperature and does not affect the presence of the kink. We now proceed to examine the attenuation data using the values of C obtained from the velocity fits in the regime $K \leq 0.15$.

B. Total attenuation

When $K=0.02$ the theory described above yields attenuation that is indistinguishable from that predicted by standard TLS theory. Substantial differences in the fits occur only when K becomes large. The velocity fits appear to imply that K cannot exceed 0.2; we display results for $K=0.15$. Figure 12 shows the comparison of the predictions of the two theories with $K=0.15$ for the total attenuation as a function of temperature. Accounting for

TABLE II. Table of values of C obtained for different values of K using the dissipative tunneling theory described in the text, with uncertainties of a few percent.

K	$C\times 10^5$
0.02	3.69
0.03	3.61
0.04	3.60
0.06	3.81
0.08	4.40
0.10	4.55
0.12	4.70
0.14	4.84
0.15	4.88
0.16	5.04
0.18	5.25

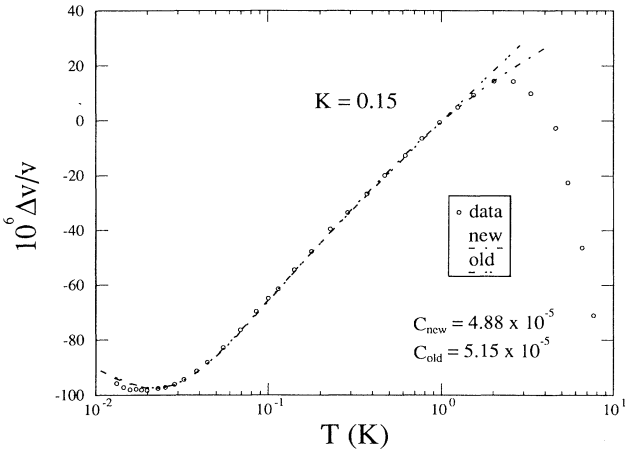

FIG. 10. Relative velocity shift $\Delta v/v$ vs temperature T for "old" and "new" calculations, for $K=0.15$. These fits are used to extract values of C for the attenuation fits shown below.

incoherent tunneling increases the relaxational attenuation relative to the resonant; qualitatively, the shape of curve is improved because the attenuation has a minimum as a function of temperature. The relaxational attenuation is larger than in standard TLS theory for two reasons. The first is that at high temperature $\alpha/(C\omega/v)$ tends to $\pi/2(1-K)$ rather than π. The second is that because the relaxation rate has a more complicated temperature dependence than previously thought, it is possible for both the coherent and incoherent TLS's to contribute substantially to the relaxational attenuation at intermediate temperature. The three separate contributions to the attenuation are shown in Fig. 13 for $K=0.15$; it is clear that crossover between incoherent and coherent tunneling increases the amount of relaxational attenuation.

Although the trend of the changes that appear in the attenuation when dissipative tunneling is taken into account is in the right direction, agreement between theory and experiment is still far from perfect. We do not expect our calculation to be perfectly quantitative, but it is of concern that the calculated relaxational attenuation is so much smaller than in the experiment. However, there is one major improvement in the comparison between theory and experiment: we no longer worry about the fact that in order to have the attenuation be consistent

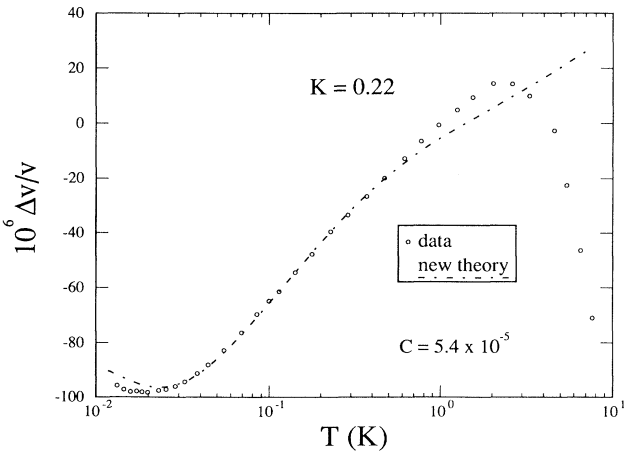

FIG. 11. Relative velocity shift $\Delta v/v$ vs temperature T calculated using dissipative tunneling theory with $K=0.02$. There is a kink in the curve that does not occur in the experimental data for $Pd_{0.775}Si_{0.165}Cu_{0.06}$; thus, we believe this value of K is too large to fit these data.

