Electron renormalization of sound interaction with two-level systems in superconducting metglasses

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The crossing of temperature dependencies of sound velocity in the normal and the superconducting state of metallic glasses indicates renormalization of the intensity of sound interaction with two-level systems (TLS) caused by their coupling with electrons. In this paper we have analyzed different approaches to a quantitative description of renormalization using the results of low-temperature ultrasonic investigation of Zr₄₁₋₂Ti₁₃.₈Cu₁₂.₅Ni₁₀B₂₂.₅ amorphous alloy. It is shown that the adiabatic renormalization of the coherent tunneling amplitude can explain only part of the whole effect observed in the experiment. There exists another mechanism of renormalization affecting only nearly symmetric TLS.

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1. INTRODUCTION

The well known tunneling model (TM) utilizes only two basic parameters for the description of low-temperature (T ≲ 1 K) behavior of the velocity v and the attenuation Γ of sound in metallic glasses. A parameter $C_0 = \rho \gamma^2 / \rho\omega^2$ is the density of states of two-level systems (TLS), γ is the deformation potential, ρ is the mass density and $\gamma$ defines the scale of variations of v and Γ in presence of TLS. A parameter $\eta = n_0 \sqrt{v_{kk}'}$ ($n_0$ is the density of electron states at the Fermi level, $v_{kk'}$ is the mean square of matrix element of electron-TLS scattering from k to k' state) determines the TLS relaxation rate due to their interaction with electron environment. According to the TM, a TLS contribution to the acoustic characteristics is determined by two additive mechanisms – the resonance and the relaxation ones. Under usual experimental conditions, $\omega ≪ T$, where $\omega$ is the sound frequency (we use the system of units where $h = k_B = 1$), the resonance contribution to the variation $\delta v(T)$ of sound velocity is always negative and represents a straight line with the unit slope in coordinates $\delta v / C_0 v$ versus ln T. The relaxation contribution is also always negative and linear with the slope $-1/2$ in the same coordinates. Thus, the resulting dependence $\delta v(\ln T) / C_0 v$ is expected to be a straight line with the slope 1/2. The attenuation of sound is determined mainly by the relaxation interaction and is virtually independent on T, whereas the resonance contribution into Γ is small ($\sim \omega / T$).

In superconducting glasses at low enough temperatures $T \ll T_c$ the relaxation interaction is frozen out. This allows to extract purely resonance contribution and therefore to verify many of TM conclusions. Acoustic measurements in superconducting metglasses $\text{Pd}_{30}\text{Zr}_{70}$, $\text{Cu}_{30}\text{Zr}_{70}$ and (Mo₁₋ₓRuₓ)₀.₈P₀.₂ carried out more than a decade ago, revealed some considerable deviations from the predictions of the TM:

i) the slope of the straight line $\delta v_n(\ln T) / C_0 v$ in the normal (n) phase is about 1/4 whereas the TM canonical slope is 1/2;

ii) at least at high frequencies (HF), the normal state line $v_n(T)$ crosses the superconducting (s) line $v_s(T)$ at $T_C \ll T_c$. From the TM point of view, this is impossible in principle;

iii) $v_s(T)$ is smaller than $v_n(T)$ just below $T_c$. This effect was observed in both low frequency (LF) vibratingreed experiments and HF experiments. According to the original TM, the sound velocity would always increase below $T_c$;

iv) the sound attenuation reveals an analogous anomaly: $\Gamma_s(T)$ exceeds $\Gamma_n(T)$ within a certain temperature interval, which is about $T/T_c > 0.8$ in HF measurements and extends down to $T/T_c > 0.05$ in LF experiments. In contrast, the TM predicts the attenuation to be nearly independent on T (with small $\delta T / T > 0$) as long as the maximum relaxation rate $\nu$ exceeds $\omega$. Thus, the attenuation in LF experiments should be insensitive to the superconducting transition at all, whereas in HF measurements $\Gamma_s$ should either be temperature-independent just below $T_c$ of decrease in metglasses with low enough $T_c$ (or at high enough frequencies).

It was supposed in Refs. that all (or most of all) deviations from the TM are related to the electron renormalization of the parameter C with respect to its bare value $C_0$. Although possible mechanisms of this renormalization were not discussed in Refs., the phenomenological consideration is rather simple. Indeed, assume that C decreases due to the interaction of TLS with the electron excitations. As a result, the slope of $v_n(\ln T)$ decreases also. On the other hand, the bare value $C_0$ should retrieve far below $T_c$. Therefore, the ratio of slopes $v_n(\ln T)$ and $v_s(\ln T)$ becomes smaller than the canonical TM value 1/2. An additional assumption that the parameter C grows more rapidly just below $T_c$ than the relaxation interaction is frozen out, leads to a simple explanation of the items iii) and iv). A connection between the item ii) and the renormalization of C
is less obvious. Nevertheless, we will demonstrate that the crossing between \( v_n(T) \) and \( v_s(T) \) at \( T_c \ll T_c \) is the most convincing evidence of reduced effective value of \( C \) in the \( n \)-phase in comparison with the \( s \)-phase.

The arguments in favor of the electron renormalization hypothesis have been already presented in comparatively early theoretical works devoted to both the general problem of tunneling with dissipation and a more detailed analysis of the TLS interaction with surrounding electrons (see Refs. 8 and references therein). However, any relations allowing to make a comparison with the experiment at a quantitative level were not derived in these works.

For the first time, a straightforward theoretical analysis of the problem of the electron renormalization of the sound-TLS interaction in metallic glasses was made in Ref. 8. It was argued that one of the reasons for the decrease of \( C \) in the presence of electrons is an adiabatic renormalization of the coherent tunneling amplitude. Moreover, in order to estimate this effect, it is not necessary to introduce any additional parameter since the renormalization \( (C_0 - C)/C_0 = \eta^2/4 \) is determined by the same interaction constant \( \eta \) (see also Ref. 9). Although the theory gives some opportunity to examine its conclusions quantitatively, such procedure was not accomplished, probably because of lack of detailed experimental data.

