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INELASTIC SCATTERING IN METALS

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The method of non-equilibrium statistical operators[1] was used to find the nonlinear corrections to Ohm's law, resulting from inelastic scattering by impurities and phonons. It is shown that the nonlinearities can be used to reconstruct the structure of the energy levels of the impurity in the metal in the case when there is no noticeable heating of the electron and impurity subsystems by the external field. It is possible also to reconstruct the phonon spectrum in the absence of phonon dragging effects. The entire analysis is based on the assumption that the phonon system is in equilibrium, and no account is taken of the electron-electron scattering.

1. INTRODUCTION

PROCESSES connected with inelastic scattering of electrons by impurities or lattice defects can be significant if the impurities (defects) have internal degrees of freedom. The latter is possible in scattering by paramagnetic impurities, by defects forming a sufficiently deep potential well, by neutral molecules that can be in different states, etc. Of particular interest in this sense is scattering by complex organic molecules, which have internal rotational degrees of freedom with low-energy states. It is clear that the act of scattering with variation of the internal state of the scattering center will occur only if the electron acquires between two collisions an excess energy eV (V is the difference of the potentials at the points between two nearest collisions) sufficient to excite the impurity. This opens up an additional channel for scattering, and the differential resistance increases. The dependence of the current on the field intensity contains information on the energy structure of the scattering center, and this information can be determined experimentally. This can be illustrated by means of the following qualitative reasoning.

Let the electron have prior to acceleration in the electric field a certain energy below which all the states are occupied. Then, when accelerated over a distance λ between two nearest collision, it acquires an energy $\Delta E = e\mathbf{E}\cdot\mathbf{n}\lambda$ (where ΔE is the energy difference that the scattering center can take on, $\mathbf{E}$, is the electric field, and $\mathbf{n}$ is a unit vector directed from the first scattering center to the second), and then the current experiences as a result of the inelastic scattering an increment

$$\Delta I \sim -\int_{-1}^{1} dx(eEx\lambda - \Delta E)\,\theta(eEx\lambda - \Delta E).$$

It is easily seen that the third derivative of the current with respect to the field intensity will then contain $\delta(eE\lambda - \Delta E)$. From the δ-like peaks of the derivatives it is possible to determine the energy spectra of the scatterer. Integration with respect to x in the direction for ΔI is the result of averaging over the solid angle of the incoming electrons. A more detailed analysis reveals an additional angular averaging, which causes the resonant structure connected with the inelastic scattering to appear not in the third but in the fourth derivative. It may turn out that the peaks of the derivatives will arise also when the electron acquires an energy sufficient to excite the impurities through 1, 2, etc. scattering acts. However, when the impurity is excited through one scattering act the expression for ΔI is subjected to an additional averaging over the angles and to a random distribution of the impurities, as a result of which only the process considered above is significant.

Inelastic scattering by static defects can be investigated experimentally at not very high temperatures and fields. At low temperatures, scattering by phonons, which strictly speaking is always inelastic, is "frozen out." However, if the fields are such that the drift velocity of the electrons becomes of the order of the velocity of sound, then phonon generation arises and leads to intense dissipation, against the background of which it is difficult to distinguish the weak nonlinearities connected with the static inelasticity. At the same time, if we investigate nonlinear effects resulting from phonon generation, then we can interpret the phonon spectra in those cases when phonon dragging can be neglected, i.e., when there exists a momentum dissipation mechanism proceeding from the electron system to the phonon system.

2. GENERAL RELATIONS

Inasmuch as in this problem deviations from Ohm's law are significant, which furthermore (as seen from the simplified reasoning) are non-analytic functions of E, we cannot use perturbation theory in powers of E, in spite of the fact that we shall henceforth assume E not to be very large. In the case when nonlinear processes are significant, it is easiest to obtain the solution of the problem by using the method of nonequilibrium statistical operators, developed by Zubarev[1].

For simplicity we shall consider a model system consisting of a gas of free electrons interacting with phonons and impurities. The Hamiltonian of such a system $\mathcal{H}$ is written in the form

$$\mathcal{H} = \mathcal{H}_0 + H_I. \tag{1}$$

Here $\mathcal{H}_0$ is the Hamiltonian of the electrons in electric field, of the phonons, and of the impurities, and

H_I is the Hamiltonian of the interaction of the electrons with the impurities and phonons:

$$\mathscr{H}_0=\sum_\sigma\int d^3r\psi_\sigma^+(\mathbf{r})\left(-\frac{\nabla^2}{2m}-e\mathbf{E}\mathbf{r}\right)\psi_\sigma(\mathbf{r})+H_{\mathrm{i}}+H_{\mathrm{ph}}, \tag{2}$$

$$H_I=\sum_\sigma\int d^3r\psi_\sigma^+(\mathbf{r})\left[\sum_l U(\mathbf{r}-\mathbf{r}_l)+g\varphi(\mathbf{r})\right]\psi_\sigma(\mathbf{r}), \tag{3}$$

$\psi_\sigma^+(\mathbf{r})$ and $\psi_\sigma(\mathbf{r})$ are the electron field operators. $\varphi(\mathbf{r})$ is the phonon field operator, H_{i} and H_{ph} are the Hamiltonians of the impurities and the phonons, $U(\mathbf{r})$ is the potential of the interaction between the electrons and the impurity, $\mathbf{r}_l$ is the location of the mass center of the l-th impurity, and g is the constant of the electron-phonon interaction.

