Defect interactions in metallic glasses: Acoustic probes

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(Received 5 October 1992; revised manuscript received 3 March 1993)

If the low-temperature properties of glasses are determined by strongly interacting defects, then in a superconducting metallic glass the apparent density of two-level-system (TLS) glass should be greater in the superconducting than in the normal state. This difference affects the acoustic properties of the material. Published measurements of sound attenuation and velocity in $Pd_{30}Zr_{70}$ exhibit anomalies consistent with an increased density of TLS's in the superconducting state, but further experiments are needed to distinguish the effects caused by changes in the interactions between TLS's as opposed to changes in the coupling between each TLS and the conduction electrons.

The striking similarity of the low-temperature properties of a wide class of amorphous materials has led to the proposal that strong strain-mediated dipolar interactions between defects in the material play a crucial role.[1–5] However, this view has gained limited acceptance, in large part because of the enormous success of two-level-system (TLS) theory[6,7] in explaining a wide variety of experimental observations. Though it is plausible that TLS theory describes the low-energy excitations of an interacting defect model,[8] the argument that such a model is necessary to understand glasses would be strengthened substantially by experimental observations that cannot be explained using standard TLS theory.

Experiments to test the idea that defect interactions are important are difficult to design, partly because the point of the defect-interaction idea is to explain *universal* properties that are independent of the microscopic details. Previous proposals have involved comparing the properties of different samples of an insulating glass system with different geometries;[9,10] they have the inherent difficulty that the effects of changing the interactions on the density of TLS's must be distinguished from other geometry-related effects.

Here the aim is to test the hypothesis that the TLS's determining the low-temperature properties of glasses are the low-energy excitations of a model of strongly interacting defects, where the density of TLS $\bar{P}$ is determined by the interaction coupling constant. To this end, it is pointed out that if interactions determine $\bar{P}$, then in a metallic glass $\bar{P}$ should be larger in the superconducting state than in the normal state. This observation is of interest because experimentally one can drive a superconductor into the normal state by applying a magnetic field larger than the upper critical field H_{c2}. One can then make a direct comparison between the low-temperature properties in the presence and absence of electron-mediated interactions between TLS's. A major advantage of examining the superconducting metallic glass system is that one can change the interactions between TLS's in a single sample at fixed temperature.

Measurements on the superconducting metallic glass $Pd_{30}Zr_{70}$ (Ref. 11) show behavior that is consistent with a larger $\bar{P}$ in the superconducting state than in the normal state. However, these measurements are not conclusive because the strong coupling of the conduction electrons to the TLS causes a renormalization of each TLS tunneling matrix element, which for the standard distribution of TLS's leads to a renormalization of the apparent $\bar{P}$ in the normal state that depends on the coupling strength between the electrons and the TLS.[12,13] Superconductivity causes the normal carriers to freeze out and removes this renormalization. The experiments in Ref. 11 were not done at temperatures low enough that this effect could be distinguished from a change in $\bar{P}$ from interactions. Nonetheless, definitive experiments are possible because the fermionic nature of the electron bath constrains the magnitude of the renormalization. For instance, if the change in $\bar{P}$ is large enough, then this can be demonstrated without reference to the details of the theory of the coupling between TLS's and conduction electrons by measuring the attenuation in the limit $\hbar\omega/k_BT \gg 1$ (where ω is the sound frequency and T is the temperature) in the normal and superconducting states. Alternatively, unambiguous evidence that $\bar{P}$ has changed can be obtained by doing measurements over a broad enough temperature range that the electron-TLS coupling constant in the normal state can be determined independently, as outlined below.

