Residual-resistivity dipole in electron transport and electromigration

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The structure of Landauer's residual-resistivity dipole is investigated using expressions which have been recently derived from linear-response theory. We calculate the local field and charge density near an impurity in a current-carrying metal. The impurity is assumed to be an isotropic scatterer and is described by an s-wave phase shift. We find that the long-range average field of the residual-resistivity dipole is exactly as obtained by Landauer in a semiclassical picture; i.e., the dipoles give the macroscopic field which drives the current past the scatterers. The charge distribution is not strongly localized, but has important Friedel oscillations extending far from the impurity. Our results are applied to the calculation of the electromigration driving force on a weak scatterer due to the presence of an s-wave scatterer in its vicinity. When the s-wave scatterer is strong, the residual-resistivity dipole contributes substantially to the force on the weak scatterer. Such contributions have not been previously considered in explicit calculations of scattering interference effects in electromigration.

I. INTRODUCTION

The residual-resistivity dipole (RRD) is a fundamental object in the theory of electronic conduction. Introduced into the theory by Landauer[1] in 1957, the RRD is the microscopic source of the electric field which must be set up in a conducting solid in order to drive current past a region containing impurities. Despite its central importance, the RRD has been generally ignored in theoretical descriptions of electron transport. One usually assumes that the macroscopic electric field arises from sources external to the system. In reality, the field has internal sources in the form of dipolar charge distributions localized around each impurity. These are the RRD's.

Analyses of electrical resistivity based on the RRD picture have not given any new answers for the resistivity[2,3]; RRD's have thus far been of conceptual value rather than practical value. As Landauer[3] has pointed out, however, a correct picture of the local dipole field would be helpful in formulating a theory for scattering by inhomogeneities which are neither macroscopic nor point-defect-like. This alone would provide motivation for studying the RRD. Further, if one is interested in the detailed nature of the local field in electrical conductivity, the field of the RRD must be considered. Since the local field acts as the driving force on an ion in a solid,[4] one finds the RRD mentioned in recent theoretical papers on electromigration,[4-9] which is the phenomenon of ion transport in the presence of an electron current.[10]

A review of the RRD with special emphasis on electromigration has been given by Landauer and Woo.[8] In summary, Landauer found that around each impurity in a conducting solid there forms a localized dipolar charge distribution which he called the RRD.[1] If one denotes the dipole moment of an RRD by $\vec{p}_0$, then the average field generated by these RRD's in a rectangular sample is given by $-4\pi N\vec{p}_0$, where N is the number of impurities per unit volume. When the value of $\vec{p}_0$ calculated by Landauer is used in this expression, there results precisely the average additional field needed to drive a current past these impurities in the first place.[1] Thus, there is no true external charge which sets up the additional field; the field arises instead from the RRD charge distributions. It was further argued by Landauer and Woo[8] that the RRD is localized to within a screening length from the impurity. This strong localization led them to predict a large RRD force on the impurity.

Up to now there have not been any explicit calculations of the RRD charge or the RRD field.[11] Local-field calculations have been made within quantum-mechanical formalism[4,12] and semiclassical formalism.[13-15] These calculations went only to terms linear in the impurity potential V. Since the RRD is proportional to the cross section,[1,8] it will not show up unless the calculations are pushed to second order in V.

One reason for the lack of activity in calculating RRD fields is obviously the extra work required to go to the next order in perturbation theory. Moreover, if one is interested only in the force on an ion in electromigration one need not explicitly examine the RRD. Instead, it turns out that one can calculate the force to all orders in V if one takes the lowest-order perturbation-theory expression for the force and replaces V by the T matrix for the impurity.[6,7] This procedure is valid for isolated impurities; for more complicated scatterers, the force can also be expressed in terms of T matrices.[6,7] Thus, the RRD need not be explicitly singled out in calculating electro-

migration forces. Nonetheless, from a fundamental point of view, it is desirable to investigate the local RRD field near an impurity. Besides illuminating some hidden assumptions in transport theory, such an investigation will clarify the picture of Landauer and Woo[8] for electromigration and other transport phenomena.[2,3]

In this paper we present an explicit calculation of the RRD charge density and the associated local electric field around an impurity in a jellium background. The calculation is performed for an impurity which is predominantly an s-wave scatterer. Calculation of the local field is formally equivalent to determining the force on a weak scatterer in the vicinity of the impurity. Our results will thus be immediately applicable to the problem of determining the electromigration force on a weak scatterer due to interference effects caused by the presence of a strong s-wave scatterer in its neighborhood. No explicit calculation of interference effects in electromigration has appeared in the literature except for the case of only weak scatterers.[16] In a study of liquid metals we again used weak-scattering expressions, but followed heuristic arguments to replace V by the T matrix.[17] These procedures, as we discuss in Sec. IV, systematically discard RRD contributions. As a result of the present study we can estimate the errors involved.

As far as the structure of the RRD is concerned, we shall see that the long-range dipole field of the RRD is precisely as described by Landauer.[1] However, the actual charge distribution of the RRD is not so localized as assumed by Landauer and Woo.[8] Instead, we find the RRD extends over a large region where it is inextricably intertwined with Friedel oscillations.