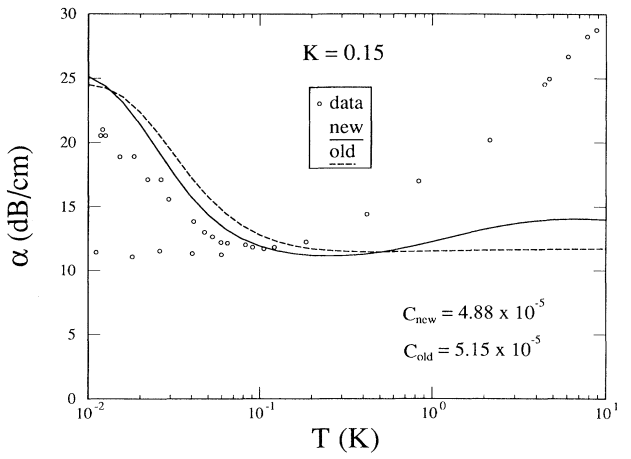

FIG. 12. Comparison of the total linear attenuation α as a function of temperature T for $K=0.15$ for the two calculations described in Sec. II, where the parameter C is determined by fitting the velocity (Fig. 11). Accounting for dissipative tunneling increases the amount of high-temperature ($T \gtrsim 0.02$ K) attenuation relative to the amount at low temperatures ($T < 0.05$ K) and improves the agreement between TLS theory and the measurements. We have assumed that the attenuation zero in the data is not known and have adjusted it arbitrarily.

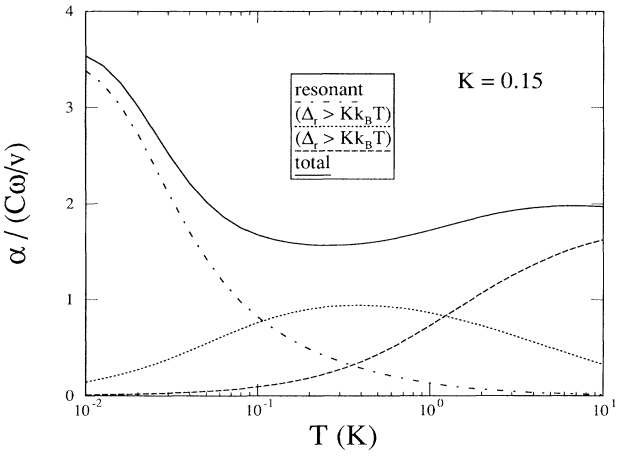

FIG. 13. Normalized attenuation $\alpha/(C\omega/v)$ vs temperature T for $K=0.15$ predicted by dissipative tunneling theory. The three contributions of the resonant attenuation, the relaxational contribution of a coherently tunneling TLS with $\hbar\Delta_r > Kk_BT$, and the relaxational contribution of the TLS in the incoherent regime ($\hbar\Delta_r < Kk_BT$), are shown separately. The combination of incoherent and coherent relaxational attenuation causes the relaxational attenuation to be larger than in the old calculation.

with the Kramers-Kronig transform of the velocity we need a substantial unsaturable attenuation even at the lowest temperatures, because the theory predicts there should be one.

There is no hope of fitting the attenuation above 1 K because the relaxational attenuation is bounded above at $\pi/2(1-K)$ at high temperatures. Therefore, if this theory applies, some other attenuation mechanism must be contributing above 0.5 K.[41]

C. Saturable attenuation

In this paper we have discussed three sources of unsaturable attenuation in metallic glasses: usual relaxational attenuation, "anomalous" resonant attenuation, and attenuation that arises because of the sensitivity of the structure factor to increases in the temperature when K is large. The first and third mechanisms both lead to an unsaturable attenuation that decreases as the temperature is decreased, whereas the second leads to attenuation that increases as the temperature is lowered. The combination of these three mechanisms could thus be consistent with the data of Ref. 9, which display an unsaturable attenuation that is roughly temperature independent between 0.01 and 0.1 K. However, we cannot make quantitative comparison of the theory to the temperature dependence of the unsaturable attenuation because we have only calculated the linear response.

In the limit of low temperature $\beta\hbar\omega \ll 1$, the anomalous piece dominates the unsaturable attenuation. Experimentally, the unsaturable attenuation appears to be about half the total attenuation at the lowest temperature (though this estimate relies on the Kramers-Kronig transform of the velocity, and therefore assumes the applicability of TLS theory with the standard distribution function). From Sec. II C 2, we estimate that the anomalous and saturable pieces are equal at $K \sim 0.15$.

IV. DISCUSSION

In this paper we have shown that strong coupling of the TLS to conduction electrons leads to qualitative changes in their linear response that affect strongly the acoustic properties of metallic glasses. The measured low-temperature acoustic properties of $Pd_{0.775}Si_{0.165}Cu_{0.06}$ are consistent with hypothesis that TLS's coupled to conduction electrons are present.