The idea that defect interactions dominate the low-temperature properties of disordered materials is based on the nearly universal value of the ratio of the phonon mean free path l to the thermal wavelength λ at temperatures $T < 1$ K that is observed for many different disordered materials ($l/\lambda \sim 150$).[14] A natural explanation for this universality arises from a model of defects with strong dipolar interactions, so that the interaction decays with the defect separation r as g/r^3. Universal features are expected to arise from such a model because in three dimensions interactions of this type give rise to a density of states per unit energy and volume of $1/g$, independent of the number of interacting objects.[3,6]

In insulators the g_{ph}/r^3 interactions are phonon mediated; the coupling constant g_{ph} is estimated to be $\gamma^2/\rho v^2$, where γ is the deformation potential, ρ is the mass density, and v is the transverse sound velocity.[3] In metals, in addition to the strain-mediated interaction, one expects the TLS's to have an interaction mediated by the conduction electrons. This RKKY-like interaction also falls off like $g_{\rm RKKY}/r^3$. The scale of the interaction constant $g_{\rm RKKY}$ is E_F/k_F^3, where E_F is the Fermi energy and k_F is the Fermi wave vector.[15,16] Fortuitously, both $g_{\rm ph}$ and $g_{\rm RKKY}$ are of order 10^5 K Å^3 for a typical metallic glass. One would expect the interactions to add, so that the total is described by an effective interaction strength $g_{\rm eff} \sim (g_{\rm ph}^2 + g_{\rm RKKY}^2)^{1/2}$. These considerations lead one to expect metallic glasses to have a density of TLS's that is somewhat lower, but still the same of order of magnitude, than insulating glasses. This expectation is consistent with thermal-conductivity measurements for $Pd_{0.775}Si_{0.165}Cu_{0.06}$, which yield a slightly higher thermal conductivity than the "universal" curve,[17] as well as acoustic measurements, which yield values of $\bar{P}$ an order of magnitude smaller for metallic glasses than for silica.[18]

Because both the phonon-mediated and electron-mediated interactions decay as $1/r^3$, at first glance it is unclear how their contributions can be separated. However, the long-range character of the RKKY coupling arises because the Fermi sea has arbitrarily low-energy excitations. In a superconductor, the RKKY interaction cuts off exponentially at separations larger than the superconducting coherence length ξ.[19] Thus, well below the superconducting transition temperature T_c, for TLS separations greater than ξ the interactions decay as $g_{\rm ph}/r^3$. Since $g_{\rm ph} < g_{\rm eff}$ and the density of TLS's, $\bar{P} \propto 1/g$, this implies that $\bar{P}$ in the superconducting state should be *greater* than in the normal state.

At a temperature T, one naively expects to probe length scales of order $\sim (g/k_B T)^{1/3}$.[20] Therefore one must change the interactions on the length scale of ≤ 100 Å if one hopes to see observable effects at 0.1 K. Fortunately, metallic glasses tend to have very short electronic mean free paths and hence short enough coherence lengths in the superconducting state that this condition is satisfied.

The most natural way to compare the normal and superconducting states is to compare the behavior at a fixed temperature below T_c in the presence and absence of a magnetic field H greater than the upper critical field H_{c2}. Determination of $\bar{P}$ in the normal state using either specific heat or thermal conductivity is difficult because in a normal metal the electrons contribute significantly to these quantities, especially at low T.[21] However, it is believed that TLS's determine the acoustic properties of both normal metals and superconductors at temperatures $T \lesssim 1$ K.[22,23] Therefore acoustic measurements in superconducting metallic glasses provide a good candidate for testing this prediction. Typically, experiments measure how the sound attenuation and velocity at a given frequency $\omega/2\pi$ depend on the temperature, and so we focus on that situation.

Interpretation of acoustic measurements is nontrivial because in the normal state most experimentally measurable quantities depend not only on $\bar{P}_N$ but also on the electron-TLS coupling constant K. This complication is not insuperable and can be addressed in two ways. The first is to use the fact that the fermionic nature of the electron bath constrains the renormalization from the TLS–normal-electron interaction.[24] Measurements of the low-temperature attenuation can be used to demonstrate a change in $\bar{P}$ without reference to the details of the theory of TLS-electron interactions. The second approach is to fit measurements over a broad temperature range to TLS theory and extract both $\bar{P}$ and K. This approach assumes the validity of the theory of TLS-electron coupling as well as the standard TLS distribution function; these assumptions can be checked because several measurements can be used to determine the three quantities $P_{\rm SC}$, P_N, and K redundantly.

