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Phonon-limited resistivity of aluminium using a first-principles pseudopotential

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Abstract. The phonon-limited resistivity of aluminium has been calculated using a local, first-principles pseudopotential which has been useful in the calculation of other properties of aluminium. This pseudopotential is obtained from the induced electron density around an aluminium ion in an electron gas. From this pseudopotential, the interionic potential, the phonons (which are calculated by the self-consistent harmonic approximation) and finally the phonon-limited resistivity have been obtained. The results are very similar to those obtained using a phenomenological, Heine–Abarenkov pseudopotential for aluminium.

1. Introduction

The pseudopotential formulation has been very useful in the calculation of the properties of solids. Pseudopotentials may be phenomenological or may be obtained from first principles. In the former case the parameters used to define the pseudopotential are usually determined by fitting some electronic properties, predicted with the pseudopotential, to experimental information such as the electrical resistivity of the metal, the shape of the Fermi surface, or spectroscopic data. It is clear at present that a pseudopotential determined in an empirical way cannot always be regarded as weak [1], so that its use in obtaining the interionic potential and, from this, the phonons to be used in the calculation of the resistivity is not justified.

In this work we use a first-principles, local pseudopotential which is constructed following a method proposed by Manninen *et al* [2], who followed the method of Rasolt and Taylor [3], with some differences.

In the approach of Rasolt and Taylor, the displaced electronic density around an ion in an electron gas is calculated using a non-linear screening theory and the full electron-ion pseudopotential. Then, a non-local pseudopotential is selected in order to reproduce, as close as possible, the non-linear displaced electronic density by linear response theory, except in the vicinity of the ion. In this way the non-linear effects are partly included in the pseudopotential. The interionic potentials calculated using these pseudopotentials have been used with success to calculate phonon dispersion curves in simple metals [4, 5], and the resistivity for Rb and Cs [6], for low temperatures.

In the method of Manninen *et al*, the starting point is also the displaced electronic density around an impurity in an electron gas, which has been calculated by non-linear screening theory. Then a local pseudopotential is defined in such a way that, in linear response theory, this displaced electronic density is reproduced exactly, except in a small

region close to the ion. In this region a modelling of the electronic density is done in order to remove all the wiggles of the density. This modelled density plays the role of the pseudodensity. The pseudopotential form factor is given in terms of the Fourier transform of this pseudodensity and a dielectric function which satisfies the compressibility theorem. In this way, some of the non-linear screening effects are also included into the pair potential calculated from this pseudopotential. Manninen *et al* obtained the pseudopotential for aluminium, considering two models to calculate the displaced electron density using non-linear screening theory. In the first model they calculated the screening of an aluminium nucleus in an electron gas. In the second they considered the nucleus embedded in a jellium vacancy. This second model gave much better results for the cohesion energy of the metal and for the equilibrium lattice constant, bulk modulus, vacancy formation energy and electrical resistivity of the liquid phase. In a more recent work, Jena *et al* [7] made a calculation of the phonon dispersion curve of aluminium using the interionic potential for this material reported by Manninen *et al* [2], for the model of the nucleus embedded in a jellium vacancy, obtaining good agreement with the experimental results.

In previous work we obtained this kind of pseudopotential, with the model of the nucleus embedded in a jellium vacancy, and used it with success to calculate the lattice specific heat of lithium [8] and aluminium [9], the pressure dependence of the lattice specific heat of lithium [10] and aluminium [9], and also to calculate the pressure dependence of the elastic constants of aluminium and lithium [11].

In this work we want to explore the applicability of the same kind of pseudopotential to the calculation of transport properties. In particular, we are interested in comparing the prediction of the phonon-limited resistivity of aluminium using this pseudopotential with the prediction made using a phenomenological, Heine–Abarenkov pseudopotential and experimental results.

In § 2 we describe briefly the method of Manninen *et al* to construct the local, first-principles pseudopotential from the displaced electron density. We also show the dielectric function used in this work and the vertex correction for the screened pseudopotential form factor used for the calculation of the phonon-limited resistivity.

Section 3 is used to describe the phonons and the expression employed for the phonon-limited resistivity. Our results and conclusions are given in § 4. We have used atomic units (i.e. the magnitude of the electronic charge = electron mass = $\hbar = 1$). The energy is given in double Rydbergs.