In the present work we test different approaches to the quantitative analysis of the sound velocity and attenuation in metal glasses using experimental results obtained on the superconducting amorphous Zr\(_{41.2}\)Ti\(_{13.8}\)Cu\(_{12.5}\)Ni\(_{10}\)Be\(_{22.5}\) alloy as an example. It is shown that the adiabatic renormalization solely does not allow to describe all the experimental results, and there exists an additional mechanism of renormalization.

II. EXPERIMENTAL RESULTS

The alloy under investigation has a high resistance with respect to crystallization in the state of overcooled melt and remains amorphous at extremely low cooling rate (\(< 10 \text{ K s}^{-1}\)). This makes it possible to obtain bulk homogeneous samples, which suit perfectly the acoustic measurements. The ultrasonic experimental technique is described elsewhere.

Figure 1 shows typical temperature dependence of the velocity of the transverse sound wave in Zr\(_{41.2}\)Ti\(_{13.8}\)Cu\(_{12.5}\)Ni\(_{10}\)Be\(_{22.5}\) in the \( n \)- and \( s \)-states. The \( n \)-state measurements were carried out at the magnetic field \( B = 1.5 \pm 2.5 \text{ T} \). In accordance with the TM, the curves \( v/(\ln T) \) represent almost straight lines in both the \( n \)- and the deep \( s \)-state. The growth of \( v_s \) below \( T_c \) reflects freezing out of the relaxation component and agrees with the TM conception, with the constant \( C_0 = (2.85 \pm 0.05) \cdot 10^{-5} \) determined from the slope of \( v_s(\ln T) \) at \( T < 0.3 \text{ K} \).

![Figure 1](image1.png)

**FIG. 1.** Temperature variations of the velocity of transverse sound in Zr\(_{41.2}\)Ti\(_{13.8}\)Cu\(_{12.5}\)Ni\(_{10}\)Be\(_{22.5}\) alloy in the superconducting and normal phases. Inset: normalized velocity of transverse (upper trace, \( C_n = 6.94 \cdot 10^{-6} \)) and longitudinal mode (lower trace, \( C_n = 2.75 \cdot 10^{-6} \)) near \( T_c \). The curves were aligned in the normal phase.

![Figure 2](image2.png)

**FIG. 2.** Temperature variations of magnetic susceptibility and sound velocity in Zr\(_{41.2}\)Ti\(_{13.8}\)Cu\(_{12.5}\)Ni\(_{10}\)Be\(_{22.5}\) in the vicinity of \( T_c \). Open and solid circles: \( B = 0 \) and \( B = 1.5 \text{ T} \). Solid lines: calculations for \( \eta = 0.65, \varepsilon = 1.2, \omega_0 = 0.5 \), \( R_0 = 0.14, T_c = 0.83 \text{ K} \). Thick vertical mark shows noise level. Experimental data was smoothed by adjacent averaging.

There are also obvious deviations from the TM: the ratio of slopes in the \( n \)- and the \( s \)-phases differs from its
canonical value 1/2 and is close to 1/4, and the curves \( v(T) \) for both phases intersect at some temperature \( T_c \). Such effects have been observed before in Pd\(_{36}\)Zr\(_{70}\). The value of \( T_c \) is frequency-dependent; particularly, \( T_c(62 \text{ MHz}) \approx 0.055 \text{ K} \) and \( T_c(186 \text{ MHz}) \approx 0.11 \text{ K} \) were found.

The inset to Fig. 1 shows the variations \( v_{s} \) and \( v_{n} \) in the vicinity of \( T_c \) and the diamagnetic response of the sample on the ac magnetic field \( H = 10^{-6} \text{ T} \) at the frequency of 22 Hz. The magnetic susceptibility \( \chi \) was measured simultaneously with \( v_{s} \) that provides coincidence of temperature scales for both measurements. The presence of two steps in \( \chi(T) \) indicates that the sample contains at least two phases with different temperatures of the superconducting transition: \( T_{cm1} \approx 0.9 \text{ K} \) and \( T_{cm2} \approx 1.0 \text{ K} \). An increase of the ac field \( H \) up to \( 10^{-5} \text{ T} \) leads to complete suppression of the anomaly in the diamagnetic response at \( T_{cm2} \), although the jump at \( T_{cm1} \) survives up to \( H = 10^{-4} \text{ T} \).

![Diagram](image)

**FIG. 3.** Normalized attenuation of the transverse sound wave versus temperature in Zr\(_{41.2}\)Ti\(_{13.8}\)Cu\(_{12.5}\)Ni\(_{10}\)Be\(_{22.5}\). Open and solid circles: \( B = 0 \) and \( B = 2.5 \text{ T} \). Solid lines: calculated dependencies for \( \eta = 0.65 \), \( T_c = 0.83 \text{ K} \). Inset: to determination of the superconducting energy gap and the parameter \( \eta \). The results are presented for three values of the background level of attenuation: \(-0.3\%\), \(0\%\) and \(+0.3\%\) of the total change of attenuation between the \( n\)- and \( s\)-phase, from bottom to top curve.

It is of interest to note that the temperature \( T_c \approx 0.83 \text{ K} \), at which one can first register a nonzero difference \( v_{s} - v_{n} \), coincides neither with \( T_{cm1} \) nor with \( T_{cm2} \). This was interpreted in Ref. [14] as a fingerprint of possible gapless superconductivity within the temperature region between \( T_{c} \) and \( T_{cm1} \). It is known that the magnetic scattering, which is the most prevalent reason of the gapless regime, also reduces the energy gap when the latter opens. However, we found the energy gap in our alloy to be close to the BCS value (see below) that allows us to reject this interpretation. Apparently, the diamagnetic anomalies are related to some surface phases with higher transition temperatures.

The temperature dependence of the sound attenuation is shown in Fig. 3. The experimental data are normalized over \( \Gamma_{n}(T_{c}) \) which can be determined from the variations of sound amplitude between \( T_{c} \) and deep \( s\)-state. The behavior of \( \Gamma(T)/\Gamma(T_{c}) \) does not show any noticeable difference with similar dependencies in other superconducting amorphous alloys and reflects evolution of the relaxation contribution to the attenuation, in accordance with the TM conception.

### III. QUALITATIVE CONSIDERATION

Before making quantitative estimations, we shall discuss qualitatively a possible origin of the peculiarities of the sound velocity in superconducting metglasses.

