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Driving force in electromigration and the residual resistivity field

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The mean force $\vec{F}$ on the electron gas due to the scattering on impurities is shown to account for the residual-resistivity field which must be applied to a sample in order to keep a stationary current. The calculation of $\vec{F}$ provides a means of estimating the driving force for the electromigration. It is also possible to obtain $\vec{F}$ by calculating the net charge flux induced by the displacement of the impurity.

In a recent paper,[1] Landauer and Woo show that the Bosvieux-Friedel calculation of the friction force[2] ignores terms (due to residual-resistivity dipoles) whose contribution is of the same order as those they explicitly considered. In the Bosvieux-Friedel paper, the charge polarization is calculated within the Born approximation; the friction force is then second order in λ the strength of the impurity potential and is compared to the residual resistivity $\Delta\rho$ calculated to the same order. The contribution of residual-resistivity dipoles to the force on the impurity is of order λ^3 however, because it is proportional to $z\Delta\rho$ [see Eq. (7) of Ref. 1], where z is the charge of the scattering impurity. Therefore, a quantum-mechanical calculation of the resistivity and of the force to order λ^3 must be performed to account for these effects. The residual-resistivity field however can be shown to be consistent with the Fiks[3]-Huntington[4] relation for the friction force.

Let us consider a current density $\vec{J}$ flowing through a pure metallic sample of resistivity ρ_0, $N Z$ conduction electrons per unit volume. The external electric field $\vec{E}^0_{\text{ext}}$ is defined by

$$\vec{E}^0_{\text{ext}} = \rho_0 \vec{J} \; . \tag{1}$$

Because of the residual resistivity $\Delta\rho$ due to the introduction of N_i interstitial impurities per unit volume, the external field must be increased by the residual-resistivity field $\Delta\vec{E}_{\text{ext}}$ in order to maintain the same current $\vec{J}$ as in the pure sample:

$$\Delta\vec{E}_{\text{ext}} = \Delta\rho\,\vec{J} = (\Delta\rho/\rho_0)\,\vec{E}^0_{\text{ext}} \; . \tag{2}$$

As a result of electron-impurity scattering and electron-electron interactions, a self-consistent screening charge builds up around each impurity and the conduction electrons are submitted to a self-consistent "internal field" contributed by the applied field, the bare impurities, and their screening charges. As the mean acceleration of the conduction electrons vanishes in the stationary state, the mean force on the electron gas due to the scattering on the impurities must exactly compensate for the force $-|e|\Delta\vec{E}_{\text{ext}}$. This effect has been analyzed by Kohn and Luttinger[5] in a system of electrons interacting with impurities [see Eq. (59) of their paper].

In order to obtain a quantitative expression of the friction force on an impurity, it is possible to use the formalism of Edwards[6] and Rousseau and Stoddart and March[7] who determined the mean-field necessary to keep a stationary current in the sample. Assuming a concentration of $N_i = 1$ impurity per unit volume and using a density matrix ρ normalized to unit volume, the condition that the mean force on the electron gas vanishes is

$$\operatorname{Tr}\rho(\vec{\phi}' + \vec{F}) = 0 \; , \tag{3}$$

where $\vec{F} = -|e|(\vec{E}^0_{\text{ext}} + \Delta\vec{E}_{\text{ext}})$ and $\vec{\phi}'$ represents the gradient of the electron-ion potential. This equation provides the mean forces $\langle\vec{F}_0\rangle$ on the electron gas of a pure metal and $\langle\vec{F}\rangle$ on the electrons of a sample containing one interstitial impurity per unit volume:

$$\langle\vec{F}_0\rangle = -\operatorname{Tr}(\rho_0\phi_0') \; , \tag{4}$$

$$\langle\vec{F}\rangle = -\operatorname{Tr}(\rho\phi') \; . \tag{5}$$

The effect of the impurity on the mean force acting on the electron gas is accordingly

$$\langle\vec{F}\rangle - \langle\vec{F}_0\rangle = -\operatorname{Tr}\rho(\phi' - \phi_0') - \operatorname{Tr}(\rho - \rho_0)\,\phi_0' \; . \tag{6}$$

The second term in this expression describes the change of force on the crystal lattice due to the change of the density matrix induced by the impurity. From a microscopic point of view this term exhibits the influence of the interstitial impurity on electron-lattice interactions and may account for a part of the temperature dependence of the effective valence of the impurity.

The first term in (6), which is the force on the electron gas due to the change in the potential ϕ brought about by the impurity, is the only term relevant to the electromigration problem in the jellium approximation. The friction force on an impurity is then

$$\vec{F}_i = \operatorname{Tr}\rho(\vec{\phi}' - \vec{\phi}_0') = \operatorname{Tr}\rho\phi'_{\text{ion}} \; , \tag{7}$$

where $\vec{\phi}'_{\text{ion}}$ is the gradient of the bare impurity potential.