2. The pseudopotential

The unscreened pseudopotential form factor, $\nu(q)$, is related to the Fourier transform of the induced charge pseudodensity, $\delta n(q)$, by

$$\nu(q) = 4\pi\delta n(q)\epsilon(q)/q^2[1 - \epsilon(q)] \quad (1)$$

where $\epsilon(q)$ is the dielectric response function of the electron gas.

$\delta n(q)$ was calculated using the induced electronic density, $\delta n(r)$, which was computed by the density functional formalism [12, 13] with a smoothing in a region near the origin [2]. In this smoothing, the conditions that the electronic charge is conserved and that $\delta n(r)$ and $(\partial/\partial r)[\delta n(r)]$ are continuous are imposed [2]. Then, equation (1) is used to obtain an effective local pseudopotential, which in linear response will give the exact induced displaced electronic density outside the region of smoothing. In this way some of the non-linear screening effects are included into the pair potential calculated from

this pseudopotential. It should be remarked that in the pseudopotential formulation, the pseudodensity must not contain wiggles near the ion, and the induced density calculated from density functional theory contains those wiggles in that region due to the orthogonalisation of conduction states to core orbitals.

From pseudopotential theory and linear response theory [14], the interionic potential is given by

$$\Phi(r) = \frac{Z}{r} \left(1 + \frac{2}{\pi Z^2} \int_0^\infty \frac{dq \sin(qr) \epsilon(q) [\delta n(q)]^2}{q(1 - \epsilon(q))} \right) \quad (2)$$

where r is the separation between the two ions and Z is the charge of the metal ion.

For the model of the nucleus embedded in a jellium vacancy, the induced electronic density is calculated by taking the difference [2]

$$\delta n(r) = n(r) - n_v(r) - 2 \sum_b |\psi_b(r)|^2 \quad (3)$$

where $n(r)$ is calculated with the total charge density corresponding to a nucleus located at the centre of a vacancy in jellium and $n_v(r)$ is the electron density around a jellium vacancy alone. Charge neutrality of the metal is a necessary condition. The bound states are represented by ψ_b .

The dielectric function used satisfies, by construction, the compressibility theorem which is important in connection with the interionic potential [2, 15]. The dielectric function is [2, 16]

$$\epsilon(q) = 1 + (4\pi/q^2)G(q) \quad (4)$$

where

$$G(q) = G_0(q)/[1 - (4\pi/k_{\text{TF}}^2)G_0(q)(1 - L)]. \quad (5)$$

$G_0(q)$ is the usual Lindhard polarisability, k_{TF} is the Fermi–Thomas screening constant and L is the ratio

$$L = (\partial\mu/\partial r_s)/(\partial E_{\text{F}}/\partial r_s). \quad (6)$$

In equation (6) μ is the chemical potential, E_{F} is the Fermi energy and

$$\mu(r_s) = E_{\text{F}}(r_s) + \mu_{\text{xc}}(r_s)$$

where $\mu_{\text{xc}}(r_s)$ is the exchange–correlation contribution to the chemical potential.

On the other hand, the screened pseudopotential form factor, $W(q)$, given by

$$W(q) = (\nu(q)/\epsilon(q))C(q) \quad (7)$$

is important in the calculation of the resistivity. The vertex correction is $C(q)$ which, in the simplest approximation for a local pseudopotential, is [16, 17]

$$C(q) = 1/[1 - (4\pi/k_{\text{TF}}^2)G_0(q)(1 - L)].$$

Using the expression of Gunnarson and Lundqvist [18] for the exchange–correlation (which is the one we used in the calculation of the induced electronic density), the corresponding value of L is

$$L = 1 - (4/q\pi^4)^{1/3} r_s \{1 + [0.6213/(r_s + 11.4)]r_s\}. \quad (8)$$

3. Resistivity and phonons

The expression used in this work for the resistivity, $\rho(T)$, as function of the temperature, T , has been derived and discussed by several authors [19, 20]:

$$\rho(T) = A \sum_{\lambda} \int_{2k_{\text{F}}} \frac{|\mathbf{q} \cdot \boldsymbol{\epsilon}(q, \lambda)|^2 |W(q)|^2 \beta q d^3 q}{[\exp(\beta\omega(q, \lambda)) - 1][1 - \exp(-\beta\omega(q, \lambda))]} \quad (9)$$

where $W(q)$ is the screened pseudopotential form factor. $\boldsymbol{\epsilon}(q, \lambda)$ is the polarisation

vector of the lattice vibration with wavevector \mathbf{q} and frequency $\omega(\mathbf{q}, \lambda)$, β is $1/k_B T$, k_B being the Boltzmann constant, and A is a constant given by

$$A = 3\Omega_0/16MV_F^2 k_F^4 \quad (10)$$

where M is the ion mass and V_F and k_F are the electron velocity and wavevector at the Fermi level, respectively.

