# Tunneling systems in polycrystalline metals: Absence of electron-assisted relaxation

R. König, P. Esquinazi,\* and B. Neppert

Physikalisches Institut, Universität Bayreuth, D-95440 Bayreuth, Germany

(Received 18 October 1994)

With the vibrating-wire technique we have investigated the acoustic properties of polycrystalline Al and Ta in the normal and superconducting states and of Pt. The measurements were performed at frequencies  $0.1 \le \nu \le 18.5$  kHz and at temperatures 0.1 mK  $\le T \le 10$  K. The relative change of the sound velocity and the internal friction resemble those of amorphous dielectrics with similar strain dependence. No difference in the acoustic properties has been observed between the normal and superconducting states indicating the absence of electron-assisted relaxation of the tunneling systems. This result is in agreement with the results for Pt. In addition, we observed in Pt similar changes of the acoustic properties after thermal treatment as those reported for amorphous superconductors. These changes indicate a decrease of the coupling constant between phonons and tunneling systems upon heat treatment.

### I. INTRODUCTION

Low-temperature acoustic properties of amorphous materials depend on the interaction of phonons with tunneling systems (TS's).1 Their investigation provides information on the density of states of tunneling systems as well as on their relaxation rate which is determined by the interaction with phonons and/or conduction electrons. The interaction between phonons and tunneling systems and its influence on sound dispersion and absorption in amorphous dielectrics or superconductors in the linear regime (i.e., strain  $\epsilon \ll \text{thermal energy } k_BT$ ) are well understood.<sup>2</sup> The influence of conduction electrons on the relaxation rate of tunneling systems has been investigated in metallic glasses by measurements of the ultrasonic attenuation and change of sound velocity.<sup>3-5</sup> The main difference in comparison with amorphous dielectrics was the observation of an anomalously large acoustic intensity necessary to saturate the ultrasonic absorption in metallic glasses. The interpretation of this remarkable difference between metallic glasses and dielectrics is based on the faster relaxation of the TS's through their interaction with conduction electrons. This interaction was considered as a Korringa-like interaction in analogy with the interaction of nuclear spins with conduction electrons.<sup>3,6</sup> In this model the maximum relaxation rate is given by

$$\tau_{e,m}^{-1} = KE \coth(E/2k_BT), \tag{1}$$

where K is a parameter related to the coupling constant between electrons and tunneling systems and E is the energy splitting of a tunneling system. Further experiments in superconductors<sup>7</sup> and theoretical work<sup>8</sup> seemed to support this interpretation.

A further development of the theory of the interaction between electrons and TS's was done by Vladár and Zawadowsky. These authors argue that the coupling between electrons and TS's is strong and leads to the for-

mation of a "bound" state, within the framework of a Kondo-like interaction, where the motions of the tunneling entities and of the electronic screening cloud around them are strongly correlated. According to this model this bound state would renormalize the energy splitting E of the TS's leading to a reduction of the density of states of tunneling systems  $\bar{P}$  below a certain "Kondo" temperature  $T_K$ . The "silent" real spin variables of the electrons provide an additional degeneracy for the interaction of electrons with tunneling systems characterizing the two-channel Kondo scattering at low temperatures. 10 Recently published work suggests that conductance signals observed in ballistic metal point contacts may be due to the two-channel Kondo scattering from tunneling systems. 11,12 Those experiments seem to test the tunneling-system-electron interaction in amorphous or disordered regions at interfaces of mesoscopic systems.

Acoustic investigations of the amorphous superconductors  $\mathrm{Pd}_{30}\mathrm{Zr}_{70}$ ,  $^{13,14}$   $\mathrm{Cu}_{30}\mathrm{Zr}_{70}$ ,  $^{14,15}$  and  $(\mathrm{Mo}_{1-x}\mathrm{Ru}_x)_{0.8}\mathrm{P}_{0.2}$  (Ref. 16) in the normal and superconducting states indicate that the density of states and/or the coupling constant between phonons and TS's may be renormalized by the presence of conduction electrons in qualitative agreement with the predictions of Vladár and Zawadowsky. These measurements showed clearly that with a Korringa-like relaxation rate only it is not possible to understand the observed temperature dependences of the sound velocity and attenuation. The temperature dependence of the acoustic properties of amorphous superconductors reveals a qualitative difference to that for the amorphous metal PdSiCu.  $^{3,17,18}$ 

The acoustic investigations of amorphous superconductors stimulated new theoretical work. Kagan and Prokof'ev<sup>19</sup> developed a theory on the interaction of conduction electrons with TS's based on the electron-polaron effect. The electron-polaron effect is due to the slow electronic excitations that do not follow the tunneling particle. They showed that, beyond the framework of perturbation theory, the density of states of TS's should

be renormalized according to their tunneling energy  $\Delta_0$ . The authors' calculations indicate that the acoustic properties of amorphous superconductors should change below the superconducting critical temperature  $T_c$  in a similar way as experimentally observed, i.e., an *increase* of the internal friction and a decrease of the sound velocity in the superconducting state in comparison to the normal state. Two free parameters in this theory make any attempt to check its predictions rather difficult: (1) the density of states of tunneling systems  $\bar{P}$  as a function of the tunneling energy  $\Delta_0$ . It is expected that in metallic glasses the function  $\bar{P}(\ln(\Delta_0))$  should decrease monotonously, but the actual function is unknown. (2) The second parameter b is proportional to the electronic density of states at the Fermi level squared,  $N^2(\epsilon_F)$ , multiplied by the interaction matrix element or coupling constant  $V_{kk'}^2$ . This parameter enters as a prefactor in the slope of the temperature dependence of the sound velocity. In the general case it is expected that b < 0.5.<sup>19</sup> Kagan and Prokof'ev showed that the renormalization of the sound dispersion would lead to a temperatureindependent sound velocity at very low temperatures, independent of the strength of the interaction.

