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Ioffe-Regel criterion and resistivity of metals

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Resistivity saturation is a general phenomenon observed in metals whose electrons have short mean free paths. The approach to saturation can be described rather well by the parallel-resistor formula $1/\rho=1/\rho_{\text{ideal}}+1/\rho_{\text{sat}}$, where ρ_{ideal} is calculated by the usual theory and ρ_{sat} represents a constant "shunt" resistor. In this work it is shown that the requirement of the minimum time between collisions (Ioffe-Regel criterion), incorporated in the distribution of free times, leads to the parallel-resistor formula.

Years ago, Ioffe[1] and Ioffe and Regel[2] used the argument based on the uncertainty relation to point out that the usual theory of electrical conduction has to break down when the mean free path (mfp) of carriers l approaches the interatomic distance a. The notion that values of $l<a$ are, in fact, impossible is known as Ioffe-Regel criterion. Similar ideas were expressed by Mott.[3] More recently the concept of minimum mfp was brought up by Fisk and Webb[4] in order to understand qualitatively what they called "resistivity saturation" of $A15$ compounds, and by Mooij[5] in his analysis of the resistivity of many transition-metal alloys. It was argued that resistivity does not readily increase beyond a certain "saturation" value ρ_{sat}, which roughly corresponds to $l\sim a$. Mott and others calculate this value[3] using the Kubo-Greenwood formula, suitable when $l\sim a$, and find what we may call $\sigma_{\text{sat}}=\rho_{\text{sat}}^{-1}=ce^2/\hbar a$, with $c=0.39$ or 0.5, depending on slight modifications of the procedure. Mott points out[3] that an almost identical result follows from a Boltzmann formula for the spherical Fermi surface $\sigma=S_F e^2 l/12\pi^3\hbar$, with $l=a$. For the case $K_F=\pi/a$ he finds from this formula $\sigma_{\text{sat}}=0.33e^2/\hbar a$. For $a=4$ Å this gives $\rho_{\text{sat}}=500\ \mu\Omega\,\text{cm}$. Experimental values of ρ_{sat} usually fall between 100 and 300 $\mu\Omega\,\text{cm}$. Mott's formula can be easily generalized to account for this variation. The condition $K_F=\pi/a$ used by Mott is equivalent to $na^3=1$, where n is electron concentration. In general, there are α electrons per cubic cell a^3, and with $na^3=\alpha$ we get

$$\sigma_{\text{sat}}=0.33\alpha^{2/3}\frac{e^2}{\hbar a} \tag{1}$$

or, in terms of n and a,

$$\rho_{\text{sat}}=\frac{1.29\times10^{18}}{n^{2/3}a}\ \mu\Omega\,\text{cm}\ , \tag{2}$$

where $[n]=\text{cm}^{-3}$ and $[a]=$ Å. The last formula shows explicitly the dependence of ρ_{sat} on n and helps one to understand the variation of ρ_{sat} values among different metals.

The recent interest in the resistivity of $A15$ compounds is in part because they display most clearly the *approach* to the value of ρ_{sat} with both thermal and static disorder. To describe that approach Wiesmann *et al.*[6] proposed a phenomenological "parallel-resistor" formula

$$\frac{1}{\rho}=\frac{1}{\rho_{\text{ideal}}}+\frac{1}{\rho_{\text{sat}}} \tag{3}$$

in which the measured resistivity $\rho=\rho_0+\rho(T)$ is presented as a parallel connection of the two branches. The "ideal" or classical branch $\rho_{\text{ideal}}=\rho_{0(\text{id})}+\rho_{e-\text{ph(id)}}$ is the resistivity as it would be if there were no limiting value of $l\sim a$. The second branch is a constant "shunt" resistor ρ_{sat}, the value of which is to be found from experiment. Formula (3) proved to be rather successful in fitting much of the $A15$ resistivity data[6–11] as well as the resistivity of Nb,[6] Chavrel phases,[12,13] and ternary borides.[14]

