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Internal friction and sound velocity of polycrystalline metals at very low temperatures*

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Abstract

We have investigated the acoustical properties of polycrystalline Ag, Al, Cu, Nb, Nb with a Cu-matrix, $Nb_{48}Ti_{52}$, Pt and Ta at frequencies of $0.1\ kHz \leqslant \nu \leqslant 8\ kHz$ and in the temperature range $0.04\ mK \leqslant T \leqslant 1\ K$. The polycrystalline samples Al, Nb, Nb with a Cu-matrix, NbTi and Ta in the superconducting state as well as normal conducting Ag, Cu and Pt show qualitatively the same acoustical behaviour (with similar strain dependence) as observed for amorphous, dielectric materials. The glass-like "anomalies" can be understood assuming the existence of tunneling systems with a broad spectrum of energy splitting and relaxation rates. When the acoustical intensity is of the order of the thermal energy, non-linear effects are observed. We show and discuss an anomalous time dependence of the sound velocity in Pt as well as a strain-dependent and temperature-independent internal friction in Cu.

1. Introduction

In the last 15 years, the term glass-like has been used to describe the anomalous behaviour of low temperature properties found in special materials (orientationally or compositionally disordered dielectric crystals as well as some alloys [1]) resembling those found in amorphous solids. The accepted explanation for the anomalous behaviour is based on the hypothesis that the disordered lattice contains two-level tunneling systems (TS) [2, 3]. These low energy excitations of still unknown origin can interact with phonons and conduction electrons [1]. Since the glass-like behaviour is observed for a large number of amorphous and polycrystalline solids, it is obvious that the tunneling entity is not a special "defect" but more likely the "interaction" between defects of different nature. Experimental [4] as well as theoretical work [5–8] has provided some hints in this direction.

Until very recently simple polycrystalline metals were ignored as candidates in the search for glass-like behaviour. Our investigations of the acoustical properties of polycrystalline metals at very low temperatures performed during the last 4 years reveal a striking and unexpected fact, namely, all polycrystalline metals investigated to date show low temperature acoustical properties very similar, sometimes even quantitatively, to those found in amorphous solids.

In this paper, we present new data on the acoustical properties of the polycrystalline metals measured to the microKelvin temperature range. For a review of the predictions of the tunneling model (TM) used to interpret the observed behaviour as well as the acoustical results of other polycrystals, we refer to our recent publications [9–14].

2. Experimental details and samples

Among the low temperature properties used to characterize the glass-like behaviour, the acoustical properties (sound velocity v and internal friction Q^{-1}) provide an excellent tool since from them one can obtain information about the density of the tunneling states and their coupling to phonons or conduction electrons. Our measurements were performed with the vibrating reed [15] and vibrating wire [10, 11] technique. The relative change in the Young's modulus sound velocity v was determined by measuring the resonance frequency $\omega/2\pi$ of the vibrating sample. The internal friction was determined from the width of its resonance curve.

The sample holders were screwed to the upper flange of a Cu nuclear refrigeration stage [16]. Temperatures were determined by Pt NMR and/or PdFe susceptibility thermometers also connected to this flange. For details on the experimental setup as well as on the sample thermalization at ultra-low temperatures, see refs. 10 and 11. Different kinds of sample clamping were used, always ensuring good thermal contact with the Cu stage.

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TABLE 1. Investigated samples[a]

Material	Dimensions	Frequency (kHz)	Manufacturer	Purity	Remarks
Ag	7×2×0.03 mm^3	0.25	GF	1 ppm 100 ppm	Vibrating reed
Al	125 μm	0.6	GF	4N	s
	5 μm	0.3		3N5	s
Cu	25 μm	0.3	LK	–	–
	125 μm	0.37	GF	3N	–
Nb	120 μm	1.48	SI	–	HNO$_3$ treated, s
Nb(Cu)	165 μm	0.5	SI	–	s
Nb$_{48}$Ti$_{52}$	20 μm	0.9	VH	–	s, one filament
	20 μm	0.6	SI	–	s, (a)
	40 μm	3.7	SI	–	s, (m)
	50 μm	0.37	SI	–	s
	50 μm	1.47	SI	–	s, with insulation
Pt	25 μm	0.76	GF	4N	RRR=460
	25 μm	0.084	GF	4N	RRR=460
	25 μm	1.5	GF	4N	RRR=460
Ta	125 μm	1.8	GF	3N	s, annealed
	125 μm	5.5	GF	3N	s, annealed

[a] GF, Goodfellow Metals (UK); LK, Lacroix+Kress; SI, Supercon. Inc., Massachusetts (USA); VH, Vakuumschmelze Hanau (D); s, superconducting; RRR, residual resistivity ratio. Except for Ag, the samples were measured with the vibrating wire technique; the written dimension represents their diameter. (a), surface treated with acid; (m), insulation mechanically removed.