A fourth approach to the interaction of tunneling systems and conduction electrons in metallic glasses has been given by Coppersmith<sup>20</sup> stimulated by the defect interaction model. 21,22 This latter model proposes an explanation of universal glasslike properties by the elastic dipolar interactions between atomic lattice defects. The interaction decays as  $g/r^3$ , with r the defect separation and g a coupling constant. This coupling constant is phonon mediated for insulators with  $g_{\rm ph} \sim \gamma^2/\rho v^2$  ( $\gamma$ is the coupling constant,  $\rho$  the mass density, and v the sound velocity); for metals it is mediated by electrons and phonons with  $g_{\rm eff} \sim (g_{\rm RKKY}^2 + g_{\rm ph}^2)^{1/2}.^{20}$  Within this model  $\bar{P} \propto 1/g$ , therefore it is expected that  $\bar{P}$  should be larger in the superconducting state than in the normal state in agreement with the observations. 13,14 However, acoustic experiments in as-quenched and annealed  $\mathrm{Zr}_{70}\mathrm{Cu}_{30}$  (Ref. 15) indicate that the density of states  $\bar{P}$ remains nearly constant upon thermal treatment whereas the coupling constant  $\gamma$  decreases. The decrease of the coupling constant  $\gamma$  with annealing leads to an increase of the thermal conductivity and a decrease of the sound attenuation and of the slopes of the temperature dependence of the sound velocity.<sup>15</sup> In this case the proportionality  $\bar{P} \propto 1/g$  does not hold, at least after the first annealing steps of amorphous metals.

From the experimental and theoretical work described above it is clear that no simple picture for the interaction between tunneling systems and conduction electrons can be given nowadays. Because the results in amorphous metals do not indicate a universal behavior of their acoustic properties [compare, for example, the results for PdSiCu (Refs. 17, 18) with those for amorphous ZrPd or ZrCu (Refs. 14, 15)] in contrast to the properties of amorphous dielectrics, it is necessary to investigate other metals where the interaction between TS's and electrons might be measurable.

Glasslike acoustic properties have been observed recently in various polycrystalline metals like Ag, NbTi, Ta,<sup>18,23</sup> Nb, Cu,<sup>24,25</sup> and Pt.<sup>25,26</sup> These investigations clearly indicate that there are basically no differences in the low-temperature acoustic properties of polycrystals and amorphous materials. The glasslike properties observed in polycrystalline metals open the possibility of studying the interaction of TS's with conduction electrons in regimes not easily available in amorphous metallic samples, for example, a large electron mean free path (larger than 1 nm) or a magnetic impurity concentration of a few ppm which can be well controlled in pure metals but not in amorphous metals.

In this experimental work the electron-TS interaction in polycrystalline metals was studied in two ways. First, we have investigated the change in the low-temperature acoustic properties for Pt after thermal treatment and compared it with the results in amorphous metals. Second, we have studied the influence of the conduction electrons on the acoustic properties of the pure superconducting metals Al and Ta by measuring them in their superconducting and normal states in the same temperature range. Our results indicate that the changes of the acoustic properties for a polycrystalline metal upon annealing are similar to those found for amorphous superconductors. The results for Pt, as well as the results in the normal and superconducting states of Al and Ta, indicate the absence of electron-assisted relaxation of the tunneling systems.

The paper is divided into three more sections. The next section is devoted to the experimental details and samples. In Sec. III the experimental results are discussed, and a short conclusion is given in Sec. IV.

# II. EXPERIMENTAL DETAILS AND SAMPLES

The study of the interaction of phonons and/or conduction electrons with tunneling systems requires a low-temperature probe that provides information on the density of states of the tunneling systems and their coupling to phonons or electrons as well. These prerequisites are well fulfilled by mechanical resonators like the vibrating-reed or vibrating wire techniques used to measure the sound velocity and internal friction of solids.<sup>2,18</sup>

All samples investigated in this work are commercially available cylindrical wires; their dimensions and characteristic properties are summarized in Table I. Sound velocity and internal friction measurements were performed using the vibrating-wire technique (see Fig. 1) as described in Ref. 18. The relative change of the resonance frequency of the vibrating-wire is determined by the relative change of the Young's modulus sound velocity  $\Delta v/v = [v(T,\epsilon)-v_0]/v_0$  ( $v_0$  is the sound velocity at an arbitrary reference temperature and strain). The internal friction was obtained from the width of the resonance curve and/or from the induced voltage.

The oscillation of the wire leads to a time-dependent strain  $\epsilon$  which has its maximum value at the fixed ends of the wire and is given by  $^{18}$ 

$$\epsilon = 28.3 \frac{ru}{l^2},\tag{2}$$

TABLE I. Metal, diameter of the wire, frequency, and pu-				
rity of the measured samples. Different resonance frequencies				
were obtained with different lengths of the wire.				