In (3), U is a function not only of the coordinates of the conduction electrons, but also of the operators describing the internal motion in the impurity. Following Zubarev, let us determine the quasiequilibrium statistical operator ρ_e, which we shall need to find the parameters of the non-equilibrium operator

$$\rho_e=e^{-S}=\exp\{-[\ln\operatorname{Sp}(e^{-\beta H})+\beta H]\}. \tag{4}$$

Here S is the entropy operator (see[2]), which does not depend on the time, since we are considering a stationary state of the system, β is the reciprocal temperature,

$$H=H_0+H_I=\sum_\sigma\int d^3r\psi_\sigma^+(\mathbf{r})\left[\frac{(-i\nabla-m\mathbf{v})^2}{2m}-e\mathbf{E}\mathbf{r}-\mu(\mathbf{r})\right]\psi_\sigma(\mathbf{r})$$
$$+H_{\mathrm{i}}+H_{\mathrm{ph}}+H_I, \tag{5}$$

where $\mathbf{v}$ and $\mu(\mathbf{r})$ are thermodynamic parameters having the meaning of the mass velocity and the chemical potential and determined with the aid of the additional requirements

$$n(\mathbf{r})=\sum_\sigma\langle\psi_\sigma^+(\mathbf{r})\psi_\sigma(\mathbf{r})\rangle_e=\sum_\sigma\langle\psi_\sigma^+(\mathbf{r})\psi_\sigma(\mathbf{r})\rangle,\quad\langle\mathbf{P}\rangle_e=\langle\mathbf{P}\rangle, \tag{6}$$

where $\langle\ldots\rangle_e$ and $\langle\ldots\rangle$ denote averages over the quasi-equilibrium and over the nonequilibrium ensembles, and

$$\mathbf{P}=-\frac{i}{2}\sum_\sigma\int d^3r[\psi_\sigma^+(\mathbf{r})\nabla\psi_\sigma(\mathbf{r})-\nabla\psi_\sigma^+(\mathbf{r})\psi_\sigma(\mathbf{r})]. \tag{7}$$

The choice of the thermodynamic parameters β, $\mathbf{v}$, and $\mu(\mathbf{r})$ in (4) and (5) is determined by the fact that a stationary homogeneous solution is sought, for which the system is characterized by a single temperature of the electrons, phonons, and impurities, an average velocity of the electron gas, and a certain chemical potential that depends on the coordinates as a result of the fact that the Hamiltonian contains a term with the electric field. Generally speaking, in sufficiently strong electric fields and at a noticeable impurity concentration, a situation is possible at which the temperature of the electrons, phonons, and impurities are different. In addition, in very strong fields it may turn out that the velocity and the temperature of the electrons in the stationary state depend on the coordinates in a plane perpendicular to the direction of the motion of the electron gas. Consequently, if it is assumed that β and $\mathbf{v}$ do not depend on the coordinates, then it is necessary to assume for metals that at least $eE \ll p_0^3/m$ (p_0 is the Fermi momentum).

To use relations (6) it is necessary to find the connection between the mean values over the quasi-equilibrium ensemble and the parameters $\mathbf{v}$ and μ. To this end we use the exact relation

$$\langle[H\mathbf{P}]_-\rangle_e=0. \tag{8}$$

Relation (8) is the consequence of the properties of the operation of taking the trace and the commutativity of ρ_e and H. On the other hand, by commuting H and $\mathbf{P}$ in (8) we obtain, taking (6) into account,

$$e\mathbf{E}N+\int d^3r\nabla\mu(\mathbf{r})n(\mathbf{r})-i\langle[\mathbf{P}H_I]_-\rangle_e=0, \tag{9}$$

where N is the total number of electrons. One of the possible solutions of (9) is

$$\mu(\mathbf{r})=\mu_0-e\mathbf{E}\mathbf{r}+i\mathbf{r}\langle[\mathbf{P}H_I]_-\rangle_e/N, \tag{10}$$

where μ_0 is the chemical potential of the system at $E = v = 0$.

As seen from (10), to determine $\mu(\mathbf{r})$ it is necessary to find the mean value

$$\langle[\mathbf{P}H_I]_-\rangle_e=\langle[\mathbf{P}H_I]_-\rangle_e^0=\mathbf{y}, \tag{11}$$

where $\langle\ldots\rangle_e^0$ is the mean value with the operator ρ_e at $v = 0$. By calculating directly the mean value in (11) we obtain an equation for $\mathbf{y}$. It is easy to see that this equation is of the form

$$\mathbf{y}=\mathbf{y}f(y^2). \tag{12}$$

One of the possible solutions of (12) is $\mathbf{y} = 0$. In principle, other solutions can also exist, but if we choose the solution with $\mathbf{y} = 0$, then $\mathbf{v}$ will have the meaning of the drift velocity of the electron gas. It is easy to see that

$$\langle\mathbf{P}\rangle_e=\langle\mathbf{P}\rangle_e^0+m\mathbf{v}N. \tag{13}$$

If $\mathbf{y} \neq 0$, then $\langle\mathbf{P}\rangle_e^0 \neq 0$, and the drift velocity, by virtue of the conditions (6), will be the quantity $\mathbf{v}+\langle\mathbf{P}\rangle_e^0/mN$. Apparently the final results do not depend on the solutions chosen for Eqs. (9) and (12), but from the computational point of view the proposed choice has considerable advantages.

We write down the non-equilibrium statistical operator in the form

$$\rho=\exp\left[-S+\int_{-\infty}^{0}dte^{\epsilon t}\dot{S}(t)\right] \tag{14}$$

Here $\epsilon = +0$ and $\dot{S}$ is the entropy production operator:

$$\dot{S}=i\beta\mathbf{v}[\mathbf{P}(t)H_I(t)]_-+\beta e\mathbf{E}[\mathbf{P}(t)-m\mathbf{v}N]/m, \tag{15}$$

where $\mathbf{P}(t)$ and $H_I(t)$ are the Heisenberg representations for the operators. If H_I is small, then the entropy-production operator is small and it is possible to expand in its terms the non-equilibrium statistical operator (see[2]). Actually such an expansion is an expansion in powers of the interaction operator[2], so that to take correct account of the orders of the perturbation-theory series it is necessary to expand consistently in H_I. A case is possible, of course, when the small quantities are the fluxes that result from the weak deviation of the non-equilibrium state from the quasi-equilibrium state. In the present case, however, we assume smallness not of the fluxes but of H_I.