II. LOCAL FIELDS AND CHARGE DENSITIES

We wish to obtain the response of an electron gas to an impurity in the presence of an applied electric field. The Kubo linear-response formalism has been applied to this problem[18] and has led to a better picture of the local fields and charge densities around the impurity.[6,7,11,18-20] For an impurity in jellium all of the linear-response results agree with each other and with Liouville-equation calculations in the relaxation-time approximation[4,19] in the regime $k_F l \gg 1$, where k_F is the Fermi wave vector and l is the mean free path.[21] This $k_F l \gg 1$ regime is the regime of physical interest for metals describable within a jellium model.

The diagrammatic Green's-function calculation of Sham[7] was the most general of the first linear-response calculations in that it explicitly included electron screening and strong scattering by the impurity. Further studies based on the diagrammatic technique have also been made.[11,19,20]

Within an independent-particle approximation these analyses lead to the physically appealing result that the local electron density $n_0(\vec{r})$ around an impurity can be obtained by simply populating the electron scattering states $\psi_{\vec{k}}(\vec{r})$ according to the distribution function $g_{\vec{k}}$ appropriate to the electron-transport problem. This result is explicitly derived by Schaich[6] and is also implicit in Sham's analysis.[7] The expression for the density is

$$n_0(\vec{r}) = \sum_{\vec{k}} g_{\vec{k}} |\psi_{\vec{k}}(\vec{r})|^2, \tag{1}$$

where the sum is over all states $\vec{k}$ and where $g_{\vec{k}}$ is the solution of the Boltzmann equation, namely

$$g_{\vec{k}} = -\tau e \vec{v}_{\vec{k}} \cdot \vec{E}_0 \delta(\epsilon_{\vec{k}} - \epsilon_F). \tag{2}$$

Here τ is the relaxation time, e is the charge of the proton, $\vec{v}_{\vec{k}}$ is the electron velocity, and $\vec{E}_0$ is the macroscopic electric field. The Fermi energy is denoted by ϵ_F and the energy of the state $\vec{k}$ is $\epsilon_{\vec{k}} = \hbar^2 k^2/2m$, where m is the electron mass. The wave function $\psi_{\vec{k}}(\vec{r})$ evolves from an incoming plane wave $\exp(i\vec{k}\cdot\vec{r})/\Omega^{1/2}$, where Ω is the crystal volume. In our model the electron propagates as a free particle, except for its interaction with the impurity. Equation (1) is of the form assumed by Bosvieux-Friedel[14] for the "dynamic polarization" due to the electron current, i.e., the "electron wind" effect. This has been pointed out by Schaich.

When the independent-particle picture is generalized to include a self-consistent treatment of the electron-electron interaction, the electron density is no longer given by Eq. (1). If screening is considered within the random-phase approximation (RPA) or, more generally, if local linear screening is assumed, one can show that the true density is not given by Eq. (1), but rather by

$$n(\vec{r}) = n_0(\vec{r}) + n_{sc}(\vec{r}), \tag{3}$$

where n is the true density and n_{sc} is the screening density. The Fourier transform of n_{sc} is related to n_0 according to the linear relation[22,23]

$$n(\vec{q}) = \frac{n_0(\vec{q})}{\epsilon(q)}, \tag{4}$$

where $\epsilon(q)$ is the dielectric function and will be assumed to be that given within the RPA.

The force $\vec{F}$ exerted on an ion, or any object which interacts with the electrons, can be written in the form[4,18]

$$\vec{F} = -\int n(\vec{r}) \frac{\partial v_0(\vec{r} - \vec{R})}{\partial \vec{R}} d^3r, \tag{5}$$

where R is the position of the ion and $v_0(\vec{r}-\vec{R})$ is the bare potential which describes the interaction of the electrons with the ion of interest. In Eq. (5) we are only considering the force arising from the local electron response $n(\vec{r})$. The force exerted by the macroscopic field $\vec{E}_0$ can be added on separately as $Ze\vec{E}_0$, where Ze is the charge of the ion. We are not concerned with this direct force here so we shall ignore it.

Using expression (4) one can transform Eq. (5) into the alternate form[23]

$$\vec{F}=\int n_0(\vec{r})\frac{\partial v(\vec{r}-\vec{R})}{\partial \vec{r}}d^3r, \tag{6}$$

where v is the screened potential. The Fourier transforms of $v(\vec{r})$ and $v_0(\vec{r})$ are related by the dielectric function, i.e., $v(\vec{q})=v_0(\vec{q})/\epsilon(q)$.

The local electric field, which we denote by $\delta\vec{E}$, can be immediately obtained by applying Eq. (6) to find the force on a test charge Q at position $\vec{R}$.[4] Taking the limit of $\vec{F}/Q$ as $Q\to 0$, this gives for the local field at position $\vec{R}$ the result

$$\delta\vec{E}(\vec{R})=\int n_0(\vec{r})\frac{\partial \phi(\vec{r}-\vec{R})}{\partial \vec{r}}d^3r, \tag{7}$$

where ϕ is the screened electrical potential. Within the RPA the transform of $\phi(\vec{r})$ is given by $\phi(\vec{q})=-4\pi e/q^2\epsilon(q)$. The total field at $\vec{R}$ is the sum of $\delta\vec{E}$ and the applied field $\vec{E}_0$.

