Electron-phonon-surface scattering in Ga

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(Received 1 July 1998)

An unambiguous experimental proof of the existence of electron-phonon-surface scattering in pure metals at low temperatures is presented. Using the surface acoustic waves technique we are able to probe the properties of a thin surface layer of metal. Observation of a nearly T^{-2} dependence of the mean free path of near-surface electrons in Ga (99.9999%) proves that precisely this scattering mechanism is dominant and clearly separates it from the volume scattering that would have a T^{-3} dependence. [S0163-1829(99)00719-5]

I. INTRODUCTION

Electron relaxation in pure metals is determined by a variety of mechanisms, which all contribute to the total scattering rate. Their relative weights depend on temperature, sample size and purity, physical quantity in question, etc. A quantity most directly connected to the electron relaxation is probably the resistivity of a metal. It is given by a sum of bulk and surface terms, where bulk resistivity is in turn a sum of impurity-related (residual) resistivity and a term related to scattering on electrons and phonons. In the extreme pure-limit impurity scattering is always the smallest term. The ratio of surface to quasiparticle scattering rates depends on the temperature-dependent mean free path $l(T)$ and on the sample size d. Surface scattering inevitably becomes important at low temperatures when $l>d$. The resistivity is given by a sum of the following contributions:[1]

$$\rho(T)=\rho_c+\delta\rho_c(T)+\rho_d+\delta\rho_d(T)+\rho_{e\text{-}e}(T)+\rho_{e\text{-}ph}(T). \tag{1}$$

Here, ρ_c is the residual resistivity due to electron scattering on impurities, vacancies, and other static defects of the lattice; $\delta\rho_c(T)$ is the temperature-dependent contribution to the impurity scattering due to thermal oscillations of impurities. For $T\ll T_D$ (T_D—Debye temperature) it has a quadratic dependence $\delta\rho_c(T)\sim T^2$ (Ref. 2). This term can show up in the ''dirty limit,'' when residual resistivity exceeds the temperature-dependent part. In this case $\delta\rho_c(T)$ can be comparable to ρ_c and larger than the electron-phonon part $\rho_{e\text{-}ph}(T)$ of the resistivity; ρ_d is the contribution of the surface scattering to the residual resistivity (size effect); and $\delta\rho_d(T)$ is the temperature-dependent part of the size effect.[3,4] According to Ref. 4 $\delta\rho_d(T)\sim T^{7/3}$. In Ref. 5 a calculation was done for an arbitrary law of electron reflection on the sample boundary, in particular taking into account the dependence of scattering probability on the direction of electron velocity relative to the surface.[6,7] The result of Ref. 5 is $\delta\rho_d(T)\sim T^{5/3}$. Just like for the $\delta\rho_c(T)$ term, the temperature dependence of $\delta\rho_d(T)$ is close to quadratic. However, the latter will dominate in the opposite limit of a pure metal.

$\rho_{e-e}(T)\sim T^2$ is the electron-electron contribution to the bulk scattering. It is very small due to smallness of the prefactor $(k_bT/E_f)^2$ (k_b is the Boltzman constant, E_f is the Fermi energy);[8,9]

$\rho_{e\text{-}ph}(T)$ is the electron-phonon contribution to the bulk scattering. At low temperatures electrons scatter on phonons mostly on small angles $\alpha\sim T/T_D$. If this angle is still sufficiently large to remove an electron from participation in the particular transport process, the effective scattering probability and resistivity will be proportional to the number of phonons $\rho_{e-ph}(T)\sim(T/T_D)^3$ (Debye law). Otherwise, several scattering events must take place. The number of necessary collisions $p\sim(T_D/T)^2$ can be taken into account by dividing the probability of each collision by a factor of p. Then $\rho_{e\text{-}ph}(T)\sim(T/T_D)^5$ (Ref. 10). The last result is derived for metals with isotropic Fermi surface (FS) but in practice is observed for a great majority of metals at low temperatures (see, e.g., Ref. 11 and references therein). In the experiments measuring total scattering frequency a T^3 dependence is observed as opposed to T^5 found for ''transport'' scattering frequency. Cubic dependence is also observed as a rule when a selected group of carriers on FS is studied (radio-frequency size effect, cyclotron resonance, etc.). There are counterexamples, however: a sharply anisotropic cylindrical FS will lead to T^2 or T^4 dependences instead of T^3 or T^5. And indeed, in bismuth and antimony with FS being a prolate ellipsoid a T^2 law is realized.[12–14] The T^3 and T^5 laws can also be violated due to U-processes (see, e.g., Ref. 15)