Using the Hamiltonian (1), we can easily write down an equation of motion for the operator $\mathbf{P}$:

$$d\mathbf{P}/dt = e\mathbf{E}N - i[\mathbf{P}H_I]_- . \tag{16}$$

Averaging (16) with the aid of the statistical operator (14), we obtain after expanding H_I in the first nonvanishing order

$$e\mathbf{E}n = -\beta \int_{-\infty}^{0} dte^{\varepsilon t} \int_{0}^{1} d\tau \langle [\mathbf{vP}(t), H_I(t)]_- [\tilde{\mathbf{P}}(\tau), \tilde{H}_I(\tau)]_- \rangle_0 . \tag{17}$$

Here and throughout we assume for simplicity that the volume of the system is equal to unity, so that the number of electrons coincides with the concentration n; $\langle \ldots \rangle_0$ denotes averaging with the quasi-equilibrium operator at $H_I(t) = 0$; A(t) are operators in the interaction representation, and

$$\tilde{A}(\tau) = \exp(-\tau\beta H_0) A \exp(\tau\beta H_0).$$

Relation (17) gives the sought connection between the field intensity **E** and the current **I** = env. As can be readily seen, the proposed method of considering the behavior of the system of electrons in the electric field is inverted relative to the traditional methods. One usually specifies the electric fields and one seeks the current produced in the system under the influence of this field. Here, on the other hand, we specify the current and find the field corresponding to this current. In this sense the theoretical calculation is in better correspondence with experiment, since to plot the current-voltage characteristics of highly conducting samples it is customary to specify the current and to measure the resultant potential difference across the sample.

3. SCATTERING BY IMPURITIES

To calculate (17) we determine the commutator

$$[\mathbf{P}(t) H_I(t)]_- = - \sum_{l,\mathbf{p},\mathbf{q},\sigma} \mathbf{q} a^{+}_{\mathbf{p},\sigma}(t) a_{\mathbf{p}+\mathbf{q},\sigma}(t) e^{i\mathbf{q}\mathbf{r}_l} \langle \mathbf{p} | U(t) | \mathbf{p}+\mathbf{q} \rangle . \tag{18}$$

Here $a^{+}_{p,\sigma}(t)$ and $a_{p,\sigma}(t)$ are the operators of creation and annihilation of electrons in states with momentum p and spin projection σ in the interaction representation, $\langle p | U(t) | p + q \rangle$ is the matrix element of the operator U(t) calculated with plane waves with momenta p + q and p,

$$\begin{aligned} a_{\mathbf{p},\sigma}(t) &= \exp[i(\mathscr{H}_{0\text{e}} - e\mathbf{ER})t] a_{\mathbf{p},\sigma} \exp[-i(\mathscr{H}_{0\text{э}} - e\mathbf{ER})t], \\ U(t) &= \exp(iH_{\text{i}}t) U \exp(-iH_{\text{i}}t), \end{aligned} \tag{19}$$

where $\mathscr{H}_0$ is the Hamiltonian of the free electron gas without the field, and

$$\mathbf{R} = \sum_{\sigma} \int d^3r\, \mathbf{r} \psi_\sigma^{+}(\mathbf{r}) \psi_\sigma(\mathbf{r}).$$

In the evolution operator in (19) it is convenient to separate the exponentials containing the kinetic energy of the electrons and the energy connected with the electric field:

$$\begin{aligned} \exp[i(\mathscr{H}_{0\text{э}} - e\mathbf{ER})t] &= \exp\left[i\left(\mathscr{H}_{0\text{э}} + \frac{e\mathbf{PE}t}{2m}\right)t\right] \\ &\times \exp(-ie\mathbf{ER}t) \exp\left[\frac{ie^2E^2t^3N}{6}\right]. \end{aligned} \tag{20}$$

Recognizing that

$$e^{-ie\mathbf{ER}t} [\mathbf{P}H_I]_- e^{ie\mathbf{ER}t} = [\mathbf{P}H_I]_-,$$

we obtain for (18) with the aid of (20) the expression

$$\begin{aligned} [\mathbf{P}(t) H_I(t)]_- = &- \sum_{l,\mathbf{p},\mathbf{q},\sigma} \mathbf{q} e^{i\mathbf{q}\mathbf{r}_l} a^{+}_{\mathbf{p},\sigma} a_{\mathbf{p}+\mathbf{q},\sigma} \langle \mathbf{p} | U(t) | \mathbf{p}+\mathbf{q} \rangle \\ &\times \exp\left[i\left(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}+\mathbf{q}} - \frac{e\mathbf{Eq}t}{2m}\right)t\right], \end{aligned} \tag{21}$$

where ϵ_p is the kinetic energy of the free electron ($\epsilon_p = p^2/2m$).

Using (17) and (21), after thermodynamic averaging and averaging over the random location of the impurities, and also after a number of simple transformations we obtain for the field intensity the expression

$$\begin{aligned} e\mathbf{E}n = 2c \sum_{\mathbf{p},\mathbf{q};n\,m} \mathbf{q}(\mathbf{qv}) |U_{nm}(\mathbf{q})|^2 \frac{f_m(1-n_{\mathbf{p}})n_{\mathbf{p}+\mathbf{q}} - f_n n_{\mathbf{p}}(1-n_{\mathbf{p}+\mathbf{q}})}{\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}+\mathbf{q}} + \omega_{nm}} \\ \times \int_{-\infty}^{0} dt \cos\left(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}+\mathbf{q}} + \omega_{nm} - \mathbf{qv} - \frac{e\mathbf{Eq}t}{2m}\right)t. \end{aligned} \tag{22}$$

Here ω_{nm} is the energy difference between the n-th and m-th energy state of the impurity, f_n is the population of the n-th energy level of the impurity, c is the impurity concentration in the metal, $U_{nm}(q)$ is the matrix element of the operator of the interaction between the electron and the impurity:

$$U_{nm} = \langle \mathbf{p}, n | U | \mathbf{p}+\mathbf{q}, m \rangle,$$

and n_p is the Fermi distribution function

$$n_{\mathbf{p}} = [1 + \exp\beta(\varepsilon_{\mathbf{p}} - \mu_0)]^{-1}.$$

It is easy to verify that if the interaction potential of the electron with the impurity is central, then the matrix element $U_{nm}(q)$ depends only on the modulus of q. For simplicity, we assume throughout a spherically-symmetrical interaction potential.

