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Lorentz Corrections in Electrical Conductivity

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In a medium consisting of a spatially uniform source of scattering supplemented by randomly distributed localized scatterers, the current distribution is spatially nonuniform. The nonuniformity arises from the fact that the current flow suffers a detour near an obstacle to the extent that the localized scatterer's cross section exceeds that of the equivalent volume of background material. Thus a typical obstacle will be exposed to an incident current which has been increased by the "detour" currents of the other scatterers. This increase in the local current incident on an obstacle results in a contribution to resistivity which is nonlinear in both the density of the obstacles and in their scattering cross sections.

INTRODUCTION

The Lorentz correction in dielectric theory arises from the fact that a polarizable atom sees an electric field which is different from the simple space average of the electric field. The polarizable atom under consideration sees this different local field because it is not at an average position but guaranteed to be at a position outside that of the other polarizable atoms. When we consider the typical residual resistivity process in metallic conductors, we find a similar situation. Usually, the resistance arises from a random array of impurities with localized potentials in addition to a uniform background of thermal scatterers. A typical impurity is guaranteed to be outside of all other impurities. We can then expect the current incident on an impurity to be modified by the current-flow distortions produced by other localized scatterers. The purpose of this paper is to point out the general form of such a correction, using an admittedly very simple physical picture.

The ordinary Lorentz correction leads to a dielectric constant which is nonlinear in atomic concentration and polarizability. In the conductivity problem we can expect a resistance which is nonlinear in scatterer concentration and in scattering cross section. Before launching into the discussion of the conductive Lorentz correction, we would like to remind the reader that there is another source for a nonlinear dependence of resistivity on the scattering cross section of a localized obstacle. This is made most clear by reference to the case of a single plane obstacle, such as a grain boundary, or a region which has to be traversed by tunneling. In the case of a barrier reflecting most of the carriers incident on it, it has always been apparent[1] that current flow and conductivity are proportional to transmission probabilities. This gives a resistance which varies as $1/(1-r)$, with the reflection probability r. For weak scatterers, on the other hand, the resistance must be proportional to the scattering rate r. It is not surprising that more complete analyses[2,3] yield a dependence of resistivity on $r/(1-r)$. The denominator $(1-r)$ insures that the resistance goes to infinity as the obstacle reflectivity approaches unity, despite the fact that the scattering rate remains finite.

This nonlinearity with r is not just an isolated and anomalous feature of the plane infinite barrier. It has been pointed out by one of us[3,4] that a single obstacle localized in three dimensions also gives a resistivity contribution which is nonlinear in its scattering cross section, though the nonlinearity is clearly less pronounced in the case of point scatterers. This particular nonlinearity, unlike the Lorentz correction, is a characteristic of an isolated localized obstacle and its interaction with the otherwise uniform environment. It results from replacing the total cross section in a nonscattering medium by that in a medium with scat-

tering. In the language of Green's functions, this corresponds to using propagators which take into account the scattering property of the medium in the calculation of the single-particle self-energy. We note that the above remarks are based on the assumption that the range of the scatterers is much smaller than the mean free path of an electron in the uniform medium.

LORENTZ CORRECTION

In this section, we consider the nonlinear dependence of conductivity on impurity concentration arising from the fact that no more than one impurity can occupy a given site. Consider an impurity added to a system in a random location. The current incident upon it, averaged over all such possible locations, is equal to the average current in the impurity-free region.

Consider a piece of metal connected to a constant-current generator. We assume that there is a uniform background scattering (say, from phonons) σ_0 per atom, so that the resistivity in the absence of impurities is

$$\rho = \frac{3\pi^2 n_0 \hbar}{k_F^2 e^2}\ \sigma_0 \quad , \tag{1}$$

where n_0 is the density of atoms and k_F is the Fermi wave number. (We assume a spherical Fermi surface.) In the absence of impurities, the current density is uniform with magnitude j_0.

Now, introduce a density n' of impurities. Assume the range of the impurity potential is small compared to the electronic mean free path. Let σ' ($> \sigma_0$) be the scattering cross section of one of these impurities in the presence of the background scattering. The current density is now no longer uniform. Each impurity will redirect some of the incident current and the current will be taken around the impurity in a fashion described in detail in earlier papers.[3,4] These earlier papers also point out that the *additional field* resulting from the localized scatterers, in the presence of a constant current flow into the sample, consists of a superposition of dipole fields centered about each of the scatterers. Let us assume that the velocity distribution incident on the scatterer deviates from equilibrium by an amount proportional to $\cos\theta$, where θ is the angle between an incident velocity class and the direction of current flow. This will be the case if the scattering can be characterized by a simple relaxation time. The current J_s scattered by each impurity is then

$$J_s = j_e(\sigma - \sigma_0) \quad , \tag{2}$$

where j_e is the (average) local incident current density, and the scattering cross sections are calculated in the usual transport-theory way, weighing the collisions by $(1-\cos\alpha)$, where α is the scattering angle. Note that j_e differs from j_0 for the reasons cited earlier.

