Acoustic properties of metallic glasses at low temperatures: Tunneling systems and their dephasing

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The low temperature acoustic properties of bulk metallic glasses measured over a broad range of frequencies rigorously test the predictions of the standard tunneling model. The strength of these experiments and their analyses is mainly based on the interaction of the tunneling states with conduction electrons or quasiparticles in the superconducting state. A series of experiments at kHz and GHz frequencies on the same sample material essentially confirms previous measurements and their discrepancies with theoretical predictions. These discrepancies can be lifted by considering more correctly the linewidths of the dominating two-level atomic-tunneling systems. In fact, dephasing caused or mediated by interaction with conduction electrons may lead to particularly large linewidths and destroy the tunneling sytems' two-level character in the normal conducting state.

The low temperature properties of disordered solids make up a fascinating thermodynamic combination comprising specific heat, thermal conductivity, and dielectric as well as mechanic susceptibilities [1–4]. The standard tunneling model [5,6] seems to account for all these properties in a material-independent, universal manner establishing its fifty-year-long success despite quantitative disagreement with some experiments. A particularly remarkable example is the unexpected crossing of the sound velocities of metallic glasses measured in the superconducting or in the normal state of the same sample [7,8]. An experiment is shown in Fig. 1. The normal state is enforced by a sufficiently strong magnetic field, otherwise the sample becomes superconducting below $T_c = 1.39$ K. A first and obviously too naive—or even incorrect—analysis within the standard tunneling model predicts the sound velocity of the superconducting state to mark an upper bound, the sound velocity of the normal state being always smaller and smoothly merging at some temperatures well below T_c.

In this paper, we discuss all questions related to the acoustic susceptibility of disordered solids at low temperatures and emphasize the importance of dephasing of the two-level atomic-tunneling systems due to their interaction with conduction electrons. We present a series of experiments at audio and radio frequencies and demonstrate that they are very well described within the tunneling model by including more

carefully extreme linewidths of the two-level systems in the normal conducting state of the metallic glass.

I. SOUND VELOCITY IN METALLIC GLASSES: STATING THE PROBLEM

The tunneling model (TM) provides a microscopic explanation for the low energy excitations present in virtually all disordered solids. It assumes that some atoms or groups of atoms in metastable configurations can occupy two different configurations modelled as particles in double well potentials and that at low temperatures the atoms can tunnel between the configurations. The overlap of the two localized wave functions generates a two-level system with eigenfunctions across both the potential wells. Coupling to phonons or strain fields requires these two-level tunneling systems (TSs) to hold an elastic dipole (more correctly: quadrupole) moment with different sizes or orientations for the two configurations. When strain is applied to the material the tunneling systems may follow this field leading to an increase of the overall susceptibility of the material, making it 'softer' than without TSs.

The softening, however, is only effective when a TS is in its ground state. At higher temperatures, when the population difference vanishes, its contribution to the susceptibility disappears. At very low temperatures all relevant TS with large enough energies are in the ground state, each adding a small contribution to the susceptibility. This so-called *resonant process* is analogous to a pendulum following in phase its drive at very low frequency. With increasing temperature both states of these TSs become equally populated which removes their contributions to the susceptibility. The material

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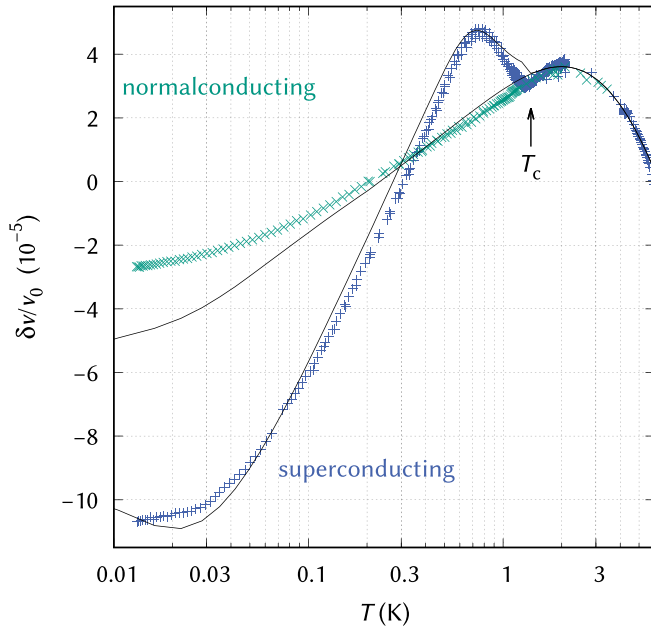‡sschnei@gwdg.de
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FIG. 1. Temperature dependence of the transverse sound velocity of the metallic glass $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ measured at 969 MHz. The superconducting transition is at $T_c = 1.39$ K. Data marked *normal conducting* is obtained in a magnetic field of 4 T. Lines show numerical calculations as explained in the text.

becomes stiffer; the sound velocity increases. This is visible in Fig. 1 where the sound velocity in the superconducting state increases with temperature between 50 mK and 500 mK. The increase may persist as long as there are TSs with even higher energies being thermally excited with rising temperature.

There is, however, another mechanism by which two-level atomic-tunneling systems contribute to the acoustic susceptibility: A *relaxation mechanism*, which becomes important for temperatures where the energy relaxation rates of the TSs are fast enough so that their occupation numbers can adjust for the modulation of the energy splitting caused by the applied strain of the sound field [9]. For relaxation rates much higher than the sound frequency the occupation of the two levels will always be close to thermal equilibrium. Due to their continuous relaxation towards the modulated equilibrium, TSs with energies on the order of the thermal energy contribute to the total susceptibility and reduce the sound velocity in a certain temperature window. This is particularly effective for TSs with asymmetric double well potentials since the lower and higher energy levels are identifiable with the tunneling particle residing preferentially in either of the two wells.

At this point, possible relaxation processes and the respective temperature dependence of the resulting relaxation rates ought to be discussed. Common for all disordered solids and well documented is the relaxation of TS due to interaction with the phonon bath. At low temperatures the one-phonon process, i.e., emission or absorption of a single resonant phonon dominates in adjusting the TSs' occupation numbers [9] whereas higher order phonon processes become important at elevated temperatures. In Fig. 1 (see also Fig. 3) phonon dominated relaxation is the reason for the sound velocity to exhibit a maximum around 2 K and to further decrease with rising temperature.

In a normal conducting metallic glass the energy relaxation rates are massively enhanced because the TSs scatter inelastically conduction electrons [10,11], which have a high density of states available around the Fermi energy. This additional relaxation channel causes the sound velocity to be reduced with respect to the always present resonant process discussed above. The reduction persists to very low temperatures although the positive temperature coefficient prevails. If the same sample, i.e., with the same distribution of TSs, became superconducting below a certain temperature, the electronic relaxation channel would rapidly vanish and, given that relaxation by phonons is already too slow, the resonant process alone would determine the temperature dependence of the sound velocity. It marks an upper bound in the absence of any relaxation mechanism. Nevertheless, as shown in Fig. 1 below 0.5 K the sound velocity in the normal conducting sample is higher than in the superconducting sample, although relaxation processes due to inelastic TS-electron scattering are present. The electronic relaxation also becomes important in the superconducting state when the density of thermally excited quasiparticles rapidly increases on approaching T_c. The related contribution to the susceptibility leads to a maximum of the sound velocity just below T_c visible in Fig. 1.