It is convenient to divide (22) into two parts: eEn = $\mathbf{A}_1$ + $\mathbf{A}_2$. One of these parts, $\mathbf{A}_1$, contains the contribution of only the elastic scattering by the impurities, i.e., scattering at which no change takes place in the internal energy state of the impurity. Contributing to $\mathbf{A}_2$ are processes accompanied by changes of the proper energy of the impurity. Elementary calculations yield for $\mathbf{A}_1$ the expression

$$\mathbf{A}_1 = \mathbf{v}mn/\tau, \tag{23}$$

where

$$\tau^{-1} = 8\pi N(0) c \sum_n f_n \int_0^1 dyy^3 |U_{nn}(2p_0y)|^2,$$

and N(0) is the density of states on the Fermi surface. The formula for $\mathbf{A}_1$ was obtained under the assumption that $v \ll p_0/m$ and $eE \ll p_0^3/2m$. These inequalities, as a rule, are very well satisfied in experiment even in the case of semimetals. If we assume in the expression for τ that all the f_n with the exception of those corresponding to the ground state of the impurity are equal to zero, then we obtain the usual expression for the relaxation time. Actually the obtained expression is a trivial generalization to include the case when the impurity is in several energy states, and the probability of observing it in the n-th state is equal to f_n ($\sum_n f_n = 1$). A much more interesting result is contained in the expression for $\mathbf{A}_2$, which can be represented in the form

$$\begin{aligned} \mathbf{A}_2 = \frac{\mathbf{v}cm^{1/2}}{\pi^2 2^{5/2}} \sum_{n\neq m} \int_0^\infty d\varepsilon \varepsilon^{-1/2} N(\varepsilon - \mu_0) \int_{-1}^{1} dxx^2 \int_0^\infty dq\, q^2 |U_{nm}(q)|^2 \int_{-\infty}^{\infty} \frac{d\omega}{\omega} \\ \times \theta\left(\left|\frac{q^2}{2m} + \omega - \omega_{nm}\right| - q\sqrt{\frac{2\varepsilon}{m}}\right) \{f_m[1 - n(\varepsilon)] n(\varepsilon + \omega_{nm} - \omega) \end{aligned}$$

$$-f_n n(\varepsilon)\left[1-n(\varepsilon+\omega_{nm}-\omega)\right]\}\int\limits_{-\infty}^{0} dt\cos\left[(\omega-qvx)t-\frac{eEqx}{2m}t^2\right]. \tag{24}$$

Here $\theta(x)$ is the Heaviside step function.

Further simplification of the expression for A_2 is possible under the assumption that $\tau^{-1} \ll |\omega_{nm}| \ll \mu_0$. Then, as can be readily seen, the main contribution in the integration with respect to ω in (24) ($\beta^{-1} \lesssim |\omega_{nm}|$) will be made by the region in which $|\omega| \ll \mu_0$, and therefore each of the terms in the curly brackets is a function with a sharp maximum in the vicinity of the Fermi energy. Under these conditions it is easy to carry out the integration with respect to ϵ and x, so that as a result we obtain the expression

$$\mathbf{A}_2=\frac{8vmN(0)p_0^3}{\pi^2}\sum_{n\neq m} f_n\int\limits_0^1 dyy^3\int\limits_{-\infty}^{\infty}\frac{d\omega}{\omega}|U_{nm}(2p_0y)|^2 \tag{25}$$

$$\times B(2p_0y,\omega)(\omega-\omega_{nm})\left[1+\operatorname{cth}\frac{\beta(\omega_{nm}-\omega)}{2}\right].$$

Here

$$B(q,\omega)=\int\limits_{-\infty}^{0} dt\cos\omega t\left\{\frac{\sin Q}{Q}+\frac{2}{Q^2}\left[\cos Q-\frac{\sin Q}{Q}\right]\right\},$$

$$Q\equiv qt(v+eEt/2m).$$

In order to see the character of the dependence of $\mathbf{A}_2$ on v in expression (25), let us consider the case when the impurity can be in only two states. It is assumed also that the energy ϵ_1 of the excited state greatly exceeds the thermal energy, and the electric field is not very strong, so that the inequality $(eEv_0)^{1/2} \ll \beta^{-1}$ is satisfied (v_0 is the electron velocity on the Fermi surface). Under these conditions, in the region where $\omega_1 - 2p_0v \gg \beta^{-1}$, we have

$$\mathbf{A}_2=8\pi mN(0)\frac{nc\mathbf{v}}{\beta p_0v}\int\limits_0^1 dyy\left(\frac{\omega_1}{2p_0v}-y\right)|U_{01}(2p_0y)|^2\exp[-\beta(\omega_1-2p_0vy)]. \tag{26}$$

We see that $\mathbf{A}_2$ is an exponentially small quantity in the entire region of variation of v from zero up to $v \approx \omega_1/2p_0$. If $\beta^{-1} \ll (eEv_0)^{1/2}$, then A_2 in this region is also exponentially small, but the argument of the exponential in (26) contains a quantity of the order of $(\omega_1 - 2p_0vy)/(eEv_0)^{1/2}$. The term preceding the exponential is likewise strongly altered in this case.