From the above considerations it is evident that an observation of a T^2 dependence of the low-temperature resistivity or any other quantity measuring electron scattering in a metal does not *per se* prove that electron-electron scattering is observed in the experiment. This remark is contrary to the common belief, but in accord with theoretical estimates of $\rho_{e\text{-}e}(T)$, which usually give a value of an order of magnitude smaller than the experimentally measured T^2 contribution. In reality, observation of a quadratic dependence $\rho(T)$ in the temperature range of 1.5–4.2 K most often means that in the extreme pure limit the contribution of $\delta\rho_d(T)\equiv\delta\rho_{e\text{-}ph\text{-}s}(T)$ is dominant. This contribution comes from the ''electron-phonon-surface'' scattering mechanism.[1,16] In a very pure metal samples electrons will necessarily collide with a surface of the crystal after scattering on a phonon if the scattering angle satisfies a condition:

$$\beta\approx\frac{k_{ph}}{k_f}\approx\frac{d}{l},$$

where k_{ph} and k_f are wave vectors of the phonon and electron. An interplay of electron-phonon and electron-surface

collisions gives additional temperature-dependent contribution to the relaxation rate and introduces a new form of a size-effect in the resistivity of a metal. Electron-phonon-surface processes were first experimentally found in tungsten,[16] then observed in copper, silver and gold,[17] aluminum,[18] molybdenum, rhenium, and ruthenium.[19]

In the present paper we report an unambiguous experimental proof of the existence of this mechanism and find it to be operational in the extremely pure gallium samples.

II. EXPERIMENTAL OBJECT

Gallium has record electron mean-free-path lengths, which can be as large as 1 cm. It is known from a large body of experimental studies that a T^5 dependence is never observed in gallium. Instead, either T^2 or T^3 dependences are measured. For larger samples of Ga a cubic dependence usually holds:

$$\frac{1}{l}=A+BT^3. \tag{2}$$

Such result was obtained for example in Ref. 20 where thermal conductivity anisotropy was measured in 99.9999%-pure Ga cylinder samples with a diameter of 1.6–4.8 mm along crystal directions *a* and *b*. A close result ($\sim T^{2.9}$) was obtained also for the *c* axis. In Ref. 22 the same dependence was obtained for both thermal and electrical conductivity along the *c* axis of a large (1 cm diameter) 99.9999% Ga monocrystal. Cubic temperature dependence is also obtained in resonance experiments. For example, Moore[23] extracted it from the cyclotron resonance linewidth, which is proportional to $1/l$ with l being the mean free path of the selected group of electrons participating in the resonance.[24,25] From the cited above experiments with comparatively massive samples we conclude that first, it is the full scattering frequency (as opposed to ''transport'') that is always measured in gallium, and second, that neither complicated FS, nor U-processes or impurity scattering and such lead to the T^3 law violation.

However, other authors obtained close to quadratic dependences of $1/l$ both in resistivity measurements on the square cross-section wires (1–0.1 mm)[26] and in size effect.[27] Those results may be, and actually were, interpreted as a manifestation of either electron-electron scattering or electron-phonon-surface scattering, provided that the ''dirty limit'' explanation is ruled out. An observation that T^2 was found on smaller samples suggests that a surface influence is important, but as discussed above, bulk-transport measurements themselves cannot tell which of the two remaining explanations is correct.

III. EXPERIMENTAL METHOD

However, it is possible to propose the method that gives an unambiguous answer to this question. This is the experiment measuring the damping coefficient of the surface acoustic wave (SAW). The specific properties of the surface wave help us to make the distinction.

First, SAW exponentially decays inside the crystal on the distance of the order of its wavelength λ. In our experimental conditions $\lambda \ll l$, therefore the wave interacts only with those electrons that move along the surface and necessarily collide with it. In this situation, if the electron-phonon-surface scattering mechanism really exists, it must lead to a quadratic $1/l \sim T^2$ dependence up to the temperature at which $l(T) \approx \lambda$. This temperature is much higher than the usual cutoff temperature for the electron-phonon-surface processes given by $l(T) \approx d$. At such a high temperature electron-electron scattering will be undoubtedly negligible and ruled out as a candidate for the explanation of the $1/l \sim T^2$ dependence.

