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Temperature Dependence of Carrier Scattering Rate in Copper from Magnetoacoustic Experiments*

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Abstract

The temperature dependence of the carrier scattering rate in copper has been determined from magnetoacoustic data. For both the (100) central belly orbit and the (110) 'dogs-bone' orbit a T^3 dependence is observed, the coefficients of the corresponding term in the scattering rate agreeing well with those obtained from radio frequency size-effect experiments. Previously reported discrepancies between the results obtained by the two methods are discussed and tentatively resolved.

Introduction

There is considerable interest in the temperature dependence of the carrier scattering rate in the noble metals. Radio frequency size-effect (RFSE) experiments have shown that below 10 K the scattering rate ν for both belly orbits and the 'dogs-bone' orbit in copper can be well represented by the equation $\nu = \nu_0 + aT^3$ (Gantmakher and Gasparov 1973). In this expression the constant ν_0 is associated with impurity scattering, while the T^3 term is due to catastrophic scattering of the electrons by thermal phonons. Effects due to electron–electron scattering, which would contribute an additional T^2 term in the preceding equation, are not observable (Gasparov and Harutunian 1976). The magnitude and anisotropy of the coefficient a are in reasonable agreement with calculations of the electron–phonon scattering in copper (Nowak 1972).

Recent magnetoacoustic experiments (Khatri and Peverley 1975) on ultrapure single crystals have also been used to analyse the carrier scattering rates in copper. Although the data are consistent with electron–phonon scattering being the dominant process, the coefficients of the T^3 term differ markedly from those obtained from RFSE experiments. For longitudinal waves propagating along [001] with a magnetic field directed along [100], the coefficient a corresponding to a (100) central belly orbit appears to be $(6\cdot0 \pm 0\cdot3) \times 10^6\ \mathrm{s^{-1}\,K^{-3}}$ compared with the value $(3\cdot1 \pm 0\cdot1) \times 10^6\ \mathrm{s^{-1}\,K^{-3}}$ obtained from RFSE data. This discrepancy is very disturbing, since it casts doubt on the utility of the magnetoacoustic technique in scattering rate studies.

The present paper reports independent measurements of the temperature dependence of the magnetoacoustic effect in single crystals of high purity copper. For both the (100) belly orbit and (110) 'dogs-bone' orbit the coefficients of the T^3 term in ν are found to be in good agreement with RFSE data. From this work and the extensive magnetoacoustic experiments of Kamm (1970), it is suggested that the above mentioned discrepancy may be due to a misinterpretation of crystal orientation and that both techniques are useful in studies of the carrier scattering rate in high purity metals.

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Experiment

The samples used in this work were spark cut from a single crystal ingot supplied by the National Bureau of Standards. This ingot had been oxygen annealed and had a resistance ratio stated to be in excess of 20 000 to 1. For most measurements a single specimen, with pairs of parallel (100) and (110) faces approximately 1 cm apart, was used. At higher frequencies separate samples, approximately 3 mm thick with normals along [100] and [110], were necessary because of the high zero-field electronic attenuation. In all cases the surfaces were carefully etched after cutting to remove the surface damage.

Magnetoacoustic measurements up to 270 MHz were made in transmission with longitudinal waves generated by 10 or 30 MHz *X*-cut transducers operating in an overtone mode. For the thinner samples a *Z*-cut delay line was used to provide adequate time separation between the transmitter pulse and the received echoes. Acoustic bonds were made with Duco cement. Data were obtained with a 12 in. (30·5 cm) Varian electromagnet at fields up to 1 kG, using a Princeton Applied Research Model 162 boxcar signal averager to obtain the amplitude of the transmitted acoustic signal. Conventional insertion loss techniques were used in calibrating the latter to give changes in relative attenuation both as a function of applied field and temperature. The sample was maintained in good thermal contact with a copper block, the temperature of which was controlled electronically and measured to an accuracy of better than 0·01 K with a calibrated germanium thermometer.