Despite these successes and despite the theoretical ground provided for (3) in the work of Chakraborty and Allen[15] which concentrated on the non-Boltzmann nature of the phenomenon, there exist certain doubts about the physical significance of the parallel-resistor formula. It is sometimes regarded as just a fitting formula. What's lacking is an intuitive understanding of why ρ_{sat} and ρ_{id} should enter in parallel, and why resistivity should

always be influenced by the presence of the shunt ρ_{sat}, even when the mfp is considerably longer than a. The aim of this paper is to resolve these difficulties and to derive (3) from a very transparent physical model based on the Ioffe-Regel criterion.

We first note that while concepts of the mean free path and the mean free time τ are most frequently used in theory, in a real solid there exist a certain distribution of free paths and times between collisions. Let probability for an electron to scatter during the time interval dt be proportional to that interval and independent of time t. If this is the case, it is easy to show that the probability dP of having no collisions during t and then collision in the interval $(t,t+dt)$ is given by

$$dP=\frac{1}{\tau}e^{-t/\tau}dt\ , \tag{4}$$

where τ is the mean time between collisions. Probability distribution dP/dt is shown in Fig. 1(a). It tells us that for any τ, more electrons scatter in the time interval dt near zero than in the interval dt at τ or at any other time. Now let us try to modify distribution (4) according to the Ioffe-Regel criterion which says that free paths $l<a$ or free times $t<\tau_0\sim a/V_F$ are forbidden (here V_F is a Fermi velocity). This implies that a new distribution dP'/dt must be such that $dP'/dt=0$ for $t<\tau_0$. At this point we notice that distribution (4) is a continuous one, while the idea of minimum time τ_0 introduces an obvious discreteness. One may be tempted to consider a discrete problem, i.e., to consider collisions only at times $t=n\tau_0$, $n=0,1,2,\ldots$. However, this would not correspond to the physical picture. Indeed, consider, for example, impurities distributed at random in a three-dimensional crystal. They cannot be closer to one another than a minimum distance of the order of a. Hence an electron after having been scattered off a certain impurity will have to travel for at least $\tau_0\sim a/V_F$ to be scattered again. Now let us assume that after the original scattering event, $n=0$, time τ_0 has passed but no further scattering corresponding to $n=1$ has occurred. In this case, at what time t can the next collision occur? The answer is by no means $t=n\tau_0$, $n=2,3,\ldots$. On the contrary, for the random distribution of impurities the next collision can be separated from the first one by any $t>\tau_0$. This point is illustrated in Fig. 2 in two dimensions; in three dimensions it is even more obvious. The same reasoning applies to phonon scattering. Therefore, we conclude that the new distribution dP'/dt must be continuous at $t>\tau_0$. As before, the probability of scattering at $t>\tau_0$ will be proportional to the time interval and independent of time. Derivation of (4) can be repeated, only now we will write $t'=t-\tau_0$ instead of t. Hence the new distribution is [Fig. 1(b)]

$$\frac{dP'}{dt}=\begin{cases}0, & t<\tau_0\\ \dfrac{1}{\tau}e^{(-t-\tau_0)/\tau}, & t\geq\tau_0.\end{cases} \tag{5}$$

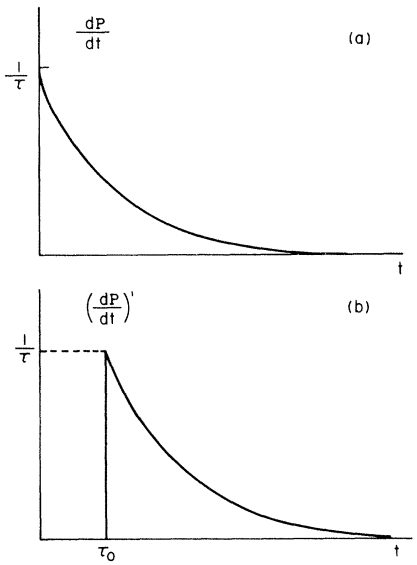

FIG. 1. (a) Distribution of times between collisions when arbitrarily small times are permitted. (b) Distribution in which times $t<\tau_0$ are forbidden.