III. MODEL CALCULATION

We consider the charge distribution around a highly-localized spherically symmetric impurity potential. The scattering wave functions can be expanded in partial waves and written in the form[24]

$$\psi_{\vec{k}}(\vec{r})=\frac{1}{\Omega^{1/2}}\left(e^{i\vec{k}\cdot\vec{r}}+\sum_{l=0}^{\infty}(2l+1)i^l P_l(\hat{k}\cdot\hat{r})\times[R_l(r)-j_l(kr)]\right), \tag{8}$$

where the P_l are the Legendre polynomials and their argument $\hat{k}\cdot\hat{r}$ is the cosine of the angle between $\vec{k}$ and $\vec{r}$. The impurity is taken to be centered at the origin. $R_l(r)$ is a radial solution of the Schrödinger equation and j_l is a spherical Bessel function. If we take the potential to be of muffin-tin form, the exterior solutions can be expressed in terms of the phase shifts δ_l as follows[24]:

$$R_l(r)=e^{i\delta_l}[j_l(kr)\cos\delta_l-n_l(kr)\sin\delta_l], \tag{9}$$

where n_l represents a spherical Neumann function. Expression (9) is valid for $r>r_{MT}$, where r_{MT} is the muffin-tin radius. It is apparent from Eqs. (1) and (2) that we are interested in the case $k=k_F$, and thus the δ_l are the phase shifts at the Fermi energy.

We now restrict our attention to impurities which scatter electrons isotropically, i.e., s-wave scatterers. (An example of such an impurity is hydrogen in copper.[25]) Since now only the $l=0$ phase shift is appreciable, the sum over l in Eq. (8) contains only the $l=0$ term. The wave function in the exterior region ($r>r_{MT}$) then becomes

$$\psi_{\vec{k}}(\vec{r})=\frac{1}{\Omega^{1/2}}[e^{i\vec{k}\cdot\vec{r}}+\psi^s_{\vec{k}}(r)], \tag{10}$$

where the scattered wave $\psi^s_{\vec{k}}(r)$ is given by

$$\psi^s_{\vec{k}}(r)=j_{l=0}(kr)(e^{i\delta}\cos\delta-1)-n_{l=0}(kr)e^{i\delta}\sin\delta, \tag{11}$$

and δ denotes the $l=0$ phase shift.

We can now evaluate $n_0(\vec{r})$ using expressions (1), (2), (10), and (11). The result is

$$n_0(\vec{r})=\frac{2k_F^2\tau eE_0\cos\theta}{\pi^2}[\Gamma_{BF}(\rho)+\Gamma_{RRD}(\rho)], \tag{12}$$

where θ is the angle between $\vec{E}_0$ and $\vec{r}$. The functions Γ_{BF} and Γ_{RRD} will be seen to describe the Bosvieux-Friedel (BF) dipole[14] and the RRD, respectively. They are given by

$$\Gamma_{BF}(\rho)=-\frac{1}{2\rho^2}\left(\frac{1}{\rho}-\sin 2\rho-\frac{\cos 2\rho}{\rho}\right)\sin\delta\cos\delta \tag{13}$$

and

$$\Gamma_{RRD}(\rho)=\frac{1}{2\rho^2}\left(1+\cos 2\rho-\frac{\sin 2\rho}{\rho}\right)\sin^2\delta. \tag{14}$$

The argument ρ is k_Fr. The above expressions are valid for $r>r_{MT}$. Since we can, in principle, imagine r_{MT} to be of arbitrarily short range, ρ can also be taken to be arbitrarily small. In the small-ρ limit, $\Gamma_{BF}\to(-\rho/3)\sin\delta\cos\delta$ and $\Gamma_{RRD}\to-\frac{1}{3}\sin^2\delta$.

In the limit of weak scattering ($\delta\ll 1$), Γ_{BF} is proportional to δ and hence proportional to the impurity potential strength. This is precisely the behavior of the dipolar distribution originally derived by Bosvieux and Friedel.[14] Thus Γ_{BF} describes the BF dipole. Γ_{RRD}, on the other hand, is proportional to $\sin^2\delta$, and hence to the cross section. This is characteristic of Landauer's RRD.[1] For large r there is a nonoscillatory component of Γ_{RRD}, namely the $\frac{1}{2}\rho^2$ term in expression (14). This leads to an asymptotic $n_0(\vec{r})$ which is consistent with a general asymptotic RRD expression derived by Schaich.[6,11] Further discussion of the BF vs RRD separation is given at the close of Sec. IV.