Second, previous theoretical papers[28,29] relate the SAW damping coefficient and the electron mean-free-path value for certain experimental geometries. In particular, when a strong magnetic field **H** is parallel to the surface, perpendicular to the acoustic wave vector **q** and inequalities $qr \ll 1$ and $ql \gg 1$ are satisfied, where r is the radius of a cyclotron orbit, the value of l does not depend on the magnetic field. So it plays the role of the transport mean free path for the near-surface electrons. We will compare the temperature dependence of l under this condition to its value in the bulk.

An experiment of this type was performed in Ref. 30 and is now confirmed for the monocrystal sample of gallium. The surface wave was excited to propagate along the *a* axis in the (ab) plane of the crystal. Using the wedge method[31] to excite the wave allowed frequency tuning in a broad range of frequencies from 10 to 140 MHz. We note that the interdigitated SAW transducers currently widely used to study the two-dimensional electron gas structures[21] would not be well suited for such tuning. The temperature was calculated from the Helium vapor pressure below 4.2 K and measured with a resistive thermometer above 4.2 K. The lower boundary for the mean-free-path was found from the number of observed Pippard oscillations in low magnetic field at 4.2 K. For the sample used it was estimated to be 1.7 mm for $q \| a$, $H \| b$. With that figure in mind we can check the validity of the $qr \ll 1$ and $ql \gg 1$ conditions for resonance electrons. The measured velocity of SAW in the (ab) plane was $V = 2.59 \pm 0.02 \times 10^5$cm/s for $T = 4.2$ K, $q \| a$. Therefore, even for the lowest frequency used, 10 MHz, $ql \approx 40$ and $ql \gg 1$ was indeed well satisfied. The magnetic field was created by an electromagnet and reached the maximum value of 7 KOe, which proved to be enough to satisfy $qr \ll 1$. According to Refs. 28 and 29, when $ql \gg 1$ a saturation of the damping coefficient as a function of H must be observed in a strong magnetic field $H \perp q$. As can be seen from the inset in Fig. 1, in our case saturation was observed already for $H > 1$ KOe. Measurements were done in a saturating field. Precise orientation of the sample in the field was performed using the Renneker tilt effect, which could be reliably observed on the surface sound.

IV. RESULTS AND DISCUSSION

In the saturating magnetic field parallel to the surface of the metal but perpendicular to the sound wave vector $(\mathbf{q} \perp \mathbf{H} \perp \mathbf{n})$, the surface wave damping is analogous to the damping in the bulk and according to Ref. 29 is determined by

$$\frac{\Gamma_H}{\Gamma_0} \sim \frac{ql}{1+(\omega\tau)^2}, \tag{3}$$

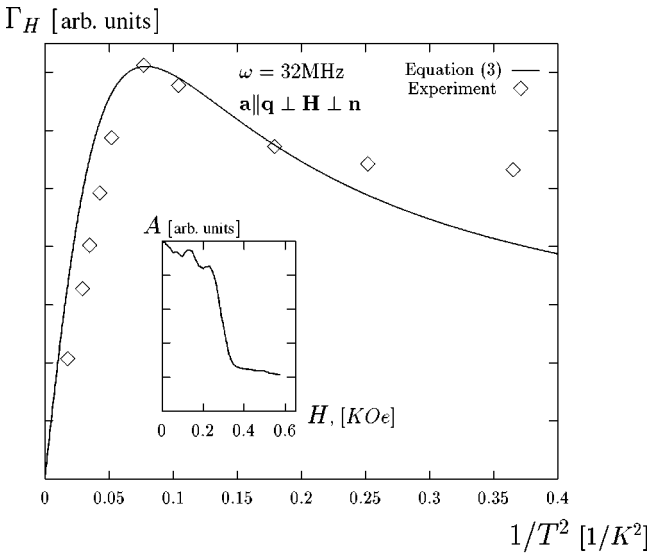

FIG. 1. Experimental plot of the temperature dependence of SAW damping coefficient in saturating magnetic field. Solid line is given by Eq. (3). Inset: example of the experimental magnetic-field dependence of transmitted SAW amplitude. Saturation effect is clearly observable.

