[48]A. J. Bray, J. Stat. Phys. 11, 29 (1974).
[49]S. N. Zolotarev, I. B. Sidorova, Yu. A. Skakov, and V. S. Solov'ev, Fiz. Tverd. Tela (Leningrad) 20, 775 (1978) [Sov. Phys. Solid State 20, in press (1979)].
[50]V. M. Nabutovskiĭ and B. Ya. Shapiro, Pis'ma Zh. Eksp. Teor. Fiz. 26, 624 (1977) [JETP Lett. 26, 473 (1977)].

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Mechanism of electron scattering in molybdenum

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The rf size effect was used to study the temperature dependence of the collision frequency $\bar{\nu}(T)$ of electrons and holes in the central sections of various sheets of the Fermi surface of molybdenum. It was found that $\bar{\nu}(T) = \alpha T^2$ at $T \lesssim 9$°K and the value of α was independent of the orbit positions on the octahedral parts of the electron and hole surfaces, and also independent of the purity and thickness of the samples. In the case of small groups (ellipsoids) the range of the quadratic dependence was shifted toward lower temperatures ($T \lesssim 5$°K). Throughout the investigated temperature range (1.2–8°K) the dependence $\bar{\nu}(T)$ for the ellipsoids was well described by a sum of two terms $\bar{\nu}(T) = \alpha T^2 + \beta T^3$. The current concepts were used to calculate the frequency of normal electron–electron collisions in various electron groups, which fitted well the experimental results. An analysis of the data obtained led to the conclusion that the quadratic term in $\bar{\nu}(T)$ of molybdenum was due to the electron–electron scattering.

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Investigations of the temperature dependence of the collision frequency of certain groups of carriers $\bar{\nu}(T)$ on different sheets of the Fermi surface of molybdenum and tungsten have been carried out using the rf size effect method and they have shown that $\bar{\nu}$ rises quadratically with temperature in the liquid helium range.[1] This quadratic law is obtained also in studies of the temperature dependences of the electrical resistivity ρ and thermal resistivity wT of these metals (for a bibliography see Refs. 2 and 3) and it is attributed to the electron–electron scattering. The only argument in support of the electron–electron collisions is the quadratic rise of ρ and wT with temperature. Usually the bulk of electron–electron collisions in transition metals is attributed to the scattering of fast s by heavy d conduction electrons.[2] However, an analysis of the electron structure of molybdenum and tungsten shows that, because of hybridization of the wave functions, the carrier velocities on different sheets of the Fermi surface of these metals differ only slightly[1,4] and, therefore, there is no justification for the use of the s–d scattering theory. This is the main reason why the interpretation of the quadratic law exhibited by molybdenum and tungsten as manifestation of the electron–electron interaction is questioned in Ref. 1. A further study of the temperature dependence of $\bar{\nu}(T)$ for molybdenum was carried out in order to obtain more information on this quadratic dependence. The results, as shown below, provided an experimental proof that the electron–electron collisions are responsible for the quadratic rise of $\bar{\nu}(T)$.

EXPERIMENTS

The rf size effect lines were deduced from the magnetic-field dependences of the first ($\partial R/\partial H$) and second ($\partial^2 R/\partial H^2$) derivatives of the resistive component of the surface impedance of molybdenum samples in the frequency range 3–8 MHz at temperatures 1.2–10°K. The methods used were described in Refs. 1 and 5.

Plane-parallel single-crystal samples of molybdenum were disks ≈6 mm in diameter and with thicknesses d in the range from 0.5 to 2 mm; they were cut by spark machining from ingots whose resistivity ratios were $\rho(293\,°\text{K})/\rho(0\,°\text{K}) \approx 2\times10^4$, 5×10^4, and 10×10^4. Next a layer about 100 μ thick, which was cold-worked in the process of cutting, was removed by grinding with silicon carbide powder of the M-7 grade and subsequent etching in a chemical polishing mixture.[6] The direction of the normal $\mathbf{n}$ to the surface of the samples was found by x-ray diffraction to within ±0.5° and it coincided with the ⟨100⟩ and ⟨110⟩ axes.

