Electrical resistivity of the alkaline earth metals Ca, Sr and Ba in the 50–300K temperature region

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Abstract. The computed values of the temperature dependent electrical resistivity is presented for the alkaline earth metals Ca, Sr and Ba. Numerical values of the mean free paths obtained using a finite mean free path approach is also reported. The structure factor has been evaluated using experimental values for the phonon spectra while local model potentials were used for the form factors. Our numerical results compare favourably with experiments.

1. Introduction

The alkaline earth metals Ca, Sr and Ba occupy group 2 in the periodic table and the calculation of some of their physical properties could be of considerable interest. Reasons for this include the knowledge that the general features of the electronic structures of these metals are intermediate between those of the simple and the transition metals. It is also known that under normal conditions of pressure and room temperature, Ca and Sr are FCC while Ba exhibits a BCC structure. Ca and Sr also exhibit a BCC structure at high temperatures. Difficulties encountered in the past in growing large single crystals of Ca, Sr and Ba have contributed to an inadequate knowledge of their experimental phonon spectrum. This has in part hampered the theoretical determination of the resistivity.

However in recent times, knowledge of the phonon spectrum has become available for Ca (Stassis *et al* 1983), Sr (Bucheneau *et al* 1984) and Ba (Mizuki *et al* 1983; Bucheneau *et al* 1985). In the present paper, we have used the Ashcroft (1963) and the Heine-Abarenkov (1964) model potentials to compute the electrical resistivity of the aforementioned alkaline earth metals in the 50–300K temperature region by applying the Ferraz-March (1979) method for finite mean free path (l) to the metals. The physical implication is that we have taken into consideration the probable distortion of the Fermi surface in these metals. We have also tried to obtain graphical plots of the variation of resistivity and l with temperature.

A brief theoretical discussion is given in §2. The determination of the structure factor is presented in §3 and §4 contains our results and conclusions.

2. Theory

Ziman (1960) and Greene and Kohn (1965) have given a general expression of the form given below for the electrical resistivity $\rho(T)$ as a function of temperature T in the pseudopotential formalism

$$\rho(T) = 3/(16nhZe^2V_f^2K_f^4) \int_0^\infty q^3s(q)|U(q)|^2\theta(q - 2K_f)dq. \tag{1}$$

In (1), m is the electronic mass, e the charge on the electron, n the number of ions per unit volume, Z the valency of the metal, K_f the Fermi wave vector, $U(q)$ the pseudopotential form factor, $\hbar = h/2\pi$ here h is the Plancks constant, V_f the Fermi velocity and $s(q)$ is the angular dependent structure factor. θ is the heaviside function. Equation (1) assumes that the Fermi surface is spherical since the q-integration is cut of at $2K_f$.

One would however prefer to leave the structure of the Fermi surface to correct itself relative to the distortion (if it exists). An attempt to take the blurring into consideration in the formulation of resistivity is given by Ferraz and March (1979). The Ferraz-March approach yields in place of (1)

$$\rho(T) = \frac{3}{16nhZe^2V_f^2K_f^4} \int_0^\infty q^3s(q)|U(q)|^2\Gamma(q, K_f, l) \tag{2}$$

where $\Gamma(q, K_f, l)$ is given by

$$\Gamma(q, K_f, l) = 2/\pi\{\tan^{-1}(ql) - 0\cdot5\tan^{-1}[(2ql)/(1 + 4(K_f l)^2 - (ql)^2]$$

$$- (\pi/2)\theta[q - (1/l^2 + 4K_f^2)^{0\cdot5}]\}. \tag{3}$$

Here the mean free path l is determined self consistently. The first step being to determine ρ using (1) i.e. with $l = \infty$. l is then recalculated using the Drude expression for the electrical resistivity i.e.

$$\rho = \hbar K_f/(ne^2l). \tag{4}$$

The iteration process begins by obtaining ρ from (1) assuming $l = \infty$, substituting in (4) to obtain l. This value of ρ is then substituted in (2) to obtain ρ and (2) and (4) are iterated until self consistent values of l and ρ are obtained.