The integral in equation (9) is over a sphere of radius $2k_F$. The pseudopotential describing electron scattering at the Fermi surface is assumed to depend only on momentum transfer \mathbf{q} . The Fermi surface is taken as spherical so that the two surface integrals describing transitions from an initial to a final state on the Fermi surface can be converted to a three-dimensional integral over \mathbf{q} . A one-phonon approximation is considered when equation (9) is derived [19, 20]. Since much of the aluminium Fermi surface is free-electron-like, it is expected that multiple-plane-wave effects might not be very important. In fact, one-plane-wave calculations with a spherical Fermi surface give a reasonable description of the experimental data of the resistivity of aluminium at high temperatures (between ~ 70 and 140 K) using a Heine–Abarenkov pseudopotential [20, 21]. At low temperatures this same pseudopotential can be used but it is necessary to consider the Fermi surface and the electron–phonon matrix elements in greater detail [21, 22]. For temperatures above 140 K we should expect that anharmonic effects become more important.

On the other hand, it is not our aim in this work to make a precise calculation of the phonon-limited resistivity of aluminium for the whole range of temperatures. For simplicity, we are interested in the temperature range (see [21]) for which the expression for the resistivity, given by equation (9), is applicable for comparison with experimental data. We are also interested in a comparison of the prediction made using our first-principles pseudopotential, and equation (9), with the results obtained with a phenomenological, Heine–Abarenkov pseudopotential, and the same expression for the resistivity. We believe that this will be sufficient to explore the applicability of our pseudopotential to the calculation of the phonon-limited resistivity of aluminium. A careful calculation of this property, for low temperatures, using our pseudopotential can be performed following the method given in [21], where a Heine–Abarenkov pseudopotential is employed.

It is clear from equation (9) that we need information about the phonon frequencies and polarisation vectors, and that we also need the screened pseudopotential form factor.

The interionic potential, given by equation (2), was obtained from the induced pseudodensity and the dielectric function from this interionic potential we calculated the phonons to be employed in the expression for the resistivity. The force constants associated with our interionic potential were calculated using the self-consistent harmonic approximation [23–25].

The resulting set of self-consistent equations which must be solved in order to obtain the phonon dispersion curve and force constants for this approximation is as follows:

$$\omega_\lambda^2(\mathbf{k})\varepsilon_\lambda^\alpha(\mathbf{k}) \sum_\beta D_{\alpha\beta}(\mathbf{k})\varepsilon_\lambda^\beta(\mathbf{k}) \quad (11)$$

where $\varepsilon_\lambda^\alpha(\mathbf{k})$ is the component α of the polarisation vector $\varepsilon_\lambda(\mathbf{k})$ and the dynamical matrix is

$$D_{\alpha\beta}(\mathbf{k}) = \frac{1}{M} \sum_l (1 - \cos(\mathbf{k} \cdot \mathbf{R}_l)) \langle \Phi_{\alpha\beta}(\mathbf{R}_l) \rangle \quad (12)$$

with

$$\langle \Phi_{\alpha\beta}(\mathbf{R}_l) \rangle = \frac{1}{(8\pi^3 \det \lambda_l)^{1/2}} \int d^3\mu \exp(-\frac{1}{2}\mu_\gamma (\lambda_l^{-1})_{\gamma\delta} \mu_\delta) \Phi_{\alpha\beta}(\mathbf{R}_l + \boldsymbol{\mu}_l) \quad (13)$$

where $\boldsymbol{\mu}_l$ is the vector describing the displacement of atom l from its equilibrium position \mathbf{R}_l , and $\Phi_{\alpha\beta}(\mathbf{R}_l + \boldsymbol{\mu}_l)$ is the tensor derivative of the interatomic potential evaluated at $\mathbf{R}_l + \boldsymbol{\mu}_l$.