It is also of interest to calculate the asymptotic expression for $\mathbf{A}_2$ at large v, i.e., in the region where $v \gg \omega_1/2p_0$. In this limit we can easily obtain from (25)

$$\mathbf{A}_2=8\pi mN(0)nc\mathbf{v}\int\limits_0^1 dyy^2\left(y-\frac{3\omega_1}{4p_0v}\right)|U_{01}(2p_0y)|^2. \tag{27}$$

From relations (23), (26), and (27) we can represent the character of the variation of the electric field with changing v. Figure 1 shows schematically a plot of E(v). At small drift velocities the E(v) plot is linear until an additional scattering mechanism appears at $v \approx \omega_1/2p_0$. Further increase of the current with increasing E is hindered by the increasing contribution of the inelastic processes. At sufficiently large v, practically all the carriers in the system have an excess energy sufficient to excite the impurity, and the E(v) curve again becomes a straight line. However, the slope of this line is larger because of the increasing resistance to the current as a result of the additional scattering. If the asymptotic straight line at large values of v is continued into the region of small E, then at E = 0 the line has an intercept v_l on the abscissa axis

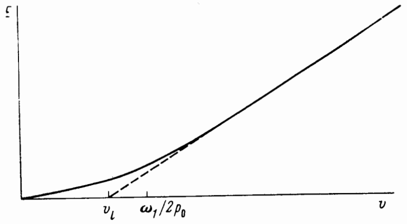

FIG. 1

$$v_l=\frac{3}{4}\frac{\omega_1}{p_0}\int\limits_0^1 dyy^2|U_{01}(2p_0y)|^2\Big/\int\limits_0^1 dyy^3|U_{01}(2p_0y)|^2. \tag{28}$$

The region where one straight section of the E(v) curve changes into the other has the dimension $\Delta v \sim p_0^{-1}\max[\beta^{-1}, (eEv_0)^{1/2}]$.

Using relations such as (28) and finding the characteristic points in the E(v) plot, we can draw definite conclusions concerning the energy spectrum of the impurity center from the current-voltage characteristics of metallic samples. However, the practical use of the current-voltage characteristics for this purpose is limited by the smallness of the matrix elements $|U_{nm}|^2$ and by the possible small spacing between the energies of the allowed transitions. Under these conditions, it will be difficult to distinguish between the individual straight-line sections of the current-voltage characteristic, and in principle the curve may not become a straight line at close values of ω_{nm} even if the distance between the neighboring ω_{nm} is $\Delta\omega > \max[\beta^{-1}, (eEv_0)^{1/2}]$. Nonetheless, it is possible to increase appreciably the resolving power of this method by investigating the derivatives of the current-voltage characteristic. The fourth derivative of E with respect to v consists of a series of sufficiently sharp "resonant" spikes at $v = |\omega_{nm}|/2p_0$, under conditions when $\beta|\omega_{nm}| \gg 1$, $\beta < (eEv_0)^{-1/2}$ or $(eEv_0)^{-1/2}|\omega_{nm}| \gg 1$, $\beta > (eEv_0)^{-1/2}$. Differentiating (25) four times and neglecting small monotonic corrections[1)], we obtain

$$\frac{\partial^4 E}{\partial v^4}=3\beta\frac{E_1}{v^4}\sum_{n,m}\omega_{nm}f_n|U_{nm}(2p_0)|^2[D(\Omega_{nm}{}^+)-D(\Omega_{nm}{}^-)]$$

$$\times\left[\sum_n f_n\int\limits_0^1 dyy^3|U_{nn}(2p_0y)|^2\right]^{-1}. \tag{29}$$

Here $E_1 = vm/\tau e$, $\Omega^{\pm}_{nm} = 2p_0v \pm \omega_{nm}$, and $D(\Omega)$ determines the form of the spike and is given by the expression

$$D(\Omega)=\frac{4}{\beta}\frac{\partial}{\partial\beta}\beta^2\int\limits_0^{\infty}\frac{dtt}{e^{2\pi t}-1}\cos[\beta^2 eEv_0t^2-\beta\Omega t]. \tag{30}$$

The function $D(\Omega)$ has a sharp maximum in the vicinity of the point $\Omega = 0$. It is convenient to introduce the parameter $\alpha = \beta(eEv_0)^{1/2}$. If $\alpha^2 \ll 1$, then it is

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[1)] The monotonic terms can be regarded as small only near the "resonant" points $v \approx |\omega_{nm}|/2p_0$, where the relative contribution is of the order of $(\beta|\omega_{nm}|)^{-1}$ or $(eEv_0)^{1/2}|\omega_{nm}|^{-1}$. Far from the spikes, their contribution can be of the same order as the terms in (29). Since the monotonic terms are of no interest from the point of view of the analysis of the energy spectrum of the impurity states, they are omitted from formula (29).

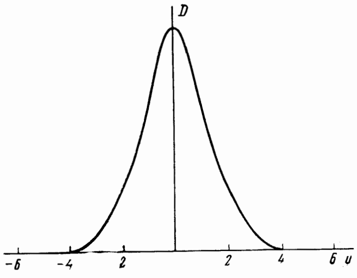

FIG. 2

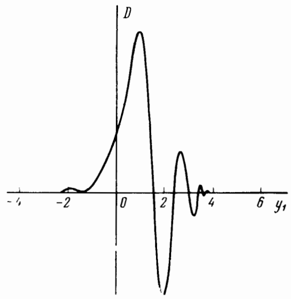

FIG. 3

possible to omit the term containing α^2 from the argument of the cosine of (30), so that the integrand is exponentially small if $t \gg (2\pi)^{-1}$, and the quadratic term still does not make a noticeable contribution. The integral obtained after neglecting the term $\alpha^2 t^2$ can be easily evaluated, and as a result we obtain for $D(\Omega)$ the expression

$$D = (y \operatorname{cth} y - 1)/\operatorname{sh}^2 y, \quad y = \beta\Omega/2. \tag{31}$$