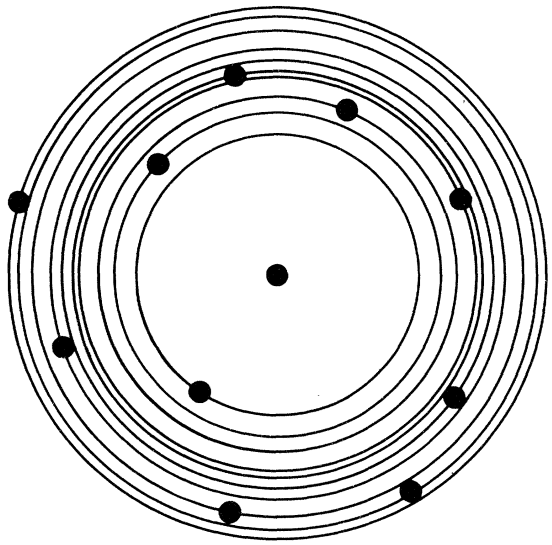

FIG. 2. Randomly distributed impurities. Circles are drawn to indicate distances from a given impurity to its neighbors.

Now let is follow the conventional procedure for obtaining σ, first using the distribution (4). In the field E electrons gain velocity $V(t)=(eE/m)t$. The drift velocity in a field E will be found as

$$V_D=\int V(t)dP=\frac{eE}{m\tau}\int_0^\infty te^{-t/\tau}dt=\frac{eE}{m}\tau,$$

the current $j=enV_D=\sigma E$ and $\sigma=\sigma_{\text{ideal}}=e^2n\tau/m$.

However, if we use the new distribution (5), we get

$$V_D'=\int V(t)dP'=\frac{eE}{m\tau}\int_{\tau_0}^\infty te^{-(t-\tau_0)/\tau}dt$$

$$=\frac{eE}{m}(\tau+\tau_0)$$

and

$$\sigma'=\frac{e^2n\tau}{m}+\frac{e^2n\tau_0}{m}=\sigma_{\text{ideal}}+\sigma_{\text{sat}}$$

which is the parallel-resistor formula.

A few words about Ioffe-Regel criterion. All agree that the usual theory of conductivity is expected to break down when $l\sim a$. One *interprets* this by saying that $l<a$ and $t<\tau_0$ are, in fact, impossible. This is the hypothesis which leads us to the parallel-resistor model. It is a hypothesis which seems to be physically justified. At least in a classical limit it is valid for both particles and waves. Indeed, if we consider a billiard ball scattering off a collection of other fixed billiard balls separated from each other by a distance $\gtrsim a$, then the existence of a minimum free path $\sim a$ is obvious. Likewise a wave of wavelength λ incident upon a collection of scattering centers will not distinguish between the two centers which are located closer than distance λ to each other. Hence, once scattered, the wave will propagate at least a distance λ before it is scattered again. For conduction electrons in a metal $\lambda=\lambda_F\sim a$ and hence $l>a$. Secondly, an important question is can we use such a concept as free time between collisions or even a semiclassical approach in general when τ approaches $\tau_0\sim a/V_F$? We believe that the answer is yes. It was already mentioned that both Kubo-Greenwood and Boltzmann formulations lead to essentially the same result for σ_{ideal}.[3] This makes one think that concepts associated with Boltzmann formulation work surprisingly well down to $t\sim\tau_0$. To further substantiate this view, let us consider materials for which detailed band-structure calculations are available, such as Nb_3Sn, Nb_3Ge, V_3Si, and Nb. Let us then estimate ρ_{sat} by substituting $\tau_0=a/\langle V(E_F)\rangle$ in place of τ. Following Allen *et al.*[16] we write $\rho=4\pi/\Omega_p^2\tau$ where Ω_p is the Drude plasma frequency $\Omega_p^2=(4\pi e^2/3)N(E_F)\langle V^2(E_F)\rangle$. Using band parameters of Klein *et al.*[17] for $A15$ compounds we obtain $\rho_{\text{sat}}=136\ \mu\Omega$ cm for Nb_3Sn, 160 $\mu\Omega$ cm for Nb_3Ge, and 122 $\mu\Omega$ cm for V_3Si. With the parameters for Nb calculated by Allen[18] we get $\rho_{\text{sat}}=110$ $\mu\Omega$ cm.