It is naturally to associate the crossing of \( v_{s}(T) \) and \( v_{n}(T) \) with a growth of \( C \) in the \( s\)-phase as a result of suppression of the electron renormalization at energies smaller than the superconducting gap. To validate this assumption, we address the expression for the resonant contribution of TLS to the sound velocity:

\[
\left( \frac{\delta v(T)}{v_{s}} \right)_{res} = P \int_{0}^{\infty} \frac{CE \tanh(E/2T)}{\omega^2 - E^2} dE. \tag{1}
\]

In the simple case of energy-independent \( C = C_{0} \), in order to avoid a formal logarithmic divergence of Eq. (1) at the upper limit, the velocity variations are usually considered with respect to some arbitrary reference temperature \( T_{0} \):

\[
\left( \frac{v(T) - v(T_{0})}{v(T_{0})} \right)_{res} = C_{0} \ln \frac{T}{T_{0}}, \quad T > \omega. \tag{2}
\]

However, if the value of \( C \) varies with energy and/or temperature, Eq. (2) is inapplicable even for qualitative estimates since the reference value \( v(T_{0}) \) may also change with \( C \). To account correctly for the changes of \( C \), it is necessary to analyze the complete integral of Eq. (1) by introducing a cutoff energy \( E_{m} \) (say, of the order of melting temperature or glass transition temperature). In the case of \( C = \text{const} \), Eq. (1) can be approximated within the logarithmic accuracy by the following piecewise-linear dependence (line 1 in Fig. 4):

\[
\left( \frac{\delta v(T)}{C_{0}v} \right)_{res} = \begin{cases} \ln(\omega/E_{m}), \quad T \leq \omega \\ \ln(T/E_{m}), \quad T \geq \omega \end{cases}. \tag{3}
\]
Here we neglect insignificant small variations in $v$ at $T \lesssim \omega$: a quadratic fall near $T = 0$ and a shallow minimum at $\omega = 2.2T$ arising from the analytical solution of Eq. (1).}

The total TLS contribution to the sound velocity in the $n$-phase is schematically shown in Fig. 4 with a line 3 which was obtained in the following way. At $T < E_k$ the line 3 contains a piece of line 3a drawn with a slope $1/2 - R$ through the point $T = \omega/\eta^2$ at the line 2. At $T > E_k$, the line 3 is a piece of line 3s drawn with a slope $1/2$ from the point $T = \omega/\eta^2$ at the line 1. A transition range arises at temperatures $T \approx E_k$.

Let us now discuss the evolution of $v(T)$ at the superconducting transition. First, we consider a low-temperature range $T \lesssim 0.3$ K. If $E_k \leq 2\Delta_s(0)$ (here and below $\Delta_s(T)$ is the superconducting energy gap), the renormalization of $C$ should be frozen out. Thus, the resonance contribution can be depicted by a low-temperature part of the straight line 1a. It is clear that the intersection of lines 3 and 1a is possible only under the condition $C < C_0$ ($R \neq 0$). From the geometry of Fig. 4 it is easy to estimate $T_{cr}$:

$$(1-S) \ln T_{cr} = R \ln E_k + (1-R-S) \ln(\omega/\eta^2) - A(T_{cr}),$$

where $S$ is the resulting slope of $v_T(\ln T)$. The parameter $A$ is introduced to account for possible shift of a background level of the sound velocity in the $s$-phase with respect to the $n$-phase normalized over $C_0$ and will be discussed below. It can be seen from Eq. (5) that $T_{cr}$ grows with $E_k$, $\omega$, and with the decrease in $\eta$.

At $E_k > 2\Delta_s(0)$ the renormalization of $C$ is only partially frozen out for $E < 2\Delta_s(0)$. Therefore, the sound velocity in the deep $s$-state can be depicted as a part of the straight line 1b (see Fig. 4). The estimate of $T_{cr}$ by Eq. (5) is also valid in this case, only $E_k$ should be replaced by $2\Delta_s(0)$.

Along this line of reasoning, we can also qualitatively explain the behavior of $v$ and $\Gamma$ at the superconducting transition. Below $T_c$, the electron renormalization rapidly reduces, and the effective $C$ grows providing the decrease in $v$ and the increase in $\Gamma$. However, a competitive effect arises simultaneously: the rate $\nu$ of the TLS relaxation on electrons falls and therefore changes $v$ and $\Gamma$ in the opposite direction. Thus, if the phonon relaxation predominates near $T_c$, the effective $\nu$ changes weakly, and the sound velocity will decrease (correspondingly, the attenuation will increase) below $T_c$, as it was observed before. If the electron relaxation prevails (for materials with lower $T_c$, like our system), the changes of $v$ and $\Gamma$ near $T_c$ may have any sign, depending upon the relations between $T_c$, $E_k$ and $\omega$.

In principle, one can propose an alternative explanation of the crossing. In our previous consideration, we silently assumed the count level of the TLS contribution into the sound velocity to be the same for both the $n$- and $s$-states. Generally, this is not the case, and the sound velocity changes at the superconducting transition with no account of the TLS-related mechanisms. For example, in pure metals a decrease in electron viscosity below $T_c$ leads to the change of dislocation contribution to the sound velocity of the order of $10^{-5}$ (Ref. 16) which is comparable with the TLS contribution but is undoubtedly

![Diagram](image-url)
absent in an amorphous metal. A more general mechanism is the change of electron contribution into elastic moduli of a metal in the s-phase. In disordered metals with a short electron mean free path, this change is usually small ($\sim 10^{-6}$) but in certain cases, for instance, in Al-15 compounds close to structural instability, it may achieve much larger values $\sim 10^{-4}$ (Ref. [3] of arbitrary sign. If we accept such a scale for the decrease in the electron contribution in the s-state of our sample, the crossing will arise without any renormalization effects. In its turn, the anomalous slope ratio may be attributed to enhanced density of states of asymmetric TLS playing the principal role in the sound attenuation. Although the latter assumption contradicts the basic TM postulate about constancy of $\eta$ within a wide range of tunnel parameters, we can not reject straight away the discussed alternative without additional argumentation presented below.