After an extensive study of the possible influence of the clamping, we can conclude that the observed temperature dependencies are not influenced by the clamping conditions, which give only a constant background contribution to Q^{-1}.

The measurements were performed as a function of temperature at constant strain or constant driving force and also as a function of strain at constant temperature. The given strain values refer to the maximum strain at the fixed ends of the sample taken at the maximum oscillation amplitude [10, 11]. Depending on the frequency and maximum oscillation amplitude, the dissipated energy was between 10^{-15} W and 10^{-10} W.

Table 1 lists the investigated samples, their resonance frequencies and some other characteristics. Typical lengths of the wires were between 4 and 20 mm. Some of the superconducting wires were treated with acid (HNO$_3$) to remove the Cu-matrix or to reduce their diameter to study possible surface contributions. Besides differences in the thermal conduction and in self-heating effects at $T<10$ mK, results were not changed by this treatment.

3. Experimental results and discussion

3.1. Glass-like behaviour of the acoustical properties

At $T<2$ K and at audio frequencies, the sound velocity v of glasses is characterized by a strain-dependent maximum in v at a temperature T_{max} and a logarithmic temperature dependence of v above and for small strains also below T_{max}. At $T>T_{\mathrm{max}}$, the internal friction Q^{-1} is nearly temperature-independent (plateau) with relatively large values and decreases at lower temperatures [1, 10, 15]. With the exception of the internal friction in Cu discussed at the end of this paper, we find those features in all polycrystalline metals listed in Table 1!

Superconducting polycrystals give us the possibility of comparing their acoustical properties with those of amorphous dielectrics since at $T \ll T_c$ (superconducting transition temperature), the number of normal conducting electrons is negligible and the TS interact only with phonons, as measurements with amorphous superconductors [18] have shown.

Figure 1 shows both acoustical properties as a function of temperature at different strains for Nb in the superconducting state. From the slope and maximum of the sound velocity as well as from the value of Q^{-1} at the plateau, we conclude that the density of states of TS and their coupling to phonons are of the same order of magnitude as observed in amorphous dielectrics, *e.g.* vitreous silica SiO$_2$ [10]. Our results are consistent with the predictions of the tunneling model for glasses. From the observed behaviour, we obtain the parameter $C=P\gamma^2/\rho v^2 \approx (0.6\pm0.1)\times10^{-4}$ (P is the density of states of TS, γ is the coupling constant between phonons and TS, and ρ is the mass density [1]) which is only a factor

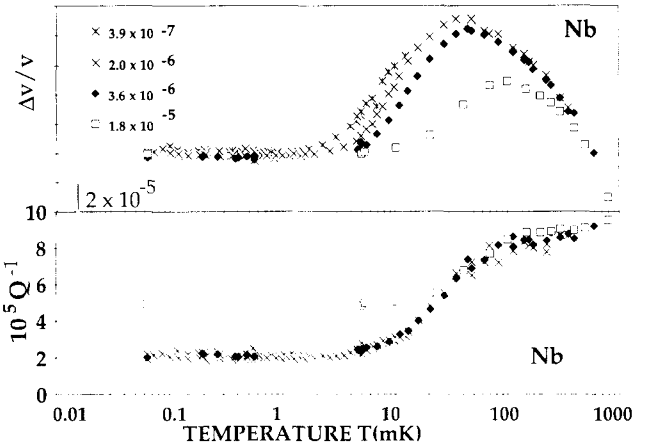

Fig. 1. Relative change of sound velocity and internal friction of Nb as a function of temperature for different applied strains. The sound velocity curves were normalized at about 1 K to allow a better comparison. In general, the sound velocity decreases with strain at $T<T_{max}$; see Fig. 3. The data at $\epsilon=1.8\times10^{-5}$ are influenced by sample heating below 20 mK.