Plots of $\Gamma_{RRD}(\rho)$ and $\Gamma_{BF}(\rho)$ are shown in Fig. 1. We note that there is no special behavior near the origin associated with Γ_{RRD}. For small ρ the $\frac{1}{2}\rho^2$ term which gave the asymptotic RRD behavior

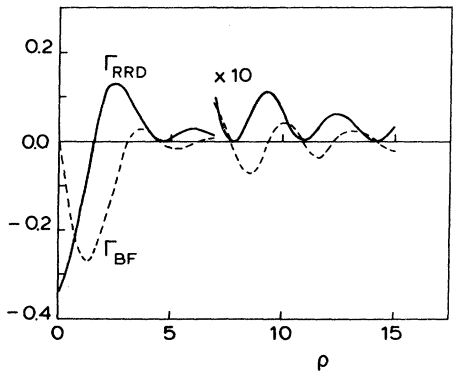

FIG. 1. Quantities Γ_{RRD} and Γ_{BF} vs ρ, where $\rho = k_F r$. The actual values of Γ_{RRD} and Γ_{BF} are obtained from the displayed values by multiplication by $\sin^2\delta$ and $\sin\delta\cos\delta$, respectively. Γ_{RRD} and Γ_{BF} are related to the unscreened density $n_0(\vec{r})$ by Eq. (12). They are related to the local electrical potential by Eqs. (12) and (25).

in Γ_{RRD} has been analytically absorbed by the Friedel oscillations of the other terms.[26]

Having obtained an expression for $n_0(\vec{r})$, we can determine the true electron density $n(\vec{r})$ by means of the Fourier-transform expression (3). For simplicity we evaluate $n(\vec{r})$ in the regime that the Thomas-Fermi screening length is shorter than other relevant lengths in the problem. In principle, this can be a valid regime. In practice, however, the screening length is not much shorter than the electron wavelength. We return to this point in Sec. IV. The Thomas-Fermi dielectric function is particularly simple in the regime of small screening length: It reduces to $\epsilon(q) = \alpha^2/q^2$, where α is the Thomas-Fermi wave vector. For a parabolic band of electrons, α equals $(4mk_F e^2/\pi\hbar^2)^{1/2}$, showing that a small screening length $1/\alpha$ can always be arranged, in principle, by appropriate choice of k_F and the effective mass m.

Using this $q \to 0$ form of the Thomas-Fermi dielectric function $\epsilon(q)$, we deduce from Eq. (4) that

$$n(\vec{r}) = -\alpha^{-2}\nabla^2 n_0(\vec{r}). \tag{15}$$

Upon evaluating $\nabla^2 n_0(\vec{r})$ with $n_0(\vec{r})$ given by Eq. (12), we find

$$n(\vec{r}) = -\frac{2k_F^2 \tau e E_0}{\alpha^2 \pi^2}\left(\frac{\cos\theta}{r^2}\right)[G_{BF}(\rho) + G_{RRD}(\rho)], \tag{16}$$

where

$$G_{BF}(\rho) = \left[-\frac{2}{\rho^3} + \left(\frac{2}{\rho^3} - \frac{4}{\rho}\right)\cos 2\rho + \left(\frac{4}{\rho^2} - 2\right)\sin 2\rho\right]\sin\delta\cos\delta \tag{17}$$

and

$$G_{RRD}(\rho) = \left[\left(\frac{4}{\rho^2} - 2\right)\cos 2\rho + \left(\frac{4}{\rho} - \frac{2}{\rho^3}\right)\sin 2\rho\right]\sin^2\delta \tag{18}$$

correspond to BF and RRD contributions, respectively. The results are plotted in Fig. 2. Note that for small ρ, $G_{BF}/r^2 \to 0$ while $G_{RRD} \to \frac{2}{3}\sin^2\delta$.

Of vital importance in discussions of the RRD, especially as it applies to electromigration,[8] is the spatial extension of the RRD. To obtain this we calculated the dipole moment of the true electron distribution associated with the RRD. The dipole moment p of all charge contained within a radius r is given by

$$p(r) = -e\int u(r - r')n(\vec{r}')z'd^3r', \tag{19}$$

where u is the step function defined by $u(x) = 1$ for $x > 0$ and $u(x) = 0$ for $x < 0$. The dipole moment is along the z direction, which is the direction of $\vec{E}_0$.

The RRD dipole moment contains only the G_{RRD} contribution to $n(\vec{r}')$ in Eq. (19). The result for the RRD dipole moment is

$$p(r) = p_0\left[\tfrac{1}{2} + \tfrac{5}{6}\cos 2\rho - \frac{1}{3}\left(\frac{2}{\rho} - \rho\right)\sin 2\rho\right], \tag{20}$$

where

$$p_0 = -\frac{2m\tau E_0}{k_F}\sin^2\delta \tag{21}$$

would be the value of the dipole moment for the RRD given by Landauer if s-wave scattering is assumed in Landauer's expression.[1] The quantity $p(r)$ is plotted in Fig. 3. We note that for $r \to \infty$,

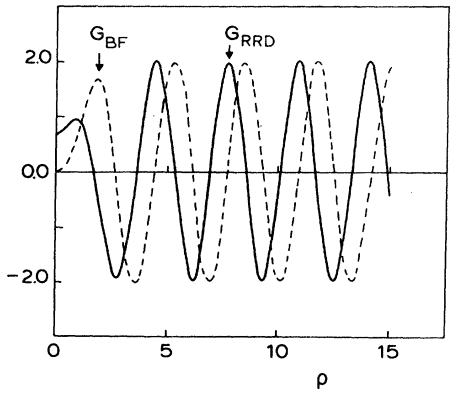

FIG. 2. Quantities G_{RRD} and G_{BF} vs ρ. These quantities are related to the screened density $n(\vec{r})$ by Eq. (16). As in Fig. 1, the values displayed do not include the $\sin^2\delta$ and $\sin\delta\cos\delta$ factors for the RRD and BF quantities, respectively.

