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INFLUENCE OF IMPURITIES AND DISLOCATIONS ON ULTRASONIC ATTENUATION IN COPPER AND DILUTE COPPER ALLOYS AT LOW TEMPERATURES

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Abstract.— We have measured the phonon-electron attenuation α_{PE} ($10 < f < 200$ MHz, long. wave in $\langle 111 \rangle$) in Cu single crystals of widely different purity ($20 < RRR < 20.000$) after suppression of dislocation damping by γ -irradiation. The α_{PE} -data are in agreement with Pippard's theory. The measurements on crystals doped with paramagnetic impurities (Fe, Mn) show an "ultrasonic Kondo-effect".

1. Introduction.— At $T < 100$ K the ultrasonic attenuation (UA) in pure Cu (impurity content typically $< 10^2$ ppm) is determined by dislocations (dislocation resonance attenuation α_{GL}) and electrons (phonon-electron attenuation α_{PE} /1/). Both effects are superimposed on a comparatively small background attenuation α_B (being mainly caused by the thermoelastic effect, sample- and sound field geometry /2/). Fig.1 (upper curve) shows as an example the attenuation of high purity Cu ($RRR \sim 1000$, sandwich buffer crystal /3/ to avoid quartz sample deformation /4/) which strongly increases below 100 K due to α_{PE} . (The sample was slightly γ -irradiated ($\Phi_Y = 10^2 \mu Ah$) to demonstrate the advantages of the buffer technique). However, by a heavy γ -irradiation which causes strong pinning of the mobile dislocation loops the dislocation attenuation α_{GL} can be completely suppressed and the $(\alpha_{PE} + \alpha_B)$ -contributions remain (lower curve). Since α_B is only slightly temperature dependent between 300 and 100 K the $\alpha(T)$ -behaviour after irradiation can be extrapolated to 4.2 K (dashed dotted curve) in order to separate α_{PE} . Subtraction of $\alpha_{PE} + \alpha_B$ finally yields the wanted dislocation attenuation α_{GL} (dashed curve in fig.1).

FIG 1

Temperature dependence of dislocation attenuation α_{GL} , background α_B and phonon-electron attenuation α_{PE}

The present paper reports on α_{PE} measurements on a wide variety of Cu crystals used for studies of dislocation damping effects. It compares measured α_{PE} with theoretical predictions by using residual resistivity

ratios RRR measured on the ultrasonic samples. The resulting Pippard factor /5/ then allows calculation of $\alpha_{PE}(T)$ and thus evaluation of α_{GL} from the $\alpha(T)$ measurement and knowledge of RRR without resort to irradiation (i.e. the second $\alpha(T)$ -run on the irradiated sample is not needed).

2. Experiment. - We measured longitudinal UA ($10 < f < 200$ MHz) in $\langle 111 \rangle$ -oriented Cu single crystals between 4.2 and 300K. Sample preparation is described elsewhere /9/. The residual resistivity ratios of the individual samples ($20 < RRR < 20.000$) were measured by the eddy current technique /6/. The γ -irradiation was produced by a Van de Graaff electron generator in an Au-target (Frenkel defect (FD) production rate 5.5×10^9 FD/cm²s /9/. The total irradiation induced defect concentration needed for complete dislocation pinning is $< 10^{-3}$ ppm. This low concentration does not influence the RRR and thus α_{PE} to any measurable extend.

3. Theory of phonon-electron absorption. - According to Pippard (free electron gas assumed /5/) the attenuation α_{PE} of longitudinal sound in a metal is determined by $a = q \cdot \ell$ with $q = |\vec{q}(111)| = 2\pi f / v_L$ the wave number, ℓ the mean free path length of the electrons and v_L the sound velocity ($5.16 \cdot 10^5$ cm/s)

$$\alpha_{PE} = A_{FE} f \frac{6}{\pi} \left\{ \frac{a}{3} \left(\frac{\arctan a}{a} - \arctan a \right) - \frac{1}{a} \right\} \quad (1)$$

$A_{FE} = \pi^2 N m v_F / 3 \rho_D v_L^2$ with N = electron concentration, m = electron mass, v_F = Fermi-velocity, ρ_D = density of Cu. For comparison with experimental results the limits of equ.(1) are useful

$$q\ell \gg 1 \text{ ("pure limit")}: \quad \alpha_{PE}/A_{FE} \cdot f = 1 \quad (2)$$

$$q\ell < 1 \text{ ("impure limit")}: \quad \alpha_{PE}/A_{FE} \cdot f = 0.5 q\ell \quad (3)$$

The "impure limit" refers to doped crystals (having small ℓ (i.e. low RRR or high T) or to measurements at low f). The "pure limit" refers to high purity crystals at lowest T measured at high frequencies.

4. Results and discussion. - Fig.2 shows $\alpha_{PE}(T)$ -curves for samples of different purity levels (e.g. RRR=20.000 refers to < 0.1 at ppm total impurity (Fe)). The strong α_{PE} increase between 60 and 20 K is due to increasing ℓ . The maximum $\hat{\alpha}_{PE}$ measured at 4.2K is determined by ℓ_0 the residual ℓ for the "impure" case ($q\ell < 1$, e.g. samples 4,6,7) where $\hat{\alpha}_{PE} \sim \ell_0 \sim RRR$. In the "pure limit" ($q\ell \gg 1$, sample 1) $\alpha_{PE} = A_{FE} f$, i.e. the maximum attenuation is given by the Pippard factor A_{FE} (equ.(2)). For comparison of magnitude the dashed curve in fig.2 shows the dislocation attenuation α_{GL} measured on sample 2: at $T > 50K$ $\alpha_{PE} \ll \alpha_{GL}$; however, at $T < 10K$ we find $\alpha_{PE} \sim 10 \alpha_{GL}$. This shows that the knowledge of α_{PE} is of fundamental importance for the evaluation of dislocation damping at these low T .