Summarizing the problem laid out in this section: Despite the increased susceptibility due to the electronic relaxation process, which in the normal conducting state persists to lowest temperatures, the material is at low temperatures stiffer, the sound velocity being higher than in the superconducting state. As both resonant and relaxation mechanisms can only increase the susceptibility and thus decrease the sound velocity, either the number of contributing TSs is reduced in the normal state or one of the mechanisms is quenched in some way. Renormalization of the density of states of TS in the normal conducting state [12–14] followed the first approach. However, only a qualitatively better description of the phenomenon could be achieved. In this paper an alternative idea is suggested: The contribution of the resonant process is reduced due to the large linewidth of TSs resulting from the strong interaction with conduction electrons. While retaining the originally proposed distribution function [see Eq. (12) further down] of the Standard Tunneling Model, a very satisfying agreement with experiments at frequencies from kHz to GHz is achieved that way.

II. THE STANDARD TUNNELING MODEL: A SHORT REVIEW

The standard tunneling model, as introduced by Phillips [6] and Anderson *et al.* [5], and its consequences for the acoustic susceptibility are briefly outlined here. For most quantities we use the notation as of Jäckle [9].

The tunneling entity is modeled as a particle in a double well potential. With the WKB method[1] the tunneling energy $\Delta = \hbar\omega_H \exp(-\lambda)$ can be calculated from the tunneling parameter $\lambda = d/2\hbar \cdot \sqrt{2mV}$, which absorbs the geometric details of the tunneling particle, being the distance d, the mass of the tunneling particle m, and the potential barrier separating

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[1]Named after G. Wenzel, H. A. Kramers, and L. Brillouin.

the two wells V. The asymmetry energy ε denotes the energy shift between the ground states of the individual wells. This TS is described by

$$\mathcal{H}_0^{\mathrm{LR}} = \frac{1}{2}\begin{pmatrix} \varepsilon & -\Delta \\ -\Delta & -\varepsilon \end{pmatrix} \qquad (1)$$

in the localized basis of the particle being in the left state $|L\rangle$ or the right state $|R\rangle$. In the energy basis $\mathcal{H}_0^{\mathrm{ge}} = E/2 \cdot \sigma_z^{\mathrm{ge}}$ with the total energy splitting $E = \sqrt{\varepsilon^2 + \Delta^2}$. σ_z^{ge} is the diagonal Pauli matrix in the energy basis $|g\rangle$ and $|e\rangle$ with the tunneling particle in superposition states of $|L\rangle$ and $|R\rangle$.

External strain $\overleftrightarrow{S}$ applied to the material changes the asymmetry energy $\delta\varepsilon = \overleftrightarrow{\gamma} \cdot \overleftrightarrow{S}$ of the TSs due to their deformation potential $\overleftrightarrow{\gamma}$. This coupling is diagonal in the localized basis $\mathcal{H}_{\mathrm{int}}^{\mathrm{LR}} = 1/2\,(\overleftrightarrow{\gamma} \cdot \overleftrightarrow{S})\sigma_z^{\mathrm{LR}}$ and yields the interaction Hamiltonian in the energy basis

$$\mathcal{H}_{\mathrm{int}}^{\mathrm{ge}} = \frac{1}{2E}\begin{pmatrix} \varepsilon & -\Delta \\ -\Delta & -\varepsilon \end{pmatrix}(\overleftrightarrow{\gamma} \cdot \overleftrightarrow{S}), \qquad (2)$$

where for the remaining part of the paper the tensorial notation of $\overleftrightarrow{\gamma}$ and $\overleftrightarrow{S}$ is dropped for simplicity. With the transverse matrix element $M = \gamma\Delta/2E$ and the longitudinal matrix element $D = \gamma\varepsilon/E$, Eq. (2) is rewritten as $\mathcal{H}_{\mathrm{int}}^{\mathrm{ge}} = 1/2 \cdot (D\sigma_z^{\mathrm{ge}} - 2M\sigma_x^{\mathrm{ge}})S$. The contribution of the TS to the susceptibility χ of the amorphous material in a sound field of frequency ω can be derived from Bloch equations or another linear response theory.

Following, e.g., Hunklinger and Arnold [1] (or Phillips [2] for an analogous discussion in the dielectric case) the response of an ensemble of N TSs to the transverse interaction in $\mathcal{H}_{\mathrm{int}}^{\mathrm{ge}}$ or the so-called *resonant contribution* results in real and imaginary parts of the susceptibility

$$\chi_{\mathrm{res}}' = -4NM^2 \tanh\left(\frac{E}{2k_{\mathrm{B}}T}\right)$$
$$\times \frac{1}{\hbar}\left(\frac{(\omega - \omega_0)\tau_2^2}{1 + (\omega - \omega_0)^2\tau_2^2} - \frac{(\omega + \omega_0)\tau_2^2}{1 + (\omega + \omega_0)^2\tau_2^2}\right), \qquad (3)$$

$$\chi_{\mathrm{res}}'' = 4NM^2 \tanh\left(\frac{E}{2k_{\mathrm{B}}T}\right)$$
$$\times \frac{1}{\hbar}\left(\frac{\tau_2}{1 + (\omega - \omega_0)^2\tau_2^2} - \frac{\tau_2}{1 + (\omega + \omega_0)^2\tau_2^2}\right), \qquad (4)$$

where $\omega_0 = E/\hbar$. The real part χ' cannot be further simplified by the rotating wave approximation as especially for low measuring frequencies the contributions at $\omega + \omega_0$ and $\omega - \omega_0$ add up and are equal for $\omega \ll \omega_0$. The linewidth $h\tau_2^{-1}$ is usually taken to be small. We will show that this is not always the case and can have a large impact on the susceptibility of a broad distribution. In contrast to the real part the loss χ_{res}'' is dominated by an ensemble subset with $\omega_0 \approx \omega$ of the broad distribution of TSs.

The longitudinal interaction in $\mathcal{H}_{\mathrm{int}}^{\mathrm{ge}}$ gives rise to the *relaxation contribution* to the susceptibility

$$\chi_{\mathrm{rel}}' = -2ND^2 \frac{\partial}{\partial E} \tanh\left(\frac{E}{2k_{\mathrm{B}}T}\right)\frac{1}{1 + (\omega\tau_1)^2} \qquad (5)$$

$$\chi_{\mathrm{rel}}'' = -2ND^2 \frac{\partial}{\partial E} \tanh\left(\frac{E}{2k_{\mathrm{B}}T}\right)\frac{\omega\tau_1}{1 + (\omega\tau_1)^2}, \qquad (6)$$

where $\partial/\partial E \tanh(E/2k_{\mathrm{B}}T)$ is the change of the occupation number difference due to variation of the energy splitting around the given energy E. TSs contribute to the longitudinal susceptibility when their energy relaxation rate τ_1^{-1} matches the frequency ω of the external driving field. In order to understand the longitudinal contribution knowledge of the energy relaxation processes is crucial. In turn, the temperature dependence of the susceptibility gives information on the dominating relaxation process in a given temperature range.