Theory

The general theory of the magnetoacoustic effect (Mertsching 1970) shows that the oscillatory component of the acoustic attenuation α for a specific extremal orbit is given by the relation

$$\alpha = Cq^{-\frac{1}{2}}\cos(qD-\tfrac{3}{4}\pi)/\sinh(\pi\nu/\omega_c)\,. \tag{1}$$

In this equation C is a constant depending on the geometry and deformation characteristics of the Fermi surface, $\boldsymbol{q}$ is the phonon wave vector, D is the orbit size in real space, ω_c is the relevant cyclotron frequency for the orbit and ν is the orbital average of the carrier scattering rate. The orbital dimension D is related to an extremal dimension Δk along the direction $\boldsymbol{q}\times\boldsymbol{H}$ in $\boldsymbol{k}$ space by the equation $D = (c\hbar/eH)\Delta k$. Therefore, from equation (1) it follows that the oscillations due to a given extremal orbit are periodic in H^{-1} and that Δk is given by the expression

$$\Delta k = (e/\hbar c)\,\lambda/\Delta(H^{-1}), \tag{2}$$

where λ is the sound wavelength and $\Delta(H^{-1})$ the relevant period in H^{-1}.

If the scattering rate is expressed in the form $\nu = \nu_0+\nu(T)$, then in the limit $\pi\nu_0/\omega_c \gg 1$ equation (1) may be rewritten as

$$\alpha = \alpha_0\exp(-\pi\,\nu(T)/\omega_c)\,, \tag{3}$$

where α_0 is the limiting oscillatory component of attenuation at absolute zero for a fixed field. Hence, a suitable plot of α/α_0 versus temperature gives $\nu(T)$, provided that the cyclotron frequency ω_c is known.

Results and Discussion

Fig. 1 shows typical magnetoacoustic data obtained at 152 MHz for longitudinal waves propagating along [011] with the magnetic field $\boldsymbol{H}$ along [100]. The oscillations are strictly periodic in H^{-1} and correspond to an extremal Fermi surface radius Δk_r along $[01\bar{1}]$ of $(1\cdot33\pm0\cdot02)\times10^8\,\mathrm{cm}^{-1}$. This value is in good agreement with the result $(1\cdot30\pm0\cdot02)\times10^8\,\mathrm{cm}^{-1}$ obtained by Kamm (1970) and assigned by him to a (100) central belly orbit. The oscillatory behaviour is essentially free from interference caused by other extrema, associated either with the four-cornered 'rosette' or off-centre belly orbits.

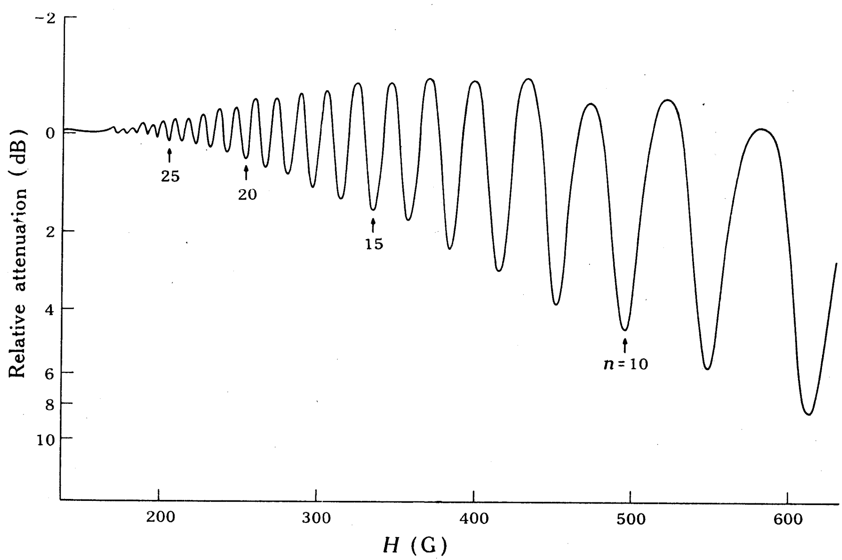

Fig. 1. Typical magnetoacoustic oscillations for 152 MHz longitudinal waves propagating along [011] in high purity copper. The temperature was 4·65 K and the field $\boldsymbol{H}$ was along [100]. Note the decrease in amplitude with increasing harmonic number n. (Note also $1\ \mathrm{G} \equiv 10^{-4}\ \mathrm{T}$.)