In the case when $\alpha^2 \gg 1$, the integral in (30) is determined in a different manner, depending on whether $\Omega \leq 0$ or $\Omega > \beta^{-1}$. If $\Omega \leq 0$, then the cosine will oscillate very rapidly with changing t, so that the main contribution to the integral will be made by small t, and therefore $\exp(2\pi t) - 1$ can be replaced by $2\pi t$. On the other hand, if $\Omega > \beta^{-1}$, the integral can be calculated by the Laplace method. The solutions that are valid in different regions of the variation of Ω can then be readily interpolated to the region where $\Omega \geq 0$. The expression obtained for D in this manner is

$$D = 2\left(\frac{2\pi}{\alpha^2}\right)^{3/2}\left\{\cos y_1^2\left[\frac{1}{2} + C(y_1)\right] + \sin y_1^2\left[\frac{1}{2} + S(y_1)\right]\right\} y_1^2 \cdot \\ \times \left(\exp\frac{2\pi y_1}{\alpha} - 1\right)^{-2} \exp\frac{2\pi y_1}{\alpha}, \tag{32}$$

where $y_1 = \Omega/2(eEv_0)^{1/2}$, and C(y) and S(y) are Fresnel integrals, defined by the expressions

$$C(y) = y\sqrt{\frac{2}{\pi}}\int_0^1 dt \cos y^2 t^2, \qquad S(y) = y\sqrt{\frac{2}{\pi}}\int_0^1 dt \sin y^2 t^2.$$

Figures 2 and 3 show plots of (31) and (32). As seen from these figures. If $\alpha^2 \ll 1$, the shape of the curve is symmetrical about the point $y = 0$, and if $\alpha^2 \gg 1$ there is a clearly pronounced asymmetry, which increases with α^2. We note that the conditions $\alpha^2 \ll 1$ and $\beta|\omega_{mn}| \gg 1$ are not completely independent. Indeed, $\alpha^2 = \beta^2 eEv_0 \sim \beta^2 p_0 v/\tau$, but in the vicinity of the spike $p_0 v \sim |\omega_{mn}|$, and consequently the conditions $\beta^2|\omega_{mn}|/\tau \ll 1$ and $\beta|\omega_{mn}| \gg 1$ should be satisfied. The latter is impossible if $\beta \ll \tau$, i.e., if the thermal time is much shorter than the relaxation time. On the other hand, when $\alpha^2 \ll 1$, the dependence on E is for all practical purposes eliminated from the argument of the cosine in (22). But this dependence is a result of the fact that the electrons are accelerated between the scattering acts, since the term with the field is due to the dynamic Hamiltonian in the absence of interaction. Consequently, if the thermal time is much shorter than the average time between successive scattering processes, then the acceleration of the electron can be neglected. In the opposite case, as seen from (32), the accelerated motion of the electrons between collisions leads to an appreciable change in the shape of the spike.

4. SCATTERING BY PHONONS

As is well known, scattering by lattice vibrations is accompanied by annihilation and creation of a phonon with energy Ω, and consequently it is alwys inelastic. At high temperatures, when $\beta\Omega \ll 1$, the inelasticity can be approximately neglected. Then the dependence of the electric field on the drift velocity is linear in a wide range of variation of v. At low temperatures, the inelasticity of the scattering becomes appreciably manifest (see, for example,[3]) even in the value of the proportionality coefficient of the linear section of E(v).

Using (17) and taking (20) into account, we obtain for the field intensity the expression

$$e\mathbf{E}n = 2g^2\sum_{\mathbf{q},\mathbf{p}} \mathbf{q}(\mathbf{qv})\,\Omega_{\mathbf{q}}\,\frac{(1+N_{\mathbf{q}})n_{\mathbf{p+q}}(1-n_{\mathbf{p}}) - N_{\mathbf{q}}n_{\mathbf{p}}(1-n_{\mathbf{p+q}})}{\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p+q}} + \Omega_{\mathbf{q}}} \\ \times \int_{-\infty}^{0} dt \cos(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p+q}} - \mathbf{qv} + \Omega_{\mathbf{q}} - e\mathbf{Eq}t/2m)\,t, \tag{33}$$

where Ω_q is the frequency of the phonon with the wave vector q and N_q are the phonon occupation numbers:

$$N_{\mathbf{q}} = [\exp(\beta\Omega_{\mathbf{q}}) - 1]^{-1}.$$

In a manner similar to that used to derive (25), we can easily obtain from (33) in the Debye model

$$e\mathbf{E}n = \frac{g^2 m^2 \mathbf{v}s}{8\pi^4}\int_{-\infty}^{\infty}\frac{d\omega}{\omega}\int_0^{q_0} q^4 dq\,(\Omega_{\mathbf{q}} - \omega)\,[N(\Omega_{\mathbf{q}} - \omega) - N_{\mathbf{q}}]\,B(q,\omega). \tag{34}$$

Here s is the speed of sound and q_0 the Debye momentum.

In the derivation of (34) it was assumed that $p_0 > q_0/2$. If $p_0 < q_0/2$, then the upper limit of the integration with respect to q will be $2p_0$ and not q_0.

In the case when $v \ll s$, we obtain from (34) a linear connection between **E** and v:

$$e\mathbf{E}n = \frac{m\mathbf{v}n}{\tau} = \frac{m\mathbf{v}n}{\tau_0}F(\gamma). \tag{35}$$

Here $\gamma = \beta\Omega_0$, Ω_0 is the Debye frequency ($\Omega_0 = sq_0$), and

$$\frac{1}{\tau_0} = \frac{g^2 p_0 m\beta^{-1}}{\pi}\left(\frac{q_0}{2p_0}\right)^4, \qquad F(\gamma) = 4\gamma^{-4}\int_0^{\gamma} dz\, z^5 e^z (e^z - 1)^{-2}.$$

At high temperatures ($\gamma \ll 1$) we have $\tau \to \tau_0$, i.e., we get the usual high-temperature expression in the Debye model. When $\gamma \gg 1$ we have $\tau^{-1} \sim \beta^{-5}$, i.e., the relation derived by Bloch. The relation (34) becomes much simpler if the inequality $eE \ll ms\Omega_0$ is satisfied. If it can be assumed that $eE \sim mv/\tau$, then the inequality acquires the clearer form $\xi = v/s \ll \tau\Omega_0$. When these inequalities are satisfied, the field dependence in $B(q, \omega)$ can be neglected and it is possible to integrate with respect to ω in (34). As a result we obtain

$$e\mathbf{E}n = \frac{g^2 m^2 \mathbf{v}\Omega_0 q_0^4}{12\pi^3}\int_0^1 dy\left\{\frac{y^2}{e^{\gamma y} - 1} + L(\xi, y) + L(-\xi, y)\right\}, \tag{36}$$

where

$$L(\xi, y) = \frac{(1+\xi)^2}{\xi} y \left[1 - \frac{3}{2}(1+\xi) y + \frac{1+3\xi}{2} y^3\right] \Big/ [\exp(\gamma(1+\xi)y) - 1].$$