The main qualitative improvement in the agreement of theory and experiment is that we now understand why the unsaturable attenuation in metallic glass is much smaller than previously expected; it is because metallic glasses display substantial unsaturable attenuation at all temperatures. We have shown that symmetric TLS's (which previously were thought to contribute only to saturable attenuation) contribute to the unsaturable attenuation. Quantitatively, we have demonstrated that, at high temperatures, accounting for dissipative tunneling processes yields a substantially different relaxation rate for a TLS; incorporating this feature improves the agreement between theory and experiment.

The agreement between theory and experiment is still not perfect. Further work is needed in order to decide whether the discrepancies arise because of the approximations used in the calculations presented here (though for the reasons outlined in the previous sections we do not expect this), whether the TLS's are not described by the standard distribution function Eq. (2.3), or whether there is an additional (non-TLS) attenuation mechanism contributing above 0.1 K. The saturation behavior at low temperature leads us to believe that $K \sim 0.2$ in this material. This value is consistent with that obtained for a single TLS with a very small Δ_r measured in a polycrystalline bismuth film.[11] However, we note that there is no reason to expect K to be the same for all TLS's in a given sample, let alone in different materials.[42]

The main prediction of this paper is that metallic glasses should display substantial unsaturable attenuation at all temperatures. This prediction can be tested directly by measuring the absolute attenuation. This is generally difficult to measure at GHz frequencies because of substantial corrections to plane-wave propagation in small samples. The simplest method conceptually to do this is to measure the attenuation in a superconducting metallic glass both with and without a magnetic field in the regime $\beta\hbar\omega \ll 1$. The unsaturable attenuation should occur in the presence of a field greater than H_{c2} but not in the superconducting state.[43] It is also possible to extract the absolute attenuation in normal metals by comparing samples of different lengths.

The presence of a distribution of TLS's in glasses makes drawing firm conclusions difficult, first because there is no fundamental basis for the distribution function, and second because the integration over the distribution of ϵ and Δ_r causes differences in the predictions of theory presented here and the one presented in Ref. 9 to be quantitative rather than qualitative. Therefore, direct measurements of relaxation rates[11] provide much more compelling evidence that the dissipative tunneling processes discussed in this paper are experimentally relevant.

We are left with many questions. The most obvious is whether the discrepancy between theory and experiment for $T > 0.1$ K arises from the failure of the TLS theory as presented in this paper, from the nature of the approximations used to calculate the acoustic properties, or from the presence some other relaxation process. An obvious way to check the third possibility is to measure the attenuation and velocity in other metallic glasses. A closely related question is whether the TLS distribution function has the form Eq. (2.3), or if some sort of generalization (such as that used in Ref. 9) is needed for metallic glasses. An answer to this question would help us understand whether there is fundamental significance to the form of the TLS distribution function.

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APPENDIX

In this appendix we show that the existence of nonsaturable resonant attenuation does not depend on the as-

sumption of a Lorentzian line shape; the logarithmic dependence of the averaged structure factor $\bar{S}(\omega)$ on the upper cutoff $\Omega_{\max}$ occurs for a large class of structure factors which have the property that the width of the resonance line is proportional to the resonant frequency.

If the linewidth is proportional to the resonant frequency Ω, then the structure factor $S(\omega)$ can be written

$$S(\omega)=\left[\frac{1}{\Omega^{\delta}}\right]F\left[\frac{\omega}{\Omega}\right] . \tag{A1}$$

We require that $\int_0^{\infty} d\omega\, S(\omega)$ be constant (independent of Ω); this condition implies that the exponent δ is unity. Thus, on general grounds one can write $S(\omega)=(1/\omega)uF(u)$, where $u=\omega/\Omega$ and $\int_0^{\infty} du\, F(u)$ =const. The Lorentzian line shape of Eq. (2.20) is described by this form, with

$$F(u)=r\eta[(u-1)^2+(r\eta)^2]^{-1} .$$

It is straightforward to show that the Ω integral of the structure factor $\int_0^{\Omega_{\max}} d\Omega\, S(\omega,\Omega)$ can be written $\int_{\omega/\Omega_{\max}}^{\infty}(du/u)F(u)$. This integral diverges logarithmically at small u for any function $F(u)$ which tends to a nonzero constant as $u\rightarrow 0$.

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the results of Yamada *et al.* (Ref. 20) imply that K should increase as the tunneling distance increases, but this increase should saturate at a distance of order the atomic spacing. Moreover, the tunneling matrix element Δ_0 depends not only on the tunneling distance but also the mass and the potential barrier. Since the tunneling distance, mass, and barrier and even the tunneling entity itself are not known, there is no compelling justification for any particular choice of a functional dependence of K on Δ_0 and ϵ.

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