The temperature-dependent part of the collision frequency $\bar{\nu}(T)$ was reduced from the temperature dependence of the amplitude of rf size effect lines $A(T)$, which was described in the $\bar{\nu} > \Omega$ range by the fairly simple expression $A \propto \exp(-\pi\bar{\nu}/\Omega)$ (Ref. 5); here, Ω is the cyclotron frequency and $\bar{\nu} = \bar{\nu}_0 + \bar{\nu}_{ee}(T) + \bar{\nu}_{ep}(T)$ is the sum of the collision frequencies with impurities and defects $\bar{\nu}_0$, with electrons $\bar{\nu}_{ee}(T)$, and with phonons $\bar{\nu}_{ep}(T)$ averaged over a number of points on a selected extremal section of the Fermi surface.[5]

The Fermi surface of molybdenum is well known and consists of an electron "jack," a hole octahedron, six hole ellipsoids, and six electron lenses.[6,7] We investigated the temperature dependences of the electron collisions on various orbits passing along the hole octahedron, the octahedral "waist" of the jack, and ellipsoids. The size effect lines were identified and the experimental results were analyzed as described in Refs. 1, 6, and 7.

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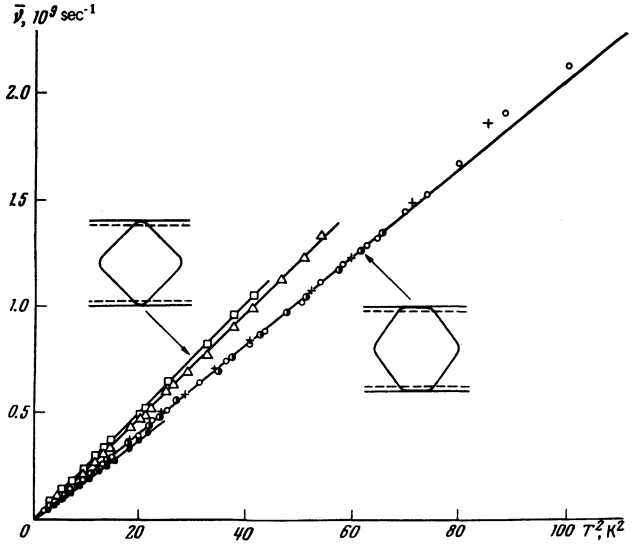

FIG. 1. Temperature dependences of the carrier collision frequency on the waist of the electron jack in the $\mathbf{H} \parallel \langle 111 \rangle$, $\mathbf{n} \parallel \langle 110 \rangle$ cases (○, ◑, ●) and on the hole octahedron (○, ◑, ●, +, △, □) in molybdenum samples of different thickness d (mm): ○) 0.515, ◑) 0.964, ●) 1.933, +) 0.52, △), □) 0.826 and of various purities as represented by $\rho(293°\text{K})/\rho(0°\text{K}) \approx 2\times10^4$ (+), 5×10^4 (△, □), 10×10^4 (○, ◑, ●). The positions of the orbits on the hole octahedron in the {100} plane are as follows: △) $\mathbf{H} \parallel \langle 110 \rangle$; □) $\mathbf{H} \parallel \langle 100 \rangle$.

Figure 1 gives the temperature dependence of the collision frequency of electrons on octahedral parts of the electron and hole surfaces, measured on samples of various purity, thickness, and orientation, and also for various positions of the orbits on the hole octahedron. We can see that in a wide temperature range (1.2–9°K) the collision frequency rises with temperature quadratically, $\bar{\nu}=\alpha T^2$, and it exhibits the following characteristic features.

1. The coefficients α are practically identical for two perpendicular sections of the hole octahedron corresponding to $\mathbf{H} \parallel \langle 100 \rangle$ and $\langle 110 \rangle$. For intermediate sections the value of α is also independent of the position on the Fermi surface. The difference between the dependences $\bar{\nu}(T)$ in the {100} and {110} planes is clearly associated with the orbit shape. In fact, as is clear from the insets in Fig. 1, the time of motion of electrons from one side of the sample to the other in the {110} plane is less than half the cyclotron period π/Ω which occurs in the formula for the amplitude of the size effect line. A conversion shows that the dependences $\bar{\nu}(T)$ coincide in these planes.

2. The values of $\bar{\nu}(T)$ for the orbits on the waist of the electron jack and on the hole octahedron coincide in the {110} plane (in the {100} plane the electron orbits have a complex shape, which makes it difficult to make a comparison with the dependence $\bar{\nu}(T)$ for the hole octahedron).

3. The change in the thickness of the samples in the {110} plane from 0.515 to 1.988 mm also depends weakly on the value of α.

4. The dependences $\bar{\nu}(T)$ obtained for samples with the resistivity ratios 2×10^4 and 10×10^4 are practically identical.