Let us consider certain limiting cases, when the integration in (36) can be carried out analytically. If $\gamma\xi \ll 1$, then we obtain the result that can be easily gotten also from (36). In the other limiting case, when $\gamma(1 - \xi) \gg 1$, we have

$$eEn = \frac{2g^2m^2\mathrm{v}}{\pi^3 s^4} \beta^{-5} \zeta(5) \left[1 + \frac{4}{(1-\xi^2)^3}\right], \tag{37}$$

where ζ is the Riemann Zeta function.

Relation (37) was obtained with exponential accuracy in $\gamma(1 - \xi)$, so that at low temperatures $(\gamma \gg 1)$ it can be used up to $\xi \sim 1$ $(\xi \lesssim 1)$. In the region when $\gamma\,|1 - \xi| \ll 1$ and $\gamma \gg 1$ we have

$$eEn = \frac{g^2m^2\mathrm{v}}{12\pi^3 s^4} \beta^{-1} \left[\Omega_0{}^3 \beta^{-1} \zeta(2) - \frac{(1-\xi)\Omega_0{}^4}{3}\right]. \tag{38}$$

If $\xi > 1$ and $\gamma(\xi - 1) \gg 1$, then

$$eEn = \frac{g^2m^2\mathrm{v}\Omega_0{}^5}{240\pi^3 s^4} \frac{(\xi-1)^2}{\xi^3} (2\xi + 1). \tag{39}$$

The asymptotic expression (39) can be used to determine the maximum value of ξ at which the formula (36) can still be used. From the inequality $eE \ll ms\Omega_0$ and expression (39), with allowance for the fact that $g^2 = 2\pi^2\kappa/p_0m$ ($\kappa \sim 1$ for metals, see, for example,[4]), we obtain the inequality

$$(1 - \xi^{-1})^2 (2\xi + 1) \ll 40 p_0{}^4 / \pi\kappa q_0{}^4.$$

The ratio p_0/q_0 can be either larger or smaller than unity. As is well known, in the isotropic case $p_0/q_0 = (z/2)^{1/3}$, where z is the number of free electrons per atom.

Figure 4 shows a plot of $E(\xi)$ at different values of the parameter γ. As seen from the figure, up to $\xi \sim 1$ the phonon scattering makes no noticeable contribution to the resistance at low temperatures. In the region where $\xi \gtrsim 1$, the resistance increases sharply. This increase is connected with the fact that the drift velocity of the electrons is larger than the phase velocity of the phonons, and the electrons begin to generate phonons. The mechanism of this reaction is similar to Cerenkov radiation.

We note that the emission of phonons by supersonic electrons was quite extensively investigated earlier in (see, for example,[5-10]). The purpose of these investigations was to determine the conditions under which carrier drift instability and coherent emission of phonons take place. It must be emphasized that the nonlinearity-inducing contribution to (34) is connected with incoherent emission of phonons in the stationary regime. The nonstationary region is outside the applicability of the present analysis. The upper limit of this region in terms of v depends strongly on the mechanism whereby the momentum transferrred to the phonon subsystem from the electron subsystem is dissipated. One of the mechanisms of such dissipation may be the scattering of phonons by defects. Actually, the results of[5-10] pertain to the case when there is practically no dissipation in the momentum subsystem. Therefore, owing to the effectiveness of only the normal processes of phonon-electron collisions, leading to dragging of phonons, the conductivity of the system will increase with increasing v. In our case the conductivity of the system decreases with increasing v. In a real situation this decrease of conductivity will continue until the momentum transferred to the phonon subsystem exceeds a certain critical value, above which a noticeable dragging of the phonons can occur. With further increase of v the phonon part of the conductivity will increase. Formula (34) does not give this increase, since it has been derived without allowance for the phonon dragging, using a stationary statistical operator. In spite of the fact that the approximation in question (in contrast to the approximations used in[5-10]) is of no interest from the point of view of technical applications, it may be of interest in connection with the possibility of investigating the phonon spectra by using the nonlinearities of the current-voltage characteristic.

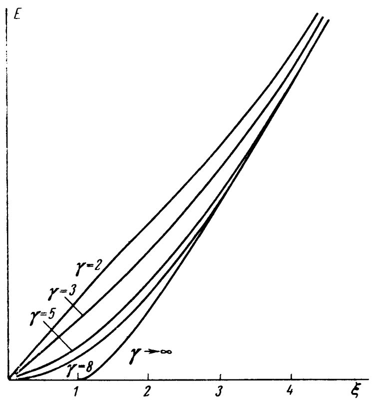

FIG. 4

It can be seen from (36) that the third derivative of E with respect to v has a δ-like singularity at v = s. This singularity is the consequence of the Debye model, for when v = s, phonons with all q begin to be generated immediately. In a real metal, there will be no singularity of this type, since each v corresponds to a certain definite q. To verify this, let us consider a model with an arbitrary isotropic dispersion law $\Omega(q)$. Relation (34) can in this case be easily changed. Using this new relation, which gives the connection between E and v at an arbitrary dispersion law, it is easy to see that at low temperatures

$$\frac{d^3E}{dv^3} + \frac{4}{v}\frac{d^2E}{dv^2} = \frac{g^2m^2}{en(2\pi)^3} \frac{q^5(v)\,\theta(2p_0 - q)}{|v - v_1(v)|}, \tag{40}$$

where v_1 is the group velocity of the phonons ($v_1 = d\Omega/dq$), and the q(v) and $v_1(v)$ relations are determined from the requirement that the drift velocity v be equal to the phonon phase velocity ($v = \Omega/q$).