In metallic glasses TSs relax to the thermal equilibrium through various processes. Independent of the material being metallic or insulating the interaction of TSs with the phonon bath is always present, and according to Jäckle [9] the one-phonon process yields the energy relaxation rate

$$\tau_{1,\mathrm{ph}}^{-1} = \left(\frac{\gamma_l^2}{v_l^5} + 2\frac{\gamma_t^2}{v_t^5}\right)\left(\frac{\Delta}{E}\right)^2 \frac{E^3}{2\pi\rho\hbar^4}\coth\left(\frac{E}{2k_{\mathrm{B}}T}\right). \qquad (7)$$

ρ is the density of the material, and indices l and t denote the phonon modes as longitudinal and transverse, respectively. Since $v_l \approx 2v_t$ and both velocities enter with fifth power in the denominator, $\tau_{1,\mathrm{ph}}^{-1}$ is by far dominated by transverse phonons. For a given ratio of Δ/E thermal TSs ($E \approx k_{\mathrm{B}}T$) have energy relaxation rates $\propto T^3$. At temperatures above 2 K multiphonon processes lead to energy relaxation rates with stronger temperature dependencies, modeled for example [15] by $\tau_{1,\mathrm{mph}}^{-1} = K_{\mathrm{mph}} \cdot T^5$.

In a metallic glass the most relevant relaxation process is caused by the TSs' interaction with conduction electrons which towards lower temperatures exceeds the phonon-induced rate by many orders of magnitude. From a microscopic point of view Black *et al.* [10,11] derived the energy relaxation rate

$$\tau_{1,\mathrm{el}}^{-1} = W_{\mathrm{el}}^2 \frac{\pi}{4\hbar}\left(\frac{\Delta}{E}\right)^2 E \coth\left(\frac{E}{2k_{\mathrm{B}}T}\right). \qquad (8)$$

The constant density of states of electronic excitations around the Fermi energy yields a linear increase $\propto T$ of the rate for thermal TS.

In the superconducting state electrons form Cooper pairs and well below T_{c} only the phonon induced rate remains. However, with rising temperature the interaction of TSs with thermally excited quasiparticles leads to a rapidly increasing rate approaching the normal conducting rate [10,16] at T_{c}

$$\tau_{1,\mathrm{qp}}^{-1} = W_{\mathrm{el}}^2 \frac{\pi}{2\hbar}\left(\frac{\Delta}{E}\right)^2 E \coth\left(\frac{E}{2k_{\mathrm{B}}T}\right)\frac{1}{1 + \exp\left(\frac{\Delta_{\mathrm{BCS}}(T)}{k_{\mathrm{B}}T}\right)}. \qquad (9)$$

This relation holds strictly only for $E \ll \Delta_{\mathrm{BCS}}$. Closely below T_{c} pair breaking effects are possible [16] and may result in corrections of our calculations.

The rates $\tau_1^{-1}(T)$ of all three processes (7), (8), and (9) are shown as solid lines in Fig. 4 further down together with experimental data extracted from the position of the maxima in the sound velocity, which marks the temperature where $\tau_1^{-1} \approx \omega$, as elucidated below. The TS-electron coupling strength W_{el} is determined from the $\delta v/v$ maximum in the superconducting sample via Eq. (9). Inserting this value for the electron-induced relaxation rate in the normal conducting state, Eq. (8), one finds rates exceeding the energy of the thermal TSs as visualized in Fig. 4.
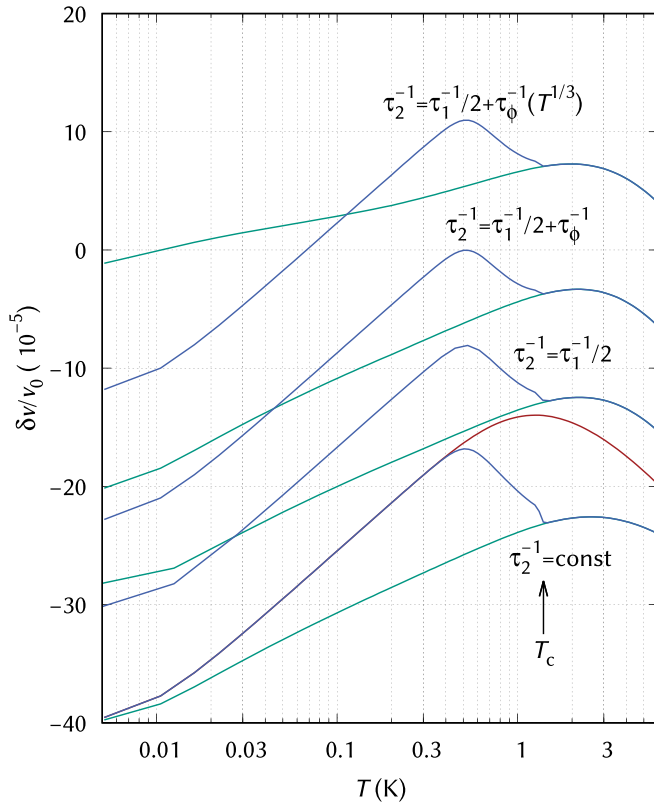
FIG. 2. Temperature dependence of the sound velocity of a metallic glass as calculated with the standard tunneling model for a measuring frequency of 200 MHz. Bottom curves comprise energy relaxation only by phonons (red) and additionally by quasiparticles (blue) and electrons (green). Constant linewidth $h\tau_2^{-1} \ll E$ is assumed. Sound velocities of normal- and superconducting states merge at low temperatures. Including the lifetime limited linewidth $\tau_2^{-1} = \tau_1^{-1}/2$ with plausible and temperature dependent τ_1^{-1} rates already produces a crossing of $\delta v/v$ of the normal state and the superconducting state (curves next to bottom). Addition of a dephasing process $\tau_\phi^{-1} \propto T$ shifts this crossing to higher temperatures. Finally, a modified dephasing model $\tau_\phi^{-1} \propto f(T)$ (see text) reproduces the experimental data (see Figs. 1 and 3) very well (topmost curves).

Obviously, the linewidth $h\tau_2^{-1}$ of these dominating, thermal TSs cannot be assumed to be small and constant and at least [17,18] $\tau_2^{-1} = \tau_1^{-1}/2$ has to be introduced when analyzing experiments measuring χ_{res}. The contribution to χ_{res} is reduced or even lost when the phase of a TS is lost within one oscillation of the external field. Including this minimal requirement in calculating expected temperature dependencies of $\delta v/v$ on the basis of Eqs. (3) and (5), however replacing N by an integration over the distribution functions $P(E, \Delta)$ as proposed in the Standard Tunneling Model [Eq. (12) further down] one finds already an answer to the major question stated above: At lowest temperatures the sound velocity in the normal state is predicted to be higher than in the superconducting state (see Fig. 2). Obviously, the short life times τ_1 already broaden the line widths of relevant TSs sufficiently to reduce their contributions to χ'_{res}, Eq. (3), leaving the material harder although both resonant and relaxation contributions are present.