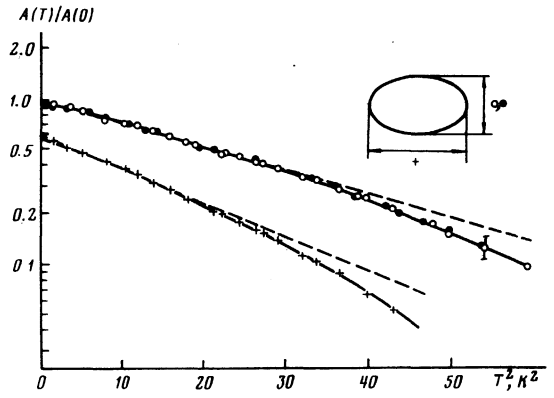

FIG. 2. Temperature dependences of the amplitudes of the rf size effect lines due to ellipsoids, plotted for different orbit positions: $\mathbf{H} \perp k_c$ (+) and $\mathbf{H} \parallel k_c$ (○, ●). The points ○ and ● denote the dependences obtained in the {100} and {110} planes for samples with the electrical resistivity ratios $\rho(293°\text{K})/\rho(0°\text{K}) \approx 5\times10^4$ and 10×10^4, respectively.

The dependence $\bar{\nu}(T)$ is more complex for the orbits on the hole ellipsoids. Throughout the investigated temperature range (1.2–8°K) it does not obey the T^2 or T^3 law. If we postulate the existence of two independent scattering mechanisms, we can describe this dependence by $\bar{\nu}(T)=\alpha T^2+\beta T^3$. The results of a least-squares analysis of the experimental dependences $\bar{\nu}(T)$ on a computer are represented by continuous curves in Fig. 2, and the coefficients α and β calculated from the effective masses[8] are listed in Table I for two mutually perpendicular sections of the hole ellipsoid. The results are plotted in Fig. 2 in units of $A(0)$ for the rf size effect line corresponding to the minimum cross section of the ellipsoid (see inset). The value of $A(0)$ is used as the variable in the calculations. The dashed curves are the dependences corresponding to the T^2 law.

However, it should be noted that the cyclotron mass m obtained for the ellipsoids in Ref. 8 differ from the results of Ref. 9. This difference is due to the superposition of the cyclotron resonance lines of the spheroids and ellipsoids. Therefore, the values of α and β obtained for the ellipsoids are not sufficiently accurate to analyze the observed anisotropy. Moreover, the precision of the determination of the coefficients α and β is low because of the narrowness of the temperature interval. Nevertheless, we should note the following features.

5. The coefficients α for the ellipsoids are close to the corresponding values for the octahedron and jack.

TABLE I.

Surface	$\bar{v}$, 10^8 cm/sec	α, 10^7 sec$^{-1}\cdot$°K^{-2}	β, 10^6 sec$^{-1}\cdot$°K^{-3}
Octahedron	0.9	2.5	–
Jack	0.6	2.5	–
Ellipsoid*			
$\mathbf{H} \perp k_c$	0.8	1.9	4.9
$\mathbf{H} \parallel k_c$	0.8	1.3	2.3

*The k_c axis corresponds to the long axis of the ellipsoid.

6. The results obtained for samples in different planes ({100} and {110}) and for different resistivity ratios $\rho(293\,^\circ\mathrm{K})/\rho(0\,^\circ\mathrm{K})=5\times10^4$ and 10×10^4 are the same.

It should be noted that the investigation reported in Ref. 1 was carried out on less pure and correspondingly thinner samples. This made it difficult to separate clearly the rf size effect lines corresponding to different sheets and sections of the Fermi surface. Therefore, the results obtained in Ref. 1 for the weak lines differ somewhat from our data; there are no differences for the strong lines.

DISCUSSION OF RESULTS

Clearly, three different scattering mechanisms are possible and these can, in principle, explain the quadratic dependence $\nu(T)$ for molybdenum: electron–phonon, inelastic electron–impurity, and electron–electron interactions. We shall now consider the characteristic features of each of these mechanisms so as to identify the one most likely to occur in our samples.