~4 smaller than for SiO_2 [10]. The different values of C which we obtain for other polycrystals [10–14] are not related to some intrinsic property or impurity concentration of them but to their thermal or mechanical history as was observed for Ag [9, 10]. It is important to remark that also in amorphous metals the C values obtained from the acoustical properties decrease after thermal annealing while the sample remains in the amorphous state [17], as observed for polycrystalline metals.

For a better comparison, we have plotted in Fig. 2 the sound velocity of amorphous SiO_2, superconducting Nb and normal conducting Pt. Surprisingly, the acoustical properties of the normal metal Pt [14] resemble perfectly those of amorphous dielectrics or superconducting polycrystals. This result disagrees with the predictions of the standard [19] and of new models [20] for the conduction electrons–TS interaction.

The dependence of the internal friction Q^{-1} shown in Fig. 1 is typical for amorphous dielectrics [1, 10]. The saturation of Q^{-1} observed at $T<5$ mK is attributed to a clamping contribution. The temperature dependence of Q^{-1} below the plateau at $T>0.1$ K and after subtraction of the background absorption does not follow the predicted T^3 dependence from the TM but a nearly linear temperature dependence. This anomalous linear T-dependence below the plateau has also been observed in SiO_2 [10] and recently in Pt [14]. A possible explanation of this behaviour based on the formation of a subsystem of collective excitations independent of phonons and due to the TS up–down interaction has been given recently by Burin and Kagan [21]. The behaviour of the internal friction shown in Fig. 1 was observed in all polycrystals investigated [10–14] with the exception of Cu (see below).

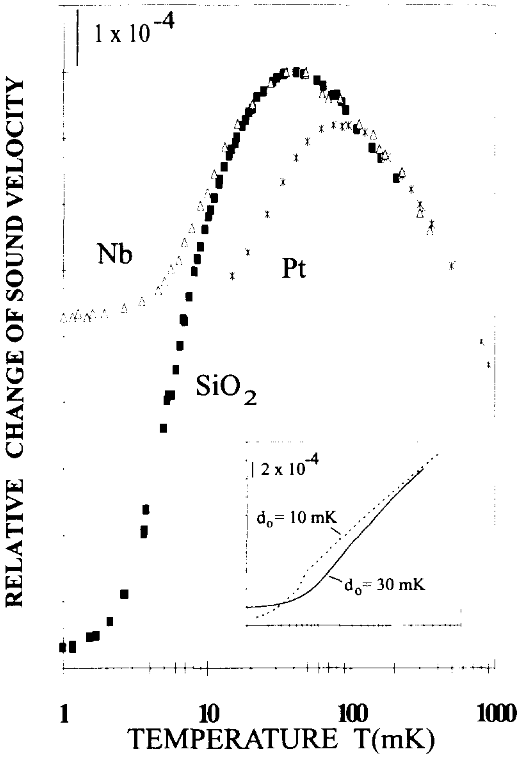

Fig. 2. Relative change of sound velocity for vitreous silica SiO_2 (0.4 kHz), polycrystalline Nb (1.5 kHz) and Pt (760 Hz). The v-scale for Nb (Pt) has to be divided by a factor of 4 (5). The inset shows numerical calculations according to the tunneling model and taking into account the time-dependent applied strain in the resonant interaction between phonons and TS. The curves were normalized at about 1 K for a better comparison. The theory predicts a decrease in the sound velocity with strain at $T<T_{max}$ [10, 11].

3.2. Strain dependence and self-heating effects

The strain-dependent behaviour in the sound velocity, see Fig. 1, *i.e.* the shift of its maximum, the change in the logarithmic slope below it and its apparent saturation at the lowest temperatures (see also Fig. 2), is observed when the energy of the sound wave or strain field $d(t)$ is of the order of the thermal energy k_BT [10]. We can explain these anomalies by introducing a change in the population number $n \alpha\, tgh(E/2k_BT)$ of the tunneling states ($E^2=(\Delta+d(t))^2+\Delta_0^2$ is the energy difference between the two levels, Δ is the asymmetry and Δ_0 the tunneling energy) taking into account the strain field $d(t)$ created by the sound wave. This change affects mainly the resonant part of the interaction between TS and phonons that accounts for the decrease in the sound velocity below the maximum. Consistent with this explanation is the negligible strain dependence of the attenuation (see Fig. 1 and also ref. 10). The inset in Fig. 2 shows the results of new numerical calculations of the sound velocity taking into account resonant interaction only and a time-dependent strain field

$d(t)=d_0\cos(\omega t)$. These numerical results agree qualitatively with those published in [10].