FIG 2 Temperature dependence of α_{PE}

FIG 3 Frequency dependence of α_{PE}

Fig.3 shows $\alpha_{PE}(f)$ for three samples of widely different RRR. Whereas the impure samples exhibit curved α_{PE} behaviour (equ.(3)) the highest purity sample 1 shows a linear $\alpha_{PE}(f)$ -dependence (equ.(2)). From its slope we obtain the Pippard factor $A_{FE} = 0.045 \pm 0.005$.

Fig.4 shows the T-dependence of the electronic mean free path $\ell(T)$ derived with help of equ.(3) and $A_{FE}=0.045$ (left scale). It can be seen that for the low sample purities shown the $\alpha_{PE} \sim f^2$ dependence is fulfilled within experimental accuracy (right scale). The data can be compared with $\ell_{IDEAL}(T)$ the "ideal" (lattice-) mean free path length. As to be expected the measured $\ell(T)$ (i.e. $\alpha_{PE}(T)$) is determined by ℓ_{IDEAL} at $T > 50\text{K}$ whereas it becomes impurity controlled at lower T. For $T \rightarrow 4.2\text{K}$ individual ℓ_0 values are measured which scale with the samples RRR values ($\ell_0 \sim \text{RRR}$).

FIG 4

T-dependence of electron mean free path length ℓ compared with ℓ_{Ideal}

Fig.5 depicting $\alpha_{PE}(f)$ for the same samples as in fig.4 shows more directly the $\alpha_{PE} \sim f^2$ behaviour (which is seen to break down for the purer sample at highest f). Since $\alpha_{PE} \sim l$ (equ.(3)) the slope 2-lines scale with RRR. The dashed line of slope 1 depicts the upper limit of applicability of equ.(3) defined by $\alpha_{PE}/f = 0.02$ (see below). From this limiting condition we obtain the experimentally more useful form $f_{LIM} [MHz] = 2 \cdot 10^4 / RRR$ which limits the ($\alpha_{PE} \sim f^2$)-range to frequencies $f \leq f_{LIM}$. For experiments outside this range ($f > f_{LIM}$ or $RRR > 2 \cdot 10^4 / 5MHz = 4.000$, $f = 5MHz$ being the lowest f for the present pulse echo technique) instead of equ.(3) the complete Pippard formula (equ.(1)) must be used for calculation of α_{PE} .

Fig.6 shows our α_{PE}/f data (left scale) fitted to the theoretical $\alpha_{PE}/A_{FE} \cdot f$ behaviour (equ.(1), right scale). The fit yields $A_{FE} = 0.045$ the same value as already derived from the data in the pure limit (dashed line). Pippards equation (1) describes the present data over almost three decades in ql within narrow limits (10%). Thus it can be used for calculation of the α_{PE} contribution and separation of the wanted dislocation resonance damping. Up to $\alpha_{PE}/f = 0.02$ (i.e. $ql < 0.9$) the "impure limit" equ.(3) can be used with sufficient accuracy.

FIG 6 Present α_{PE} data fitted to Pippard equation (equ. (1)) to obtain Pippard factor A_{FE} (≤ 0.045).

FIG 5

$\alpha_{PE}(f)$ in the $ql < 1$ limit defined by $\alpha_{PE} \leq 0.02 f$ (dashed line)

5. Ultrasonic Kondo-Effect.— Our results on crystals doped with paramagnetic impurities (Fe, Mn) exhibit on "anomalous" $\alpha_{PE}(T)$ dependence (attenuation maximum) at $T < 30K$ (c.f. fig.1, sample 7). Fig.7 shows this for a Cu+50ppmFe and a Cu+60ppmMn crystal. We plot the inverse attenuation vs T since according to equ.(3) $f^2/\alpha_{PE} \sim 1/\ell$ is directly proportional to the electrical resistivity ($\rho \sim 1/\ell$). Both results in fig.7 are in accordance with measured $\rho(T)$ -curves /7,8/ on dilute CuFe, CuMn which show Kondo behaviour. Table 1 (comparing our results with the resistivity-data) shows close agreement between the observed temperature T_{KMIN} (Kondo-minimum temperature) and Kondo-slope $SL_K = [1/x(4.2)] \cdot dx/d\log T$ with $x = \Delta\rho(T)/c$ (c =impurity concentration) for the electrical resistivity data and $x = f^2/\alpha_{PE}(T)$ for the present ultrasonic attenuation data.

FIG 7 T-dependence of ultrasonically measured resistivity $\sim f^2/\alpha_{PE}$ showing Kondo-minimum.

TAB.1

		Ultrasonic Attenuation	Electrical Resistivity
CuFe	$T_{KMIN}[K]$	22	21 /7/
	SL_K	0.149	0.153 /7/
CuMn	$T_{KMIN}[K]$	17	16.2 /8/
	SL_K	0.176	0.178 /8/

The close agreement between T_K and SL_K (measured by attenuation and resistivity) indicates that the same phenomenon is observed. To our knowledge this is the first ultrasonic measurement of Kondo scattering.

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