A. Electron–phonon scattering

Under conditions corresponding to the rf size effect (anomalous skin effect) the correction to the electron distribution function Δf is localized in a small region of the Fermi surface and it moves, together with an electron, on this surface. The dimensions of this region vary from δ/d to $(\delta/d)^{1/2}$ depending on the position of the Fermi surface (here, δ is the depth of the skin layer).[10] At low temperatures the angle of scattering of electrons by phonons is fairly small $\vartheta_{ep}\approx q/k_F$ (here, $q\approx k_BT/\hbar s$ is the phonon wave vector), so that $\bar\nu_{ep}(T)$ depends strongly on the position of the Fermi surface[5,11] since it is governed by the local properties of electrons on this surface. We usually have $\delta/d \ll \vartheta_{ep}\lesssim(\delta/d)^{1/2}$; as a result, we find that $\bar\nu_{ep}(T)=\beta T^3$, where the coefficient β depends on the sample thickness d. In particular, in the case of copper when d is increased from 0.36 to 1.882 mm, the coefficient β increases approximately by a factor of 1.4 (Ref. 12). The observed quadratic dependence of $\bar\nu(T)$ for molybdenum is in conflict with the cubic law typical of the electron–phonon scattering. However, the specific nature of the wave functions of electrons in transition metals may alter considerably the matrix element of the electron–phonon scattering.[1] In any case, the above features of this scattering, namely the strong dependence of α on the position of the Fermi surface and on the sample thickness, should clearly be retained. We can see, as reported above, that the coefficient α of molybdenum have just the opposite properties.

B. Inelastic electron–impurity scattering

Kagan and Zhernov[13] showed that inelastic (accompanied by phonon emission or absorption) electron scattering by vibrations of an impurity results in a nonlinear temperature dependence of the scattering probability and impurity electrical resistivity $\rho_0(T)$ of metals. At low temperatures the value of $\rho_0(T)$ rises quadratically, passing through a maximum at $T=T_{\max}\approx(0.1\text{–}0.2)T_D$ (T_D is the Debye temperature), and falls at higher temperatures. The coefficient in front of T^2 in the $T<T_{\max}$ range is proportional to the impurity concentration.

Clearly, we have to reject also this mechanism as a possible explanation of the experimentally observed quadratic dependence $\bar\nu(T)$, because the coefficient α is not affected when the electrical resistivity changes by a factor of 5.

C. Electron–electron scattering

We can easily show that the frequency of electron–electron collisions measured in the rf size effect is given by[1,14]

$$\nu_{ee}(T)=\frac{1}{3\pi^2}\frac{e^4}{\hbar^3}\frac{1}{m_1}\oint\frac{dk_1}{v_{\perp,1}(\mathbf{k})}\times\iiint(M_{12}^{34})^2\delta(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4-\mathbf{K})\frac{dS_2\,dS_3\,dS_4}{v_2v_3v_4}(k_BT)^2, \tag{1}$$

where M_{12}^{34} is the matrix element of the electron–electron interaction; v_i is the Fermi velocity; dS_i is an element of the Fermi surface on which integration is carried out; $\mathbf{K}$ is the reciprocal lattice vector; m is the cyclotron mass in a given orbit; k_B is the Boltzmann constant. It follows from the law of conservation of momentum the angle of the electron–electron scattering is large ($\vartheta_{ee}\sim1$) and, consequently, each electron–electron collision event is effective irrespective of the thickness of the sample d. Moreover, the square of the matrix element in Eq. (1) is integrated over all the initial states of the electron being scattered $\mathbf{k}_2$ and over the final states of the interacting electrons $\mathbf{k}_3$ and $\mathbf{k}_4$, and it is averaged over points on the orbit of the electron being scattered $\mathbf{k}_1$. Thus, it follows that $\bar\nu_{ee}$ should depend weakly on the position of the orbit on the Fermi surface, since it is governed by the states on the whole surface.

We shall consider a model of the Fermi surface comprising two spherical sheets I and II in which the electron wave functions ψ are in the form of unmodulated plane waves and the scattering occurs on the screened Coulomb potential $(e^2/r)\exp(-gr)$. Allowing for the normal electron–electron collisions ($|\mathbf{K}|=0$), we can calculate the integral in Eq. (1) in accordance with Ref. 15 and show that

$$\nu_{ee}(T)=\frac{4\pi}{3}\left(\frac{e}{\hbar}\right)^4\frac{k_2k_3k_4}{v_2v_3v_4}\frac{I}{k_1g}(k_BT)^2, \tag{2}$$

where

$$I=\left(\frac{\Delta k}{\Delta k^2+g^2}+\frac{1}{g}\operatorname{arctg}\frac{\Delta k}{g}\right)\Bigg|_{\Delta k_{min}}^{\Delta k_{max}}.$$

For the interband collisions the values of $\Delta k_{\min}$ and $\Delta k_{\max}$ are

1) $k_1<k_{II}$, $\Delta k_{max}=|\mathbf{k}_{II}|+|\mathbf{k}_I|$, $\Delta k_{min}=|\mathbf{k}_{II}|-|\mathbf{k}_I|$,
2) $k_1>k_{II}$, $\Delta k_{max}=2|\mathbf{k}_{II}|$, $\Delta k_{min}=|\mathbf{k}_I|-|\mathbf{k}_{II}|$.