We first discuss experiments that can demonstrate a change in $\bar{P}$ independent of the details of the TLS–conduction-electron coupling. These experiments rely on the fact that in metals K must be less than $\frac{1}{2}$, which in turn constrains the renormalization of $\bar{P}$ from the TLS-electron interaction to be less than a factor of 2. One experiment that can demonstrate a change in $\bar{P}$ is measuring the low-temperature ($\hbar\omega/k_B T \gg 1$) attenuation in the normal and superconducting states. In the superconductor the resonant attenuation is entirely saturable, but in the normal state, even as $T \to 0$, the attenuation is partly unsaturable. Therefore it is important to determine independently the attenuation zero, which can be done by measuring the saturated attenuation as $T \to 0$ in the superconducting state. The saturable attenuation in the superconducting state, $\alpha_{\rm sat,SC}$, is related to $\bar{P}_{\rm SC}$ by

$$\alpha_{\rm sat,SC}(T \to 0) \to \frac{\pi\gamma^2\omega}{\rho v^3} \bar{P}_{\rm SC} \; . \tag{1}$$

In the normal state, the low-intensity attenuation depends on both $\bar{P}_N$ and K, but it satisfies the inequality[25,26]

$$\alpha_{{\rm tot},N}(T \to 0) \geq \frac{\pi\gamma^2\omega}{\rho v^3} \bar{P}_N(1-K) \; . \tag{2}$$

Equations (1) and (2) imply that

$$\frac{\bar{P}_N}{\bar{P}_{\rm SC}} \leq \frac{\alpha_{{\rm tot},N}}{\alpha_{\rm sat,SC}(1-K)} \leq 2\frac{\alpha_{{\rm tot},N}}{\alpha_{\rm sat,SC}} \; , \tag{3}$$

where the last inequality follows because K, which determines the change in the apparent $\bar{P}$ caused by TLS-electron interactions via Eq. (A6), is bounded above by $\frac{1}{2}$. Thus, if as $T \to 0$ the saturable attenuation in the superconducting state is more than a factor of 2 greater than the total attenuation in the normal state, then the change cannot arise entirely from the TLS-electron interaction.

We now discuss how ultrasonic data can be analyzed to extract both the TLS-electron coupling constant K and the density of TLS's, $\bar{P}$. A discussion of the acoustic properties of normal and superconducting metallic glasses summarizing the key results in Refs. 13 and 25 is given in the Appendix; here, we focus on the experimental quantities needed to determine $\bar{P}$ in the normal and superconducting states $\bar{P}_N$ and $\bar{P}_{\rm SC}$. The relevant experimental quantities as well as which parameters they help determine are summarized in Table I. Assuming that metallic glasses have TLS's described by the standard distribution function, the most direct means of comparing $\bar{P}_N$ and $\bar{P}_{\rm SC}$ involves three measurements: the low-temperature ($\hbar\omega/k_B T \gg 1$) saturated attenuation in the superconducting state, the low-temperature linear (low-

TABLE I. Summary of experimentally measurable quantities that can be used to extract information about $\bar{P}_N$ and $\bar{P}_{SC}$.

