[3]M. Gomez, S. P. Bowen, and J. Krumhansl, Phys. Rev. 153, 1009 (1967).
[4]D. Walton, Phys. Rev. Letters 19, 305 (1967), and in Localized Excitation in Solids, edited by R. F. Wallis (Plenum, New York, 1968), p. 395.
[5]J. P. Harrison, P. P. Peressini, and R. O. Pohl, Phys. Rev. 171, 1037 (1968), and 180, 926 (1969); G. Höcherl and H. C. Wolf, Phys. Letters 27A, 133 (1968); R. A. Herendeen and R. H. Silsbee, Phys. Rev. (to be published).

## ANISOTROPY OF THE ELECTRON-PHONON SCATTERING RATE IN Cu

J. F. Koch* and R. E. Doezema
Department of Physics and Astronomy, University of Maryland, College Park, Maryland 20742
(Received 26 December 1970)

The temperature dependence of the electron-scattering rate in Cu has been investigated in the range 2 to 20°K with magnetic-field–induced surface quantum states. From the temperature dependence we obtain the electron-phonon scattering rate at points on the Cu Fermi surface. Over the limited range of our present explorations we observe a scattering-rate anisotropy of nearly a factor of 6, the highest scattering rate being on the necks.

We report some results of measurements of the anisotropy of the electron-scattering rate due to thermally excited phonons in Cu. Such "point-by-point" determination of the electronic mean free path on the Fermi surface has recently come to challenge the capabilities of solid-state physicists. Interest in this topic is amply evidenced in the recently published proceedings[1] of an international conference which give an up-to-date account of the status of such investigations. Particular interest attaches to Cu (more generally the noble metals) as a metal with a relatively simple and thoroughly studied Fermi surface geometry, and yet with an expected anisotropy of scattering which is sufficiently large to be readily accessible experimentally. Some first-order attempts at theory and experiment on relaxation-time anisotropy in Cu are reflected in a calculation by Ziman[2] and the experimental determination of cyclotron-orbit relaxation times by Häussler and Welles.[3] It is our intent by communicating early results on the relaxation-time anisotropy of Cu to demonstrate convincingly how such data can be obtained from the electron surface-state resonance spectrum[4] and to stimulate the theoretical effort to calculate and understand such data in Cu as would warrant the more extensive experimental effort to provide a more detailed map of point-by-point relaxation times for the entire Fermi surface.

To measure local values of the electron relaxation rate on the Fermi surface requires an effect that arises from a unique, readily identifiable group of electrons. As such, the microwave resonance spectrum due to surface electron states is eminently suited for this purpose and, given the present state of the art of sample preparation, provides a quite generally applicable method for the study of relaxation-time anisotropy. The determination of lifetimes, as is typical for resonance phenomena, however, requires detailed calculation and fitting of theoretical curves to experimental results.

In Cu samples (RRR$\approx$20 000) with carefully electropolished surfaces we observe a rich spectrum of microwave resonances (~35 GHz) in fields 0-200 Oe in all of the major symmetry planes of Cu. The angular variation, polarization dependence, and position in field of the resonances[5] make for unambiguous identification of where on the Fermi surface the signal originates. For conditions typical of our experiments, the classical skipping electron trajectory that corresponds to the $n=1$ quantum mechanical state penetrates the metal to a depth $z_1\approx 0.1\ \mu$ and has an arc length $d\approx 30\ \mu$. The electron consequently traverses an angular region on the Fermi surface on the order of only a small fraction of a degree. A resonance spectrum arises for every stationary value of the parameter $(K/v_F{}^3)_\perp$, on the zone of the Fermi surface where $v_z$, the component of velocity normal to the surface, vanishes. $K$ is the radius of curvature of the Fermi surface in $k$ space and $v_F$ the Fermi velocity; both are to be taken in the plane perpendicular to the magnetic field. While of course the resonance signal is dominated by skipping electrons with the extremum value of $(K/v_F{}^3)_\perp$, neighboring electrons at slightly different values of the resonance parameter do contribute some additional broadening and distortion of the resonance line. To gain some perspective on the characteristic

length of strip contributing to the resonance signal let us consider the locally spherical ($K = 0.37 \times 10^8\,\mathrm{cm}^{-1}$) portion of the Cu Fermi surface in the vicinity of the [100] point. The variation of the resonance field [$H \propto (K/v_F{}^3)_\perp{}^{1/2}$] as a function of angular position away from the [100] point (and referred to the center of the Fermi surface) is readily computed from known Fermi-surface parameters. For a fractional linewidth $\Delta H/H \approx 0.05$ it would appear that electrons within ±6° of [100] have resonance frequencies within this linewidth. Because of the parabolic variation of the resonance parameter, there are, of course, always more electrons at the extremal value and hence the effective width should be somewhat less than this number, perhaps ±3°. Thus our "point-by-point" measurement really selects electrons on a small rectangular strip of the Fermi surface with width of typically $\frac{1}{2}$ deg, but with a length more nearly 6°, depending on the sharpness of the $(K/v_F{}^3)_\perp$ extremum and the experimentally observed linewidth.