Finally,

$$(\lambda_l)_{\alpha\beta} = \frac{1}{MN} \sum_{\mathbf{k}, \lambda} (1 - \cos(\mathbf{k} \cdot \mathbf{R}_l)) \varepsilon_{\lambda}^{*\alpha}(\mathbf{k}) \varepsilon_{\lambda}^{\beta} \coth[(1/2)\beta\omega_{\lambda}(\mathbf{k})] / \omega_{\lambda}(\mathbf{k}) \quad (14)$$

where N is the number of ions and the sum is over the first Brillouin zone.

To solve the set of self-consistent equations (11), (12), (13) and (14), we start with the frequencies generated by the harmonic approximation as the first trial. Then the convergence procedure is followed.

To calculate all the phonon frequencies and polarisation vectors entering the expression for the resistivity (equation (9)) from the force constants obtained in the phonon dispersion curve, we followed the method of Gilat and Raubenheimer [26]. This method consists of solving the secular equations associated with the dynamical matrix only at a relatively small number of points in the irreducible first Brillouin zone. Then, by means of linear extrapolation the other phonon eigen-frequencies are extracted from within small cubes, each centred at one point. These cubes can be arranged to fill the entire irreducible first Brillouin zone and thus can yield the complete frequency distribution of the crystal. Simple translations of the vectors \mathbf{q} are used to complete the integration region up to $2k_F$.

4. Results and discussion

In order to calculate the resistivity we started by obtaining the induced density shown in equation (3) using the density functional formalism. For this it is necessary to calculate the displaced electronic density around an aluminium nucleus embedded into a jellium vacancy and also the displaced electronic density around a vacancy alone. After this a smoothing of the density near the ion is done in order to construct the displaced electronic pseudodensity. In figure 1 we show the displaced electronic density calculated using equation (3) and the corresponding smoothed density.

The next step was to calculate the Fourier transform of the pseudodensity. This was achieved using the asymptotic form for $\delta n(r)$ given by

$$\delta n(r) = B \cos(2k_F r + \varphi) / r^3$$

where the constants B and φ were obtained using the last points in our calculation of $\delta n(r)$. This asymptotic form was taken for distances larger than $R_{\max} = 15.04a_0$, where a_0 is the Bohr radius ($a_0 = 0.529 \text{ \AA}$). The accuracy of the Fourier transform was tested by taking the inverse Fourier transform of $\delta n(q)$, and the resulting difference with respect to the original values of $\delta n(r)$ was less than 0.1% for each point.

With $\delta n(q)$ and the dielectric function defined in § 3 we could calculate the interionic potential using equation (2). From this interionic potential we found the force constants by the self-consistent harmonic approximation [23–25]. Using these together with the method of Gilat and Raubenheimer [26] we obtained the phonon frequencies and polarisation vectors to be used in equation (9) to calculate the phonon-limited resistivity. The results are shown in figures 2 and 3.

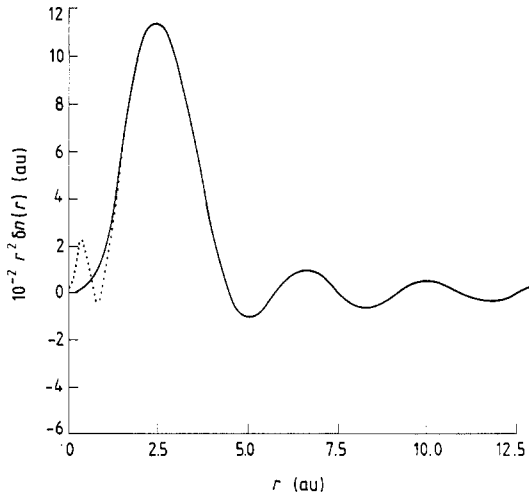


Figure 1. Displaced electronic density (\cdots) and displaced electronic pseudodensity obtained by smoothing the displaced electronic density (—).

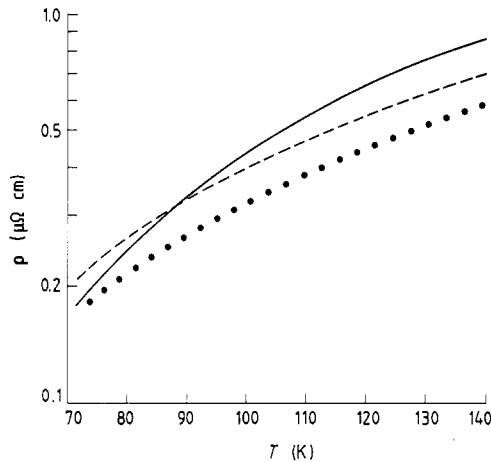


Figure 2. Resistivity of aluminium for temperatures between 70 and 140 K: experimental results [27] (—); result of this work (---); result from equation (9) using a Heine-Abarenkov pseudopotential (taken from [21]) (\cdots).