Useful quantities Phase	Quantity	Gives information on
Superconductor	$\Delta v/v$ (low T)	$\bar{P}_{SC}$
	α_{sat} ($T\to 0$)	attenuation zero
	α_{linear} ($T\to 0$)	$\bar{P}_{SC}$
	relaxational parts have significant phonon contributions, and so not easy to extract useful information	
Normal metal	α_{sat} ($T\to 0$)	$\bar{P}_N, K, E_{max}$
	α_{linear} ($T\to 0$)	$\bar{P}_N, K, E_{max}$
	$\alpha_{plateau}$	$\bar{P}_N$
	$\Delta v/v$ (high T)	$\bar{P}_N, K$
	$\Delta v/v$ (low T)	need to fit $\bar{P}_N, K, E_{max}$

intensity) attenuation in the superconducting state, and the normal-state attenuation in the "plateau" region. The saturated attenuation of the superconductor as $T\to 0$ can be used to determine the zero of attenuation. (As is discussed in the Appendix and Ref. 25, the saturated attenuation in the normal state *cannot* be used to set this zero.) The difference between the linear attenuation and saturated attenuation as $T\to 0$ in the superconductor is $\pi(\omega/v)C_{SC}$, where $C_{SC}=\bar{P}_{SC}\gamma^2/\rho v^2$, γ is the deformation potential, ρ is the mass density, and v is the sound velocity. The value of the attenuation in the plateau region in the normal state (which is all unsaturable) is $(\pi/2)(\omega/v)C_N$. Thus, assuming the attenuation zero is the same in the normal state as in the superconducting state, then these measurements are sufficient to determine $\bar{P}_{SC}/\bar{P}_N$.

Other measurements can be used to check the consistency of this determination. For example, at low enough temperature the variation of the relative velocity shift $\Delta v/v$ with T in the superconducting state yields direct information about C_{SC}; in particular, there is a temperature range where $\Delta v/v$ is linear in $\ln(T)$ with slope C_{SC}. In the normal state, the coupling between TLS's and electrons leads to more complicated behavior; other measurements tend to involve not only C_N, but also the TLS-electron coupling constant K. There is a temperature region where the velocity shift $\Delta v/v$ in the normal state is linear in $\ln(T)$ only when K is not too large (<0.2 or so).[25] If this regime exists and if the relaxational contribution can be neglected, the slope is $C_N(1-K)$.[13,25] If K is not too large, at temperatures where the relaxational contribution is important $\Delta v/v$ is linear in $\ln(T)$ with slope $C_N(\frac{1}{2}-K)$ over a substantial temperature range. In the normal state, the resonant attenuation that is dominant at low temperatures has a magnitude that depends on K, C_N, as well as a high-energy cutoff parameter for the TLS distribution function E_{max} (though the dependence on E_{max} is logarithmic). For $K\lesssim 0.2$, the value of the attenuation as $T\to 0$ is $C_N(\omega/v)(1-K)$. The attenuation as $T\to 0$ is partially unsaturable; the ratio of saturable to unsaturable depends on K, C_N, and E_{max}.

Assuming that metallic glasses have TLS's described by the standard distribution function, then detailed fits can be made to both the attenuation and velocity temperature dependences over a broad temperature range, so that K, C_N, and C_{SC} can be determined reliably.[25,27]

Previous measurements[11] of 620-MHz sound in $Pd_{30}Zr_{70}$ show that the slope of $\Delta v/v$ is greater in the superconducting state than in the normal state by a factor of about 4 over the temperature range 0.1–0.5 K. However, it is likely that the measurements in the normal state are in the regime where the slope of $\Delta v/v$ is $C_N(\frac{1}{2}-K)$, so that the results are consistent with $\bar{P}$ remaining unchanged and the change in the slope arising entirely from TLS–conduction-electron coupling effects. Extraction of K or C_N from attenuation measurements reported in this reference is difficult: The plateau is not well enough developed to determine C_N, and the temperature range does not extend low enough to probe the resonant attenuation. Therefore one cannot extract the $T\to 0$ behavior or do a detailed fit to determine C_N and K including both resonant and relaxational processes.