It is in fact sensible to accept explicitly a dephasing process due to the TS-electron interaction as already suggested by Black [10] when briefly discussing the longitudinal coupling between TS and excitations of the electron bath. A more general formulation of dephasing of two-level systems [19] considers explicitly the importance of the spectral density of the bath in the low energy limit $E \ll k_B T$. This leads to a dephasing rate

$$\tau_{\phi,\mathrm{el}}^{-1} = W_{\mathrm{el}}^2 \frac{\pi}{2\hbar} \left(\frac{\varepsilon}{E} \right)^2 k_B T \tag{10}$$

for the spectral density of the electronic bath in the normal conducting state, which may be modified as

$$\tau_{\phi,\mathrm{qp}}^{-1} = W_{\mathrm{el}}^2 \frac{\pi}{\hbar} \left(\frac{\varepsilon}{E} \right)^2 k_B T \frac{1}{1 + \exp\left(\frac{\Delta_{\mathrm{BCS}}(T)}{k_B T} \right)} \tag{11}$$

for the superconducting state. The magnitude of τ_ϕ^{-1} is determined by the same coupling constant W_{el} as τ_1^{-1}, and τ_ϕ^{-1} has the same temperature dependence as τ_1^{-1} of thermal TSs with $E \approx k_B T$. But in contrast to τ_1^{-1}, dephasing τ_ϕ^{-1} due to this process vanishes for symmetric[2] TSs with $\Delta = E$. However, slightly asymmetric TSs are sufficiently affected as well, and their contribution to χ'_{res} is also reduced. Calculation of $\delta v(T)/v$ with [17,18] $\tau_2^{-1} = \tau_1^{-1}/2 + \tau_\phi^{-1}$ produces a small but noticeable shift to higher temperature of the crossing between normal and superconducting states (see Fig. 2).

The calculated influence of dephasing can be amplified by replacing the linear T dependence in Eqs. (10) and (11) by $f(T) = T \cdot (T/T_0)^{\alpha-1}$. With $0 < \alpha < 1$ dephasing τ_ϕ^{-1} varies weaker than linearly with T and is effectively enhanced towards lower temperatures. Combination of large τ_1^{-1} and strong dephasing τ_ϕ^{-1} weakens the contribution of symmetric and slightly asymmetric TSs to χ'_{res}, and the calculated $\delta v(T)/v$ exhibits a further reduced temperature variation in the normal conducting metal shifting the intersection with the superconducting state closer to T_c (Fig. 2 topmost curves). To this point the suggested temperature dependence $f(T)$ of the electron-mediated dephasing is purely phenomenological. We will come back to this later.

Calculation of the response to an applied sound field of an ensemble of atomic-tunneling systems in a disordered solid requires both resonant and relaxation contributions to the susceptibility to be integrated over the distribution of parameters of all TSs. The standard tunneling model [5,6] suggests a flat distribution in asymmetry energy ε and tunneling parameter λ yielding the distribution [2]

$$P(E, \Delta)\,dE\,d\Delta = \frac{P_0 E}{\Delta \sqrt{E^2 - \Delta^2}}\,dE\,d\Delta \tag{12}$$

of TSs with energy splitting E and tunneling energy Δ, where P_0 is constant. The susceptibility of a macroscopic sample is

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[2]A $(\varepsilon/E)^2$ dependence of τ_ϕ^{-1}, originating from the TSs' dipolar interactions, has been observed in dielectric echo experiments on individual TSs in the oxide films of Josephson junction qubits [20].

given by

$$\langle \chi \rangle = \int_0^{E_{\max}} \mathrm{d}E \int_{\Delta_{\min}}^{E} \mathrm{d}\Delta \, (\chi_{\mathrm{res}} + \chi_{\mathrm{rel}}) P(E, \Delta). \qquad (13)$$

The real part of $\langle \chi \rangle$ yields the change of sound velocity and the imaginary part the absorption of sound waves, as quantified in the next paragraph.

We briefly discuss the role of the integration limits, because in contrast to the known analytic solutions [2] they do not appear in our final results. The integration limit $E_{\max}$ vanishes when with Eq. (3) the linewidth is adequately taken into account. At high temperatures the energy relaxation rate caused by interaction with phonons leads to a linewidth $h\tau_2^{-1}$ increasing faster than linearly with energy E. TSs with $h\tau_2^{-1} \geqslant E$ do not contribute to χ'_{res} and this limits the increase of the sound velocity above a certain temperature thus $E_{\max} \to \infty$ can be taken. Already for the one-phonon process the energy-relaxation limited linewidth $h\tau_2^{-1} = h\tau_1^{-1}/2$ scales $\propto E^3$, for multiphonon processes the linewidth increases even stronger with exponents from E^5 to E^7. The upper bound for TSs energies contributing to χ'_{res} is of the order of 10 K and almost independent of the prefactors in Eq. (7). This differs from introducing a high energy cutoff for the distribution of TSs. Although unimportant for χ'_{res} high energy TSs are still present and are responsible for the relaxation contributions to the susceptibility at higher temperatures.

The lower cutoff for the tunneling energy $\Delta_{\min}$ is required in analytic calculations to avoid the divergence of the integral for $\Delta \to 0$. For the transverse susceptibility this is canceled by Δ in the matrix element M representing the overlap of the localized wave functions and thus defining the two-level character of a TS with given energy E. The longitudinal susceptibility vanishes when the temperature dependence of the relaxation rate is taken into account: Asymmetric TS with $\Delta \to 0$ become too slow to relax within the time scale of the experiment set by the measuring frequency and they no longer contribute to $\langle \chi \rangle$.

III. EXPERIMENTAL METHODS

The TSs' contribution to the real part of the generalized susceptibility $\langle \chi \rangle'$ is related to the variation of the sound velocity $\delta v = v(T) - v_0$ normalized to the velocity at a reference temperature $v_0 = v(T_0)$ as [1,2]

$$\frac{\delta v}{v_0} = -\frac{1}{2} \frac{1}{\rho v_0^2} \langle \chi \rangle'. \qquad (14)$$

The imaginary part $\langle \chi \rangle''$ is related to the loss, and ultrasonic measurements yield the attenuation α per unit length. Normalized to the energy loss per wave number results in the loss tangent [1,2]

$$\tan \delta = \frac{\alpha v_0}{\omega} = \frac{1}{\rho v_0^2} \langle \chi \rangle''. \qquad (15)$$

In experiments with vibrating reeds or other acoustic resonators the corresponding quantity is the inverse quality factor $Q^{-1} = \tan \delta$ defined as the energy loss per cycle divided by the total energy stored in the resonator.