Thermodynamic treatment shows that the electron contribution variations in the s-state are independent on the sound frequency and lead to a jump in derivatives $dv_i/dT$ at $T = T_c$ for both longitudinal and transverse modes proportional to $\delta^2 T_c/\delta e_i^2$ where $e_{i,t}$ are corresponding deformations. A small jump of $v_i$ itself can be also expected at $T = T_c$. As the temperature decreases, the electron contribution changes as the density of a superfluid condensate, so that its variations become negligibly small at $T \ll T_c$. By making use of Eq. (5) at $R = 0$ and the measured values of $S \approx 0.28$, $C_0$, $\eta \approx 0.65$ and $T_c(26$ MHz), the corresponding shift of the sound velocity between the n- and the deep s-state can be estimated as $\delta v/v \sim 5 \cdot 10^{-5}$. This value is comparable with the resulting velocity change $3 \cdot 10^{-5}$ for t-mode (see Fig. 1) between $T_c$ and the maximum in $v(T)$, i.e., the electron and the TLS contributions appear to be of the same order but have opposite signs. However, since the normalized TLS contribution is independent on the polarization, the data presented in the inset in Fig. 1 demonstrate that the same independence should take place for the electron contribution, i.e., the condition $(1/\gamma^2) \delta^2 T_c/\delta e^2 \approx (1/\gamma^2) \delta^2 T_c/\delta e^2$ must be satisfied. The latter does not follow from theory and can be only a result of random coincidence that is hardly possible. Thus, we conclude that the scale of temperature variations of the TLS contribution much exceeds that connected with the electron mechanisms. Furthermore, since the velocity shift $A$ is frequency-independent, the following expression derived from Eq. (5)

$$(1 - S) \ln T_{cr}(\omega_1)/T_{cr}(\omega_2) = (1 - R - S) \ln \omega_1/\omega_2$$

shows that in the absence of renormalization ($R = 0$) $T_{cr}$ must be proportional to $\omega$ that contradicts our experimental data. In that way, the absence of such proportionality is the most clear evidence of renormalization of the parameter $C$ irrespectively of whether a certain additional sound velocity shift between n- and s-phases exists or not.

## IV. SOME RESULTS OF THE TUNNELING MODEL

In this Section we present a brief overview of basic results of the TM, which describe the behavior of $v(T)$ and $T(T)$ in glasses with account of the dependence of $C$ on tunnel parameters and were used in our numerical calculations. We also discuss modifications introduced into given relations for a more exact account for the TLS-electron coupling.

The main postulate of the TM is a statement of the existence of double-well potentials in glasses with the tunnel coupling between wells. The density of states of TLS is constant in the space of parameters $\zeta$, $\ln \Delta_0$ where $\zeta$ is the asymmetry of the double-well potential and $\Delta_0$ is an amplitude of the coherent tunneling. In order to determine the response of the TLS ensemble on an external field, it is necessary to perform an averaging over $\xi$ and $\ln \Delta_0$, which appears to be more convenient in variables $E = \sqrt{\xi^2 + \Delta_0^2}$ and $u = \Delta_0/E$. In this representation, the TLS density of states is independent on $E$:

$$g(E,u) = \frac{\pi}{u \sqrt{1 - u^2}} \equiv g(u).$$

The relationships which determine the TLS contribution to the sound velocity and attenuation read:

$$\left(\frac{\delta v(T)}{v}\right)_{\text{res}} = -\int_0^{E_m/T} \tan \left(\frac{\zeta}{2}\right) \frac{d\zeta}{\zeta} \int_0^1 C(\zeta, u) g(u) u^2 du$$

$$\left(\frac{\delta v(T)}{v}\right)_{\text{rel}} = -\frac{1}{2} \int_0^{E_m/T} \frac{d\zeta}{\cosh(\zeta/2)} \int_0^1 C(\zeta, u) g(u) \times (1 - u^2) \frac{u^2}{\omega^2 + u^2} du,$$

$$\left(\frac{\Gamma v}{\omega}\right)_{\text{rel}} = \int_0^{E_m/T} \frac{d\zeta}{\cosh^2(\zeta/2)} \int_0^1 C(\zeta, u) g(u) \times (1 - u^2) \frac{\omega^2}{\omega^2 + u^2} du.$$
\[ \nu = \frac{\pi \eta^2}{2} u^2 T J(\varepsilon). \]  

In the n-phase \( J(\varepsilon) = J_n(\varepsilon) = (\varepsilon/2) \coth(\varepsilon/2) \), \( \nu \approx \eta^2 T u^2 \), and the relaxation interaction is essential for \( T > \omega \). In the s-state it is necessary to use a function \[ J_s(\varepsilon, \Delta) = \frac{1}{2} \int \limits_\Delta^\infty d\varepsilon' \frac{f(-\varepsilon')}{\sqrt{\varepsilon'^2 - \Delta^2}} \left( \frac{\varepsilon' - \varepsilon}{\sqrt{\varepsilon'^2 - \Delta^2}} - \frac{\Delta^2}{\varepsilon'^2 - \Delta^2} \right) \frac{f(\varepsilon' - \varepsilon)}{f(\varepsilon)} \Theta(\varepsilon^2 - \Delta^2) \theta(\varepsilon' - \varepsilon) + (\varepsilon \rightarrow -\varepsilon), \]  

where \( f(x) \) is the Fermi function, \( \Theta(x) \) is the step Heavyside function, and \( \Delta = \Delta_s(T)/T \). This integral coincides with \( J_n(\varepsilon) \) for \( \varepsilon \gg 2\Delta \), it has a discontinuity at \( \varepsilon = 2\Delta \), and \( J_s(\varepsilon, \Delta) \rightarrow 2 f(\varepsilon) \) for \( \varepsilon \ll 2\Delta \). A rapid fall in \( J_s \) below \( T_c \) leads to freezing out of the relaxation interaction when the maximal relaxation rate \( (u = 1) \) becomes smaller than \( \omega \).

A more complicated picture was revealed beyond the perturbation theory. Just at \( T = 0 \) in the n-phase, the bare amplitude of the coherent tunneling \( \Delta_0 \) is renormalized due to an adiabatic part of the interaction between the TLS and electrons:

\[ \Delta_0^* \propto \Delta_0 \left( \frac{\Delta_0}{\omega_0} \right)^4 \frac{\eta^2}{4 - \eta^2}, \]  

where \( \omega_0 \) is of the order of the Debye energy.