The experiments proposed here determine the combination $C=\bar{P}\gamma^2/\rho v^2$, where γ is the deformation potential, ρ is the mass density, and v is the sound velocity. The quantities ρ and v can be measured independently, and so the acoustic experiments measure $\bar{P}\gamma^2$, as opposed to the desired quantity $\bar{P}$. The coupling constant γ can be determined in the superconducting state by doing phonon-echo experiments,[28] but this cannot be done in the normal state because phonon echoes are not observed.[22] In principle, γ can be extracted by fitting the temperature dependence of the attenuation and velocity at higher temperatures,[11,22] but in practice this fit has substantial uncertainty. Nonetheless, γ is unlikely to be much different in the normal and superconducting states because it is of order 1 eV (Ref. 29) and the superconducting energy gap is $\sim 10^{-4}$ eV.

Unfortunately, a null result ($\bar{P}_{SC}=\bar{P}_N$) is not definitive because $\bar{P}$ depends on both g_{ph} and g_{RKKY}, and so changing the RKKY interaction changes $\bar{P}$ substantially only if g_{RKKY} is comparable to or larger than g_{ph}. However, the large difference between the values of $\bar{P}$ obtained for insulating and metallic glasses[18] leads one to expect a substantial effect if interactions are important in determining $\bar{P}$. If a change in $\bar{P}$ is observed, this implies that the long-range nature of the interdefect interaction is crucial, for superconductivity only affects the RKKY interaction at length scales greater than the superconducting coherence length ξ.

Changes in $\bar{P}$ arising from interactions should exhibit several systematic trends. The temperature at which changes in $\bar{P}$ become substantial should depend on the inverse cube of the superconducting coherence length ξ. Whether the effect arises because of a change in coupling strength can also be verified by investigating different materials and determining the relative sizes of the strain-mediated and RKKY interactions. The RKKY interaction is more likely to be important in materials with small values of $\gamma^2/\rho v^2$ and large values of K. Because it is possible to change the interactions at a fixed temperature by applying a magnetic field, different arrangements of defects should be obtained if the history of the sample is changed. For instance, cooling a sample in a magnetic field and then turning off the field could yield results different than cooling a sample in zero field.

The experiments described above should provide a useful complement to experiments on insulating glasses. Watson[10] has compared the thermal conductivity of vitreous silica to that of Vycor glass with pore diameters between 20 and 75 Å as well as to low-density silica aerogels of density $\sim 40\%$ and $\sim 10\%$ that of fused silica down to 70 mK. She found that the thermal conductivity in the Vycor is substantially similar to that of the vitreous silica, indicating that the presence of holes does not qualitatively alter the low-temperature thermal conductivity, though the holes do affect the behavior at temperatures where the thermal wavelength of the phonons is comparable to the pore size. These results could be consistent with the idea that long-range strain-mediated interactions between defects play a crucial role if the interactions on long length scales are not substantially changed by the holes. A detailed comparison of the different systems is difficult because there is some question as to whether the deformation potential γ is affected by the presence of holes in the material. Fu[9] has also proposed experiments in optical fibers because the defect interactions cut off exponentially at separations greater than the fiber diameter. However, for a fiber with a diameter of 1 μm, observable effects are only expected for temperatures below $\sim 10^{-7}$ K. The TLS–conduction-electron coupling in metallic glasses leads to complications in data interpretation that do not arise for insulating glasses, but because the experiments can be done on a single sample at fixed temperature, the metallic glass system is a promising candidate to search for a change in $\bar{P}$.

To summarize, this paper discusses how experiments on superconducting metallic glasses can be used to test the idea that defect interactions are important in determining the low-temperature properties of glasses. By comparing the acoustic properties in the presence and absence of a magnetic field, it should be possible to extract the density of TLS's, $\bar{P}$, in the presence and absence of RKKY interactions between defects, though the strong electron-TLS coupling effects in the normal state must be accounted for when interpreting the data. If defect interactions determine $\bar{P}$, then $\bar{P}$ should be larger in the superconducting state than in the normal state.