Experimentally found values for $A15$ compounds lie between 120 and 160 $\mu\Omega$ cm,[6–10,19] and in Nb ρ_{sat} was found to be 230 $\mu\Omega$ cm.[6] We see a rather impressive agreement between experimental and calculated values, especially for $A15$'s. Even if agreement is somewhat fortuitous (for we do not really expect that $\tau\sim\tau_0$ should mean precisely $\tau=\tau_0$), it supports the view that the Boltzmann formulation predicts correct values of resistivity for scattering times as short as τ_0.

We note that the reasoning used in the derivation of (3) applies to the electronic thermal conductivity K as well. Hence we expect to find $K=K_{\text{ideal}}+K_{\text{sat}}$ in the same class of metals where resistivity saturation is observed: Not a surprise, perhaps, for it follows also from the Wiedermann-Franz law $K=L\sigma T$. Note that in a "good" metal (one with long mfp), at $T>\theta_D$, $\tau\propto 1/T$ and $K=K_{\text{ideal}}=\pi^2K_B^2nT\tau/3m$ is independent of temperature, as experiment confirms. However, in a saturated regime, $K=K_{\text{sat}}=\text{const}\cdot T$. Substituting σ_{sat} from (1) and $L=\pi^2/3(K_B/e)^2$ into the Wiedemann-Franz law we obtain $\text{const}=1.09a_0K_B^2n^{2/3}/\hbar$. We speculate that lattice thermal conductivity, which is proportional to the mfp of phonons, will also saturate in disordered systems or at high temperatures. The saturation value should correspond to the minimum free path of phonons, which is likely to be of the order of phonon wavelength, i.e., $\sim a$ at high temperatures and $\sim\theta_D^a/T$ at $T<\theta_D$. In fact there are reports of the anomalously large thermal conductivity in disordered alloys.[20]

Arguments used above in the derivation of (3) are very general. They can be applied to a metal with a single band as well as to one with many bands. This can not be said about other treatments of the parallel-resistor model which depend on the presence of many bands[15] and *s-d* scattering.[21]

Finally, Mooij found that when ρ approaches ρ_{sat}, quite universally $d\rho/dT$ becomes negative, i.e., conduction becomes "activated."[5] This phenomenon probably reflects new (non-Boltzmann) physics appearing when l approaches a and, as was argued by Girvin and Jonson[22] and by Imry,[23] can be due to incipient Anderson localization. Even be-

fore this theoretical work appeared, Mooij[5] expressed the idea that partial localization may be the cause of negative $d\rho/dT$ in disordered alloys. This behavior can be formally included in the parallel-resistor model by allowing for the number of nonlocalized electrons n in (2) to decrease at low temperatures. This will result in a temperature-dependent ρ_{sat} and $d\rho/dT < 0$.

In conclusion, we argue that the particular way in which metals approach the saturated regime can be understood on the basis of the Ioffe-Regel criterion. The parallel-resistor formula (3) which describes this approach can be derived and a simple physical picture gained when the whole distribution of free times between collisions is considered and the minimum time τ_0 is introduced. In particular, it becomes clear that the validity of (3) is based on the fact that a relatively large number of electrons always have short free paths (times) and therefore conduction is sensitive to the details of the distribution in that region.

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