From Fig. 2 it can be seen that for harmonic number $n > 15$ the data are well represented by $\nu(T) = aT^3$, corresponding to catastrophic electron–phonon scattering as in the case of RFSE experiments. Even for n as low as 10 there is no evidence for the T^5 dependence, characteristic of diffusive type scattering, reported by Khatri and Peverley (1975). Fig. 3 shows the corresponding values of $a = \nu(T)/T^3$ as a function of harmonic number, using the relevant values of ω_c for copper (Koch *et al.* 1964). For n greater than $\sim$20 the data clearly reach an asymptotic value of $a = (2\cdot7\pm0\cdot1)\times10^6\,\mathrm{s}^{-1}\,\mathrm{K}^{-3}$. The measurements of Gantmakher and Gasparov (1973) on a 0·53 mm thick [001] sample for a central belly orbit with $\boldsymbol{H}$ parallel to [100] give $a = 3\cdot13\times10^6\,\mathrm{s}^{-1}\,\mathrm{K}^{-3}$. Extrapolation to the thick sample-limit $(\pi v_0/\omega_c \gg 1)$, using the method of Johnson *et al.* (1976), gives $a = 2\cdot7_2\times10^6\,\mathrm{s}^{-1}\,\mathrm{K}^{-3}$. As can be seen from Fig. 3, the dashed line, which corresponds to this corrected value, fits the present data quite well in the asymptotic limit.

Fig. 4 shows a best fit of the data to equations (1) and (3) using a total scattering rate $\nu = 3\cdot2_5\times10^9\,\mathrm{s}^{-1}$. At the temperature of measurement, namely 4·65 K, the phonon contribution to the scattering rate is $\nu(T) = 0\cdot27\times10^9\,\mathrm{s}^{-1}$, so that the computed impurity scattering rate $\nu_0 = \nu - \nu(T)$ is $2 \cdot 9_8 \times 10^9\ \mathrm{s}^{-1}$. This value is to be compared with the value $\nu_0 = 2 \cdot 9_7 \times 10^9\ \mathrm{s}^{-1}$, deduced by Johnson *et al.* (1976), for the specimens of comparable purity used in the RFSE experiments of Gantmakher and Gasparov (1973).

From Fig. 4 it can be seen that the approximation corresponding to equation (3) produces negligible error for harmonic numbers in excess of 15. At higher temperatures, when the phonon scattering rate becomes comparable with that due to the

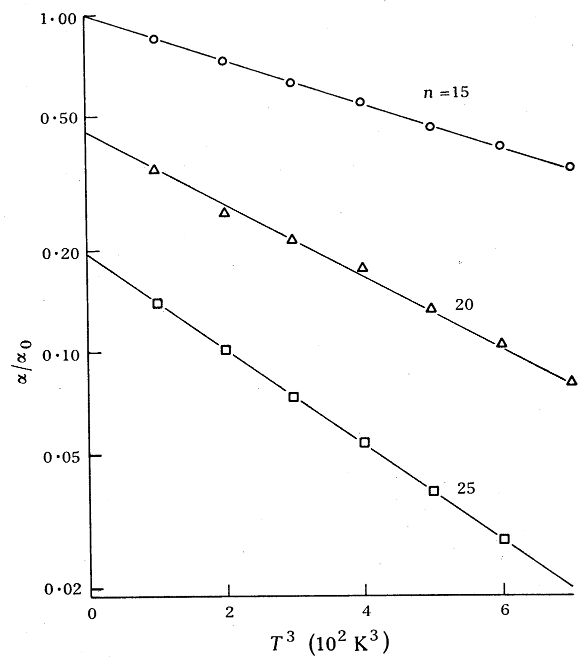

Fig. 2. Reduced attenuation α/α_0 versus T^3 for longitudinal wave magneto-acoustic data at 152 MHz with $\boldsymbol{q}$ along [011] and $\boldsymbol{H}$ along [100]. Plots are shown for harmonic numbers $n = 15$, 20 and 25 (the latter two plots being displaced vertically for clarity).