The samples measured in this work are made from one rod (diameter $\approx 3.5\,\mathrm{mm}$) of the bulk metallic glass $\mathrm{Zr}_{59}\mathrm{Ti}_3\mathrm{Cu}_{20}\mathrm{Ni}_8\mathrm{Al}_{10}$ fabricated at IFW Dresden. Details on the fabrication of the metallic glass sample and its properties can be found elsewhere [21]. At low frequency (1.1 kHz) quality factor and variation of the sound velocity—determined by tracking the resonance frequency—are measured by a capacitively driven vibrating reed [22–24] machined as a stripe (rough size $0.3\,\mathrm{mm} \times 2\,\mathrm{mm} \times 12\,\mathrm{mm}$) [25] from one part of the rod. The reed is clamped to a copper sample holder to provide thermalization in a dilution cryostat. Details of the setup used in this work are described elsewhere [26]. To investigate the normal conducting state of the material the reed is exposed to a magnetic field of 4 T, sufficient to reliably suppress superconductivity of the metallic glass. In order to minimize possible eddy currents and other disturbing effects the sample is mounted such that the direction of its oscillation lies parallel to the magnetic field vector.

For the high frequency experiments cylinders were cut from the same sample rod. Their two end faces were lapped plain and parallel.[3] RF sputtered zinc oxide transducers of thicknesses between $3.6\,\mu\mathrm{m}$ and $1.2\,\mu\mathrm{m}$ were employed to generate and detect ultrasonic pulses between 200 MHz and 1.7 GHz with a phase sensitive homodyne technique [26]. Sample cylinders were mounted in a copper clamp and the transducers were contacted by spring loaded probe pins. Longitudinal polarization at 1.44 GHz was measured in reflection with only one transducer. All measurements with transverse polarization were carried out in transmission with a transducer at either end. This setup allows for the use of cryogenic amplifiers and reduced noise.

IV. RESULTS AND DISCUSSION

Measurements of the temperature dependence $\delta v/v$ of the sound velocity of transverse ultrasound are shown in Fig. 3. At these frequencies in the GHz range all features caused by TSs with a broad distribution of parameters as discussed in the previous sections are clearly observed. Although both resonant and relaxation mechanisms contribute to the acoustic susceptibility the sound velocity at the lowest temperature is higher in the normal state than in the superconducting state. The onset of the relaxation process by quasiparticles in the superconducting state is testified by the pronounced maxima between 0.6 and 0.9 K depending on frequency. Maxima at 2 K demonstrate the frequency independent transition from electron dominated to phonon dominated relaxation which has been found and described in earlier experiments [7,27]. Over some temperature ranges the sound velocity varies logarithmically with temperature. The 'slopes' of the $\log T$ dependencies are a measure of the TSs' density of states as will be explicated below.

In a first step of analyzing the data the frequency dependence of the $\delta v(T)/v$ maximum in the superconducting state of the sample is considered. At temperatures below the maximum, the contribution of the longitudinal susceptibility (*relaxation process*) is negligible and $\delta v(T)/v$ is solely determined by the transverse susceptibility (*resonant process*). The

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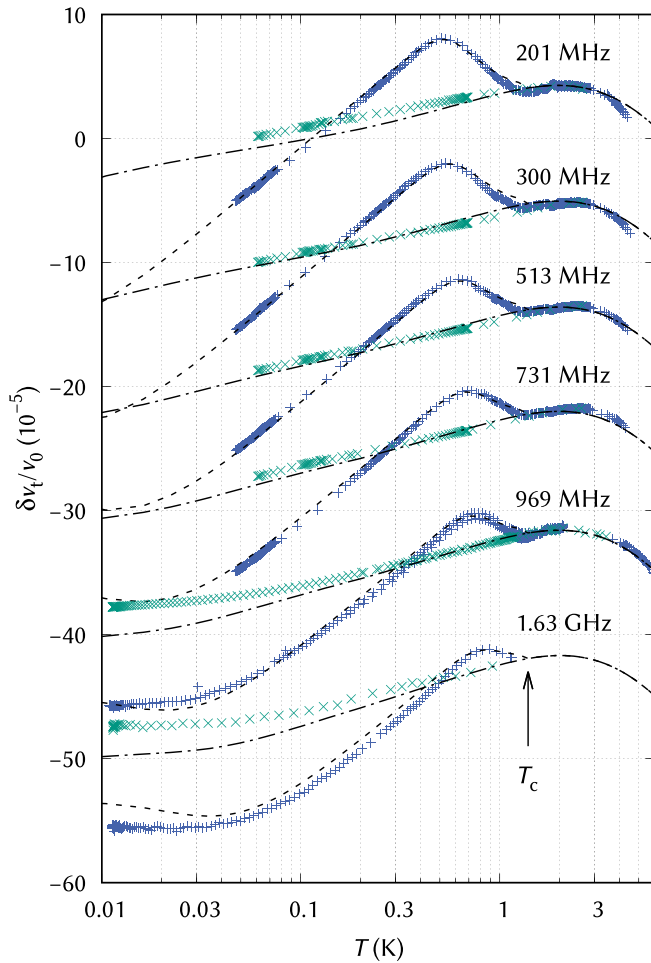[3]Lapping was done by Stähli-Läpptechnik, Weil im Schönbuch, Germany

FIG. 3. Relative temperature variation of the transverse sound velocity of glassy $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ measured at frequencies between 200 MHz and 1.63 GHz. Blue crosses are data without magnetic field where the sample is superconducting below $T_c = 1.39$ K. A magnetic field of 4 T keeps the sample normal conducting at all temperatures (green ×). Dashed lines result from numerical calculations using always the same parameters as detailed in the text. Sets of data and curves for different frequencies are shifted for clarity.
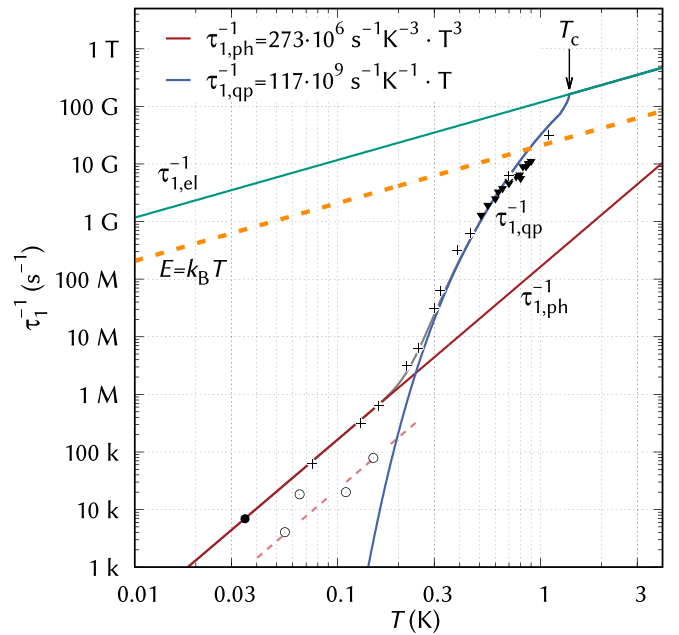