In the n-phase for \( T \neq 0 \) an ensemble of TLS can be divided into three energy intervals:

1. \( T \ll E^* = \sqrt{\xi^2 + \Delta_0^2} \) - coherently tunneling TLS.
2. \( E^* < T < 4E/\pi \eta^2 \) - incoherently tunneling TLS
3. \( T > 4E/\pi \eta^2 \) - low-energy TLS are relevant. In this region, the amplitude of incoherent tunneling is also \( \tilde{\Delta} \) of Eq. (14) but the factor \( (1 - \tilde{u}^2) \) in Eqs. (9) and (10) is absent because the incoherent transitions between the broadened levels take place with energy variations even in the symmetric case. The corresponding relaxation frequency varies as

\[ \nu_3 \propto \frac{2}{\pi \eta^2} T \tilde{u}^2 \sqrt{\frac{\varepsilon^2}{J(\varepsilon)}} \]  

One can think that a part of TLS with \( \tilde{E} < \sqrt{\omega T} \) should decrease its contribution to \( \Gamma \) and \( (\delta u/v)_{\text{rel}} \) due to the fall of \( \nu_3 \) for small \( \varepsilon \). However, a numerical analysis shows that this fall is compensated by the growth of influence of the symmetric TLS. As a result, partial contribution to \( \Gamma \) and \( (\delta u/v)_{\text{rel}} \) from the interval 3 virtually does not change in comparison with the original TM. The contribution of TLS from the second interval remains the same also. Only the contribution from the coherently tunneling TLS, undergoing the adiabatic renormalization, experiences an essential change. The main postulate of the TM concerning the constancy of \( \bar{\rho} \) in the space of variables \( \xi \), ln \( \Delta_0 \) remains valid. However, \( g(u^*) \), along with the parameter \( C \), acquires an additional factor \( (1 - \eta^2/4) \) under transformation to the variables \( E^* \), \( u^* \) because of a nonlinear dependence between \( \Delta_0^* \) and \( \Delta_0 \), Eq. (13). The latter changes rapidly at the superconducting transition to the linear one of the type of Eq. (14), where \( \Delta_0^* \) substitutes for \( T \), and all TM relations are restored.

V. ANALYSIS OF THE EXPERIMENTAL DATA

A. Determination of \( \Delta_s(0) \) and \( \eta \)

For the frequencies used in our experiments, a rapid freezing of relaxation interaction begins at the temperature well below \( T_c \) (see Fig. 3). In this case the renormalization of \( C \) is also frozen out and the sound attenuation \( \Gamma_s \) is described by Eq. (10) with \( C = C_0 \). According to Eqs. (10) and (11), the low-temperature part of \( \Gamma_s(T) \) should be a straight line in coordinates \( \log(\Gamma_s(T)/T) \) versus \( T^{-1} \):

\[ \frac{\Gamma_s(T)}{\Gamma_n(T_c)} = \frac{2\pi \eta^2}{3\omega} \frac{T}{e^{-\Delta_s(0)/T}}. \]

This allows us to use the sound attenuation for a simple evaluation of \( \Delta_s(0) \) and \( \eta \) from its low-temperature dependence plotted in the inset to Fig. 3. Since this construction is very sensitive to the reference level of the attenuation, we also present two additional curves for level variations of \( \pm 0.3\% \) of the whole signal change between the n- and s-states. Within this range, a large enough temperature interval exists where each curve can be well approximated by a straight line with the slope determining \( \Delta_s(0) = 1.45 \pm 0.05 \) K. If we accept \( T_c = 0.83 \) K, this value agrees well with the BCS relation \( \Delta_s(0)/T_c = 1.76 \) which was used in all further calculations.

The value of \( \eta = 0.55 \pm 0.15 \), determined by crossing of the approximating straight lines with the ordinate axis for different curves in the inset to Fig. 3, reveals a large spread due to its exponential dependence on the position of the crossing point. A more accurate estimate of \( \eta \) can be obtained from the numerical analysis of the attenuation within the whole temperature region of Fig. 3. By matching the most sharp part of \( \Gamma_s(T) \), calculated from Eq. (10) at \( T_c = 0.83 \) K, with the experimental dependence, we found \( \eta = 0.65 \pm 0.05 \), in agreement with previous rough estimate.
B. Sound attenuation near \( T_c \)

The analysis performed above shows that the temperature dependence of the sound attenuation in \( \text{Zr}_{31.2}\text{Ti}_{13.8}\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{2.5} \) can be rather well described by the original TM. However, within some temperature range just below \( T_c \), the behavior of \( \Gamma_s \) reveals anomalies which find no explanation in the original TM.

\[
\frac{\Gamma(T)}{\omega C_0} = 0.46 - 0.02 T \quad \text{at } T < 0.8 \quad \text{and } \quad 0.46 - 0.05 T \quad \text{at } T > 0.8
\]

\[
54.3 \text{ MHz} \quad T_c
\]

**FIG. 5.** Comparison of sound attenuation near \( T_c \) at 54 MHz with model calculations. Open and solid circles: \( B = 0 \) and \( B = 1.5 \text{ T} \). Lines: related calculation for the original TM (set 1 of lines) and the TM with account for the adiabatic renormalization of the coherent tunneling amplitude, \( \eta = 0.65, \epsilon_b = 1.2 \) (set 2). Thick vertical mark shows noise level, data was smoothed by adjacent averaging.

Figure 5 shows variation of \( \Gamma(T) \) in the vicinity of \( T_c \) obtained with higher resolution than in Fig. 3. The main peculiarities of \( \Gamma_s(T) \) can be discovered by means of a comparison of the experimental curves with those calculated in the frame of the original TM (line set 1 in Fig. 5). According to the calculation, a fall in the attenuation begins just at \( T_c \) with growing slope at low temperatures. The experimental dependence has quite different behavior: \( \Gamma_s(T) \) does not vary at \( T_c \) within the experimental resolution and tends to exceed \( \Gamma_n(T) \) at lower temperatures. A more prominent excess of \( \Gamma_s(T) \) over \( \Gamma_n(T) \) starting just at \( T_c \) has been previously observed in \( \text{Pd}_{30}\text{Zr}_{70} \) alloy.