where Γ_0, Γ_H are SAW damping coefficients without magnetic field and in the saturating field accordingly, τ is a relaxation time of electrons. From Eq. (3) it is seen that SAW damping in the saturating magnetic field must have maximum as a function of temperature at the point determined by:

$$\omega\tau = 1. \tag{4}$$

We measure a series of curves $\Gamma(H)$ at $\omega = \text{const}$ and different T and extract from them the saturation value $\Gamma_H(T)$ at $\omega = \text{const}$. From Fig. 1 it is seen that the experimental curve for $\Gamma_H(T)$ indeed has a maximum as discussed above. At the maximum point T_* we have $\tau(T_*) = 1/\omega$, thus maximum damping will occur at different temperatures for different SAW frequencies. From the family of the experimental curves $\Gamma_H(T)$ taken at different ω we reconstruct the $\tau(T)$ dependence by plotting $1/\omega$ as a function of T_*. Results are presented in Fig. 2 and show a quadratic dependence

$$\frac{1}{\tau} = a + bT^2 \tag{5}$$

of the collision frequency of charge carriers in gallium on the temperature in the range from 1.6 to 7.2 K.

A least-squares fit of Fig. 2 gives the values $a = 0.25 \times 10^8$, $b = 0.14 \times 10^8$. When used in the formula (3) they determine a theoretical curve shown as a solid line in Fig. 1. Numerical differences between the theoretical and experimental curves could be anticipated knowing that Eq. (3) was derived with certain approximations, one of them being spherical FS. However, this formula reflects the key feature: a maximum in the $\Gamma_H(T)$ curve at the point $\omega\tau = 1$ which, as we see, does not disappear for an FS of a complicated shape.

Furthermore, when $(\omega\tau)^2 \ll 1$, Eq. (3) takes the form

$$\frac{\Gamma_H}{\Gamma_0} \sim ql. \tag{6}$$

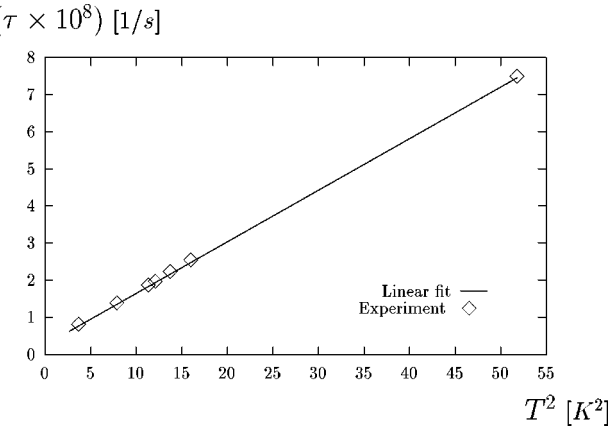

FIG. 2. Temperature dependence of the electron relaxation time in Ga: $\tau^{-1} \sim T^2$.

For the high end of the temperature interval investigated in our experiment and lower SAW frequencies the condition $(\omega\tau)^2 \ll 1$ is satisfied. In that case formula (6) applies and if indeed $l \sim 1/T^2$, Γ_H should start as a linear function of $1/T^2$. Experimental points in Fig. 1 indeed show linear behavior for small argument, however it extends to a larger interval than Eq. (3) would predict.

From the qualitative agreement of experimental and theoretical curves we conclude that the electron-phonon-surface mechanism is the source of the T^2 contribution to the scattering frequency. This conclusion is consistent with the sample-size dependence discussed in Sec. II. If the T^2 dependence was due to the FS shape or other bulk effects discussed before, it would be observed in the samples of an arbitrary size.

V. CONCLUSION

Our experiment leads to the following picture. Bulk samples of high-purity gallium switch to cubic temperature dependence of the scattering rate already for $T \sim 1.5 - 4.2$ K. This established fact rules out a hypothesis that complicated Fermi surface structure, presence of umklapp processes, scattering on impurities, and so on can be a reason of T^2 dependence observed for smaller samples. In contrast, according to the SAW experiment, the scattering rate of the near-surface carriers remains quadratic up to 7.2 K, where the electron-electron scattering mechanism is definitely excluded. Therefore, it is proved unambiguously that electron-phonon-surface mechanism exists and is indeed responsible for the nearly quadratic temperature dependence of the inverse electron mean free path.

We believe that this result obtained for gallium is universal and holds for all metals. We anticipate that any metal of high purity will demonstrate a close to quadratic dependence of the inverse mean-free-path of near-surface electrons and confirm the defining role of the electron-phonon-surface scattering mechanism.

ACKNOWLEDGMENT

This paper was partially supported by the Fund of Fundamental Research of Ukraine, Grant No. N2.4/211.

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