FIG. 4. Energy relaxation rate of TS in the metallic glass as extracted from the onset of the relaxation process related to respective maxima of $\delta v/v$. The energy relaxation rates τ_1^{-1} of the fastest, symmetric thermal TS due to interaction with phonons (red), thermally activated quasiparticles (blue), and electrons in the normal conducting state (green) are adjusted to the data. The dashed (orange) line indicates the energy splitting of thermal TS with $E \approx k_B T$. Low frequency data are determined by vibrating reed experiments, high frequency by pulsed ultrasound. Full symbols denote data of samples machined from a bulk metallic glass rod of $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$; open symbols result from of a splat quenched sample of $Zr_{46.8}Ti_{8.2}Cu_{7.5}Ni_{10}Be_{27.5}$. For crosses see text.

contribution of the longitudinal susceptibility becomes important when the TSs' energy relaxation rates become larger than the frequency of the applied external field, $\tau_1^{-1} \gtrsim \omega$, yielding a maximum in $\delta v(T)/v$ when the contributions per temperature interval of the two mechanisms cancel each other. Thus we may plot the frequency of the ultrasound versus the temperature of the maximum in Fig. 4 to extract the temperature dependence of the energy relaxation rate of thermal TSs. The resulting rate varies rapidly with temperature between 0.5 and 0.9 K and is clearly identified with the relaxation rate of Eq. (9) caused by the TSs' interaction with quasiparticles and plotted as a blue line in Fig. 4. In a better quantitative analysis respective maxima in $\delta v(T)/v$ are extracted from numerical calculations including all contributions. Plotted as black crosses in Fig. 4, these calculated maxima clearly confirm this analysis and yield an interaction strength $K_{el} = W_{el}^2 k_B \pi / 2\hbar = 117 \times 10^9 \text{ s}^{-1} \text{ K}^{-1}$ in the superconducting state.

As already claimed in the previous section, transfer of the same value $K_{el} = 117 \times 10^9 \text{ s}^{-1} \text{ K}^{-1}$ to the normal conducting state results in relaxation rates Eq. (8) of the dominating thermal TSs which considerably exceed their energy splitting divided by h as depicted by the green line in Fig. 4. Thus their contribution to the transverse susceptibility is reduced, the sound velocity remains at higher values. Continuing this line of argument it seems plausible to introduce a regular dephasing based on TS-electron interaction with respective rates as given in Eqs. (10) and (11). In a last step the linear temperature dependence of these rates is replaced by $f(T) = T \cdot (T/T_0)^{\alpha-1}$ and it turns out that with $\alpha = 1/3$ the overall best agreement of numerical calculations with experiment is achieved. Apart from taking into account the polarization of the deformation being transverse or longitudinal, all relevant parameters concerning distribution and dynamics of the TSs are the same for the calculated graphs of Figs. 3 and 5.

Although it is evident that dephasing plays the key role in understanding the dynamics and susceptibility of TSs in metallic glasses our analysis has some deficiencies. In the normal state both resonant and relaxation contributions make up the total temperature dependence of $\delta v/v$. Experimentally $\delta v/v$ is a straight $\log T$ which is not perfectly reproduced by our calculation. This is noticeable with the 201 MHz data in
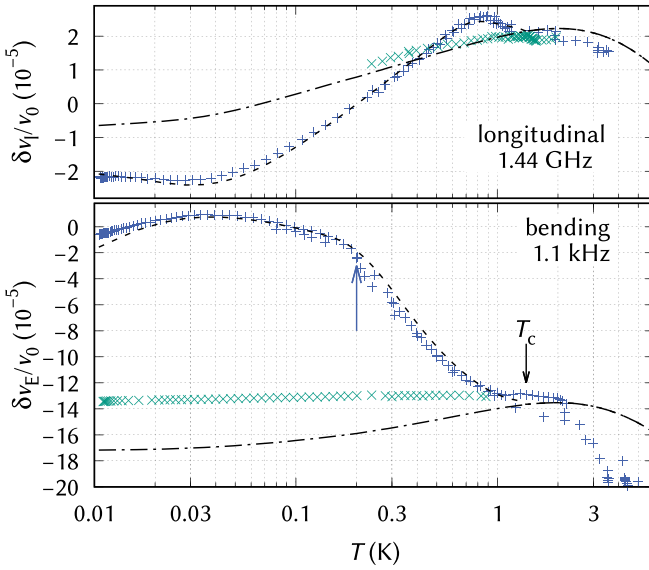
FIG. 5. Temperature dependence of the longitudinal sound velocity at 1.44 GHz (upper panel) and of the Young's modulus velocity measured by a vibrating reed [30] at 1.1 kHz (lower panel). Arrow indicates the transition from phonon to quasiparticle dominated relaxation. Dashed lines show numerical calculations.

Fig. 3, however most clearly visible for the 1.1 kHz data in Fig. 5. We will come back to this point.

In high frequency experiments a minimum of the sound velocity is expected at a temperature where $h\nu \approx 2.2\,k_BT$. In a small temperature range with $h\nu \gtrsim E \approx k_BT$ a negative contribution to the susceptibility χ'_{res} leads to an inverted temperature dependence before this mechanism becomes ineffective [1] at even lower temperatures where $k_BT \approx E \ll h\nu$. In our measurements (see Fig. 3) these minima do not appear as pronounced as predicted. The reasons might be that particularly in the superconducting state the sample was not cooled sufficiently at lowest temperatures and that in the normal state this effect is additionally smeared out by the relaxation contribution. For high frequencies, the condition $\omega\tau_1 \gtrsim 1$ may be reached after all at lowest temperatures in the normal conducting state, leading to a flattening of the relaxation contribution to $\delta v(T)/v$.

Figure 5 shows two more measurements of the sound velocity change versus temperature, one with longitudinal ultrasound at 1.44 GHz and a vibrating reed experiment at 1.1 kHz probing essentially longitudinal deformation as well. In the superconducting state the low frequency experiment exhibits a maximum around 35 mK which is identified as the onset of the relaxation contribution. Since $T \ll T_c$ the quasiparticle induced rate is certainly negligible. The one-phonon process dominates the energy relaxation of TSs and symmetric TSs have a relaxation rate $\tau_1^{-1} = 1.67\,\omega$ at the maximum. We may thus add another important data point to Fig. 4 (full circle) and with Eq. (7) we may extract the deformation potential of the TS with respect to transverse phonon modes from this experiment. We find $\tau_1^{-1} = 273 \times 10^6\,s^{-1}\,K^{-3} \cdot T^3$ for the fastest TSs corresponding to a deformation potential $\gamma_t = 0.49\,eV$. Further independent measurements of another vibrating reed sample fabricated from splat-quenched $Zr_{46.8}Ti_{8.2}Cu_{7.5}Ni_{10}Be_{27.5}$ (Vitreloy 4) [28,29] allow us to de-

termine relaxation rates between 0.6 kHz and 12 kHz in the same way (open circles in Fig. 4). The frequency dependent shift supports the T^3 increase of the relaxation rate as predicted by Eq. (7), however with a smaller γ_t.

In Fig. 4, the relaxation rate exhibits at 200 mK a transition from phonon-dominated energy relaxation of the TSs to relaxation dominated by the interaction with quasiparticles. This is clearly reproduced in the $\delta v(T)/v$ measurement at 1.1 kHz (see Fig. 5) as a distinct change of the temperature dependence marked by an arrow.

Quantitative inspection of the sound velocity measurements (Figs. 3 and 5) in the superconducting state allows us to extract the density of states of TSs. Analytical integration of the susceptibility's resonant contribution χ_{res}^{-1} with $P(E, \Delta)$ yields a logarithmic temperature dependence of $\delta v/v$ for $T > h\nu/2.2k_B$ in agreement with experiments as long as the contribution of relaxation is negligible [1]

$$\left.\frac{\delta v(T)}{v}\right|_{res} = \frac{P_0\gamma_{l,t}^2}{\rho v_{l,t}^2} \cdot \ln\frac{T}{T_0}. \tag{16}$$

Numerical calculations reproduce this behavior most clearly for the measuring frequencies between 201 and 513 MHz. The magnitude of the logarithmic slope $C_{l,t} = P_0\gamma_{l,t}^2/\rho v_{l,t}^2$ contains the mass density of the material [21] $\rho = 6592\,kg/m^3$ and the transverse and longitudinal sound velocities $v_t = 2182\,m/s$ and $v_l = 4741\,m/s$ which were determined independently. From the transverse ultrasound data in the superconducting state of Fig. 3 we extract an average and frequency independent $\ln(T/T_0)$ slope of $C_t = 5.9 \times 10^{-5}$, and knowing γ_t from the vibrating reed measurement we find $P_0 = C_t\rho v_t^2/\gamma_t^2 = 3.0 \times 10^{44}\,J^{-1}m^{-3}$. Analog evaluation of Fig. 5 yields $C_l = 2.07 \times 10^{-5}$ for longitudinal ultrasound. The vibrating reed experiment allows us to extract a value $C_E = 4.17 \times 10^{-5}$ (related to the Young's modulus deformation field) from the total $\delta v(T)/v$ decrease between 40 mK and T_c dominated by the relaxation contribution to χ' and also but minor importantly by a decreasing resonant contribution. C and P_0 values resemble those of other dielectric and metallic glasses [23,31,32].

Closer inspection of $\delta v(T)/v$ in the superconducting state reveals that the $\ln(T/T_0)$ behavior has a slight upward bend. Quantitatively for the 969 MHz measurements (see also Supplemental Material [33]), and quite similar for other frequencies, the slope increases from $C_t = 5.2 \times 10^{-5}$ in the temperature range between 100 mK and 200 mK to about $C_t = 6.2 \times 10^{-5}$ between 300 mK and 500 mK, still well below the maximum (see Fig. 1 in Supplemental Material [33]). This suggests an equivalent increase of less than 20% of the TSs' density of states P_0 in the corresponding energy range $E \approx k_BT$. A reduced density of states at lower energies might be explained by models invoking a so-called *dipole gap* arising from dipole-dipole interaction between TSs [34,35], which becomes especially important at very low temperatures and frequencies. However, thermal conductivity measurements on another Zr based superconducting bulk metallic glass do not support the idea of a reduced density of states of TSs towards lower energies [36,37]—there seems to be no clear answer yet for this class of disordered matter.

In contrast to the slight temperature dependence of the slope in the superconducting state, a clear $\log T$ temperature dependence of $\delta v/v$ is always observed in the material's normal conducting state. It is further observed that the slope depends on frequency. It decreases by 15% when lowering the measuring frequency from 731 MHz to 201 MHz. The extremely small, though still $\log T$ temperature dependence of $\delta v/v$ at 1.1 kHz is remarkable (Fig. 5) and requires a delicate balance of resonant and relaxation contributions. Previous vibrating reed measurements on glassy PdZr and CuZr alloys [7] show an overall quite similar behavior, however, even a negative slope of $\delta v(T)/v$ is observed in a temperature range where a positive slope is expected. Quantitatively, this frequency dependence is not understood. It seems to be plausible, however, that short dephasing times disturbing the TSs' coherence and eventually leading to localization are more sensitively detected the lower the measuring frequency is. In extreme cases the still present relaxation contribution may dominate and lead to a negative temperature dependence of total $\delta v/v$.

In this last section, the attenuation of transverse ultrasound is discussed. As for the temperature dependence of the sound velocity, the attenuation due to TSs comprises both resonant and relaxation contributions, and numerical calculation requires integration of the corresponding imaginary parts of the susceptibility, χ''_{res} and χ''_{rel}.

The resonant absorption (transverse interaction) selects from the broad distribution those TSs for which $E = \hbar\omega$ and its temperature dependence reflects the respective occupation number difference $\propto \tanh(E/2k_B T)$. The temperature dependence of the relaxation absorption (longitudinal interaction)—in particular its increase when coming from low temperatures—is determined by the dominating relaxation process, caused by interaction with phonons, electrons, or quasiparticles and by the measuring frequency. In any case, $\tan\delta$ arrives at a frequency and temperature independent plateau [9,16] when the energy relaxation of the fastest TSs attains the condition $\omega\tau_1 \approx 1$.

Figure 6 shows measurements at 969 MHz and 1.63 GHz together with respective theoretical curves which are calculated with parameters γ_t and W_{el} as extracted from the $\delta v/v$ measurements. P_0 was magnified by 20%. The same parameters are used for all frequencies (more frequencies shown in Fig. 2 in Supplemental Material [33]). Constant values have been subtracted from the complete experimental data sets of the two frequencies to adjust for the zero line given by the calculation at the same time accounting for background losses. Resonant and relaxation contributions are easily identified, particularly in the superconducting state. There, the measured resonant absorption exhibits the correct temperature dependence however is smaller than predicted by roughly a factor two. This discrepancy may easily be explained by the fact that our calculations were carried out for the limit of small driving fields leaving the thermal occupation undisturbed whereas experimentally this limit was not achieved. With decreasing T the condition for a 'weak' drive becomes more and more challenging in the superconducting state as the energy relaxation time τ_1 becomes very long. Thus resonant TSs may well be partially saturated at the given ultrasound intensity resulting in reduced absorption. This is supported by
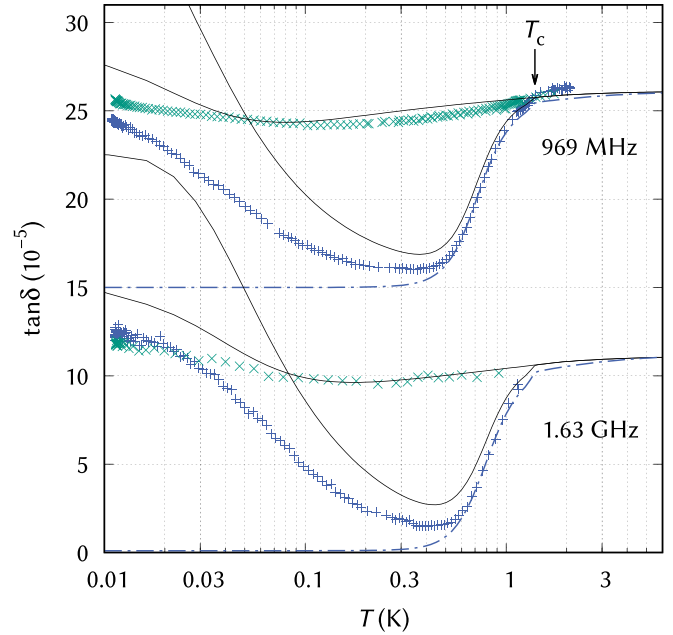

