ISOTOPIC EFFECT IN THE RESIDUAL ELECTRICAL RESISTANCE OF METALS

I. Ia. POMERANCHUK

The existence of various isotopes in a metal leads to a difference in the zero vibrations, depending on the different masses of these isotopes. This in turn leads to the appearance of a non-regular perturbing field which is capable of scattering the conduction electrons. Thus, even in crystals without physical defects or chemical impurities, a residual electrical resistance may arise. The magnitude of this resistance is estimated.

IF several different isotopes are found in the lattice of a metal, scattering of the conduction electrons is brought about by deviation of the zero vibrations of a given isotope from their average value. This effect leads to a finite residual electrical resistance in the metal at absolute zero, under conditions in which the resistance due to chemical and physical impurities is absent. In other words, in the purification of the metal, its residual resistance ought to decrease at first and then approach a constant limit. The scattering of interest to us is a result of the combined action of the following terms in the Hamiltonian, which we shall regard as a perturbation:

(1) The additional kinetic energy of the isotope contributes a term to the Hamiltonian (the origin of the coordinates coincides with the atom of the isotope at its equilibrium position)

$$-\frac{M-\overline{M}}{2}\dot{\mathbf{u}}(0)^2 \tag{1}$$

$$= -\sum_{f_1 f_2 l_1 l_2}\frac{M-\overline{M}}{2}(a_{f_1 l_1}-a_{f_1 l_1}^+)(a_{f_2 l_2}-a_{f_2 l_2}^+)(\mathbf{j}_1\cdot\mathbf{j}_2)\frac{\hbar V\sqrt{\omega_1\omega_2}}{2\overline{M}N};$$

M is the mass of the isotopic atom, $\overline{M}$ the average mass of the atoms of the metal:

$$\overline{M} = \sum_s c_s M_s, \quad \sum_s c_s = 1, \tag{2}$$

c_s is the concentration of the isotope with mass M_s, $\mathbf{u}(0)$ the vector of the displacement of the isotopic atom from its equilibrium position, and $\mathbf{j}$ the polarization vector.

(2) We express the interaction of the electron with the vibrations of the lattice V (considering an idealized isotopic model of the metal without sound dispersion) in the usual way:

$$V(\mathbf{x}) = V_0\sum_{fj}\sqrt{\frac{\hbar\omega}{2\overline{M}s^2 N}}(a_{fj}e^{i\mathbf{f}\mathbf{x}/\hbar}+a_{fj}^+e^{-i\mathbf{f}\mathbf{x}/\hbar}); \tag{3}$$

V_0 is the energy of atomic order, and s the speed of sound. Equations (1) and (3) give the scattering of the electrons in the third approximation of perturbation theory. Equation (1) contains matrix elements of three types: (a) those corresponding to phonon scattering $(\sim a_{f_1}, a_{f_2}^+)$, (b) those corresponding to the emission of two phonons $(\sim a_{f_1}^+, a_{f_2}^+)$, and (c) those corresponding to the absorption of two phonons $(\sim a_{f_1}, a_{f_2})$. The process of scattering under the action of (1) and (3) is therefore obtained from the summation of three chains of processes which are graphically illustrated in the drawing (the initial and final states of the electron are characterized by the momenta $\mathbf{p}_0$ and $\mathbf{p}$ respectively).
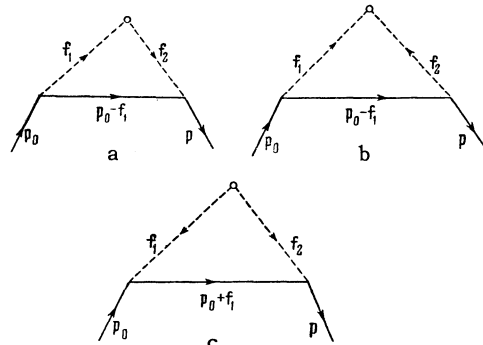
The matrix element arising under the action of the chain (b) has the form

$$-\sum_{l_1 l_2}\int V_0^2\,\frac{\hbar^2\omega_1\omega_2}{4\overline{M}^2 s^2 N^2}(M-\overline{M})(\mathbf{j}_1\cdot\mathbf{j}_2)$$

$$\times\left\{\frac{1}{[E(\mathbf{p}_0)-E(\mathbf{p}_0-\mathbf{f}_1)-sf_1][-sf_1-s|\mathbf{f}_1-\mathbf{q}|]}\right\}\frac{\Omega\,d^3f_1}{(2\pi\hbar)^3}. \tag{4}$$