For the details of calculating and comparing theoretical and experimental line shapes we refer the reader to published work.[6] The scattering rate $\Gamma$ is a parameter of the calculation, chosen to fit the experimental curve with regard to line shape and width. Of course only when the resonance-parameter anisotropy (the $k_y$ broadening of Koch and Jensen[7] and Ref. 1) is properly accounted for would $\Gamma$ represent the correct total scattering rate due to various causes. Under certain restrictive circumstances, as has been shown,[7] such a calculation can be carried out. However, since the present work focuses on the changes in this parameter $\Gamma$ with temperature $T$, we have thought it safe to ignore the complication of $k_y$ broadening, and have fitted the resonance curves within the cylindrical approximation as discussed in Ref. 6. As a check on this procedure, a calculation was carried out for the neck point including $k_y$ broadening. The coefficient of the temperature-dependent part of $\Gamma$ was found to change only negligibly, while as expected the constant part was altered substantially.

The total scattering rate $\Gamma$ contains various contributions due to physically distinct causes. There is expected a temperature and field-independent part due to scattering by bulk impurities, strains, and dislocations in the crystal. Surface irregularities and consequent nonspecular reflection are expected to be field dependent[8] and possibly dependent on temperature,[9] because the vibration of surface atoms would act as a dynamic diffraction grating. However in our analysis of the data there is little evidence for such a mechanism, as is apparent from the good fit to a $T^3$ dependence in Fig. 1, and we ignore this possibility in the present work. Finally there remains the temperature-dependent scattering due to collisions with thermally excited phonons, on which we focus our attention in the present work.

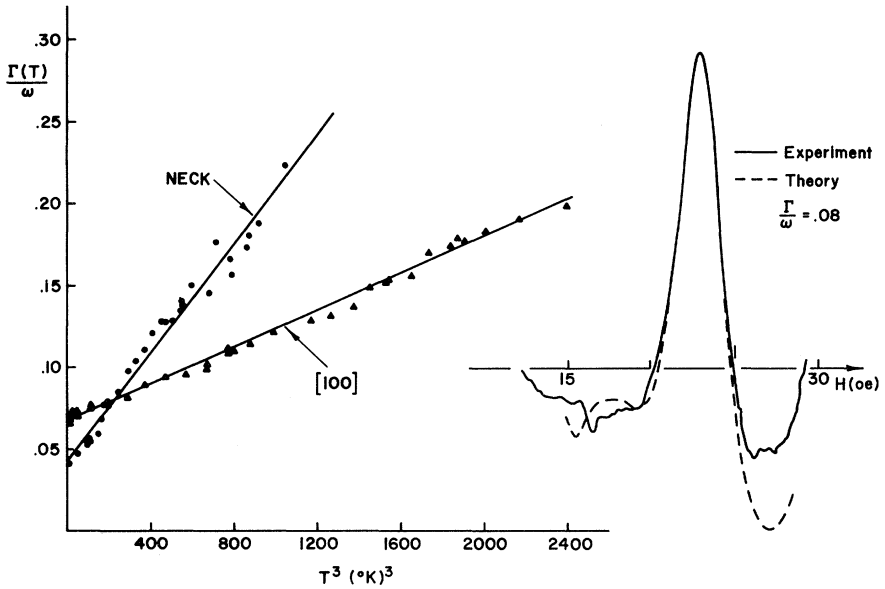


FIG. 1. Variation of $\Gamma$ with $T^3$ for a point on the neck and the [100] point. $\omega$ is $2\pi(35.91\times10^9)$ sec$^{-1}$ for the neck and $2\pi(35.78\times10^9)$ sec$^{-1}$ for the [100] point. Insert: $dR/dH$ trace ($R$ is the surface resistance) at the 1-2 transition for the [100] point in a (100) Cu sample at 6.62°K. Even better fit is obtained by including $k_y$ broadening, especially on the high-field side of the peak.

Moreover, since our experimental measurements range over temperatures 2-20°K, where the average phonon wavelength ($\lambda \approx hv_s/kT$) is much less than the depth of electron penetration, the measured scattering rate is expected to be typical of bulk phonon scattering with no influence of special surface phonon modes.[10] Because even the very slight displacement in electron momentum incurred in a long-wavelength phonon collision is sufficient to destroy the phase coherence of the electron skipping motion, we expect that every scattering event is inherently equally effective. Thus phonon scattering is expected to vary simply as the number of thermally excited phonons available at a given temperature, i.e., as $T^3$, and with a strength proportional to the anisotropic electron-phonon coupling for the point of the Fermi surface under consideration. Thus for a given resonance, such as the $n=1$ to $m=2$ transition, we expect to fit a linewidth parameter $\Gamma(T)$ of the form

$$\Gamma(T) = \Gamma_0 + \gamma T^3,$$

with $\Gamma_0$ reflecting the temperature-independent part due to impurity and defect scattering, surface scattering, and to some extent the $k_y$ broadening discussed earlier. $\gamma$ represents the anisotropic coupling to the phonons.

For each of the chosen points on the Fermi surface we obtain a set of experimental traces of the resonance spectrum (particularly the $n=1$, $m=2$ transition) over as wide a range of temperatures as possible. Fitting the 1-2 transition carefully by a set of computed curves with different values of $\Gamma$, we ascertain a value of $\Gamma$ for each temperature point. The data in Fig. 1 reflect the variation of $\Gamma$ with $T^3$ for a point on the neck and the [100] point. The scatter of data points is indicative of the accuracy to which such data points can be reproduced. Alternatively, we have analyzed such data in the form $\Gamma_0 + \gamma T^x$, with $x$ chosen to minimize the mean-square deviation of the data points. We have found $x = 3.0 \pm 0.2$, thus a posteriori justifying the assumption of a temperature-independent surface scattering. Throughout our work temperatures were determined using a carbon resistor (calibrated at 2.4, 4.2, and 20.4°K) mounted on the back surface of the specimen.

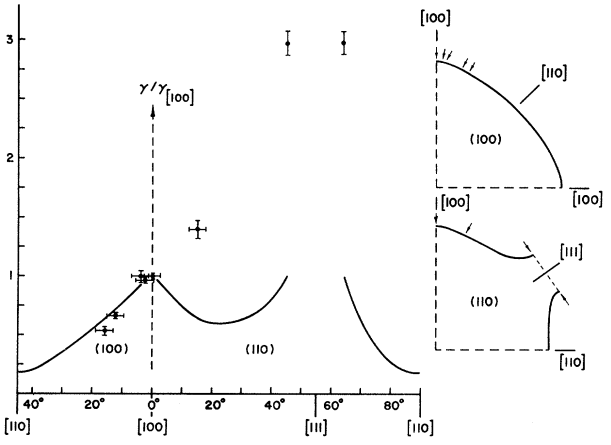


FIG. 2. Experimental values of $\gamma$ relative to $\gamma_{[100]}$. The solid curve is the theoretically expected variation of $\gamma$ taken from Lee. Insert: The arrows indicate the points on the Fermi surface where the scattering rate has been measured in the present work.

As indicated in Fig. 2, the seven points chosen for the present study are a neck point, the [100] point, four others away from the [100] along a (100) section, and one away from [100] along a (110) section. Table I summarizes the essential results for $\Gamma_0$ and $\gamma$, and gives the exact location of each of the points in terms of the angle away from the [100] point referred to the center of zone. Figure 2 also contains a plot of the experimental values of $\gamma$ with experimental uncertainties as indicated. The fact that our measurement really provides a value averaged over several degrees is indicated by the horizontal error bars. For comparison the theoretically expected[11] variation of $\gamma$ is sketched in.

The considerable anisotropy of phonon scattering in Cu is immediately apparent from the data. By far the strongest phonon scattering occurs in the neck region, where we find $\gamma = (3.8 \pm 0.1) \times 10^7$ $\text{sec}^{-1}/(°\text{K})^3$. This value is found to be isotropic within experimental uncertainty around the entire neck, and thus allows a meaningful comparison with that found by Häussler and Welles[3] from cyclotron resonance. Their result is $\gamma = 2.7 \times 10^7$

Table I. Results of fitting the observed scattering rate to $\Gamma = \Gamma_0 + \gamma T^3$.

| Zone | Angle away from [100] (deg) | $\Gamma_0$ ($10^{10}$ $\text{sec}^{-1}$) | $\gamma$ [$10^7$ $\text{sec}^{-1}/(°\text{K})^3$] |
|---|---|---|---|
| (100) | 0 | 1.52 ± 0.02 | 1.26 ± 0.02 |
| | 2.5 | 1.56 ± 0.02 | 1.22 ± 0.02 |
| | 4 | 1.45 ± 0.03 | 1.29 ± 0.04 |
| | 12.5 | 1.27 ± 0.02 | 0.84 ± 0.02 |
| | 16 | 1.12 ± 0.02 | 0.67 ± 0.04 |
| (110) | 15 | 1.08 ± 0.04 | 1.76 ± 0.08 |
| Neck | ... | 0.96 ± 0.05 | 3.8 ± 0.1 |

$\sec^{-1}/(°K)^3$, and given their estimated error of a factor of 2, agrees with the present result.