impurities, the two curves become indistinguishable over the entire range of ω_c shown in the graph. Clearly the use of equation (3) in analysing the temperature dependence of low-order oscillations will produce a lower effective scattering rate compared with the limiting value, in apparent agreement with experiment (Peverley 1975). However, the detailed calculations shown in Fig. 4 indicate that the magnitude of this effect is much too small to explain the experimentally observed behaviour.

For propagation along [011], the present work shows that essentially pure oscillations, corresponding to a central belly orbit, persist for field directions within $\pm 10°$ from [100]. More complex oscillatory behaviour occurs for other field orientations, making it impossible to carry out the preceding analysis in general. A similar situation exists for propagation along [001], even when the field is along the symmetry directions. For $\boldsymbol{H}$ parallel to [100] the oscillations due to the central belly orbit are obscured by those arising from the 'rosette', while for $\boldsymbol{H}$ along [110] the oscillations due to the 'dogs-bone' orbit are mixed with much stronger effects due to off-centre

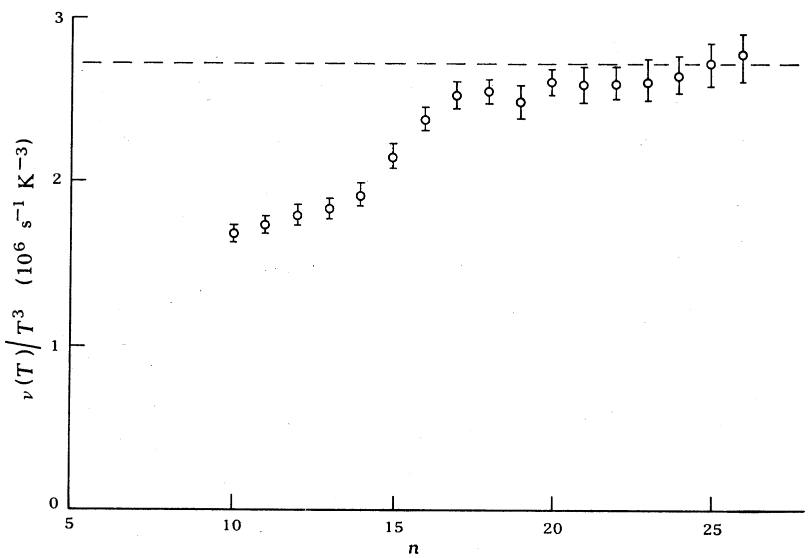

Fig. 3. Plot of the coefficient $a = \nu(T)/T^3$ as a function of harmonic number n for the data corresponding to Fig. 2. The dashed line gives the corrected value obtained from RFSE data for this field orientation.