The author thanks B. Golding and C. Yu for useful conversations.

APPENDIX

This appendix discusses the low-temperature acoustic properties of metallic glasses. We assume, as is usual,[11,13,22,23,25] that at low temperatures the sound attenuation in both superconducting and normal metals arises predominantly because of the coupling between phonons and TLS's. Both the sound attenuation and velocity are affected by the TLS's; these quantities are related by a Kramers-Kronig transformation.[30]

The sound propagation probes the dynamics of the TLS's. These dynamics are strongly affected by the conduction electrons,[13,22,23,25,31] and so normal metals and superconductors are expected to exhibit different behavior.

We make the standard assumptions that an isolated TLS with asymmetry ε and tunneling matrix element Δ_0 is described by the Hamiltonian

$$H_{\mathrm{TLS}} = \tfrac{1}{2}\varepsilon\sigma_z - \tfrac{1}{2}\hbar\Delta_0\sigma_x \ , \tag{A1}$$

where

$$\sigma_z = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}, \quad \sigma_x = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} .$$

The isolated TLS has two energy eigenstates with energies $\pm E/2$, where $E = [(\hbar\Delta_0)^2 + \varepsilon^2]^{1/2}$. The number of TLS's which have given values of ε and Δ_0 is described by a distribution function of the usual form,

$$P(\varepsilon, \Delta_0) = \frac{\bar{P}}{\Delta_0} \ . \tag{A2}$$

The acoustic properties are used as a probe of the TLS dynamics; we assume the usual strain coupling between phonons and TLS's:

$$H_{\text{TLS-ph}} = \gamma\sigma_z e \ , \tag{A3}$$

where e is the strain field at the site of the TLS's and γ is the deformation potential ($\gamma = \tfrac{1}{2}d\varepsilon/de$).[23]

At high enough temperatures, the TLS-phonon coupling affects the TLS dynamics, but below 1 K one expects the coupling to electrons affect the TLS dynamics much more than the coupling to phonons. The coupling between the TLS's and electrons is described by the Hamiltonian

$$H = H_{\mathrm{TLS}} + \sigma_z \sum_{kk'\eta} V_{kk'} c^{\dagger}_{k\eta} c_{k'\eta} + H_e \ , \tag{A4}$$

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 electrons are described by H_e; for noninteracting fermions, $H_e = \sum_{k\eta} \xi_k c^{\dagger}_{k\eta} c_{k\eta}$. The coupling between the TLS's and the electrons is described quite generally by a parameter K, which for an s-wave potential ($V_{kk'} = V$) and a Fermi sea of free fermions is determined solely by the scattering phase shift of electrons;[32] in the weak-coupling limit, $K = \tfrac{1}{2}(n_0 V)^2$, where n_0 is the electronic density of states at the Fermi level.[33] It is known that in metallic glasses K must be less than $\frac{1}{2}$.[24]

It is customary to divide the effects of the TLS's on phonons into resonant and relaxational pieces. The former arises from transitions induced by phonons between the two energy levels of the TLS's, whereas the latter arises from the phonon changing the TLS asymmetry energy and the TLS reequilibrating via an incoherent tunneling process. Both contributions are substantially different in the normal state than in the superconducting state. We first discuss the resonant piece, which is dominant at low enough temperatures.

In insulating or superconducting glasses, the resonant attenuation is entirely saturable, so that high-intensity sound is not attenuated. This property is useful because it is very difficult to measure absolute attenuation, and one can use the saturated low-temperature value as the attenuation zero. The magnitude of the saturable attenuation is directly proportional to $\bar{P}$: $\alpha_{\rm res}(T\to 0)=\pi\omega\bar{P}\gamma^2/\rho v^3$, where γ is the deformation potential, ρ is the mass density, $\omega/2\pi$ is the sound frequency, and v is the sound velocity. Experimentally, one must be sure that the power is low enough that one is measuring the linear response as well as that the temperature T is low enough that the resonant attenuation dominates. This latter point can be checked by making sure the attenuation depends on T as $\tanh(\hbar\omega/2k_BT)$.