Here $\hbar\omega_1 = sf_1$ and $\hbar\omega_2 = sf_2 = s|\mathbf{q}-\mathbf{f}_1|$, where $\mathbf{q} = \mathbf{p}_0 - \mathbf{p}$ is the change in the momentum of the electron. Since only the order of magnitude of the effect is of interest, we shall not consider the difference in the velocities of longitudinal and transverse sound. In the case of chain (c) we obtain a

matrix element that differs from (4) only in that, instead of the expression which appears in the curly brackets, we have

$$\left\{\frac{1}{[E(p_0)-E(p_0+f_1)-s|f_1+q|][-sf_1-s|f_1+q|]}\right\}. \quad (5)$$

In the integration of (4) and (5), the first factor can pass through zero. It is necessary to get around the singularity in a way as corresponds to the function $\delta_+(x)$. In this case, two terms arise — an integral in the sense of the principal value, and a term containing $i\delta[E(p_0)-E(p_0-f_1)-sf_1]$ in case (4), and $i\delta[E(p_0)-E(p_0+f_1)-s|f_1+q|]$ in case (5). The part containing the principal value corresponds to $|E(p_0)-E(p_0\mp f_1)|$ a value on the order of atomic energies, which is significantly greater than sf_1 or $s|f_1+q|$. Therefore, the principal value due to the chain (2) and (3) is the same in each case. Since the function $\delta[E(p_0)-E(p_0-f_1)-sf_1]$ appears with the factor i, it makes a contribution to the imaginary part of the transition matrix element. However, the sum of the imaginary parts entering from all the chains must vanish, since the matrix element itself must be real. Electrons are scattered by an isotopic atom by the presence of a small perturbing field close to the isotope. This field is produced by the deviations of the zero-point vibrations of the given isotope from their mean amplitude, taking into account the fact that the mean of the zero-point vibrations is small in comparison with the lattice constant a and, moreover, that the difference of the zero-point vibrations of the isotope is doubly small because of the smallness of $(M-\overline{M})/\overline{M}$, and because the perturbing field acting on the electron is very small. The scattering in such a field $V_{eff}(x)$ must be described by the Born approximation. The scattering matrix element with transfer of momentum q is expressed by the integral

$$\int V_{eff}(x)e^{iqx/\hbar}d^3x,$$

which is a real number, since $V_{eff}(x)=V_{eff}(-x)$.

Thus there remains in (4) only the principal value of the integral, which is the same for the chains (b) and (c). The matrix element corresponding to the chain (a) has the form

$$-\sum V_0^2\int\frac{\hbar^2\omega_1\omega_2}{4\,\overline{M}^2s^2N^2}\frac{\Omega\,d^3f_1}{(2\pi\hbar)^3}$$

$$\times\left\{\frac{1}{[E(p_0)-E(p_0-f_1)-sf_1][E(p_0)-E(p_0-f_1)-s|f_1-q|]}\right\}$$

$$\times(j_1\cdot j_2). \quad (6)$$

In the integral here, both factors in the curly brackets must vanish; therefore, we have, first,

an integral in the sense of the principal value, and second, two terms proportional to

$$i\delta[E(p_0)-E(p_0-f_1)-sf_1]$$

$$\text{or } i\delta[E(p_0)-E(p_0-f_1)-s|f_1-q|].$$

In accord with what was said above, we shall omit these.