It is important to stress that the apparent saturation of the sound velocity at the lowest temperatures and at low enough strains ($\epsilon<10^{-5}$ for Nb, see Fig. 1) is not related to self-heating effects (that affect both acoustical properties at larger strains ($\epsilon>10^{-5}$), see Fig. 1) but can be explained by the population change produced by the strain field (see inset in Fig. 2). The exclusion of self-heating effects is reinforced by: (a) the slope of the sound velocity as a function of strain is independent of temperature at $\epsilon<3\times10^{-6}$ (see Fig. 3); (b) the strain independence of Q^{-1} (Fig. 1); and (c) by the agreement of results of a Nb wire without and with a Cu-matrix that enlarges enormously the thermal conductivity of the sample.

Figure 3 shows that even at $T\sim0.1$ mK and at small strains, the sound velocity in Nb decreases with strain with the same slope as measured at much higher temperatures. The s-shape of the strain dependence of the sound velocity shown in Fig. 3 can be explained by a superposition of three non-linear effects as discussed in [11]. The observed logarithmic decrease in the sound velocity with strain can be well accounted for by the change in the population difference of the TS [11].

The sound velocity in Nb closely follows a linear temperature dependence at temperatures below 20 mK (apparent saturation of v in the semilog plot of Fig. 1). This dependence can be extracted also from Fig. 3 taking the sound velocity at constant strain. This nearly linear T-dependence in the non-linear regime can also be accounted for by a change in the population as described before. The observed strain- and T-dependence of the sound velocity in the non-linear regime disagree with the dependence $\Delta v/v\propto(T/d_0)^2$ obtained in [22].

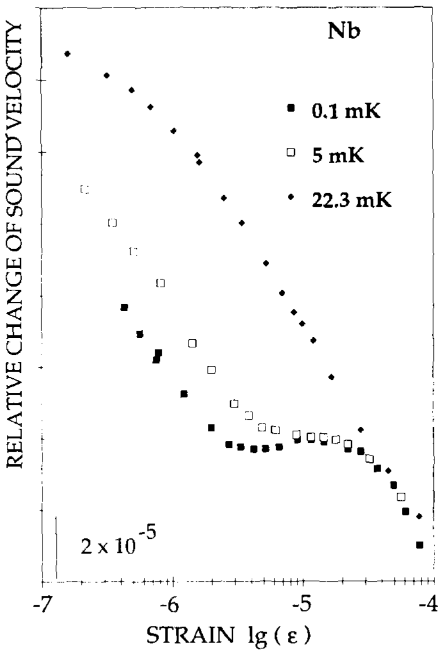

Fig. 3. Strain dependence of the sound velocity of Nb at three different temperatures. The s-shape of the curves can be understood taking into account three non-linear processes [10, 11].

3.3. Time-dependent effects

Time dependence of the acoustical properties at $T<10$ mK has been observed in the amorphous materials SiO_2 [10] and PdSiCu [10, 23] and has been attributed to the relatively large thermal resistance of the dielectric material or to the weak coupling of the nuclei in the sample to the cooling stage. Recently, we have observed a novel time dependence of the acoustical properties of Pt. The Pt-wire was soft soldered to a thick Cu-wire that was the sample holder. Other thermal (*e.g.* made with Stycast) and electrical contacts for other wires (see Table 1) have been already used and show no measurable influence on the wire thermalization. Figure 4 shows the relative change in sound velocity as a function of time for Pt measured after a relatively rapid ($t<100$ s) change in temperature. We observe a strong time dependence of the sound velocity, with a relaxation time strongly decreasing with temperature.