As we have already mentioned, the expression used for the resistivity is adequate for a comparison with experimental data for temperatures between 70 and 140 K (see [21]). In figure 2 we show the results from our first-principles calculation compared with experimental results [27] and with the calculation of [21] from a Heine-Abarenkov pseudopotential for the same range of temperatures, using the same expression for the resistivity given in equation (9). This figure shows a reasonable agreement between our prediction and the experimental results and the results from the Heine-Abarenkov pseudopotential. We have already said that it is not our aim in this work to make a precise calculation of the phonon-limited resistivity of aluminium for the whole range of temperatures. We want only to assess the suitability of our pseudopotential for the calculation of this property of aluminium.

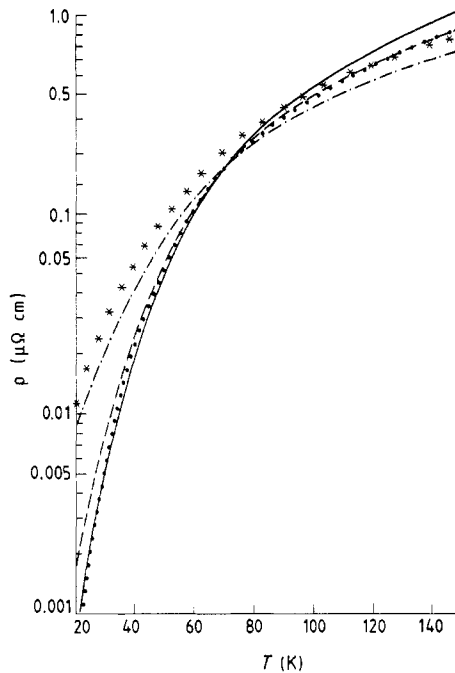


Figure 3. Resistivity of aluminium for temperatures between 20 and 140 K: experimental results [27] (—); result obtained from equation (9) using our pseudopotential (***) . The chain curve is calculated by equation (9) and a Heine–Abarenkov pseudopotential [21]. The dotted curve is the result obtained using the scattering-time approximation, 15 plane waves for the electron–phonon interaction, the Heine–Abarenkov pseudopotential and a non-spherical Fermi surface [21]. Finally, the broken curve refers to a calculation similar to that of the dotted curve; the only difference is that the expression for the resistivity is from a variational approximation [21].

In figure 3 we show experimental values for the resistivity of aluminium for temperatures between 20 and 140 K, taken from [27]. We also show the predictions made using equation (9) with our pseudopotential and with a Heine–Abarenkov pseudopotential (the latter is taken from [21]), for the same range of temperatures. We can see that these two are very similar. Also in figure 3 we show a more careful calculation, from [21], of the resistivity. In this calculation, the approximation of a spherical Fermi surface is no longer taken and, for the electron–phonon matrix element, 15 plane waves have been used to describe the electronic states. For the solution to the Boltzmann transport equation two approaches were taken. One was the scattering time approximation and the other was obtained from a variational principle [21]. The Heine–Abarenkov pseudopotential is again used in both approximations. The scattering-time approximation gave better results for temperatures below 70 K. However, for higher temperatures they are practically identical and both results are very similar to ours.

From the foregoing we can say that our pseudopotential is adequate for the calculation of the phonon-limited resistivity of aluminium. A reasonable agreement with experimental results can be seen for the range of temperatures for which equation (9) is expected to be adequate (between 70 and 140 K) [21].

On the other hand, within the approximations implicit in equation (9) (which are not valid at low temperatures), the results obtained with our first-principles pseudopotential

and a Heine–Abarenkov pseudopotential are very similar and the latter were used successfully in [21] for a careful calculation of the phonon-limited resistivity of aluminium.

Finally, it is reasonable to expect that our first-principles pseudopotential could also be used successfully in a careful calculation of the phonon-limited resistivity of aluminium for a much wider range of temperatures.

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