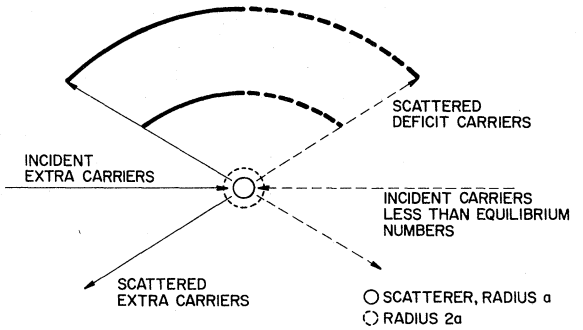

FIG. 1. Symbolic representation of current disturbance near a localized scatterer. After scattering by the obstacle, the carriers typically move a mean free path l in unscattered flight, and then are subject to a more macroscopic flow, as indicated by the heavy arcs.

In the vicinity of an obstacle, the current flow is disturbed. The total current

$$j = j_i + j_s$$

consists of the incident current j_i plus the effects due to the scattering, shown symbolically in Fig. 1. Here j_s denotes the deviation in current flow from the incident current pattern. Since the current across any plane not parallel to the direction of j_i in Fig. 1 must be the total incident current, we must have

$$\int j_s d\tau = 0 \quad , \tag{3}$$

where the integral extends over the whole sample. It now becomes convenient to break the whole sample into two regions which are separated by the dotted line in Fig. 1. This is a sphere with twice the obstacle radius. The center of another obstacle, exposed to the current disturbances j_s of the obstacle shown in Fig. 1, must be outside of this dotted line. If within the crude approximations of this discussion we identify the current incident on an obstacle with the current incident on its center, then we are interested in the current j_s outside of the dotted line. We then have

$$-\int_I j_s d\tau = \int_O j_s d\tau \quad , \tag{4}$$

where I indicates an integration over the volume inside the dashed sphere of radius $2a$ and O indicates the volume outside of this sphere.

Figure 1 shows that the current due to the localized scattering, $j_e(\sigma' - \sigma_0)$, is carried around the obstacle. If we assume that the mean free path l is large compared to the scatterer radius a, then the scattered "extra" and "deficit" carriers shown in Fig. 1 as emanating from the obstacle volume will almost all pass through the dashed line of radius $2a$. Thus j_s carries a current $j_e(\sigma' - \sigma)$

around the outside of the dotted sphere. The exact value of the right-hand term in Eq. (4) depends on which part of the scattered current passes through which part of the sphere of radius $2a$. As an order-of-magnitude approximation, however, the current typically gets carried a distance $2a$ in going from the left-hand side of the sphere of radius a to the right-hand side. Thus

$$\int_O j_s d\tau = j_e(\sigma' - \sigma_0)\eta d \ , \tag{5}$$

where η is an undetermined coefficient of order unity and we have written d for $2a$. An exact calculation of η from the existing semiclassical theories[3,4] would be relatively meaningless in any case. Unfortunately, at this time, a quantum-mechanical version of this viewpoint exists only for the one-dimensional case.[5] [A direct uncritical application of Eq. (4.9) of Reference 3 does yield $\eta = 1$.]

If we now consider a density n' of scattering centers, an additional center inserted into the space outside of all the dotted spheres will see an average current increment, due to all the j_s contributions from the other scatterers, of $n' j_e(\sigma' - \sigma_0) \times \eta d$. Thus

$$j_e = j_0 + n' j_e(\sigma' - \sigma_0)\eta d \ . \tag{6}$$

Solving for j_e, we find

$$j_e = \frac{j_0}{1 - n'(\sigma' - \sigma_0)\eta d} \ . \tag{7}$$

Without the Lorentz correction we have a resistivity:

$$\rho = \frac{3\pi^2\hbar}{k_F^2 e^2}\left[n_0\sigma_0 + n'(\sigma' - \sigma_0)\right] \ . \tag{8}$$

The Lorentz correction increases the current incident on the extra scattering power, $\sigma' - \sigma_0$, and therefore raises the final right term in Eq. (8), giving

$$\rho = \frac{3\pi^2\hbar}{k_F^2 e^2}\left(n_0\sigma_0 + \frac{n'(\sigma' - \sigma_0)}{1 - n'(\sigma' - \sigma_0)\eta d}\right) \ . \tag{9}$$

The result in Eq. (9) can alternatively be pictured in terms of the model in which the extra potential due to each localized scatterer consists of a dipole field around each localized scatterer[3,4] which is proportional to the localized scattering power $(\sigma' - \sigma_0)$ and the local current j_e.

COMMENTS

The specific expression given in Eq. (9) is valid for a degenerate spherically symmetric Fermi gas. The factor

$$\frac{1}{1 - n'(\sigma' - \sigma_0)\eta d} \ , \tag{10}$$

by which the local current is increased, however, has a broader validity and does not depend, for example, on the degeneracy of the Fermi gas. This expression only assumes that there is a highly localizable source of scattering which reroutes the current around the obstacle involved. The long-range screened Coulomb fields, typical of impurities in semiconductors, do not meet this qualification.