For the intraband collisions[1)] we have $|\mathbf{k}_{II}|=|\mathbf{k}_I|=k$, $\Delta k_{\max}=2k$, $\Delta k_{\min}=0$.

The following combinations of the initial and final states are generally possible:

$$\mathbf{k}_1^{\mathrm I},\mathbf{k}_2^{\mathrm I}\to\begin{cases}\mathbf{k}_3^{\mathrm I},\mathbf{k}_4^{\mathrm I}\\ \mathbf{k}_3^{\mathrm I},\mathbf{k}_4^{\mathrm{II}}\\ \mathbf{k}_3^{\mathrm{II}},\mathbf{k}_4^{\mathrm I}\\ \mathbf{k}_3^{\mathrm{II}},\mathbf{k}_4^{\mathrm{II}}\end{cases},\quad \mathbf{k}_1^{\mathrm I},\mathbf{k}_2^{\mathrm{II}}\to\begin{cases}\mathbf{k}_3^{\mathrm I},\mathbf{k}_4^{\mathrm I}\\ \mathbf{k}_3^{\mathrm I},\mathbf{k}_4^{\mathrm{II}}\\ \mathbf{k}_3^{\mathrm{II}},\mathbf{k}_4^{\mathrm I}\\ \mathbf{k}_3^{\mathrm{II}},\mathbf{k}_4^{\mathrm{II}}\end{cases}. \tag{3}$$

In the presence of a large number of sheets, the number of possible combinations rises correspondingly.

We shall replace each of the sheets of the Fermi surface of molybdenum by a sphere with the average radius. Then, the jack consists of a central sphere surrounded by six smaller spheres (spheroids). In this way we find that the Fermi surface consists of 20 spherical sheets of different radii. Clearly, the number of possible electron–electron collisions on this Fermi surface is considerably greater than on a single-sheet surface. If we sum the contributions of these sheets to $\bar{\nu}_{ee}$ by means of Eq. (2) and consider only the combinations of Eq. (3) (for each pair of surfaces), we find that $\bar{\nu}_{ee} \approx 1\times10^7 T^2$ sec$^{-1}\cdot$°K^{-2} for all the sheets of the Fermi surface of molybdenum. In general, this value is overestimated several times because the formulas for $\bar{\nu}_{ee}(T)$ are obtained in the Born approximation.[15,16] Nevertheless, the agreement with the experimental data for such a rough model of the Fermi surface is very striking. However, it should be stressed that, in fact, the calculations of $\bar{\nu}_{ee}(T)$ must be carried out allowing for the real nature of the wave functions of electrons in the Fermi surface of the metals, exactly as has been done for the electron–phonon interaction in molybdenum.[4]

It is well known that the conductivity σ is given by the following expression:[15]

$$\sigma = \frac{e^2}{6\pi^2 h}\int \frac{v(\mathbf{k})\,dS}{\nu_{tr}(\mathbf{k})}, \tag{4}$$

where $\nu_{tr}(\mathbf{k})$ is the transport collision frequency. In the case of compensated metals, which include molybdenum, the contribution to $\sigma(T)$ is made by the normal electron–electron collisions as well as by the umklapp processes.[1,17] In fact, as pointed out earlier, the quadratic dependence $\sigma(T)$ has been observed in a large number of investigations.[2,3] The available data make it easy to estimate the transport collision frequency averaged over the Fermi surface: $\langle\nu_{tr}\rangle(T) = e^2 S_F \langle v\rangle / 6\pi^2 h\sigma \approx 3\cdot10^7 T^2$ sec$^{-1}\cdot$°K^{-2}. In fact, this estimate is very rough for such a complex Fermi surface as that of molybdenum, but nevertheless the observed agreement is very good. It should be noted that investigations of the temperature dependence of the attenuation of ultrasound in molybdenum[18] also support the quadratic dependence of the collision frequency averaged over the Fermi surface and the coefficient is similar: $\langle\nu\rangle_q = 4\cdot10^7 T^2$ sec$^{-1}\cdot$°K^{-2}.

All the above considerations are based on the theory of the electron–electron scattering which allows for the Coulomb repulsion of carriers in the scattering process. It is shown in Ref. 19 that allowance for the mutual attraction of electrons because of the exchange of virtual phonons partly compensates the Coulomb repulsion. As a result, the T^2 law applies only for $T < T_A$ (where $T_A \ll T_D$ is some characteristic temperature), whereas for $T > T_A$ dependence $\nu_{ee}(T)$ is described by the T^6 law. The coefficient in front of T^2 is found to be $(T_A/T_D)^4$ times less than for the Coulomb scattering. However, these theoretical conclusions are in conflict with the experimental results. In fact, the T^2 law in the dependence $\rho(T)$ for transition metals is observed in a wide temperature range and the T^6 law has not been observed. Moreover, the value of T_A for some metals[14] are even greater than T_D and do not agree with the results of Akhiezer *et al.*[19] Clearly, the model of Akhiezer *et al.*[19] has to be refined.

It follows from our experiments that the quadratic dependence of the collision frequency in molybdenum is due to a strong contribution of the electron–electron scattering. The value of ν_{ee} is in agreement with theoretical estimates. Further studies of this interaction in molybdenum and other metals are definitely desirable because the electron–electron interaction has a number of very interesting properties.

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[1)]There is a misprint in Eq. (2) of Ref. 14: the numerator should contain k^3 and not k^2.

[1]V. V. Boĭko, V. F. Gantmakher, and V. A. Gasparov, Zh. Eksp. Teor. Fiz. **65**, 1219 (1973) [Sov. Phys. JETP **38**, 604 (1974)].
[2]N. V. Volkenshtein, V. P. Dyakina, and V. E. Startsev, Phys. Status Solidi B **57**, 9 (1973).
[3]J. C. Garland and D. J. Van Harlingen, Phys. Rev. B **10**, 4825 (1974).
[4]J. Yamashita and S. Asano, Prog. Theor. Phys. **51**, 317 (1974).
[5]V. F. Gantmakher and V. A. Gasparov, Zh. Eksp. Teor. Fiz. **64**, 1712 (1973) [Sov. Phys. JETP **37**, 864 (1973)].
[6]V. V. Boĭko and V. A. Gasparov, Zh. Eksp. Teor. Fiz. **61**, 2362 (1971) [Sov. Phys. JETP **34**, 1266 (1972)].
[7]V. V. Boĭko, V. A. Gasparov, and I. G. Gverdtsiteli, Zh. Eksp. Teor. Fiz. **56**, 489 (1969) [Sov. Phys. JETP **29**, 267 (1969)].
[8]M. Surma, J. Lebech, and K. Saermark, Solid State Commun. **17**, 1359 (1975).
[9]R. Herrmann and H. Krüger, Phys. Status Solidi **41**, 99 (1970).
[10]V. F. Gantmakher, Rep. Prog. Phys. **37**, 317 (1974).
[11]V. A. Gasparov, Zh. Eksp. Teor. Fiz. **68**, 2259 (1975) [Sov. Phys. JETP **41**, 1129 (1975)].
[12]V. A. Gasparov, Proc. of European Physical Society Study Conf. on Transport Properties of Normal Metals and Alloys below Θ_D, Cavtat, Yugoslavia, 1977, p. 1.
[13]Yu. Kagan and A. P. Zhernov, Zh. Eksp. Teor. Fiz. **50**, 1107 (1966); **60**, 1832 (1971) [Sov. Phys. JETP **23**, 737 (1966); **33**, 990 (1971)].
[14]V. A. Gasparov and M. A. Arutyunyan (Harutunian), Solid State Commun. **19**, 189 (1976).
[15]J. M. Ziman, Electrons and Phonons, Oxford University Press, 1960 (Russ. Transl., IIL, M., 1962).
[16]C. A. Kukkonen and H. Smith, Phys. Rev. B **8**, 4601 (1973).
[17]V. F. Gantmakher and I. B. Levinson, Zh. Eksp. Teor. Fiz. **74**, 261 (1978) [Sov. Phys. JETP **47**, 133 (1978)].
[18]D. P. Almond, D. A. Detwiler, and J. A. Rayne, Phys. Lett. A **54**, 229 (1975).
[19]A. I. Akhiezer, I. A. Akhiezer, and V. G. Bar'yakhtar, Zh. Eksp. Teor. Fiz. **65**, 342 (1973) [Sov. Phys. JETP **38**, 167 (1974)].

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