For an estimate of the order of magnitude, we can also discard the principal value of the integral in chain (a), since the energy differences in the curly brackets of (6) correspond to energies of atomic magnitudes. One of the differences in (4) and (5) was $\hbar\omega_1+\hbar\omega_2\sim\Theta\ll V_0$ (V_0 is of the order of several electron volts). We therefore have, in order of magnitude, the following matrix element (neglecting the chain (a) and assuming $E=p^2/2m$, $m=$ effective mass of the electron) at $q=0$:

$$\mathcal{M}=-\frac{3}{2}V_0^2a^3\frac{M-\overline{M}}{(2\pi\hbar)^3N\overline{M}^2s}\int\frac{f|f-q|}{f+|f-q|}\frac{d^3f}{E(p_0)-E(p_0-f)}$$

$$=-\frac{3}{2}V_0^2a^3\frac{M-\overline{M}}{(2\pi\hbar)^3N\overline{M}s}\frac{\pi mf_0^3}{3p_0}F\left(\frac{f_0}{2p_0}\right);$$

$$F\left(\frac{f_0}{2p_0}\right)=\ln\frac{2p_0-f_0}{2p_0+f_0}-\frac{2p_0}{f_0}-\frac{8p_0^3}{f_0^3}\ln\frac{4p_0^2-f_0^2}{4p_0^2}; \quad (7)$$

f_0 is the maximum momentum of the phonon, p_0 is the momentum of the electron on the Fermi surface. We assume that $2p_0>f_0$. Substituting $(2\pi^2)^{1/3}\hbar/a$ in (7) in place of f_0, we find

$$\mathcal{M}=-\frac{V_0^2(M-\overline{M})m}{8\,\overline{M}^2sp_0N}F\left(\frac{f_0}{2p_0}\right). \quad (8)$$

We thus find the total probability of scattering W in a medium consisting of s isotopes with concentration c_s, neglecting the dependence of $\mathcal{M}$ on q:

$$W\approx\frac{F^2}{64\pi}\frac{a^3V_0^4(M-\overline{M})^2m^2}{\overline{M}^4s^2\hbar^4}\frac{dp_0}{dE}=\frac{V_0^4F^2(\overline{M^2}-\overline{M}^2)a^3m^3}{64\pi\overline{M}^4s^2\hbar^4p_0}; \quad (9)$$

$$\overline{M^2}-\overline{M}^2=\sum_s c_sM_s^2-\left(\sum_s c_sM_s\right)^2.$$

The corresponding free path l is equal to:

$$l=\frac{dE}{dp}\frac{1}{W}\approx\frac{64\pi\overline{M}^4s^2\hbar^4p_0^2}{(\overline{M^2}-\overline{M}^2)V_0^4F^2m^4a^3}. \quad (10)$$

Frequently, the quantities that enter here can be expressed in terms of the electrical conductivity σ at high temperatures ($T\gg\Theta$), which in our model is equal to

$$\sigma=4(4\pi)^{1/3}e^2n_e^2\hbar^3a\overline{M}s^2/m^2V_0^2T \quad (11)$$

(T in energy units, n_e is the number of conduction electrons in 1 cm^3). Substituting $m^2V_0^2/\overline{M}s^2$ from (11) in (10), we find:

$$l \approx (36)^{1/3}\pi^{2/3} \frac{\overline{M}^2}{\overline{M^2} - \overline{M}^2} \frac{\sigma^2 T^2}{n_e^2 \nu_e^{4/3} a s^2 F^2 e^4} \; ; \qquad (12)$$

$$(\nu_e = n_e a^3).$$

For $\dfrac{\overline{M^2} - \overline{M}^2}{\overline{M}^2} \sim 10^{-4}$, which corresponds to natural tin, we obtain l of the order of several millimeters. If we introduce the path brought about by the impurities as

$$l_i = a/\epsilon \qquad (13)$$

(ϵ is the concentration of impurities) then (12) corresponds to the following effective concentration ϵ_0:

$$\epsilon_0 \approx \frac{1}{(36)^{1/3}\pi^{2/3}} \frac{\overline{M^2} - \overline{M}^2}{\overline{M}^2} \frac{n_e^2 \nu_e^{4/3} a^2 s^2 F^2 e^4}{\sigma^2 T^2} \; . \qquad (14)$$

For $\epsilon \gg \epsilon_0$, the residual resistance is proportional to ϵ. For $\epsilon < \epsilon_0$, it no longer depends on ϵ.

We can improve the estimate of l if we introduce data on the electronic specific heat.

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