3. Structure factor

Although considerable success has been obtained in the experimental determination of the structure factor for a lot of liquid metals (Waseda 1980), the situation is not the same for the solid phases of the metals. One may however use the standard (Van Hove 1954) one phonon approximation for the structure factor given by

$$s(\vec{q}) = \hbar^2/(MK_BT)\sum_p[\vec{q}\cdot\hat{e}_p(\vec{q})]^2/[(e^{\beta\omega}p^{(\vec{q})} - 1)(1 - e^{-\beta\omega}p^{(\vec{q})})]. \tag{5}$$

Here $\omega_p(\vec{q})$ and $\hat{e}_p(\vec{q})$ are respectively the frequency and polarization vectors of a

phonon of branch (p) and wave vector $\bar{q}$, $\beta = \hbar/K_BT$, K_B is the Boltzman constant and T is the temperature.

The $\omega_p(\bar{q})$ are obtained using the Born-Von Karman model in which they are expressed in terms of experimental values of the interplanar force constants $\Phi_n(T)$ (Woods *et al* 1962) as

$$\omega^2(T) = M^{-1} \sum_{n=1}^{N} \Phi_n(T)[1 - \cos(n\pi q/q\max)] \tag{6}$$

where M is the mass of the atom and the sum over n is taken over N planes of atoms.

The structure factor $s(q)$ is obtained using

$$s(q) = 1/(4\pi) \int s(\bar{q}) \, d\Omega. \tag{7}$$

For $\bar{q}$ in (5) we have used [100], [110], and [111] directions since these are symmetry directions for which experimental values of force constants can easily be obtained.

The structure factor $s(q)$ in (7) was evaluated using a modified form of the Houston's (1948) 3-term integration procedure.

4. Results and discussion

We present for the first time the numerical results for the temperature variation of resistivity for the alkaline earth metals Ca, Sr and Ba using the self consistent theory explained above.

The parameters R_C and A used in the Ashcroft and Heine-Abarenkov potentials are chosen in such a way that the self consistent values of the calculated resistivity reproduce the experimental results at the highest temperature under consideration. The values are shown in table 1 below and could be used for calculation of other properties which require the use of the model potentials.

As stated earlier, the calculation of the resistivity depends on the model potential and the structure factor. For the determination of the phonon frequencies in (6), we have used experimental force constants provided in (Stassis *et al* 1983), for Ca, (Bucheneau *et al* 1983) for Sr and Ba and (Mizuki *et al* 1985) again for barium. The reason for using two different tables of force constants in Barium is to test two different methods of determining the lattice dynamics of the metals investigated, one using polycrystalline samples (Bucheneau *et al* 1984), and the other (Stassis *et al* 1983; Mizuki *et al* 1985) using single crystals. However as can be observed from figure 3, the results are not remarkably different.

In figures 1, 2, 3 we show plots of temperature variation of electrical resistivity for Ca, Sr and Ba respectively. For the first two metals we have used the two model

Table 1. R_C and A in the Ashcroft and Heine Abarenkov potentials respectively.

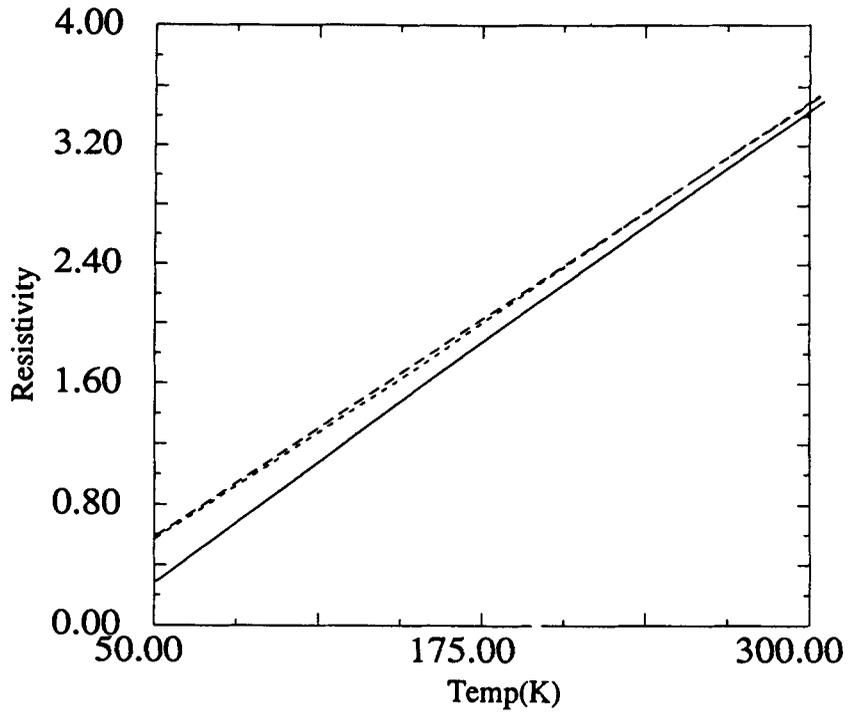|	Ca	Sr	Ba
R_C(a.u)	0·7	0·6	1·56
A (a.u)	0·865	0·2	—