In normal metals the resonant attenuation differs from that of a superconductor in two major ways. One important feature of the normal state is that the effective matrix element Δ_r of a TLS that determines the low-energy properties is renormalized because of the TLS-electron interaction:[34]

$$\Delta_r=\Delta_0\left[\frac{\Delta_0}{\omega_c}\right]^{K/(1-K)}, \tag{A5}$$

where ω_c is a cutoff frequency expected to be of order the Debye temperature.[35] Because of this, the distribution function describing the renormalized parameters becomes[13]

$$P(\varepsilon,\Delta_r)=\frac{\bar{P}}{\Delta_r}(1-K)\ . \tag{A6}$$

Thus one expects the TLS-electron coupling to cause the *apparent* value of $\bar{P}$ to be smaller in the normal state than in the superconducting state even if $\bar{P}$ itself is unchanged. However, since $K\le\frac{1}{2}$, this effect cannot change the apparent $\bar{P}$ by more than a factor of 2.

The second change is that the resonant attenuation develops an unsaturable component even as $T\to 0$.[25] This occurs because the coupling between the TLS's and conduction electrons results in a linewidth that is proportional to the energy splitting, so that TLS's of all energies contribute to the attenuation at a given frequency.[36] The ratio of the unsaturable and saturable resonant pieces is given by[25]

$$\frac{\alpha_{\rm unsat}}{\alpha_{\rm sat}}\approx 0.7K\ln\left[\frac{E_{\max}}{\hbar\omega}\right], \tag{A7}$$

where K is the TLS-electron coupling constant and $E_{\max}$ is the upper cutoff of the TLS distribution function. (This expression is valid when K is not too large.[25]) If one assumes that the attenuation zero is given by the saturated attenuation in the superconducting state, then one can estimate K fairly accurately because the dependence of this ratio on the (unknown) upper cutoff is weak—changing $E_{\max}$ from $200K$ to $300K$ changes the logarithm by less than 10% for a sound frequency of 1 GHz. It might be possible to determine $E_{\max}$ itself by measuring the attenuation at two different frequencies.

In the normal state, as $T\to 0$ the total attenuation is bounded below by $\pi C_N(1-K)$. The $1-K$ factor arises because of the renormalization of Δ_0 by the conduction electrons described in Eqs. (A5) and (A6). If $K\le 0.2$, then the attenuation is within a few percent of this lower bound; if $K\ge 0.2$, $\alpha(T\to 0)$ has a nontrivial (but monotonic) dependence on K. Nonetheless, if the attenuation zero and K have been determined, then C_N can be determined by numerical fitting of the total attenuation.[25] Thus knowing both the amount of total attenuation as well as what fraction of the attenuation is unsaturable is sufficient to determine both K and $\bar{P}$ to reasonable accuracy.

The resonant contribution to the velocity shift also provides important information. In the superconducting state, the resonant contribution to the velocity, which dominates at low temperatures, has the form[37]

$$\frac{\Delta v_{\rm res}}{v}=C_{\rm SC}\left[\Psi\left[\frac{1}{2}+\frac{\hbar\omega}{k_BT}\right]-\ln\left[\frac{\hbar\omega}{k_BT}\right]\right], \tag{A8}$$

where ψ is the digamma function. If $\hbar\omega/k_BT\ll 1$, the velocity shift depends logarithmically on temperature: $\Delta v/v=C_{\rm SC}\ln(T/T_0)$, where T_0 is a reference temperature. Thus the temperature dependence of $\Delta v/v$ yields $C_{\rm SC}$; this value can be checked for consistency with that obtained from the magnitude of the unsaturated attenuation. In the normal state, the relaxational component is important at much lower temperatures than in the superconducting state; in addition, the resonant contribution to the velocity has a component arising from the unsaturable attenuation. Even if $\hbar\omega/k_BT\ll 1$, $\Delta v_{\rm res}/v$ for 1-GHz sound is not linear in $\ln(T)$ if K is larger than 0.2 or so. If $K\lesssim 0.2$ and the relaxational contribution is negligible, then one finds the simple result $\Delta v/v=C_N(1-K)\ln(T/T_0)$, but numerical fitting is needed to determine whether there is a temperature range where this form is valid.[25]