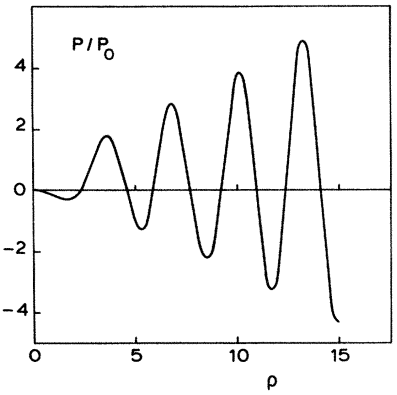

FIG. 3. Plot of the RRD dipole moment for the screened density $n(\vec{r})$. The quantity p/p_0 is the dipole moment for all charge up to radius ρ and normalized to the value p_0, which is the value of the RRD dipole moment calculated by Landauer.

our $p(r)$ diverges as $r\sin 2k_F r$. The conclusion is that there is no localized dipole moment associated with the RRD distribution.

We now calculate the local field $\delta\vec{E}$ according to Eq. (7). The electrical potential ϕ to be used in that equation is the Thomas-Fermi potential $\phi(\vec{r}) = -(e/r)\exp(-\alpha r)$. It is convenient to integrate Eq. (7) by parts and obtain for the local field the result

$$\delta\vec{E}(\vec{R}) = -\int \phi(\vec{r}-\vec{R})\frac{\partial n_0(\vec{r})}{\partial\vec{r}}d^3r. \tag{22}$$

Again, for simplicity, we assume that the screening length $1/\alpha$ is much smaller than the scale of variation of $n_0(r)$. Equation (22) immediately reduces to

$$\delta\vec{E}(\vec{R}) = \frac{4\pi e}{\alpha^2}\frac{\partial n_0}{\partial\vec{r}}\bigg|_{\vec{r}=\vec{R}}. \tag{23}$$

Equation (23) implies that the local electrical potential is proportional to n_0. The local potential Φ at any position is related to the local field at that position according to the defining equation

$$\delta\vec{E}(\vec{r}) = -\nabla\Phi(\vec{r}), \tag{24}$$

where $\nabla = \partial/\partial\vec{r}$ and where we have now used the variable $\vec{r}$ rather than $\vec{R}$ to denote a general point in space where the fields are to be calculated. It follows from Eqs. (23) and (24) that

$$\Phi(\vec{r}) = -4\pi e\alpha^{-2}n_0(\vec{r}). \tag{25}$$

Thus, the quantities Γ_{BF} and Γ_{RRD} appearing in Eqs. (12)–(14) and displayed in Fig. 1 are actually the BF and RRD components of the local potential field. For $r\to\infty$ the average field is clearly dominated by the term $(\frac{1}{2}\rho^2)\sin^2\delta$ in Γ_{RRD} since the term involving $\cos 2\rho$ averages to near zero over a volume containing several electron wavelengths. When the leading asymptotic term in Γ_{RRD} is used in Eq. (25), it is easily seen that the resulting potential field is precisely the electrostatic field arising from a dipole whose value is $p_0\hat{z}$, where p_0 is defined by Eq. (21) and $\hat{z}$ is a unit vector in the direction of $\vec{E}_0$. This verifies Landauer's picture for the long-range average field of the RRD.

It is of interest to have an explicit expression for the electric field $\delta\vec{E}$. The component of the local field in the z direction is easily obtained from Eqs. (24) and (25) and the $n_0(\vec{r})$ expression (12). We find

$$\delta\vec{E}_z = \frac{8ek_F^3\tau E_0}{\pi\alpha^2}\left(\frac{\Gamma(\rho)}{\rho}\sin^2\theta + \frac{d\Gamma(\rho)}{d\rho}\cos^2\theta\right), \tag{26}$$

where in Eq. (26) the RRD field is obtained by inserting Γ_{RRD} for Γ, while the BF field is obtained by inserting Γ_{BF} for Γ. The results for Γ/ρ and $d\Gamma/d\rho$ are plotted in Fig. 4. The BF field is regular for $r\to 0$. There is an apparent divergence in $\Gamma_{RRD}(\rho)/\rho$ since $\Gamma_{RRD}(0)$ does not vanish. However, recall that our expressions apply for $r \geq r_{MT}$ only. Inside the muffin tin the interior solutions $R_l(r)$ are well behaved and δE_z is, of course, finite.

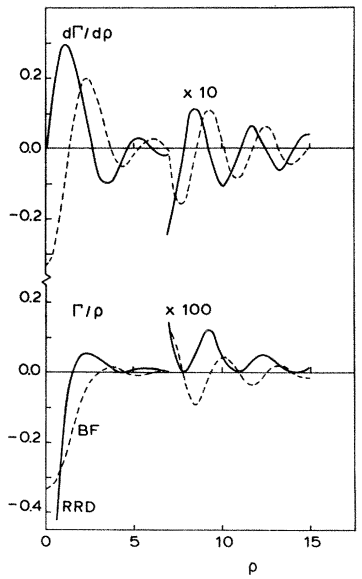

FIG. 4. Quantities $d\Gamma/d\rho$ and Γ/ρ vs ρ, showing RRD and BF contributions. These quantities are related to the local electric field by Eq. (26). As in Fig. 1, the values displayed do not include the $\sin^2\delta$ and $\sin\delta\cos\delta$ factors for the RRD and BF quantities, respectively.