Figure 1. Graphs of resistivity in $\mu\Omega$-cm vs temperature (K) for calcium. ——Experiment; ---Calculated using Ashcroft potential; -- Calculated using Heine-Abarenkov potential.
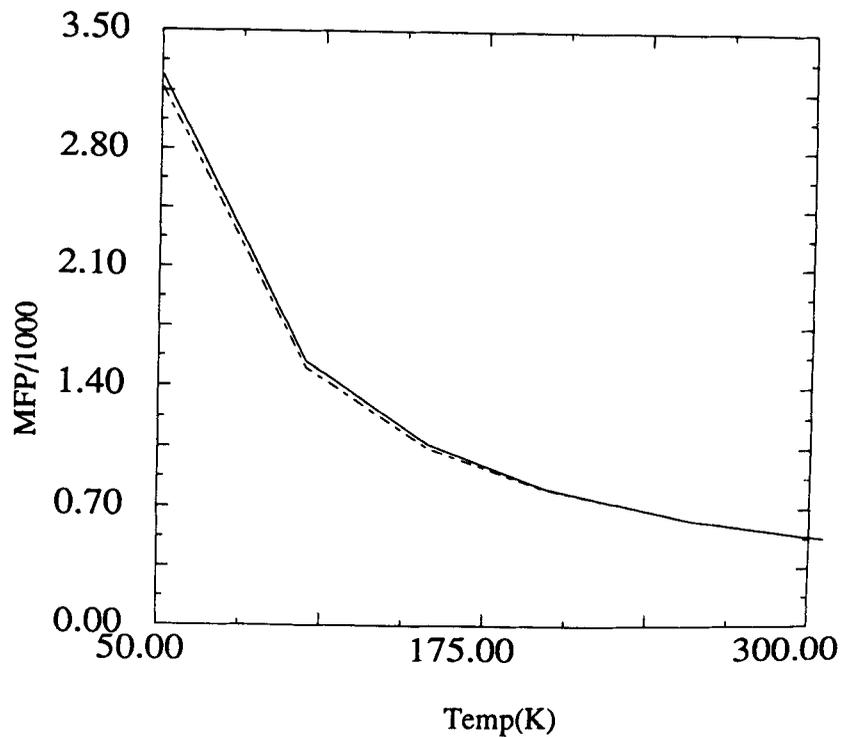

Figure 2. Graphs of calculated mean free path (atomic units) vs temperature (K).—Ashcroft Potential; ---Heine Abarenkov potential.
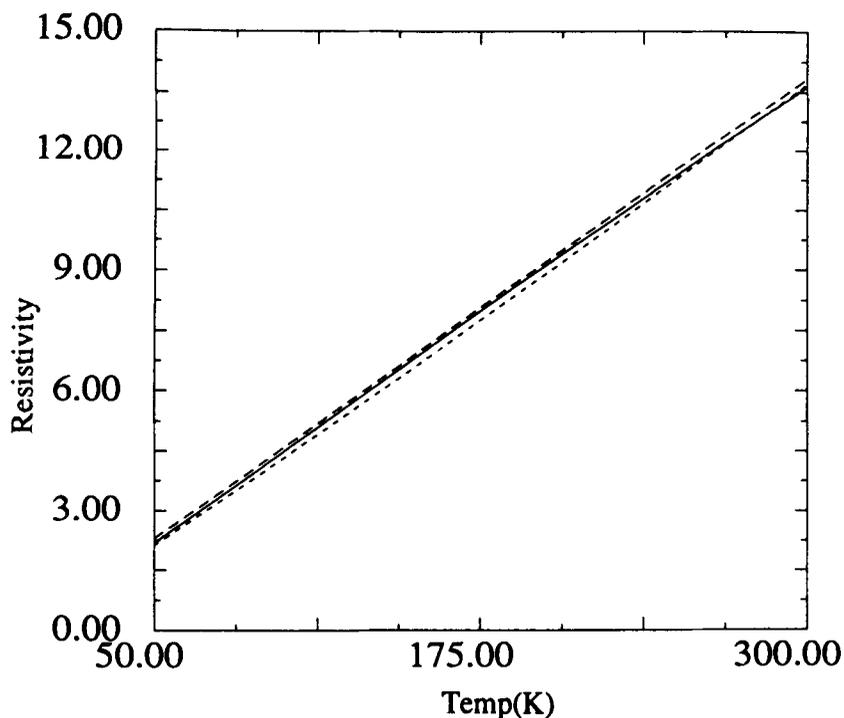
Figure 3. As in figure 1 but for strontium.