FIG. 6. Temperature dependence of the attenuation $\tan\delta$ of transverse ultrasound in the GHz frequency range. Values are determined by the squared ratio of the amplitudes of the first transmitted ultrasound pulse and a fixed reference signal. Solid lines are calculated for weak excitation using values for W_{el} and γ_t derived from the temperature dependence of the sound velocity. For P_0 values see text. The acoustic intensity for the experiment was not at the weak excitation level. Dash-dotted lines show calculations of only the relaxation contribution of the superconduction sample. Experimental data of both frequencies is vertically shifted to adjust for the zero line of the calculated pure relaxation contribution. 969 MHz data are shifted by another 15×10^{-5}.

the observation of distorted pulse shapes indicating that steady state conditions are not achieved within the duration (typically 500 ns) of a single ultrasound pulse. For a comparison, ultrasound echo experiments [38] on amorphous $Pd_{30}Zr_{70}$, the first direct measurements of energy relaxation and phase coherence times of a superconducting metallic glass, yield $\tau_1 \approx 30\,\mu s$ and $\tau_2 \approx 5\,\mu s$ at 20 mK for TS resonant with the ultrasonic frequency of 0.86 GHz. Phonon echo experiments on $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ are presented in the Supplemental Material [33,39–41].

The relaxation absorption in the superconducting state is characterized by a steep rise between 0.5 and 1 K caused by the rapidly increasing quasiparticle density when approaching T_c. Measurement and calculation agree very well. In the normal conducting state the absorption depends only weakly on temperature. As expected, the plateau of the relaxation absorption extends to low temperatures due to the slow variation of the dominating contribution of the electron bath to τ_1^{-1}. Below 200 mK, a clear however small increase of the absorption is observed indicating a resonant contribution. This behavior is remarkably well reproduced by our calculations. At first glance, one is led to expect a rather large resonant absorption since fast relaxation times would leave the TSs preferentially in their ground states except for extremely high ultrasonic intensities. However, caused by short dephasing times or, respectively, large linewidths $h\tau_2^{-1}$ the resonant con-

tribution to χ'' is largely suppressed. This corresponds to the reduced slope of $\delta v(T)/v$.

V. CONCLUSIONS

This paper deals with low temperature ultrasound experiments on metallic glasses and their interpretation in terms of the elastic susceptibility of atomic two-level tunneling systems. It turns out that the standard tunneling model in its simplest version fails to describe the experiments properly, which is most evident for the temperature dependences of the sound velocity in the sample's superconducting state in comparison with its normal state. The simple analysis does not predict the experimentally observed crossing.

In previous publications it was suggested that interaction with conduction electrons might in general reduce the density of states (DOS) of the tunneling systems thus leading to an overall smaller temperature variation of the sound velocity in the normal state compared to the superconducting state. This idea, however, is ruled out by the relaxation contribution to the attenuation $\tan \delta$ which obviously has the same absolute value in the normal state and the superconducting state below T_c, however still in the plateau region (this is even better visible at frequencies between 201 and 731 MHz in Supplemental Material [33]). In the present study, we show that another basic assumption of the treatment within the standard tunneling model is not fulfilled: Caused by their coupling to the bath of electronic excitations the tunneling systems no longer have linewidths that are much smaller than their energy. This does not affect the relaxation contribution however considerably reduces the resonant contribution to the susceptibilities in the normal conducting metallic glass.

Our numerical calculations employ the 'classical' approach for the resonant response of ensembles of two-level systems as given by Eqs. (3) and (4) although this might be inadequate for extremely broad linewidths caused by very high dephasing rates τ_ϕ^{-1}, which rather lead to localization and destruction of the quantum two-level nature. With $\tau_\phi^{-1} \propto T(T/T_0)^{-2/3}$, which emphasizes dephasing towards lower temperatures, we achieve very good overall agreement of our calculations with experiment, at least in the GHz frequency range. It is obvious that dephasing has to be consequently incorporated in such calculations. Usually, $\tau_2^{-1} \ll E/h$ is taken for granted. However, strong dephasing removes a considerable fraction of TS from contributing to χ_{res}. A similar idea has been put forward for insulating glasses [42]. It was suggested that strain mediated TS-TS interaction causes loss of coherence and a transition to incoherent tunneling, which finally leads to a reduced contribution to χ_{res}.

Here, we have to ask: Which mechanism provides the temperature dependence of τ_ϕ^{-1} in a metallic glass, only in the normal state? It is unlikely that electronic excitations are directly responsible for a dephasing rate varying other than linearly with temperature. Instead, we propose a dephasing mechanism that involves other TSs surrounding those which contribute to the resonant part of the susceptibility. We further propose that this TS-TS interaction is indirect and mediated by conduction electrons. Since the electron wave functions themselves become more coherent with decreasing temperature an increasing number of TSs will be within the volume defined by the respective phase coherence length and contribute to the dephasing of those resonant TSs. It is thought that both the TS-electron and the associated electron-TS interactions are longitudinal and combine to produce the suggested indirect TS-TS interaction.

It is evident that the key for understanding the low temperature acoustic susceptibility of metallic glasses are the extremely high dephasing rates of TSs, the dynamics of which are largely determined by their interaction with conduction electrons. It is as well evident that the experiments at audio frequencies call for an additional explanation. As stated above, the straight $\log T$ dependence of $\delta v/v$ in the normal state at any frequency deserved particular attention. In any case, already Black[4] expected a region of *unusual behavior* when $E < h\tau_2^{-1}$.

Last but not least we like to note that the analysis of our measurements supports quite generally the assumptions of the standard tunneling model including in particular the suggested distribution of relevant parameters. In this sense our work seems to constitute a good example of a 'smoking gun' experiment as was recently asked for by Leggett and Vural [43]. While the energy space is explored by the temperature from 10 mK up to several Kelvin, the dynamics of these tunneling systems is measured at kHz and at GHz frequencies. Moreover, the dynamcis (τ_1 and τ_2) of TSs with same energy E are vastly different whether the sample is superconducting or normal conducting.

ACKNOWLEDGMENTS

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[4]Fig. 8.7 in the book chapter [10].

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