IV. DISCUSSION

We have evaluated the microscopic field and charge density around an s-like impurity scatterer in a jellium background. The field given by Eq. (26) gives precisely the asymptotic behavior obtained by Landauer in a semiclassical analysis.[1] A superposition of such dipole fields gives the extra field needed to drive electrons past a region containing impurities.

If one considers a conductor in which the only source of scattering is impurity scattering, then one is led to the conclusion that there is, in fact, no true external field. Rather, there is only a self-consistent field internally generated by a superposition of RRD's. It is this superposition which yields the macroscopic electric field in a metal. To a certain extent, then, "exact" quantum-mechanical derivations of transport phenomena are less than rigorous since in such treatments an *external* field is taken as the perturbation in the system Hamiltonian. (This, of course, was also assumed here since the expressions used in Sec. II were based on the "exact" Kubo formalism.)

As we have discussed elsewhere,[4] there are standard tricks to short-circuit the internally generated RRD field. Among these tricks are the assumption of toroidal geometry, periodic boundary conditions, or an "infinite medium," i.e., a medium where average dipole fields are taken to vanish by "symmetry." None of these methods is free of objections.[4,7] Nonetheless, the procedure used here and outlined elsewhere[4] is sensible. First we focus on a small region (within a mean free path) around the impurity. Everything outside this region is treated as setting up an external field within the region. This enables us to obtain a local field using Kubo theory. Finally, we find that our local field summed over all impurities is precisely the field assumed in the first place. This closes the self-consistency loop. Although this procedure gives no new results for the conductivity in a homogeneous medium, it does imply that great care is needed in treating inhomogeneous media when the scale of the inhomogeneity is comparable to a mean free path.

Perhaps the most surprising feature of our calculation is the elusive nature of the RRD charge distribution. It is not localized within the order of screening lengths or electron wavelengths. Instead, the RRD is hidden, chameleonlike, within the Friedel oscillations which extend to large distances around the impurity. At large distances it *appears* as though the average RRD field is emanating from a dipole at the origin. If one were to ignore all oscillatory terms involving $\cos 2\rho$ and $\sin 2\rho$ in Eqs. (13) and (14), then it would also appear as if the RRD were a dipole of strength p_0 at the origin. But this is misleading since there is no RRD singularity at the origin; the oscillatory terms combine with the would-be singular term to give smooth behavior with no localized dipole near the impurity. This is seen in the figures.

We note from expression (26) and Fig. 3 that the field of the RRD near the impurity is opposite to the external field. Also, the true charge density $-en(\vec{r})$ due to the RRD near the impurity is in the opposite sense to that expected for a localized dipole moment $p_0\hat{z}$ near the impurity. The BF field near the impurity is opposite to $\vec{E}_0$ for an attractive potential ($\delta > 0$) and in the direction of $\vec{E}_0$ for a repulsive potential ($\delta < 0$). The BF force on the impurity is thus always in the direction of the electron wind. It is clear from the figures that the structure in the RRD and BF fields cannot simply be attributed to localized dipoles. The Friedel oscillations are an essential part of the near- and far-field structure.

We now discuss implications for the driving force in electromigration. Our results apply to the calculation of the force exerted on a weak, very localized scatterer due to the presence of a strong s-like scatterer in its vicinity. This force corresponds to an interference effect between two scatterers and has thus far been calculated only from weak-scattering expressions.[16,17]

The force on the weak scatterer can be simply obtained from Eq. (6) by first integrating by parts. The result, analogous to Eq. (22), is

$$F_z = -\int V(\vec{r}-\vec{R})\,\frac{\partial n_0(\vec{r})}{\partial z}\,d^3r, \tag{27}$$

where V is the (screened) potential of the weak scatterer. We now assume a very localized scatterer modeled by a delta-function potential $V = A\delta(\vec{r}-\vec{R})$, where A is the strength parameter. With this form Eq. (27) becomes

$$F_z = -\frac{A\alpha^2}{4\pi e}\,\delta E_z, \tag{28}$$

where δE_z is the local field at the position of the weak scatterer and is given by Eqs. (23) or (26). Equation (28) gives the force on the weak scatterer correct to linear order in the strength of the weak scatterer. The discussion we have given for δE_z (see Fig. 3) thus applies to the force F_z.

We comment on the apparent large RRD contribution to the force F_z and the field δE_z as $r \to 0$. This behavior arises from the $1/\rho$ divergence in the term $\Gamma_{\mathrm{RRD}}/\rho$ in Eq. (26). The divergence appears in the RRD term and not the BF term because in the exterior solution given by Eq. (11)

the "singular" $n_{l=0}$ term contributes to Γ_{RRD}, while the "well behaved" $j_{l=0}$ term contributes to Γ_{BF}. If we wish to claim that $r \to 0$ is a physically relevant domain for our expressions, we must also choose $r_{MT} \to 0$ since we have used solutions valid for $r > r_{MT}$ only. The condition $r_{MT} \to 0$ and the condition that only s-wave scattering is appreciable places severe restrictions on the impurity potential and the resulting value of δ. For example, to satisfy these conditions for a spherical-well model potential we need a very deep potential and small δ. As a result of small δ, the BF contribution would dominate the force on an isolated impurity. In fact, it is generally true that the force on an isolated impurity is in the direction of the electron wind, i.e., in the direction of the BF force. This has been established from T-matrix expressions[6,7] which are rigorous consequences of the expressions (1) and (6) used here.

We emphasize that a weak scattering treatment of the s-wave scatterer would give a spatial variation of the field exactly like the BF field which we have calculated. The lowest-order perturbation-theory result simply replaces the $\sin\delta\cos\delta$ factor in Γ_{BF} by the factor δ. The RRD fields do not, however, appear in lowest-order perturbation theory. Therefore, to the extent that the RRD fields are comparable to, or exceed, the BF fields in Fig. 4, one must go beyond perturbation theory and use the strong-scattering theory presented here. Typically $k_F r_{MT} \sim 1$ for metals, so that $\rho > 1$ is the appropriate region in the figures. In that region the BF and RRD terms are comparable, except that Γ_{BF} contains the factor $\cos\delta\sin\delta$ while Γ_{RRD} contains the factor $\sin^2\delta$. As an example, if $\delta \sim 0.3$, the RRD fields are around 30% of the BF fields. The most dramatic differences show up at resonance ($\delta = \pi/2$), where the BF contribution vanishes entirely. Furthermore, the structure of the RRD fields and BF are always different. It is not possible to simulate RRD contributions simply by changing δ or choosing a "better" potential or pseudopotential.

The distinction between the RRD and BF contributions to the force F_z also arises in T-matrix expressions. A T-matrix expression which is equivalent to Eq. (27) is[6]

$$F_z = 2\pi i \sum_{\vec{k},\vec{k}'} g_{\vec{k}} \left(\frac{\partial}{\partial R_z} \langle \vec{k}|T^{(-)}|\vec{k}'\rangle \right) \langle \vec{k}'|T^{(+)}|\vec{k}\rangle \, \delta(\epsilon_{\vec{k}} - \epsilon_{\vec{k}'}), \tag{29}$$

where R_z is the z coordinate of the impurity on which the force is to be calculated and $T^{(+)}$ and $T^{(-)}$ denote the T matrix for the localized impurity complex. (The $\pm$ signs denote electron or hole propagators in the defining equation for T.)

If Eq. (29) is applied to the problem of a strong s-wave scatterer and a weak second scatterer, expression (28) is obtained to lowest order in V. If one approximates the T matrix in a single-site picture, one writes $T^{(\pm)} = t^{(\pm)} + V$, where $t^{(\pm)}$ represents the T matrix of the isolated strong scatterer. Using this in Eq. (29) and taking the real part of that expression, we find that F_z is precisely the result we have obtained for the BF contribution alone. The RRD field is completely lost in this procedure. The RRD contribution requires one to include multiple scattering terms in $T^{(\pm)}$ when evaluating Eq. (29), i.e., terms to all orders in the strong scatterer and first order in V. We have already seen that such terms are important, especially at resonance. We expect that these RRD-related terms would be important corrections in single-site calculations of both electromigration forces and resistivities for clusters of strong scatterers in both solid and liquid metals.[17,27]

We comment on our approximations. The central approximation is the use of linear screening. As far as we are aware, this approximation is common to all previous self-consistent treatments of fields and charges in electron transport. A discussion of linear versus non-linear screening has been given elsewhere[20] in the context of Kubo formalism. An improved treatment of screening is not expected to lead to substantial changes in any of our results.

The particular assumption of short screening length simplified the calculations and allowed us to obtain analytic expressions for fields and charge densities. Expressions which are valid independent of any screening-length assumptions include Eq. (12) for $n_0(\vec{r})$ and Eq. (28) for the force on a weak very localized scatterer. [In using Eq. (28) for the force, one must use the δE_z expression (26) even though this expression does not give the actual local field if the screening length is not small.]

When the screening length is not small ($\alpha \lesssim k_F$), the Friedel oscillations we have found in $n(\vec{r})$ and $\delta E(\vec{r})$ will be somewhat washed out. In general, one would have to average our calculated $n(\vec{r})$ or $\delta E(\vec{r})$ over the region containing the Thomas-Fermi screening potential. For example, the true local field would be given by

$$\delta\vec{E}(\vec{r}) = \frac{\alpha^2}{4\pi} \int \frac{e^{-\alpha|\vec{r}-\vec{r}'|}}{|\vec{r}-\vec{r}'|} \, \delta\vec{E}_c(\vec{r}')d^3r', \tag{30}$$

where $\delta\vec{E}_c$ represents the calculated value for $\alpha \to \infty$, i.e., expression (26). The relation (30) also holds between the true density $n(\vec{r})$ and the calculated density $n_c(\vec{r})$, where $n_c(\vec{r})$ is given by

Eq. (16). The major effect of the integration is to average our results over a distance of order $1/\alpha$, and hence to diminish somewhat the amplitude of the Friedel oscillations in fields and charge densities. The qualitative picture we have found for the RRD would not change very much.