The proximity of \( \Delta_s(0)/T_c \) to the BCS value, as well as the lowered temperature of superconducting transition of the main volume of the sample in comparison with \( T_{cm1} \) (see Fig. 2) shows that the explanation of \( \Gamma_s(T) \) features by the magnetic depairing is irrelevant. Let us now discuss the applicability of the concept of the electron renormalization of \( C \) to the description of the behavior of \( \Gamma_s(T) \).

Indeed, Fig. 1 shows that the renormalization of \( C \) really takes place that follows from the crossing of \( \nu_n(T) \) and \( \nu_m(T) \) at \( T_{cr} \ll T_c \). However, the ratio of slopes of \( v(T) \) in the \( n \)- and \( s \)-states points out that the scale of renormalization is rather large: \( \delta C/C_0 \sim 0.22 \), i.e., more than twice possible maximal contribution \( \eta^2/4 \sim 0.1 \) of the adiabatic renormalization of \( \Delta_n \). Hence there should be an additional mechanism of renormalization. Moreover, an incomparability between the scale of \( \delta C/C_0 \) and the magnitude of the anomalies of \( \Gamma_s(T) \) indicates that this mechanism involves TLS which do not contribute to the relaxation attenuation at \( T \sim T_c \). Recall that the main contribution to the attenuation comes from TLS with \( \nu_{opt} \approx \Omega \) or \( \nu_{opt} \approx \sqrt{\omega/\eta T} \). One of possible additional mechanisms of the renormalization is associated with the fluctuational modulation of a barrier in the double-well potential. This mechanism affects only almost symmetric TLS (\( u \sim 1 \)) and does not give a contribution into \( \Gamma_n(T) \).

According to Ref. 3, the adiabatic renormalization of \( \Delta_n \) should involve all TLS with \( E \gtrsim T \). This condition places coherently tunneling TLS into the range where a cutting factor in the denominator of Eq. (10) is relevant. In spite of that, their partial contribution to \( \Gamma(T) \) can be essential in the scale of Fig. 5.

In the original TM, the value of \( \Gamma(T_c) \) for \( \omega \ll T_c \) is close to \( 1/2 \) for the scale used in Fig. 5. The decrease in \( C \) shifts \( \Gamma_n(T) \) towards smaller values. In order to analyze the shift of the experimental dependence with respect to the calculated one, we need the accuracy better than 1% for the absolute value of the attenuation, which is beyond the accuracy of our experiments. Therefore, in Fig. 5 we discuss only the relative position of \( \Gamma_n(T) \) and \( \Gamma_s(T) \) curves, which was measured much more precisely.

For numerical calculations we used the following energy dependence of the parameter \( C \):

\[
\frac{C}{C_0} = 1 - \frac{\eta^2}{4} \Theta(\varepsilon - \varepsilon_b) [1 + (2f(\Delta) - 1)\Theta(2\Delta - \varepsilon) - B]. \quad (17)
\]

This approximation is quite reasonable, since for \( \varepsilon^* < 1 \) the coherent amplitude \( \Delta^* \) decreases exponentially. Here \( \varepsilon_b \sim 1 \) is a fitting parameter which confines the range of coherently tunneling TLS. The last factor in the second term takes into account that for \( \varepsilon < 2\Delta \) the contribution comes only from the normal excitations. The last term in Eq. (17) takes into consideration an additional renormalization due to the symmetric TLS; its origin will be discussed below. A contribution of \( B \) to the sound attenuation is negligibly small and at this stage we assume \( B = 0 \) for the sake of simplicity.

The results of simulation are plotted in Fig. 5 (lines 2). The difference between lines 1n and 2n reflects a contribution from the adiabatic renormalization of \( \Delta_n \) to the sound attenuation in the \( n \)-state. We matched almost temperature-independent part of \( \Gamma_s(T) \) at \( \eta = 0.65 \) with measured curves and obtain quite reasonable value
of \( \varepsilon_b = 1.2 \pm 0.1 \). The calculated dependence of \( \Gamma_s(T) \) varies similarly to the predictions of the original TM just below \( T_c \). Then \( \Gamma_s(T) \) undergoes a break, changing the sign of \( d\Gamma/dT \) at \( T = 2\Delta_s(T)/\varepsilon_b \). These features arise due to exploiting a step approximation in Eq. (17). The superconductivity does not affect the renormalization of \( C \) if \( 2\Delta_s(T) \) is smaller than the value of \( E = T\varepsilon_b \). Obviously, a smoothed energy dependence of the cutoff factor in Eq. (17) would decrease the variation of \( \Gamma_s(T) \) at \( T_c \) and eliminate the break. The same result would also occur due to a possible broadening of the superconducting transition in an amorphous sample.

In this way, the evolution of \( \Gamma_s(T) \) in the vicinity of \( T_c \) is determined by two factors: a fall due to the decrease in the relaxation rate \( \nu \) and a growth because of freezing out of the \( C \) renormalization. The first factor is frequency-dependent in contrast to the second one. Therefore, the resulting variations of \( \Gamma_s \) should also depend on frequency. When \( \omega \) decreases, the temperature range where \( \Gamma_s(T) > \Gamma_n(T) \) should be extended and vice versa. In particular, if \( \eta, T_c, \) and \( \varepsilon_b \) are fixed, the increase in frequency by an order of magnitude (see, for example, Ref. [2] where the measurement frequency was about of 600 MHz) has to mask the action of the second factor utterly. This frequency increase leads to \( \Gamma_s(T) \) always lower than \( \Gamma_n(T) \). Besides, \( d\Delta_s(T)/dT \) grows as the temperature decreases. However, the experiments in Ref. [2] were carried out in metglass with \( T_c = 2.6 \) K where \( \nu \) is essentially determined by phonons and depends weakly on the state of electron subsystem. Under these circumstances, the freezing out of the renormalization should give even stronger effect than observed in our case.