As

$$\vec{\phi}'_{\text{ion}} = (i/\hbar)[\vec{p}, \phi_{\text{ion}}] , \tag{8}$$

where $\vec{p}$ is the one-particle momentum operator, (7) shows that $\vec{F}_i$ is due to the momentum transfer $\langle d\vec{P}/dt\rangle_{\text{imp}}$ to the electron gas occurring in electron impurity collisions:

$$\vec{F}_i = -\text{Tr}\rho\left(\frac{d\vec{p}}{dt}\right)_{\text{imp}} = -\left\langle\frac{d\vec{P}}{dt}\right\rangle_{\text{imp}}, \tag{9}$$

where $\vec{P}$ is the momentum operator of one unit volume of the electron gas.

In the $\vec{r}$ representation, (7) can be written

$$\vec{F}_i = \int d^3r\, n_\epsilon(\vec{r})\vec{\nabla}\phi_{\text{ion}} , \tag{10}$$

where $n_\epsilon(\vec{r})$ is the total change in the electronic density brought about by the presence of the electric field. This charge density is made of two parts:

$$n_\epsilon(\vec{r}) = n_1(\vec{r}) + n_2(\vec{r}) . \tag{11}$$

(i) $n_1(\vec{r})$, the bare polarization charge, is due to the scattering of electrons which are displaced by the field, on the self-consistent potential of the impurity; (ii) $n_2(\vec{r})$ is the reaction of the electron gas to this external charge.

The expression (10), a consequence of the Feynman-Hellmann theorem, has been used previously by Bosvieux-Friedel,[2] Sorbello,[8] Gerl,[9] and Kumar and Sorbello.[10] It shows that $\vec{F}_i$ arises from Coulomb interactions between the self-consistent polarization charge n_ϵ and the bare ion charge. In a free-electron gas, it is equivalent to write (10) as the interaction between the screened impurity potential ϕ_{scr} and the bare polarization charge n_1:

$$\vec{F}_i = \int d^3r\, n_1\vec{\nabla}\phi_{\text{scr}} . \tag{12}$$

This equivalence can be demonstrated by writing (10) in the reciprocal space: the integral contains a product involving $n_\epsilon(q)$ and $\phi_{\text{ion}}(q)$, where $n_\epsilon(q) = n_1(q)/\epsilon(q)$, $\epsilon(q)$ being the dielectric function of the electron system. This product can also be written as $n_1(q)$ multiplied by $\phi_{\text{scr}}(q) = \phi_{\text{ion}}(q)/\epsilon(q)$. As shown in Ref. 11, n_1 can be easily calculated to first order in λ by summing up all contributions from the waves scattered by the impurity, and it is straightforward to relate $\vec{F}_i$ to the residual resistivity $\rho_i = N\Delta\rho/N_i$ of the impurity

$$\vec{F}_i = -Z[\rho_i/(\rho_0+\Delta\rho)]\,|e|(\vec{E}^0_{\text{ext}} + \Delta\vec{E}_{\text{ext}}) ,$$

or

$$\vec{F}_i = -Z(\rho_i/\rho_0)\,|e|\,\vec{E}^0_{\text{ext}} . \tag{13}$$

Then the force contributed by N_i impurities per unit volume on the electron gas

$$\vec{F} = -N_i\vec{F}_i = \bar{n}(\Delta\rho/\rho_0)\,|e|\,\vec{E}^0_{\text{ext}}, \tag{14}$$

where $\bar{n} = ZN$ is the number of free electrons per unit volume, can be interpreted as due to the internal field $-(\Delta\rho/\rho_0)\vec{E}^0_{\text{ext}}$. This force cancels exactly that due to the residual-resistivity field (2).

Another way of attacking the problem of the effective valence Z_i^* of an interstitial impurity is to come back to the definition of Z_i^* given by the thermodynamics of irreversible processes:

$$Z_i^* = z_i - (J_e/J_i)_{\vec{E}=0} , \tag{15}$$

where z_i is the true ionic charge and $(J_e/J_i)_{\vec{E}=0}$ is the *total* electron flux associated with a unit flux of the impurity, in the *absence of an applied electric field*. The equivalence between (15) and the definition in terms of a force follows from Onsager's relations. The electron flux $\vec{J}_e$ is made of two parts: a convective flux $\vec{J}_e^1$ due to the permanent reconstruction of the screening cloud by the electron gas and a flux $\vec{J}_e^2$ due to the scattering of electrons on the screened impurity. In the case of a slowly moving interstitial of velocity $\vec{v}_i$, the flux $\vec{J}_e^1 = z_i\vec{v}_i$ (Ref. 12) compensates exactly for the charge carried by the bare ion and the direct electrostatic force on the interstitial ion vanishes. This point is demonstrated in some details in Ref. 13. The flux $\vec{J}_e^2$ can be calculated through a linear response formalism to first order in the velocity $\vec{v}_i$ and any order in the impurity potential, and the usual relationship between the force $\vec{F}_i$ and the residual resistivity of the impurity is recovered. This way of tackling the problem of the friction force and the direct electrostatic force as well avoids some difficulties with the definition of the fields and the charges on which they act.

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