We now discuss the relaxational attenuation, which arises from incoherent tunneling of the TLS's. In the superconducting state near T_c, the behavior is complex because the normal carrier freezeout affects the renormalization of Δ_0 (Ref. 13) as well as the density of quasiparticles;[31] roughly speaking, in the superconducting state the elimination of the renormalization of the tunneling matrix element tends to increase the attenuation, while the quasiparticle freezeout causes the relaxation rate of the TLS's to decrease, which tends to decrease the attenuation. Detailed calculations of the temperature dependence of the attenuation near T_c are difficult. When the quasiparticles have frozen out, one must also consider relaxation via coupling to phonons, but one expects all sources of relaxational attenuation to be very small well below T_c.

In the normal state, the detailed temperature dependence of the relaxational attenuation is complex, but at high enough temperatures it saturates at the value[13,25]

$$\alpha_{\rm relax} \to \frac{\pi}{2}\frac{\omega}{v}\frac{\bar{P}_N \gamma^2}{\rho v^2} \ . \tag{A9}$$

This result follows because the $(1-K)$ factor renormalizing the TLS distribution function in Eq. (A6) is canceled by a factor of $(1-K)^{-1}$ that arises from the dependence of the incoherent relaxation rate on the renormalized tunnel splitting Δ_r.[13,25] If this asymptotic value is reached at a temperature low enough that other contributions to the attenuation are unimportant, then one has a direct measure of $\bar{P}_n$.

We now discuss the relaxational contribution to the velocity shift. In superconductors this contribution is not expected to be important well below T_c because quasiparticles freeze out; at higher temperatures one needs to consider the detailed nature of the coupling between phonons and TLS's (including phonon-assisted tunneling). Therefore simple extraction of $\bar{P}_{\rm SC}$ from this contribution is not possible. In the normal state, the relaxational contribution is much more likely to be dominated by the coupling to electrons and also should be substantial down to much lower temperatures than in a superconductor. It is important to determine whether the changes in the velocity arise from both resonant and relaxational processes or from resonant processes only. If the relaxational contribution to the velocity has saturated and K is not too large, then $\Delta v/v = C_n(\frac{1}{2}-K)\ln(T/T_0)$ for $\hbar\omega/k_B T \ll 1$. Determination of the temperature range where this form applies as well as extraction of values for C_N and K requires numerical fitting of the data.

In summary, extracting information about $\bar{P}$ in the normal and superconducting states can be done using acoustic measurements over a broad temperature range. If the standard TLS distribution function applies to metallic glasses, the necessary parameters can be extracted with confidence; both the resonant and relaxational contributions to both the velocity and attenuation are determined by the same parameters, and so K, $\bar{P}_{\rm SC}$, and $\bar{P}_N$ are determined redundantly. Even if a modified distribution function is necessary to fit the data,[22,38] it should be possible to fit numerically the resonant attenuation in the normal and superconducting states and extract $\bar{P}_N$ and $\bar{P}_{\rm SC}$.

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$$\int_0^{\Omega_{\max}} d\Omega \frac{\Gamma(\Omega)}{(\omega-\Omega)^2+\Gamma^2(\Omega)} = \int_0^{\Omega_{\max}} d\Omega \frac{\eta\Omega}{(\omega-\Omega)^2+\eta^2\Omega^2} .$$

This integral diverges logarithmically as the upper cutoff $\Omega_{\max}\rightarrow\infty$.

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