C. Sound velocity

The resonance contribution to \( \nu(T) \) is completely determined by coherently tunneling TLS with \( \varepsilon \gtrsim 1 \). Therefore, the account of adiabatic renormalization of \( \Delta_0 \) in the \( n \)-state will lead to \( \delta \nu/C_0\nu \nu_{\text{res}} = (1 - \eta^2/4)\ln T \) independently on the magnitude of \( \varepsilon_b \) in Eq. (17). From Eqs. (9) and (17), we estimate a magnitude of the relaxation contribution as \( \delta \nu/C_0\nu \nu_{\text{rel}} = -1/2 \left[ 1 - \eta^2/4(1 - \tanh(\varepsilon_b/2)) \right] \ln T \). Using the values of \( \eta \) and \( \varepsilon_b \) obtained before, we get \( \delta \nu/C_0\nu \nu_{\text{rel}} = 0.42 \ln T \) for the total change in the \( n \)-state, whereas the slope of the experimental dependence \( \delta \nu/C_0\nu \nu_{\text{rel}} = 0.28 \ln T \) differs from the original TM coefficient 0.5 much more. Thus, the mechanism of the adiabatic renormalization of \( \Delta_0 \) can solely explain less than a half of the whole effect, and, as it was mentioned above, an additional origin of the electron renormalization of \( C \) has to exist. It must affect mainly the symmetric TLS which do not participate in the relaxation attenuation. Besides, both mechanisms can be considered as additive because the scale of \( C \) renormalization is small.

The authors of Ref. [2] studied the effect of electron density fluctuations on the barrier height of the interwell potential. It was argued that below some critical temperature \( T_k \), almost symmetric TLS and a surrounding electron cloud can form a strongly-correlated (bound) state similar to the Kondo state. This effect has an energy threshold, \( E < E_k(T_k) \). Fluctuations also lead to the renormalization of the tunnel amplitude similar to Eq. (13):

$$\sum_0 = \Delta_0(T/D)^m,$$

where \( D \) is of the order of the Fermi energy. The exponent \( m \) depends on \( \eta \) and is close to \( 0.1 \div 0.2 \) for \( \eta \sim 0.65 \).

The spectrum transformation of Eq. (18) does not reflect in the renormalization of the parameter \( C \) since it does not change the density of the TLS states within the space of new variables \( \pi, \bar{E} \). These questions were not considered in Ref. [2]. Nevertheless, one can conclude that the renormalizing factor would depend on \( u = \Delta_0/E \) in Eq. (18) since, according to Ref. [2], \( \sum_0 \rightarrow \Delta_0 \) if \( u \rightarrow 0 \). This nonlinear relation between \( \Delta_0 \) and \( \Delta_0 \) means the effective renormalization of TLS density of states similar to the adiabatic renormalization. One can also expect a reduced value of the deformation potential in the bound state.

Thus, the slope \( \nu(C_0\nu)_{\text{res}} \) is determined by the relation \( C/C_0 = 1 - \eta^2/4 - R_s \), where \( R_s \) describes the contribution from symmetric TLS in \( n \)-phase. The latter does not change the slope of \( \nu(C_0\nu)_{\text{rel}} \), therefore \( R_s \) is not a fitting parameter. Its value for given \( \eta \) is unambiguously determined by the resulting slope \( S \) (with the account for a small correction related to the contribution of adiabatic renormalization of \( \Delta_0 \) in \( \nu(C_0\nu)_{\text{rel}} \)). Particularly, the estimates presented above give \( R_s \approx 0.14 \).

For computing, we model \( B \) in Eq. (17) by simplest step function, introducing a conventional boundary \( u_b \) of the “symmetric” TLS:

$$B = \frac{R_s}{\sqrt{1 - u_b^2}} \Theta(u - u_b) \left[ 1 + (2f(\Delta) - 1) \Theta(2\Delta - \varepsilon) \right].$$

The meaning of two last factors in Eq. (19) is clear from preceding discussion. The first factor represents the “real” renormalization by the symmetric TLS because the renormalizing correction appears with the weight \( 1 - u_b^2 \) under the integration over \( u \) in Eq. (8).

A comparison between calculated and experimental dependencies for \( u_b = 0.5 \) is presented in Fig. 6. Here we use the following procedure. The experimental points for the frequency of 62 MHz were taken from Fig. 1 and normalized by \( C_0 \). It is impossible to measure mutual position of \( \nu(T) \) for different frequencies with necessary accuracy of \( 10^{-7} \). However, according to Eq. (3), the value \( \nu(C_0\nu)_{\text{res}} \) does not depend on frequency for \( \omega \ll T \). That is why the experimental points in the \( s \)-state for both frequencies 186 MHz and 62 MHz are placed together at temperatures far below \( T_c \). The positions of the calculated dependencies are determined by the upper
limit of integration in Eq. (7). Finally, one can also compare them with experimental dependencies after matching of $(\delta v(T)/C v)_{\text{res}}$ in the s-state at $T \ll T_c$. 

![Diagram](image_url)

**FIG. 6.** Comparison of the experimental change of sound velocity for different frequencies and magnetic fields (circles: 62 MHz, triangles: 186 MHz; open symbols: $B = 0$, solid symbols: $B = 2.5$ T) with the calculations. The following set of parameters is used: $\eta = 0.65$, $\epsilon_b = 1.2$, $u_b = 0.5$, $R_s = 0.14$, $T_c = 0.83$ K.

Figure 6 shows satisfactory agreement between the calculated and the experimental dependencies. Some difference arises only in the temperature range from $T_c$ to 0.4 K (see also Fig. 2). The calculated curve in the s-phase is noticeably steeper at $T \sim T_c$ than the experimental one. An estimate shows that a more smooth energy dependence of the adiabatic renormalization of $\Delta_0$ leads only to insignificant decrease in the slope of $(\delta v/C v)_{\text{s}}$ at $T \sim T_c$. The most probable reason for deviations of the computed dependencies from the experimental ones near $T_c$ is the smearing of superconducting transition and a contribution from small thermodynamic corrections to $v(T)$ in the s-phase. A small contribution can also come from a residual influence of phonon relaxation which may cause the excess of the calculated slope of $(\delta v/C v)_{\text{s}}$ over the experimental one near $T_c$ (Fig. 2).