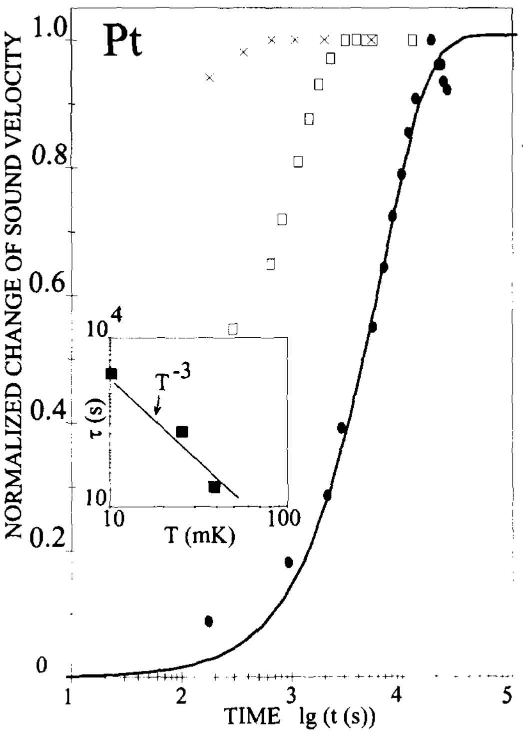

Fig. 4. Time dependence of the sound velocity in Pt at three rapid ($t\sim100$ s) temperature changes: from 5 mK to 40 mK (×), 26 mK (□) and 10 mK (●), respectively. The inset shows the temperature dependence of the relaxation time obtained from a fit using eqn. (1).

It can be shown that the resonant interaction between TS and phonons is proportional to the population number $n(E, T)$ of TS [1]. The temperature dependence of the sound velocity at $T < T_{\max}$ is due to this interaction. If a sample is cooled or heated rapidly from a "charging" temperature T_1 to a measuring temperature T_0, the time-dependent relative change in the sound velocity in the relaxation time approximation and within the TM is given by

$$\frac{\Delta v}{v} = -\frac{\gamma^2}{\rho v^2} \int\int P(\Delta, \Delta_0) \left(\frac{\Delta_0}{E}\right)^2 \frac{1}{E} \times [n(E, T_1) \exp(-t/\tau(T_0, E)) + (1 - \exp(t/\tau(T_0,E)))n(E, T_0)] \, \mathrm{d}\Delta \, d\Delta_0 \qquad (1)$$

where $P(\Delta, \Delta_0) d\Delta d\Delta_0 = (P/\Delta_0) d\Delta d\Delta_0$ and $\tau(T, E)$ is the relaxation time between TS and phonons and/or conduction electrons. Expression (1) should not be confused with the logarithmic time dependence of the sound velocity in the non-linear regime predicted in [22].

We have fit the observed time dependence to three different kinds of relaxation rates in (1): (a) TS-phonons, (b) TS-electrons and (c) an only temperature-dependent relaxation rate. With reasonable values for the coupling constant between TS and phonons (or electrons), we obtain qualitatively the observed time dependence but shifted to lower times by several orders of magnitude. For example, an effective relaxation time $\tau \sim 0.1$ s at 10 mK is obtained for the TS-phonon interaction instead of ~2500 s observed experimentally (see Fig. 4). Note also that the acoustical properties of Pt contradict the use of the standard TS-electron relaxation rate. The fit of eqn. (1) to the data, shown in Fig. 4, was obtained assuming an TS-energy-independent relaxation time. The fit to the three curves gives a T^{-3} dependence for $\tau(T)$ in (1) (see inset in Fig. 4).

The acoustical properties indicate that the TS in Pt interact only with phonons (see Fig. 2 and ref. 14). Because the TS-phonon relaxation time is several orders of magnitude smaller than the one measured between 10 and 40 mK (inset in Fig. 4), we interpret this relaxation time as due to the phonon thermalization in the sample, *i.e.* the TS act as a microscopic thermometer of the phonon temperature. This thermalization can be due, for example, to the phonon–electron interaction. A T^{-3} dependence for the energy-loss lifetime for the conduction electron–phonon interaction has been observed in Cu-films at milliKelvin temperatures [24] and for other metals at higher temperatures [25]. However, the absolute value of the relaxation time obtained here for Pt is three to five orders of magnitude larger than that measured in other metals [24, 25].

At present we are investigating whether the observed relaxation time depends on the amount or kind of defects (*i.e.* TS) in Pt or on the applied strain. Preliminary results on a Pt wire with smaller defect concentration and with the same kind of thermal coupling to the nuclear stage indicate a smaller and strain-independent time constant.