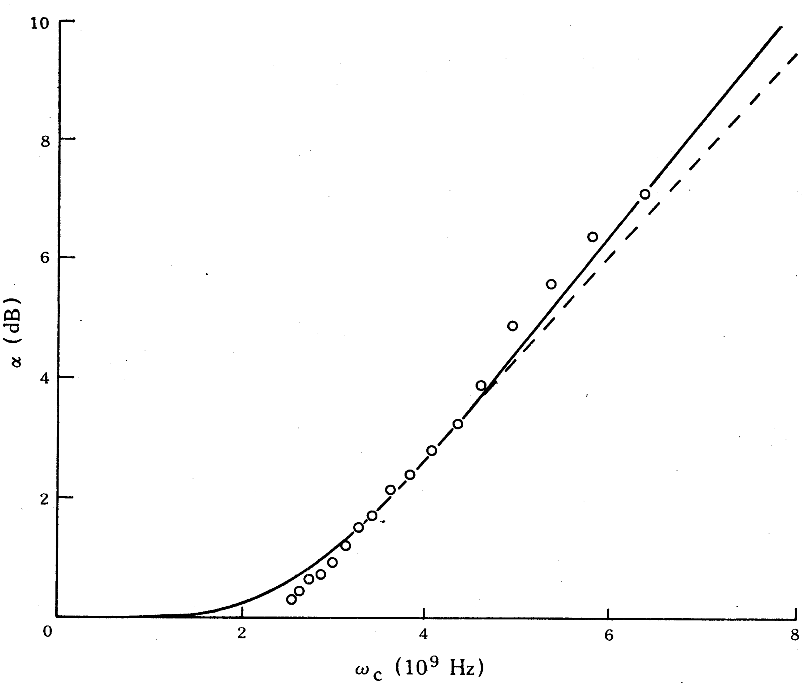

Fig. 4. Fit of the data corresponding to Fig. 1 to equation (1) (full curve) and equation (3) (dashed curve) for a total scattering rate $\nu = 3{\cdot}2_5 \times 10^9\ s^{-1}$.

belly orbits. According to Kamm (1970) the latter are damped out relatively quickly, so that at low fields only the 'dogs-bone' oscillations persist. In the present work, measurements for $\boldsymbol{q}$ along [001] and $\boldsymbol{H}$ along [110] have substantiated this behaviour and analysis of the oscillations using equation (2) leads to a value of $\Delta k_r = (1\cdot 18 \pm 0\cdot 02) \times 10^8\ \mathrm{cm}^{-1}$. This result agrees well with the accepted dimension of the 'dogs-bone' orbit along $[1\bar{1}0]$. Preliminary measurements for the temperature dependence of these oscillations yield $a = (6\cdot 2 \pm 0\cdot 2) \times 10^6\ \mathrm{s}^{-1}\,\mathrm{K}^{-3}$, which is in good agreement with the value $6\cdot 3_7 \times 10^6\ \mathrm{s}^{-1}\,\mathrm{K}^{-3}$ obtained for the 'dogs-bone' orbit from RFSE data (Gantmakher and Gasparov 1973), after correction for finite specimen size.

The present results for $\boldsymbol{q}$ along [001] differ from those reported by Khatri and Peverley (1975). In particular, their data for $\boldsymbol{H}$ along [100] appear to match very closely those obtained in the present work for $\boldsymbol{H}$ along [110]. Furthermore, they obtain a value of $a = (6\cdot 0 \pm 0\cdot 3) \times 10^6\ \mathrm{s}^{-1}\,\mathrm{K}^{-3}$, which is consistent with both our value and that obtained from RFSE for the 'dogs-bone' orbit. This circumstance leads us to suggest that, owing to a misinterpretation of crystal directions, their magnetic field direction is in error by 45°. With this hypothesis, the previously mentioned discrepancy between magnetoacoustic and RFSE data for copper is resolved. As noted above, the value of $a = 6\cdot 0 \times 10^6\ \mathrm{s}^{-1}\,\mathrm{K}^{-3}$ would then be assigned to the 'dogs-bone' orbit. For $\boldsymbol{q}$ along $[01\bar{1}]$ their shear-wave data yield $a = (2\cdot 9 \pm 0\cdot 2) \times 10^6\ \mathrm{s}^{-1}\,\mathrm{K}^{-3}$, which is in reasonable agreement with both our value obtained with longitudinal waves and the RFSE results for a (100) *central* belly orbit. This reinterpretation has the added attraction that it removes the necessity of assuming that the oscillations for this configuration are due to off-centre orbits, in disagreement with the work of Kamm (1970).

The present work shows that the dependence of $v(T)/T^3$ on harmonic number is in fact real but cannot be attributed to the assumption $\pi v_0/\omega_c \gg 1$. Further theoretical consideration of this effect is clearly necessary. Notwithstanding this problem and the very real limitation that it can only be used if there is a dominant set of oscillations, the temperature dependence of the magnetoacoustic effect can give reliable information about the carrier scattering in metals.

Acknowledgments

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