Sample	$\begin{array}{c} {\rm Diameter} \\ {\rm [\mu m]} \end{array}$	Frequency [kHz]	Purity
Platinum		0.084	4N
Platinum	25		
	25	0.76	4N
	25	1.39	4N
	25	1.50	4N
Aluminum	5	0.32	3N5
	25	7.7	4N
	125	0.60	4N
Tantalum	125	1.88	3N
	125	4.42	3N
	125	5.45	3N
	125	5.61	3N
	125	18.5	3N

where u is the maximum amplitude of the oscillation in the middle of the loop, r is the wire radius, and l its length. The strain denoted in the figures represents the maximum strain calculated with Eq. (2) and at  $T\simeq 0.05$  K. Its relative error is about 25% due to errors in the wire geometry. In the temperature range of our measurements the vibration amplitude u does not change strongly with temperature; therefore most of the measurements as a

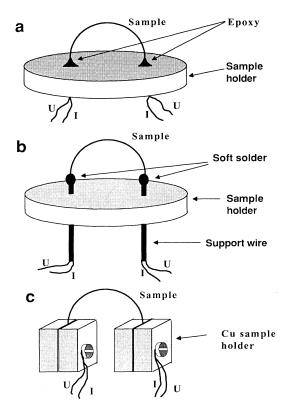


FIG. 1. Three sample holders used in our experiments to study a possible clamping contribution to the acoustical properties. For details see text.

function of temperature have been performed at constant driving force. In this case the strain  $\epsilon(\propto u)$  in all measured samples changes by less than 30% with temperature. Measurements with constant amplitude of vibration were performed to confirm the results obtained at constant driving force. We observed very good agreement between these different sets of data. In addition, the strain dependence of the acoustic properties was investigated at different temperatures.

Typical sample lengths of a few millimeters result in resonance frequencies from  $\sim 0.1~\rm kHz$  to  $\sim 18.5~\rm kHz$  (see Table I). Because recording complete resonance curves is usually a very time-consuming measurement, most data were taken by locking in on the resonance frequency of the wire and then monitoring the change in resonance frequency and the induced voltage as a function of temperature.

Since the diameters of our samples are very small (see Table I), resulting in a poor thermal conduction at the lowest temperatures, energy dissipation due to the vibration has to be kept to a minimum level to avoid self-heating effects. This was of particular importance for the samples in the superconducting state. In addition, attention was taken to ensure good thermal contact between the wire and the sample holder.

To study carefully a possible contribution of the sample clamping to the measured properties, various sample holders were used that differed by the way the sample was clamped to the sample holder. Figure 1 shows three typical sample holders: (a) The wire is fed through two tiny holes in a Ag or Cu sample holder and fixed with a small amount of glue (either GE varnish or Stycast 2850 FT); (b) The sample wire is soldered to a strong supporting wire of a much larger diameter which is electrically isolated from the main body of the sample holder; (c) Each end of the sample is clamped between two small Cu blocks; both pairs of blocks are electrically isolated from each other and from the main body of the sample holder. To avoid squeezing the sample between the Cu blocks, small slits are scratched into each block where the sample ends are fixed. The experiments were performed in two different cryostats: a dilution refrigerator enabling measurements in fields up to 8 T and temperatures to 40 mK and a Cu nuclear refrigerator<sup>27</sup> extending the accessible temperature range below 1 mK.

Contributions of the sample clamping to the damping should be observable, for example, by comparing the strain dependence of the internal friction for different sample holders. In Fig. 2 we present the strain dependence of  $Q^{-1}$  for a 25- $\mu$ m Pt wire attached to different sample holders as shown in Fig. 1(a) ( $\nu = 84 \text{ Hz}$ ) and Fig. 1(b) ( $\nu = 1504$  Hz). The measurements were performed at a constant temperature  $T \sim 15$  mK. Although both wires were differently attached to their sample holder, they show about the same magnitude of the internal friction and in particular the same strain dependence. We note that the clamping has no influence on the relative change of the sound velocity. Therefore we can neglect contributions from the sample clamping to the acoustic properties in the strain range used in our experiments. Moreover, we note that measurements of the strain de-

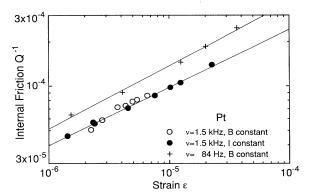


FIG. 2. Strain dependence of the internal friction for a 25- $\mu$ m Pt wire attached to different sample holders: (+) holder of Fig. 1 (a) ( $\nu = 84$  Hz) and ( $\circ, \bullet$ ) holder of Fig. 1(b) ( $\nu = 1.5$  kHz).

pendence of the internal friction of Pt measured at  $\nu \sim 1504$  Hz and performed in small magnetic fields B < 40 mT (see Fig. 2) confirm that a change of the magnetic field is equivalent to a change of the excitation current.

The study of the influence of heat treatment was investigated on a Pt wire that was annealed in vacuum at a temperature of  $\sim 1300$  K for 5 h. To avoid any further bending of the wire when mounting it on the sample holder, the heat treatment was performed with the wire already in the shape of a semicircular loop with a diameter according to its final position on the sample holder.

Thermometry was based on the superconducting transitions of five samples in a NBS superconducting fixed-point device. A Pt NMR thermometer as well as a PdFe susceptibility thermometer were calibrated against this NBS reference and used in the lower millikelvin or microkelvin range, respectively. Higher temperatures were measured by means of a calibrated Ge resistor positioned in the compensated part of a superconducting solenoid used for the measurements of the acoustic properties in the normal state of the superconducting wires. The superconducting critical temperatures of the wires at zero applied field are  $T_c=1.1~{\rm K}$  (4.48 K) for Al (Ta).

# III. RESULTS AND DISCUSSION

## A. Platinum

## 1. Sound velocity

We have already reported recently some results on the temperature and time dependence of the sound velocity of pure Pt.  $^{25,26}$  The sound velocity and internal friction of one of the investigated Pt samples measured at 760 Hz showed the typical behavior of amorphous dielectrics or amorphous or polycrystalline superconductors at audio frequencies: a maximum in the sound velocity at  $T\sim0.1$  K and logarithmic temperature dependences above and below it. As in amorphous SiO<sub>2</sub> the maximum in the

sound velocity is shifted to higher temperatures for larger strains <sup>18,28</sup> while the internal friction is strain independent with the typical temperature dependence and magnitude as for amorphous dielectrics. <sup>26</sup>

However, some of the Pt wires show a strain-dependent contribution to the internal friction; see Fig. 2. Our results indicate that this contribution is superimposed on a strain-independent internal friction (due to the linear interaction of phonons with tunneling systems) resembling the behavior of  $Q^{-1}(T,\epsilon)$  observed in  $\operatorname{Cu},^{25}$  which might be interpreted as due to dislocation-mediated attenuation. To investigate in more detail the acoustic properties of Pt we have studied three samples: two as-received wires at frequencies of 84 Hz and 1.5 kHz and, from the same batch, an annealed wire at 1.4 kHz.