potentials while for Ba, we have used only the Ashcroft potential, the reason for this being that the fit for A obtained using the Heine Abarenkov potential was too high.

The experimental results are taken from (Cook *et al* 1975) for Ca (Cook and Van Der Meer 1973) for Sr and Ba.

A close observation of our graph indicates that the results are very good for Sr, reasonable especially at high temperatures for Ca but poor for Ba.

We attribute deviations where they exist to the possibility that the local pseudopotential formulation used could be too simple for the metals. One reason for this being that we have not taken into consideration the proximity of d bands to the Fermi level in other words the effect of hybridization with d states. It is possible that using a non local pseudopotential that takes these factors into consideration could yield better results.

Also presented in figures 4–6 are the computed variations of mean free path with temperature for the elements under investigation. This is in contrast to our earlier work on alkali metals (Akinlade and Hussain 1989) in which we used the value of the mean free path at just one point to compute the thermopower.

One observes that l tends to have a very high value at low temperature which implies that the average distance between successive collisions is long, after which l tends to a constant value at high temperatures. Thus it is reasonable to assume at high temperatures that the finite mean free path is constant.

It is also observed that l decreases with increase in atomic number of the metal. l is highest in Ca and least in Ba, the ratio being about the order of magnitude of 10.
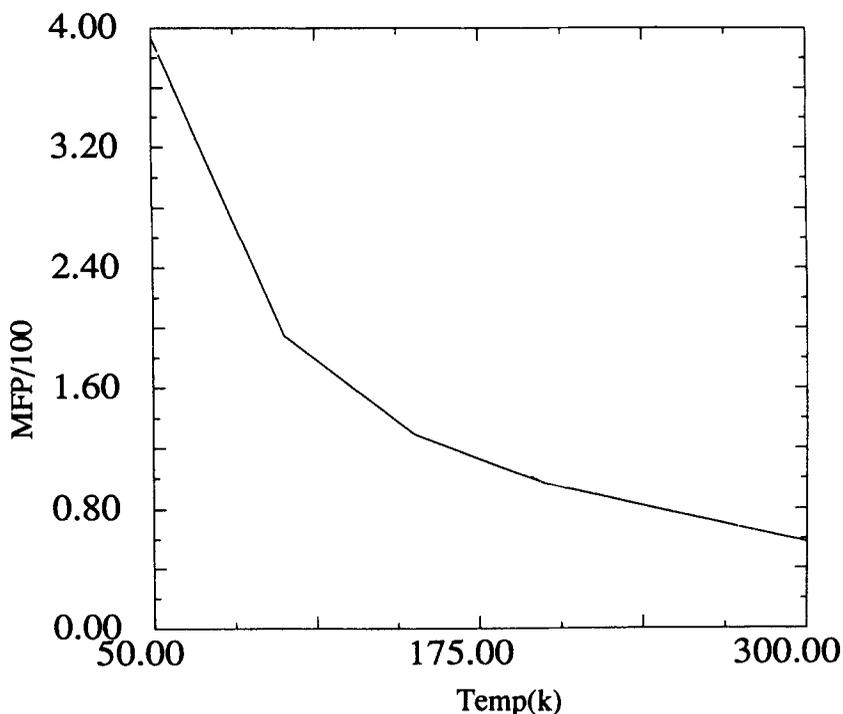
In conclusion we state that the fair results demonstrate the adequacy of the Ferraz-March approach for the class of metals investigated.
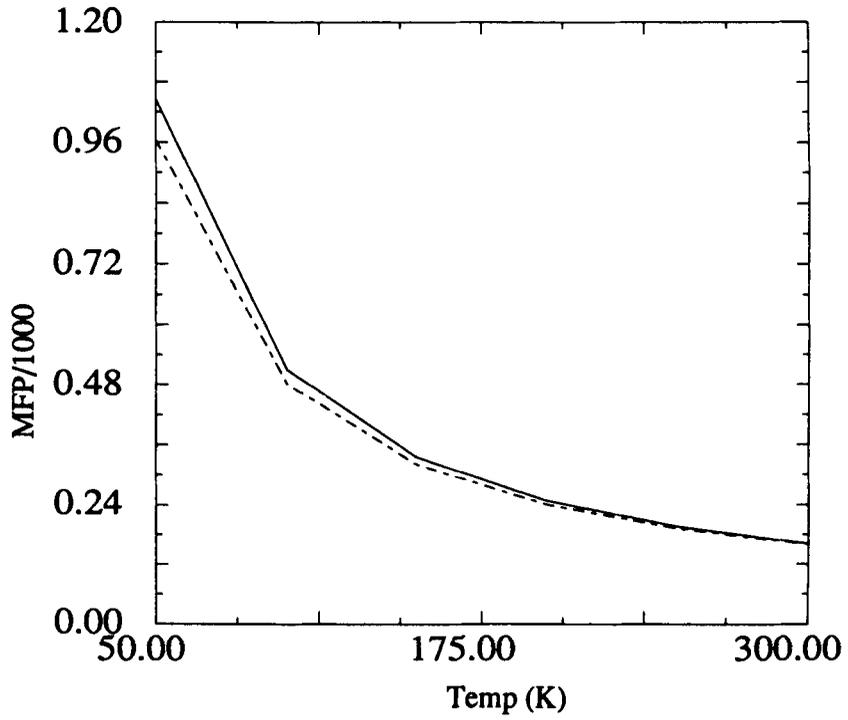
Figure 4. As in figure 2 but for strontium.
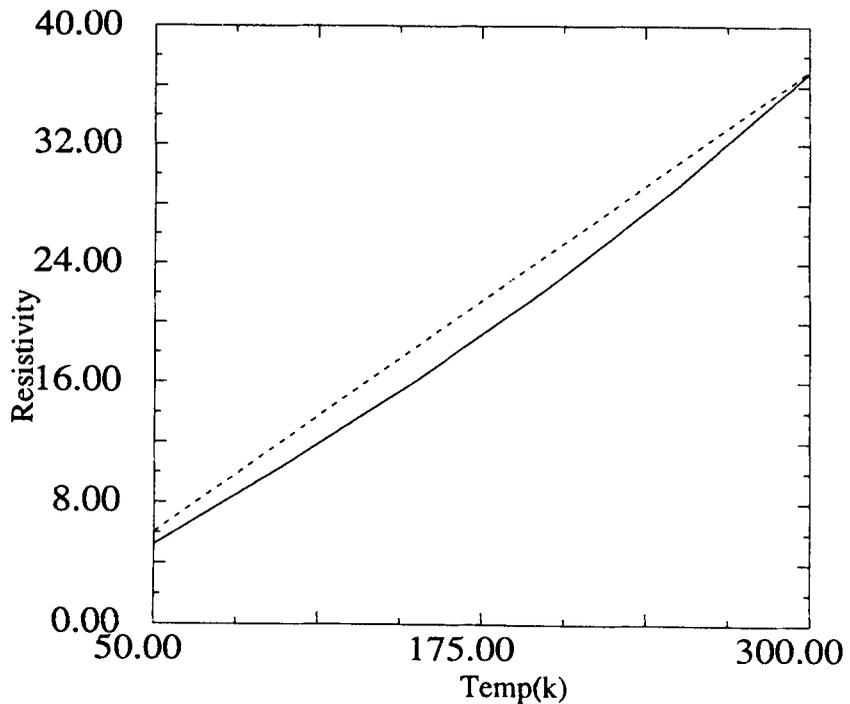
Figure 5. Graphs of resistivity in $\mu\Omega$-cm vs temperature (K) for barium. ——Experiment; ---Calculated using Heine-Abarenkov potential.

Figure 6. Graph of mean free path (atomic units) vs temperature (K).——Heine Abarenkov potential.

Acknowledgements

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