The account for the symmetric TLS shifts up the resulting dependence $(\delta v/C v)_{\text{n}}$ at $R_s \ln 1.36u_b$ without changing the slope of $(\delta v/C v)_{\text{rel}}$, as follows from computation of Eq. (9) with renormalization of Eq. (19) for $\omega/\eta^2 T \ll u_b^2 \ll 1$. This shift plays the same role as the parameter $A$ in Eq. (5). Therefore, according to Eq. (5), the fitting of $u_b$ is reduced in fact to matching of the computed $T_{cr}$ with the experimental value at given $\eta$, $R_s$, and $S$. The fitted value of $u_b = 0.5$ is smaller than $u_b \approx 0.7$ accepted in Ref. [5]. However, the latter is determined by the bare magnitude of $\Delta_0$, whereas the integration in Eq. (8) is performed over the renormalized variable $\overline{\omega}$. Therefore, according to Eq. (18), $\overline{R}_s$ will be smaller than the bare value.

The account for the thermodynamic correction also will lead to an increase in $u_b$; formally, $u_b$ can be made as much close to 1 as one likes. Unfortunately, there is no clear way to separate the electron contribution on the background of the TLS effects. Note that a weak temperature dependence in Eq. (18) will introduce a correction into the slope $(\delta v(\ln T)/C v)_{\text{rel}}$ too. Then the value $R_s$ in Eq. (19) should be decreased by a factor of $(1 + m)$.

Thus, our assumption about an additional mechanism renormalizing the contribution of almost symmetric TLS to the sound velocity gives a satisfactory description of the sound velocity behavior in both the n- and s-states with a help of only one extra parameter $u_b$. Note that a thorough calculation of $\Gamma_s(T)$ for $u_b = 0.5$ with the additional parameter $B$ in Eq. (16) leads to practically the same dependencies as shown in Fig. 5 for $B = 0$.

**VI. CONCLUDING REMARKS**

Using the results of the acoustic measurements obtained on the superconducting glass Zr$_{41.2}$Ti$_{13.8}$Cu$_{12.5}$Ni$_{10}$Be$_{22.5}$ we have carried out a quantitative analysis of different theoretical approaches to the electron renormalization of sound interaction with TLS. A convincing evidence for the renormalization is the crossing of the lines $v_n(T)$ and $v_s(T)$ at some temperature $T_{cr} \ll T_c$, in combination with the absence of proportionality between $T_{cr}$ and $\omega$. Assuming the simplest model renormalization of the interaction parameter $C$ in the space of tunnel variables, it is possible to describe quantitatively the behavior of the sound velocity and attenuation exploiting the original tunnel model. It is sufficient to use the adiabatic renormalization of the coherent tunneling amplitude to fit the dependence $\Gamma_s(T)$ with the experiment. However, the behavior of the sound velocity can be described only with the help of an additional mechanism of the renormalization affecting only almost symmetric TLS. This is the main result of our consideration. The additional mechanism can be presumably related to rebuilding of the interwell potential due to fluctuations but this approach has not been developed enough to consider it to be incontrovertible.

The analysis carried out in this work allows us to evaluate several parameters using the experimental dependencies of $\Gamma(T)$ and $v(T)$. We would like to emphasize that most of them are not fitting parameters in a common sense because it is not necessary to vary the whole set of them simultaneously to determine each of them. We utilize the following sequence of parameter evaluation. First of all, from the temperature dependence of the sound attenuation in the s-state we determine the energy gap $\Delta_s(0)$ and the parameter $\eta$. Using the lat-
ter together with the experimental $\Gamma_s(T)$, $v_s(\ln T)$, and $T_{cr}$, we evaluate the parameters $\varepsilon_b$, $R_s$, and $u_b$ consequently. In order to evaluate any parameter, we utilize only the values which have been already found during previous steps. In this way, $\eta$ is the central parameter which determines all others. Therefore a question may rise: could an error in the determination of $\eta$ lead to essential redistribution of the contributions from different mechanisms? Under assumption that the electron contribution into $v_s(T)$ does not change below $T_c$, the needed value of $u_b \sim 0.3$ is nonrealistically small already for $\eta = 0.67 \div 0.68$. On the other hand, for $\eta \lesssim 0.6$ the best fit yields lower $T_c$. In this way, the studied alloy does not give a large choice for the variation of the parameters.

In principle, it would be possible to manage without the concept of additional influence of electrons on the symmetric TLS in the analysis of elastic properties of our metglass and reduce all effects to the adiabatic renormalization of the tunnel amplitude, if we accept $\eta \approx 1$ assuming its preliminary estimate from low-temperature “tail” of the attenuation (Sec. V, A) to be unreliable. However, in this case the overall shape of calculated $\Gamma_s(T)$ can be adjusted to the experimental dependence only at $T_c = 0.9$ K, revealing noticeable deviations from the experimental data at $T \ll T_c$. Besides, the experimental value of $T_{cr}$ could be obtained only under assumption that in s-state at $T = T_{cr}$ the electron contribution is lowered by $\delta \varepsilon/\varepsilon \approx 1.5 \cdot 10^{-5}$ (this estimate follows from Eq. (5)) that is comparable with the TLS contribution. Within this approach, the resulting variations of $v(T)$ near $T_c$ would be determined by three equipollent mechanisms: the change of the TLS relaxation rate, the freezing of the adiabatic renormalization, and the evolution of electron contribution. In our opinion, it is impossible to expect proper mutual compensation of their partial contributions, for both the longitudinal and transverse modes, providing regular variation of $v(T)$ observed at the superconducting transition.

It interesting to notice that the parameter $E_k$ which appears in the fluctuation model is not so important in our evaluation process. Our analysis does not demand also the absence of this parameter. Most probably, the condition $E_k > 2\Delta_s(0)$ is satisfied in the alloy used in our investigation because, as follows from Eq. (5), the introduction of $E_k < 2\Delta_s(0)$ at constant $T_{cr}$ should be accompanied by a decrease in $u_s$. So we believe that there is no need to use $E_k$ for the description of low-temperature features of the sound velocity and attenuation in our alloy. The introduction of $E_k$ can be probably more fruitful for the analysis of the metglass elastic properties at higher temperatures.

VII. ACKNOWLEDGMENTS

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