3.4. Amplitude-dependent internal friction

While the sound velocity in Cu shows the same glass-like behaviour as in Ag [12], the internal friction shows an "anomalous" strain and temperature dependence. We have measured two polycrystalline Cu wires with 25 μm (Cu no. 1, 300 Hz) and 125 μm (Cu no. 2, 370 Hz) diameter in the temperature range 0.06 mK $< T <$ 900 mK and in the strain range $7 \times 10^{-4} \geqslant \epsilon \geqslant 3 \times 10^{-7}$. Our results shown in Fig. 5 indicate that at $T <$ 300 mK $Q^{-1} \propto \epsilon^n$ with $n = 0.41 \pm 0.02$ for $\epsilon > 10^{-6}$ independent of temperature, *e.g.* Q^{-1} ($T <$ 300 mK) $\cong 5 \times 10^{-4}$ for $\epsilon \cong 5 \times 10^{-6}$. For lower strains Q^{-1} deviates from this dependence (see Fig. 5); this deviation is larger the higher the temperature.

The behaviour of Q^{-1} in Cu may be explained by tunneling of pinned dislocations. At sufficiently low temperatures and high stress, the escape of dislocations from their pinning centers can be more effective via tunneling than by means of thermal activation [26–28]. Unlike the temperature-dependent phonon assisted tunneling rate used in the tunneling model to explain the glass-like low temperature properties [1], we have to assume only a stress-dependent rate to explain the observed behaviour in Cu. The form of that rate might be, for example, $\propto \exp(-E_0/2\sigma V)$ [28], where E_0 is the

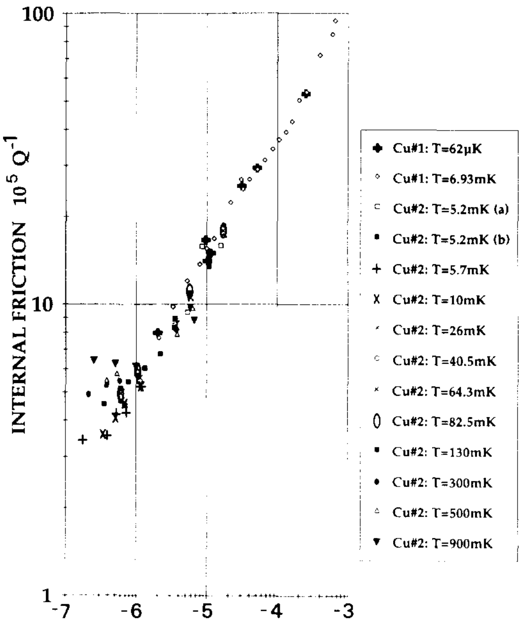

Fig. 5. Strain dependence of the internal friction of two Cu-wires at different temperatures. (a) Applied magnetic field = 9 mT; (b) 27.4 mT. Wire's diameter 25 μm (no. 1) and 125 μm (no. 2).

binding energy, σ the local stress and V an atomic volume. Within this picture, it is possible to understand the observations qualitatively: the strain dependence as well as the temperature independence and the start of the thermally activated regime at the low stress region (Fig. 5). To understand the results quantitatively, two points should be clarified: a possible binding energy distribution and the background contribution to Q^{-1} due to TS which should be present according to the temperature dependence of the sound velocity [12]. A similar strain dependence of Q^{-1} superimposed on the temperature-dependent and strain-independent TS contribution to Q^{-1} has recently been observed in two Pt wires.

4. Conclusions

With the vibrating wire and vibrating reed technique and nuclear demagnetization, we have measured the acoustical properties of several polycrystalline metals at very low temperatures. Our results indicate that there are basically no differences in the low-temperature, low-frequency acoustical properties of polycrystals and amorphous materials. The glass-like anomalies are by far more universal than expected. Without question this result is more significant than the deviations from the predictions of the standard tunneling model.

As in amorphous materials, the acoustical properties of polycrystals can be understood within the tunneling model and a phonon-assisted relaxation rate. The influence of conduction electrons to the tunneling rate remains still an open question. The observed non-linear effects in the sound velocity can be understood taking into account the dependence of the population of the TS on the acoustical intensity. Time-dependent effects were measured and indicate that TS acts as a microscopic thermometer of the phonon temperature. The behaviour of the internal friction in Cu (as well as in Pt) over more than three orders of magnitude in temperature and strain may be explained by dislocation tunneling. We are confident that the observed similarity in the acoustical properties of very different kinds of materials will trigger new experiments which might contribute to the understanding of the nature and interaction of the tunneling entities in disordered media and polycrystalline materials.

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