Equation (40) can be written in a different manner, so as to reveal the connection between q and v:

$$q^5(v) = \frac{5en(2\pi)^3}{g^2m^2} \theta(s - v) \left\{ \frac{d^2E}{dv^2}\bigg|_{v=s} - \frac{d^2E}{dv^2} + 4\int_v \frac{dv}{v}\frac{d^2E}{dv^2} \right\}. \tag{41}$$

It is easy to see that with the aid of relation (41) it is possible to reconstruct the phonon spectrum in metals from the experimental plots of d^2E/dv^2 against v. Indeed, if it is assumed that relation (41) gives the implicit v(q) dependence, then the dispersion law is determined from the equality $\Omega(q) = qv(q)$. Relations (40) and (41) were obtained without allowance for the Umklapp processes. In this approximation, the second derivative is equal to zero at $v < v_0$, where v_0 is determined ($v_0 < s$) either by the equation $q(v_0) = \overline{q}$ if $p_0 > \overline{q}/2$ or the equation $q(v_0) = 2p_0$ when $p_0 < \overline{q}/2$

($\bar{q}$—momentum on the boundary of the first Brillouin zone). Thus, the entire information concerning the phonon spectrum is contained in a relatively small section where v varies from v_0 to s. In the Debye model, this section vanishes, since $v_1 = s = v_0$ and $q_0 = \bar{q}$. As a result, a δ-like singularity appears in relation (40); this singularity was discussed earlier. We note that if $p_0 > \bar{q}/2$ and Unklapp processes are taken into account, then the section from $\bar{v}_0$ to v_0 ($q(\bar{v}_0) = 2p_0$) contains information concerning these processes.

The resolution of this method of reconstructing the phonon spectrum can be easily determined by considering the errors introduced by the employed approximations. If $\beta^{-1} > (e E\bar{q}/m)^{1/2}$, then the error in the frequency will be $\Delta\Omega \sim \beta^{-1}$, and if the inequality is reversed we have $\Delta\Omega \sim (eE\bar{q}/m)^{1/2}$.

5. CONCLUSION

Let us assess the experimental possibilities of investigating nonlinearities in metals. The main problem arising here is the heating of the samples. It is therefore necessary to determine the power P at a given measured energy ΔE. The power P is usually drawn away from the surface of the sample by a cooling medium, so that it can be assumed that the surface has a specified temperature β_0^{-1}. If it is assumed that the conductivity depends little on the temperature (the main contribution to the conductivity is made by impurity scattering), then, using the Wiedemann-Franz law to determine the thermal conductivity of a cylindrical sample with radius a, we obtain the temperature distribution over the cross section

$$\beta^{-2}(r) = \beta_0^{-2} + \frac{3}{8\pi^2}(\Delta E)^2\frac{a^2 - r^2}{\lambda^2}, \qquad (42)$$

where $\lambda = v_0\tau$ is the mean free path.

It is seen from (42) that from the point of view of decreasing the temperature gradient in a sample it is desirable to increase the mean free path. However, when $a \sim \lambda$, a residual resistance arises as a result of scattering of the electrons by the sample boundary. Therefore, in the best case $a/\lambda \sim 1$ and, if heating in the center of the wire to a value $\Delta\beta^{-1} \sim \beta_0^{-1}$ is permissible, we get from formula (42) that $\Delta E \sim 5\beta_0^{-1}$, It is also of interest to estimate the resultant dissipated power, $P \sim V(\Delta E)^2 n/8\mu_0\tau$ (here V is the volume of the sample). For a wire of diameter $2a \sim 10^{-2}$ cm we have $\tau \sim 10^{-10}$ sec in sufficiently pure substances and $P \sim V\beta^{-2} \times 2 \times 10^4$ W, where V is in cm^3 and β^{-1} in degrees Kelvin. If $\beta^{-1} \sim 1$°K and the sample length is 10^{-1} cm, then $P \sim 0.2$ Watt. The chosen volume of the sample $V \sim 10^{-5}$ cm^3 is dictated by the following considerations: It can be assumed that the sensitivity of the method is of the same order as in tunnel investigations of inelastic tunneling with impurity molecules located in the region of the potential barrier[11-13] [2)]. It is then quite easy to register 10^{-10}—10^{-11} impurities (see[7]). If it is assumed that $c \sim 10^{16}$ cm^{-3} at $\tau \sim 10^{-10}$ sec, then $Vc \sim 10^{11}$.

The obtained estimate $\Delta E \sim 5\beta_0^{-1}$ is quite stringent and it might seem that it would not permit an investigation of the energy region of interest, which reaches 10^{20}K for impurities, and all the more will not permit investigations of phonon singularities (10^2—10^{3}°K). Nonetheless, if we use a pulse measurement procedure, then the region of attainable ΔE increases by two or even three orders of magnitude, so that apparently an experimental observation of the described effects entails no particular difficulty. We note that even in the continuous measurement procedure the situation can be greatly improved by using semimetals or degenerate semiconductors. In semimetals and in semiconductors, however, effects of phonon dragging may exert a strong influence, which was not taken into account in the present investigation. In addition, a noticeable temperature difference may arise between the impurity, the electron, and phonon subsystems. The occurrence of a temperature difference between different subsystems, the dragging of phonons, and the influence of electron scattering can greatly distort, under certain conditions, the obtained results. Allowance for these effects will be the subject of further investigations.

Let us make also a few remarks concerning formulas (30) and (32). As seen from the foregoing estimates, the case $\alpha^2 \gg 1$ is quite difficult to realize in experiment, all the more since relation (30) and (32) were derived without allowance for the proper lifetime τ_i of the excited impurity states. Under conditions when the impurity is in the metal, this time can be sufficiently small and comparable with β. If $\tau_i < \beta$, then it is necessary to consider in lieu of α the quantity $\sim \tau_i(eEv_0)^{1/2}$.

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