Figure 3 shows the relative change of sound velocity for Pt at 1.5 kHz at three different strains obtained from the change in resonance frequency of the vibrating wire. As in the measurement at 760 Hz,<sup>26</sup> the sound velocity shows a strain-dependent maximum and a logarithmic temperature dependence above and below it, in agreement with the temperature dependence observed in amorphous dielectrics.<sup>18</sup> From these similarities we interpret the behavior of the acoustic properties in terms of the interaction of phonons with TS's.

Below  $\sim 4$  mK the sound velocity saturates and remains constant to the lowest temperatures. This saturation of the sound velocity as well as the shift of the maximum with strain has been observed in amorphous  ${\rm SiO_2}.^{18}$  These effects cannot be explained by self-heating of the sample because (i) the maximum of the sound velocity shifts to higher temperatures for larger strains; (ii) in spite of a large difference in thermal conductivity the saturation of the sound velocity in Pt occurs at similar temperatures and strains as in  ${\rm SiO_2}$ , Ta, and NbTi in the superconducting state;  $^{18}$  (iii) the temperature-dependent part of the internal friction is not dependent on strain.  $^{18,23,26}$ 

This nonlinear effect occurs when the energy of the sound wave or strain field,  $d(t) = \epsilon(t)\gamma$ , is of the or-

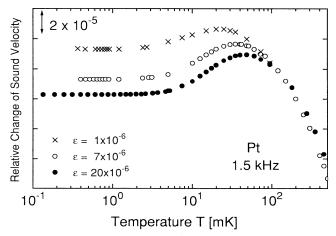


FIG. 3. Relative change of sound velocity as a function of temperature for Pt (as-received) at 1.5 kHz and at three different strains.

der of the thermal energy  $k_BT$ , leading to a change of the population number of the tunneling states,  $n \propto \tanh(E/2k_BT)$ , with an energy difference between the two states  $E^2 = [\Delta + d(t)]^2 + \Delta_0^2$  ( $\Delta$  is the asymmetry of the double-well potential and  $\Delta_0$  is the tunneling energy). Rigorous theoretical approaches that account for the nonlinear acoustic properties, within the same concepts, provide similar results. Since  $\frac{29,30}{2}$ 

Because the temperature dependences of the acoustic properties resemble those of amorphous insulators, we conclude that the conduction electrons in Pt do not influence the relaxation rate of the tunneling systems. Therefore, as in amorphous dielectrics we assume that the slope of the sound velocity above its maximum (where the nonlinear effects vanish) is proportional to the density of states of tunneling systems  $\bar{P}$  multiplied by the coupling constant  $\gamma^2$  as given by the one-phonon or direct process. Within  $\sim 15\%$  the three as-received Pt samples studied show the same strain-independent slope of  $\Delta v/v$  vs T at  $T>T_{\rm max}$ ; see Fig. 4. If we assume that  $\Delta v/v=-(C/2)\ln(T/T_0)$  above the maximum  $(C=\bar{P}\gamma^2/\rho v^2, {\rm and}\ T_0$  is an arbitrary reference temperature) we obtain  $C=(7\pm 1.5)\times 10^{-5}$ . This value is only about a factor of 4 smaller than for vitreous silica. 18

In Fig. 4 we note that the position of the maximum  $T_{\rm max}$  of the sound velocity does not scale with the phonon frequency  $\omega$  as the standard tunneling model predicts, i.e.,  $\omega/T_{\rm max}^3 \simeq {\rm const}$ ; compare, for example, the results obtained at 1.5 kHz and at 760 Hz. The reason for this

FIG. 4. Relative change of sound velocity as a function of temperature for Pt (as-received) at three different frequencies. The applied strains were (×)  $3.5 \times 10^{-6}$ , ( $\diamondsuit$ )  $8.2 \times 10^{-6}$ , ( $\spadesuit$ )  $1.6 \times 10^{-5}$ , ( $\bigtriangleup$ )  $1.1 \times 10^{-5}$ .

disagreement is not understood at present; we point out only that the sample measured at 760 Hz was taken from a different batch than the one used for the 84 Hz and 1.5 kHz measurements. Since  $T_{\rm max}$  is very sensitive to strain, it might be possible that the strain distribution of the wires is different from that calculated through the resonance amplitude u [maximum strain at the wire surface  $\epsilon \propto ru/l^2$ ; see Eq. (2)]. It is also possible that the clamping and/or the bending of the sample measured at 760 Hz produced an extra strain distribution and a maximum total strain which is not taken into account in Eq. (2) and should be added to that produced by the vibration. Measurements of  $T_{\rm max}$  at different frequencies but similar strains in polycrystalline NbTi showed the expected dependence, i.e.,  $T_{\rm max} \propto \omega^{1/3}$ .

At 84 Hz we cannot resolve a maximum in the sound velocity within experimental resolution. Taking as reference the measurements at 1.5 kHz with  $T_{\rm max} \simeq 25\,(40,50)$  at strain  $\epsilon=1\,(7,20)\times 10^{-6}$ , we expect  $T_{\rm max}\sim 16$  mK at  $\epsilon=8.2\times 10^{-6}$ . According to the above given explanation we expect a saturation of the sound velocity to start already at a temperature of  $\sim 8.2\times 10^{-6}\,\gamma\sim 80$  mK with  $\gamma\sim k_B10^4$  K taken from the temperature of the maximum in the sound velocity. This estimate indicates that a maximum in the sound velocity at 84 Hz cannot be observed because it is expected to be located in the nonlinear region.