V. CONCLUSIONS

The RRD has been found to play an important role in the local field and electromigration driving forces. Our calculations give the first quantitative description of the RRD. The long-range (average) RRD field is exactly as described by Landauer. We found that the RRD does not arise from a very localized charge distribution around the impurity; rather, the RRD is extended and contains important contributions from long-range Friedel oscillations. Since our expressions are valid for distances up to a mean free path from the impurity, we conclude that the sources of the RRD are effectively distributed over a distance on the order of a mean free path from the impurity.

The RRD contributions to the electromigration driving force has been calculated for the case of a weak scatterer near a strong scatterer [Eq. (28)]. This RRD contribution is missing in previous calculations based on perturbation theory or single-site T matrices. RRD corrections are appreciable, and near a scattering resonance they become dominant.

Finally, we comment on the separation of RRD and BF terms throughout our analysis. The total charge density (BF + RRD) is, of course, the relevant physical quantity. The fact that the charge density neatly separates into parts which we define as BF and RRD quantities is convenient, but not essential, in our investigation. The BF and RRD densities are precisely defined in Eqs. (13) and (14) according to whether they exhibit $\sin^2\delta$ behavior (RRD) or $\sin\delta\cos\delta$ behavior (BF). The RRD quantities are thus proportional to the cross section as in Landauer's treatment.[1] The BF quantities are identical to the perturbation-theory results of Bosvieux and Friedel[14] when one invokes the weak-scattering limit ($\delta \ll 1$) and replaces $\sin\delta\cos\delta$ by δ in our expressions. Note that the RRD distribution does contain some "interference" terms and Friedel oscillations as well as the r^{-2} asymptotic behavior,[26] and is thus not precisely the quantity which has been envisioned by Landauer.[1-3] The separation of the interference terms and the Friedel oscillations from the RRD distribution is only possible in the asymptotic limit.[6] However, such asymptotic separations have no meaning close to the impurity, i.e., there is no meaningful analytic continuation of the asymptotic form to the $r \to 0$ limit. Our calculations show that there is no localized dipolar charge near the impurity on a microscopic scale. However, on the macroscopic scale there are electric fields set up which appear to emanate from a microscopically localized dipolar charge. These are our main conclusions and they are not dependent on the fact that we have made a particular separation between BF and RRD quantities. We emphasize this point because the BF and RRD separation is not so neatly made when the scattering potential is more complicated as, for example, when several phase shifts are appreciable. One could, in that case, still follow the general analysis of this paper, but without explicitly separating the charge density into RRD and BF quantities.

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[16]Some interference effects are considered in the weak-scattering regime in Ref. 14. Interference effects have been calculated within the framework of pseudopotential theory by R. S. Sorbello [J. Phys. Chem. Solids 34, 937 (1973)] for the case of solid metals and by D. Stroud [Phys. Rev. B 13, 4221 (1976)] for the case of liquid metals.

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[21]In Ref. 19 Schaich criticizes the particular form assumed in Ref. 4 for the collision term in the Liouville equation. The collision term is generally approximated by $-(\rho_T-\tilde{\rho})/\tau$, where ρ_T is the density matrix and $\tilde{\rho}$ is the density matrix to which ρ_T relaxes. In Ref. 4 $\tilde{\rho}$ was taken to be the equilibrium density matrix in the absence of the field. In Ref. 19 Schaich uses a form for $\tilde{\rho}$ which leads to a solution for ρ_T, which conserves particles at every point in space, i.e., the particle trajectories remain continuous even in the presence of collisions with the background. Since local conservation of particles describes physically correct behavior, Schaich's form for $\tilde{\rho}$ is preferable to the form used in Ref. 4. However, for large $k_F l$ (or large $\epsilon_F\tau$) both choices for $\tilde{\rho}$ give virtually identical fields and forces. The point is that the leading contribution to ρ_T turns out to be proportional to τ and hence the collision term is of zero order in τ. Since there are other terms in the Liouville equation (for the off-diagonal elements of ρ_T) which are linear in τ, they completely dominate the collision term for $\tau\to\infty$. The form assumed for $\tilde{\rho}$ is thus unimportant in the regime of physical interest, i.e., for $k_F l \gg 1$. (A similar conclusion holds for the collision term in the semiclassical analysis, e.g., the Boltzmann-equation analysis of Ref. 13.) Note, however, that imposition of the $\tau\to\infty$ limit and the consequent ignoring of the collision term effectively excludes us from addressing the problem of the electrostatic screening of the direct field; the latter is formally of order $1/k_F l$ times the dynamic screening considered here (see Ref. 20).

[22]This result can be drived from Sham's analysis (Ref. 7) by considering his response functions with the force vertex replaced by the electron-density vertex. The insertion of random-phase approximation bubble diagrams just before this vertex has the effect of replacing the unscreened density response $n_0(\vec{q})$ by the screened density response $n_0(\vec{q})/\epsilon(q)$. An explicit derivation of Eq. (4) can be found in the self-consistent treatment of Ref. 4 within the framework of a one-particle density matrix in a Hartree approximation.

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