At the [100] belly point the scattering rate is nearly 3 times smaller than that of the neck. Furthermore we observe a rapid decrease of scattering away from the [100] point, reducing $\gamma$ by nearly a factor of 2 only 16° away from [100]. Thus over the limited range of our present explorations we observe a scattering rate anisotropy of nearly a factor of 6. The results follow qualitative expectation, when we consider the distortion of the Fermi surface due to the lattice potential as prima facie evidence for electron-phonon coupling. Of the distortions, those of the neck region are most marked. Likewise the [100] point bears the marks of the lattice-potential influence. The local radius of curvature is only approximately $\frac{1}{3}$ that of the free-electron sphere, but increases to a free-electron-like value away from [100] on the (100) section.

For a more quantitative comparison we may turn to Lee's work,[11] who in fitting to cyclotron resonance and de Haas–van Alphen data has derived results for the strength of the anisotropic electron-phonon interaction. Assuming this to be proportional to the electron-scattering rate one arrives at the theoretical curves of Fig. 2, where the scattering rate is plotted relative to the value at the [100] point. The absolute value has not been determined to allow comparison. While the curve correctly gives the experimentally observed decreased scattering with distance away from the [100] on the (100) section, it fails badly at the neck point.

Using the present experimental values of scattering rate near [100] and bravely extrapolating along the theoretical curve of Fig. 2, we can attempt to derive a cyclotron-orbit relaxation time to compare with Ref. 3. We find $\gamma_{\text{belly orbit}} = 0.67 \times 10^7\ \sec^{-1}/(°K)^3$ to compare with the earlier result of $(0.29 \pm 0.12) \times 10^7\ \sec^{-1}/(°K)^3$, indicating that the scattering rate should decrease faster toward [110] than the curve of Fig. 2. The intrinsically interesting variation of electron scattering at points along the orbit, as reflected in our measurements, is of course inaccessible in such a cyclotron-resonance measurement.

The five points along the (100) section were measured in a single sample, and one might be tempted to draw some conclusion regarding anisotropic impurity scattering from the variation of the $\Gamma_0$ term. It would seem to indicate impurity scattering decreases with position away from [100]. However, this conclusion is premature because of the slight change in $\Gamma_0$ and the possibility of serious errors in $\Gamma_0$ because of the neglect of $k_y$ broadening in the calculated curves.

The present work and the very limited number of points that have been studied give at best a sketchy picture of the scattering-rate anisotropy that can be observed in Cu. A more complete point-by-point exploration of the electron-phonon scattering, and a more thorough and detailed calculational effort to include $k_y$ broadening, are intended for future work. We hope that the present results provide a stimulus for further theoretical consideration of the anisotropic mean-free-path problem.

---

*On sabbatical leave, currently at the Institute for Physical Problems, Moscow, U.S.S.R.

[1]Physik Kondensierten Materie 9 (1969). For a discussion of electron surface states see J. F. Koch, ibid., pp. 146-157.

[2]J. Ziman, Phys. Rev. 121, 1320 (1961).

[3]P. Häussler and S. J. Welles, Phys. Rev. 152, 675 (1966).

[4]R. E. Prange and T. W. Nee, Phys. Rev. 168, 779 (1968); see also review articles: M. S. Khaikin, Advan. Phys. 18, 1 (1969); and J. F. Koch, in Electrons in Metals, Simon Fraser Summer School Lectures Solid State Physics 1967, edited by J. F. Cochran and R. R. Haering (Gordon and Breach, New York, 1968), Vol. I, p. 253.

[5]R. E. Doezema and J. F. Koch, to be published.

[6]T. W. Nee, J. F. Koch, and R. E. Prange, Phys. Rev. 174, 758 (1968).

[7]J. F. Koch and J. D. Jensen, Phys. Rev. 184, 643 (1969).

[8]J. F. Koch and T. E. Murray, Phys. Rev. 186, 722 (1969).

[9]V. F. Gaidukov, private communication, and based on results of transport studies in whiskers.

[10]R. E. Prange, Phys. Rev. (to be published).

[11]M. J. G. Lee, Phys. Rev. (to be published).