Figure 5 shows the relative change of sound velocity of the annealed wire (1.4 kHz) at two applied strains. For comparison we have included in this figure the results

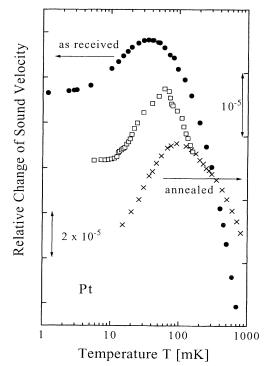


FIG. 5. Relative change of sound velocity as a function of temperature for Pt: (•) as-received, 1.5 kHz and  $\epsilon = 7 \times 10^{-6}$ ; annealed, 1.4 kHz: ( $\square$ ) 2.6 × 10<sup>-6</sup> and (×) 4 × 10<sup>-6</sup>.

obtained before thermal treatment. The heat treatment influences the sound velocity in two important ways: (1) The slope of the relative change of sound velocity taken above  $T_{\rm max}$  decreases by a factor  $\sim 4$ , and (2) at similar strains the position of the maximum is shifted to higher temperatures. These two observations are similar to the changes of the sound velocity obtained in amorphous Zr<sub>70</sub>Cu<sub>30</sub> (Ref. 15) after annealing. Because  $\Delta v/v \propto \bar{P}\gamma^2$ , we can ask whether the decrease of the slope of the relative change of sound velocity due to the thermal treatment is mainly due to a decrease of the density of states  $\bar{P}$  of tunneling systems or of the coupling constant  $\gamma$ . According to the tunneling model, the shift of the maximum  $T_{\max}$  at equal strains and phonon frequencies is related solely to a change in the coupling constant  $\gamma$ , i.e.,  $\gamma^2 T_{\rm max}^3 \sim {\rm const}$  since  $T_{\rm max}$  occurs at  $\omega \tau \sim 1$  and  $\tau \propto T^{-3} \gamma^{-2}$ . Taking  $T_{\rm max} \sim 40$  mK at  $\epsilon \sim 7 \times 10^{-6}$  before thermal treatment and  $T_{\rm max} \sim 90$ mK at  $\epsilon \sim 4 \times 10^{-6}$  after thermal treatment, we obtain  $\gamma$  after annealing,  $\simeq 0.3\gamma$  before annealing. Therefore, it seems reasonable to conclude that the change in the slope of the sound velocity above  $T_{\text{max}}$  after annealing is mainly caused by the reduction of the coupling between phonons and tunneling systems as was observed in amorphous superconductors. 15

Figure 6 shows the relative change of sound velocity as a function of applied strain at two different temperatures below the maximum in the nonlinear regime for the annealed wire. The strain has been changed by applying different excitation currents. We obtain the same strain dependence by changing the applied magnetic field at constant ac current through the wire. The sound velocity decreases nearly logarithmically with strain as observed in other amorphous and polycrystalline metals  $^{18,23,28}$  at fixed temperature and in agreement with numerical calculations  $^{23,30}$  based on the strain dependence of the tunneling splitting energy  $E(\epsilon)$ .

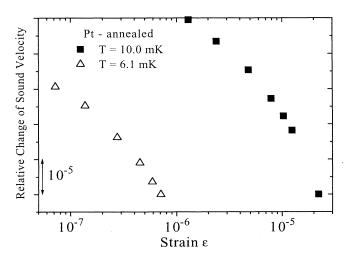


FIG. 6. Relative change of sound velocity as a function of strain for Pt (annealed) at 1.4 kHz and at two constant temperatures.

## 2. Internal friction

Figure 7 shows the temperature dependence of the internal friction of Pt in the as-received and annealed states obtained at similar strains  $\epsilon(T=10~{\rm mK})\simeq (6\pm2)\times 10^{-6}$ . The observed temperature dependence resembles that of amorphous dielectrics or amorphous superconductors in the superconducting state and polycrystalline metals. We note that the as-received sample does not show a temperature independent internal friction ("plateau") above 70 mK as was observed for other Pt samples. His deviation might indicate that the distribution of TS's or their density of states  $\bar{P}$  for this Pt sample is not energy independent as assumed in the standard tunneling model. A maximum in the internal friction instead of a true plateau is also observed in amorphous materials.  $^{14,15}$ 

The internal friction decreases by a factor of 4 after annealing in very good agreement with the decrease of the slope in the temperature dependence of the sound velocity; see Figs. 5 and 7. Due to the weak logarithmic strain dependence of the internal friction (see Fig. 2) small differences in the applied strain cannot explain the observed decrease in the internal friction after annealing. It is also important to note that the maximum or beginning of the plateau at  $T_p$  in the internal friction shifts to higher temperatures with annealing:  $T_p \sim 70(100)$ mK before (after) annealing. This result indicates that  $\gamma$  decreases  $\sim 40\%$  after thermal treatment ( $\gamma$  after annealing,  $\sim 0.7\gamma$  before annealing) in agreement with the results in amorphous Zr<sub>70</sub>Cu<sub>30</sub>. 15 The change in the coupling constant after annealing obtained from the internal friction is only in qualitative agreement with the change obtained from the sound velocity. The quantitative discrepancy might be related to the difficulty of measuring the true shift of  $T_{\text{max}}$  upon annealing from the sound velocity temperature dependence due to its sensitivity to the applied strain.

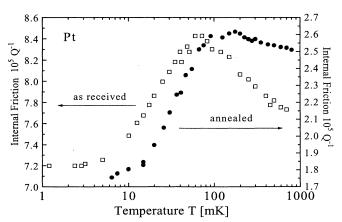


FIG. 7. Internal friction as a function of temperature for Pt: ( $\square$ ) as-received (left scale),  $\nu=1.5$  kHz,  $\epsilon=7\times10^{-6}$ , ( $\bullet$ ) annealed (right scale),  $\nu=1.4$  kHz,  $\epsilon=4\times10^{-6}$ .

### B. Sound velocity and internal friction of aluminum

Figure 8 shows the relative change of sound velocity for Al in the superconducting  $(B=4.7~\mathrm{mT},~\epsilon=7\times10^{-5})$  and normal  $(B=10.7~\mathrm{mT},~\epsilon=9\times10^{-5})$  states at 7.7 kHz. The typical temperature dependence of amorphous insulators is observed. It is interesting to mention that a glasslike thermal conductivity  $\kappa \propto T^2$  at  $T \ll 1~\mathrm{K}$  was measured in Al in the superconducting state.<sup>31</sup>

From the slope of the sound velocity above its maximum we obtain  $C = (6 \pm 2) \times 10^{-5}$ , a value similar to that obtained for Pt  $(C \simeq 7 \times 10^{-5})$  and again only a factor 4 smaller than for amorphous  $\mathrm{SiO}_2$ . As shown in Fig. 8 the temperature dependence of the relative change of sound velocity does not depend on the normal or superconducting state of the sample. The small difference observed below 0.1 K is not attributed to a contribution of the conduction electrons but to the small difference in the applied strain (see below).

Figure 9 shows the temperature dependence of the sound velocity for Al at 323 Hz. The two upper curves were taken in the superconducting state using magnetic fields below the critical field  $B_c$ . To check the reproducibility of the results, measurements in the superconducting state were performed with the same strain  $\epsilon \simeq 7 \times 10^{-7}$  by adjusting the ac current through the wire at two applied fields B=0.8 mT and B=3 mT. As seen in Fig. 9 excellent reproducibility has been achieved within experimental error. In the same figure we have plotted the results for the normal state of Al but with applied strain  $\epsilon=2\times 10^{-6}$ . As observed in amorphous SiO<sub>2</sub> and other polycrystalline metals<sup>17,18</sup> the sound velocity shows a maximum that shifts to higher temperatures and larger strains and saturates at lower temperatures.

Figure 10 shows the temperature dependence of the internal friction of Al in the normal and superconducting states at 7.7 kHz. The temperature dependence as well as the magnitude of the internal friction resembles

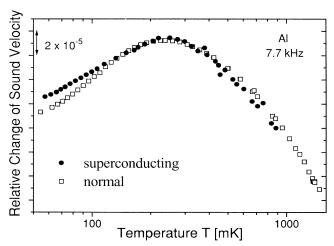


FIG. 8. Relative change of sound velocity as a function of temperature for Al (25  $\mu \rm m$  diameter) at 7.7 kHz: (•) applied magnetic field B=4.7 mT (superconducting state), strain  $\epsilon \simeq 7 \times 10^{-5};$  (□) B=10.7 mT (normal state),  $\epsilon \simeq 9 \times 10^{-5}.$ 

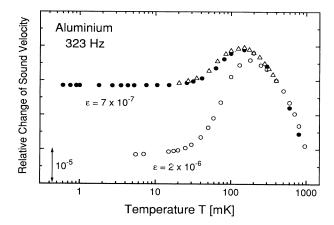


FIG. 9. Relative change of sound velocity as a function of temperature for Al (5  $\mu$ m diameter) at 323 Hz: ( $\circ$ ) applied magnetic field B=33 mT,  $\epsilon=2\times10^{-6}$ ; ( $\bullet$ ) B=3 mT,  $\epsilon=7\times10^{-7}$ ; ( $\triangle$ ) B=0.8 mT,  $\epsilon=7\times10^{-7}$ .

those found in amorphous insulators and polycrystalline metals. <sup>18</sup> In agreement with the sound velocity (see Fig. 8), the internal friction does not show any difference in the normal and superconducting states, reinforcing the experimental evidence that in this metal, as well as in Pt, the conduction electrons do not interact with tunneling systems. We should point out that Al has a density of states of conduction electrons at the Fermi level which is one order of magnitude smaller than in Pt. This is of particular importance since in all the models for the interaction between electrons and tunneling systems the coupling constant is proportional to the electronic density of states at the Fermi level.

From the plateau measured at T>0.2 K and without subtracting any background contribution to the internal friction we obtain  $C\simeq 1.2\times 10^{-4}$ , only a factor of 2 larger than the one obtained from the sound velocity; this difference may come from the clamping contribution to the internal friction.

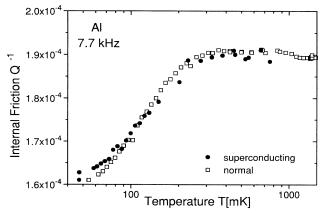


FIG. 10. Internal friction as a function of temperature for Al (20  $\mu m$  radius) at 7.7 kHz: ( $\bullet$ ) applied magnetic field B=4.7 mT (superconducting state),  $\epsilon \simeq 7 \times 10^{-5}$ ; ( $\square$ ) B=10.7 mT (normal state),  $\epsilon \simeq 9 \times 10^{-5}$ .

## C. Tantalum

As pointed out above the acoustic properties of polycrystalline Al and Pt strongly resemble those observed in amorphous dielectrics. If Eq. (1) also holds for normal metals, not only different temperature dependences for the acoustic properties are expected, but a clear difference of the strain dependence (at constant temperature) in the nonlinear regime should occur as well. The reason is that the main nonlinear effects are clearly observed when the period of the sound wave is shorter than the relaxation time of the tunneling systems, i.e.,  $\omega \tau > 1$ . If Eq. (1) holds for typical values of the parameter K, then  $\omega \tau < 1$  at T > 0.1 mK; therefore we expect much smaller changes of the sound velocity with strain as compared with insulating glasses or polycrystalline metals in the superconducting state as calculated in Ref. 30.

Figure 11 shows the relative change of sound velocity as a function of strain for Ta in the normal and superconducting states at  $T=45~\mathrm{mK}$  in the nonlinear regime. No difference is observed within experimental error in clear disagreement with theoretical expectations.

For the Ta sample discussed in Refs. 18, 23 the slope of the temperature dependence of the relative change of sound velocity as well as the magnitude of the internal friction was a factor of 10 smaller than in  $SiO_2$  due to the previous thermal treatment of the sample in agreement with the results for Pt reported above and with previous results for Ag. 18 For the Ta sample investigated in this work the relative change of sound velocity between applied strains  $10^{-6}$  and  $1.5 \times 10^{-5}$  is about  $6 \times 10^{-5}$  (see Fig. 11) in comparison with  $\sim 4 \times 10^{-5}$  reported in Refs. 18 and 23. This difference can be attributed to a larger coupling contant and/or density of states of tunneling systems ( $\propto \bar{P}\gamma^2$ ). Although the Ta wire was cut from the same batch as the sample measured previously,  $^{23}$  the difference in the coupling constant and/or in the density of states of tunneling systems might be produced by the bending of the wire with a smaller radius of the loop, needed to obtain larger resonance frequencies (18.5 kHz

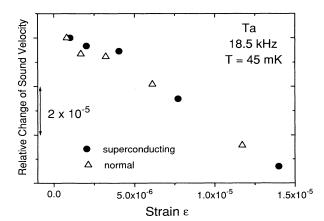


FIG. 11. Relative change of sound velocity for Ta as a function of strain in the normal ( $\triangle$ ) and superconducting ( $\bullet$ ) states at T=45 mK and 18.5 kHz.

compared to 5.5 kHz in Refs. 18 and 23. The systematic creation of tunneling systems through cold work of the metal has been reported for pure Ag. 18

# IV. CONCLUSIONS

The acoustic properties of polycrystalline Al, Pt, and Ta are identical to those found in amorphous insulators or superconductors at  $T \ll T_c$  at kHz phonon frequencies. The strain dependence of the sound velocity also resembles that found for amorphous SiO<sub>2</sub>. Therefore, we conclude that in these metals the interaction between tunneling systems and conduction electrons is absent or very weak. We observe in Pt similar changes of the acoustic properties after thermal treatment as those reported for amorphous superconductors. These changes indicate that, at least at the first annealing steps, the coupling constant between phonons and tunneling systems decreases.

Taking into account the results for amorphous superconductors in the normal state, amorphous PdSiCu, polycrystalline superconductors, Ag and Cu, and the results presented here, we can identify three different temperature dependences or behaviors.

- (1) The first one is the one observed in amorphous insulators, amorphous superconductors at  $T \ll T_c$ , and the metals Al, Ta, Pt, Nb, and NbTi in the superconducting state: well-defined maximum in the sound velocity, logarithmic slopes of the same magnitude in the temperature dependence below and above its maximum, and an internal friction with the typical plateau and with a decrease at low temperatures. In these materials we observe that the maximum in the sound velocity as well as the temperature dependence below the maximum is strongly dependent on strain.
- (2) A second one we identify for amorphous superconductors in the normal state and polycrystalline Ag and Cu. The temperature dependence of the acoustic properties, although qualitatively similar to that in (1), has strong quantitative differences that we believe are of importance: The sound velocity is weakly temperature dependent below 0.5 K, and the slope of the temperature dependence of the sound velocity for these metals is at least one order of magnitude smaller than those found in amorphous insulators and the polycrystalline metals described above. Measurements performed in Ag and Cu showed a maximum in the sound velocity at  $T_{\rm max} \sim$ 0.1 K and below  $T_{\text{max}}$  a logarithmic temperature dependence that extends into the  $\mu K$  region. In contrast to the behavior described in (1), the temperature dependence of the sound velocity below the maximum is much weaker than above it. The internal friction also shows an anomalously weak temperature dependence. A qualitatively different behavior as in (1) is observed for the strain dependence of the sound velocity: Ag and Cu show no strain dependence in the investigated strain range.<sup>24,34</sup>
- (3) The temperature and strain dependences of the acoustic properties of amorphous PdSiCu remain still unique.<sup>3,24,18</sup> An analysis of the data indicates that they can be explained assuming a Korringa-like relaxation

time for the electron–tunneling-system interaction<sup>3,32</sup> including the nonlinear response of tunneling systems with dissipation.<sup>33</sup>

We have no explanation for the different behaviors observed. We might speculate that some intrinsic parameters like the electron mean free path influences the interaction between electrons and tunneling systems and/or between the tunneling systems. In fact, the electron mean free path is the electronic property with the largest difference between an amorphous and a polycrystalline metal. Taking into account the low-temperature electrical resistivity, PdSiCu has the smallest electron mean free path, i.e., less than 1 nm. It might be possible that an effective enhancement of the relaxation rate of tunneling systems (as in PdSiCu) or the formation of a bound state occurs

only when the electron mean free path is smaller than some characteristic length of the interaction between tunneling systems.

### ACKNOWLEDGMENTS

We gratefully acknowledge discussions with Yu. Kagan. The authors are indebted to Frank Pobell for his collaboration and support, and for a careful reading of the manuscript, and to A. Betat for his collaboration with the measurements of the Pt sample. This work was supported by the German Research Society under Grant No. DFG Es 86/2-2.

- \* Present address: Fakultät für Physik, Universität Leipzig, Linnéstrasse 5, D-0341 Leipzig, Germany.
- <sup>1</sup> W. A. Phillips, J. Low Temp. Phys. **7**, 351 (1972). P. W. Anderson, B. I. Halperin, and C. Varma, Philos. Mag. **25**, 1 (1972).
- <sup>2</sup> S. Hunklinger and A. K. Raychaudhuri, in *Progress in Low Temperature Physics*, edited by H. Brewer (Elsevier, New York, 1986), Vol. IX.
- <sup>3</sup> B. Golding, J. E. Graebner, A. B. Kane, and J. L. Black, Phys. Rev. Lett. **41**, 1487 (1978).
- <sup>4</sup> P. Doussineau, P. Legros, A. Levelut, and A. Robin, J. Phys. (Paris) Lett. **42**, L265 (1978).
- <sup>5</sup> For reviews, see *Glassy Metals I*, Topics in Applied Physics Vol. 46, edited by H. Beck and H. J. Güntherodt (Springer, Berlin, 1981).
- <sup>6</sup> J. L. Black, in Ref. 5.
- <sup>7</sup> G. Weiss, S. Hunklinger, and H. v. Löhneysen, Phys. Lett. 85A, 84 (1981).
- <sup>8</sup> J. L. Black and P. Fulde, Phys. Rev. Lett. 43, 453 (1979).
- <sup>9</sup> K. Vladár and A. Zawadowsky, Phys. Rev. B 28, 1564 (1983); 28, 1582 (1983); 28, 1596 (1983).
- <sup>10</sup> G. Zaránd and A. Zawadowsky, Phys. Rev. Lett. **72**, 542 (1994).
- <sup>11</sup> D. C. Ralph and R. A. Buhrman, Phys. Rev. Lett. **69**, 2118 (1992).
- <sup>12</sup> D. C. Ralph, A. W. Ludwig, Jan von Delft, and R. A. Buhrman, Phys. Rev. Lett. **72**, 1064 (1994).
- <sup>13</sup> H. Neckel, P. Esquinazi, G. Weiss, and S. Hunklinger, Solid State Commun. 57, 151 (1986).
- <sup>14</sup> P. Esquinazi, H.-M. Ritter, H. Neckel, G. Weiss, and S. Hunklinger, Z. Phys. B 64, 81 (1986).
- <sup>15</sup> P. Esquinazi and J. Luzuriaga, Phys. Rev. B **37**, 7819 (1988).
- <sup>16</sup> F. Lichtenberg, H. Raad, W. Moor, and G. Weiss, in Phonons '89, edited by S. Hunklinger, W. Ludwig, and G. Weiss (World Scientific, Singapore, 1990), p. 471.
- <sup>17</sup> P. Esquinazi, R. König, F. Pobell, F. Dietzel, G. Weiss, and

- S. Hunklinger, in *Phonons '89*, edited by S. Hunklinger, W. Ludwig, and G. Weiss (World Scientific, Singapore, 1990), p. 423.
- <sup>18</sup> P. Esquinazi, R. König, and F. Pobell, Z. Phys. B 87, 305 (1992).
- Yu. Kagan and N. V. Prokof'ev, Solid State Commun. 65, 1385 (1978); Sov. Phys. JETP 70, 957 (1990).
- <sup>20</sup> S. N. Coopersmith, Phys. Rev. B **48**, 142 (1993).
- <sup>21</sup> M. W. Klein, B. Fischer, A. C. Anderson, and P. J. Anthony, Phys. Rev. B **18**, 5887 (1978); M. W. Klein, Phys. Rev. Lett. **65**, 3017 (1990).
- <sup>22</sup> C. C. Yu and A. J. Leggett, Comments Condens. Matter Phys. 14, 231 (1988).
- <sup>23</sup> R. König, P. Esquinazi, and F. Pobell, J. Low Temp. Phys. 90, 55 (1993).
- <sup>24</sup> P. Esquinazi, R. König, and F. Pobell, *Phonon Scattering in Condensed Matter VII*, Springer Series in Solid-State Sciences Vol. 112 (Springer, New York, 1993), p. 317.
- <sup>25</sup> P. Esquinazi, R. König, D. Valentin, and F. Pobell, J. Alloys Compounds 211-212, 27 (1994).
- <sup>26</sup> R. König, P. Esquinazi, and F. Pobell, Physica B **194-195**, 417 (1994).
- <sup>27</sup> K. Gloos, P. Smeibidl, C. Kennedy, A. Singsaas, P. Sekowski, R. Mueller, and F. Pobell, J. Low Temp. Phys. 73, 101 (1988).
- <sup>28</sup> J. Classen, C. Ens, C. Bechinger, G. Weis, and S. Hun-klinger, Ann. Phys. (Leipzig) 3, 315 (1994).
- <sup>29</sup> D. A. Parshin, Z. Phys. B **91**, 367 (1993).
- <sup>30</sup> J. T. Stockbürger, M. Grifoni, and M. Sassetti, Phys. Rev. B 51, 2835 (1995).
- <sup>31</sup> K. Gloos, C. Mitschka, F. Pobell, and P. Smeibidl, Cryogenics 30, 14 (1990).
- <sup>32</sup> S. N. Coppersmith and B. Golding, Phys. Rev. B 47, 4922 (1993).
- <sup>33</sup> J. Stockbürger, M. Grifoni, M. Sassetti, and U. Weiss, Z. Phys. B **94**, 447 (1995).
- <sup>34</sup> D. Valentin, Ph.D. thesis, Bayreuth Universität, 1993.