Experimental comparisons will be difficult. As we increase the concentration to bring the correction factor (10) away from unity we will be likely modifying many other aspects of the material, e.g., carrier densities. Thus there are other sources of nonlinearity in the dependence of resistivity on obstacle density.

One place where the Lorentz correction can play a role is in electromigration.[6,7] In the presence of current flow, carriers pass their momentum preferentially on to localized scatterers, giving the scattering centers a drift velocity. The Lorentz factors will enhance this effect. The nonuniformity of the associated electric field[3,4] may also be relevant, if there are any direct electrolytic forces. The existence of direct electrolytic forces on an impurity screened by a dense and highly mobile electron gas is, however, open to questions.

In dielectric theory the Lorentz factor can diverge, and this has on occasion been used to explain ferroelectricity. Expression (10) can diverge if we have a relatively dense ($n'a^3 \sim 1$) obstacle array, and if the scatterers are relatively impenetrable, i.e., $\sigma' - \sigma_0 \sim a^2$. Whether the expression really can have a pole or not will depend on the exact value of η. This tendency for the Lorentz factor to become very large, however, is reasonable and expected. As we fill space more and more with relatively impenetrable scatterers, the resistance must go to infinity. This obvious effect is, however, not represented correctly by the usual elementary impurity-resistance equations of the form of Eq. (8), and demonstrates the need for the Lorentz correction. The point where the resistance goes to infinity (in a rigorous theory, not necessarily in our approximation) may in fact occur somewhat before the obstacles are dense enough to fill the sample completely and block all carrier motion. Before this point is reached, the passages around the obstacles may, in typical percolation-theory fashion,[8] cease to provide a completely continuous path through the whole sample, even though they still permit some local motion.

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Higher-Derivative Photoemission Spectra and the Relativistic Band Structure of Gold

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Following a prescription by Ehrenreich and Hodges, a relativistic band structure for Au has been constructed by the insertion of spin-orbit coupling into an originally nonrelativistic interpolation scheme. The bands are used to compute the energy distribution of the joint density of states $D(E, \hbar\omega)$ and its second derivative with respect to energy $D''(E, \hbar\omega)$. Peaks in $-D''(E, \hbar\omega)$ are found to correlate quite well in energy location with structure in experimental higher-derivative photoelectron-energy spectra measured on cesiated Au. The comparison is made with previous data and with supplementary spectra presented here. Profile changes in the experimental spectra on varying the photon energy $\hbar\omega$ are also reasonably well accounted for. The density of states computed from the same band structure shows d-band peaks at 2.65, 4.00, 4.50, 5.05, 6.35, 6.85, and 7.60 eV below the Fermi level. From the derivative spectra at the lower photon energies, the $L_6^-(L_{2'}) \rightarrow L_6^+(L_1)$ band gap in Au is estimated to be 4.0 eV.

I. INTRODUCTION

Several relativistic band calculations on Au have been performed recently,[1-3] and they confirm that relativistic effects are quite large in a heavy element such as Au. Experimental information from optical and photoemission studies is now sufficiently detailed that the relativistic effects should be taken into account in any realistic interpretation. Christensen and Seraphin[1] have laid particular emphasis on this point, and it has been considered also by Kupratakuln.[2] Indeed, an attempt made in an earlier paper by the author[4] to interpret the photoemission spectra of Au using a nonrelativistic band structure was significantly less successful than similar analyses on the lighter metals Cu and Ag.

This paper presents a reanalysis of the photoemission data from Au using an interpolated band structure into which spin-orbit coupling has been inserted. It is assumed that the relativistic effects other than spin-orbit coupling (mass-velocity and Darwin terms) can be absorbed into the parameters of the original nonrelativistic interpolation scheme. In fact, we follow very closely the prescription set down by Ehrenreich and Hodges.[5] The interpolated band structure is used to compute the energy distribution of the joint density of states (EDJDOS) and its second derivative with respect to energy. These results are then compared with higher-derivative photoelectron-energy spectra taken on cesiated Au. It is found that the inclusion of spin-orbit splitting does bring about a significant improvement in agreement with experiment and permits a more reliable identification of structure. Good agreement for photon energies greater than about 9.0 eV has been reported also by Christensen,[6] who has computed the EDJDOS from a relativistic augmented-plane-wave (APW) calculation.

New higher-derivative photoelectron-energy spectra on cesiated Au are presented here in order to supplement those presented in the earlier paper,[4] particularly in the lower photon-energy region. The new spectra show some interesting profile changes on varying the photon energy. It is shown that these are also consistent with the predictions of the interpolated band structure.

II. BAND STRUCTURE OF GOLD

A. Interpolation Scheme

The band structure used here is obtained from the interpolation scheme of Hodges, Ehrenreich, and Lang[7] (HEL). As pointed out by them and by Ehrenreich and Hodges,[5] the original nonrelativistic HEL scheme can be converted into a relativistic scheme in an approximate manner by expressing the model Hamiltonian as follows: