Magnetic susceptibilities of liquid binary alloys

M Iwamatsu, S K Lai, R A Moore and S Wang
Guelph Waterloo Program for Graduate Work in Physics, Waterloo Campus, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada

Received 26 March 1982, in final form 28 June 1982

Abstract. A description of the concentration dependence of the magnetic susceptibilities of a number of liquid binary alloys of simple metals is effected in three stages. First, a previous pseudopotential formalism is extended to enable the calculation of the conduction electron diamagnetic susceptibilities in the pure liquid metals, Li, Na, K, Rb, Cs, In and Pb. These results are then combined with much earlier calculated values of the diamagnetic susceptibilities of the core electrons and with observed values, existing in the literature, of the total magnetic susceptibilities to infer the conduction electron paramagnetic susceptibilities of the above pure liquid metals. Second, the expressions for the two diamagnetic contributions are modified permitting the determination of their concentration dependence in a number of nearly-free-electron (NFE) like liquid binary alloys. The Landau theory of a Fermi liquid is used to infer the concentration dependence of the paramagnetic contribution. Third, Li–Pb, which is not NFE like over the entire concentration range is treated separately as a special but closely related case. The observed results for the concentration dependence of the total magnetic susceptibility are favourably reproduced, both for those alloy systems exhibiting smooth variations and for those with large variations.

1. Introduction

It has been observed (Uemura et al 1978) that the total magnetic susceptibility, $\chi$, is almost linearly dependent on concentration for some liquid binary alloys of simple metals, such as Na–Cd and Na–Tl. However, on going to other liquid binary alloys, such as Li–Pb and Na–Bi, the observed (Hackstein et al 1980) concentration dependence of $\chi$ is anomalous in that $\chi$ changes dramatically at particular concentrations. It seems to us that this varied behaviour in $\chi$ has not yet been described theoretically and hence we present a possible explanation in this work.

We start with $\chi$ written in its usual form

$$\chi = \chi_i + \chi_d + \chi_a$$

(1.1)

where $\chi_i$ is the diamagnetic susceptibility of the ion cores, $\chi_d$ is the diamagnetic susceptibility of the valence electrons and $\chi_a$ is the paramagnetic susceptibility of the valence electrons. Since there is no \textit{a priori} reason to expect the concentration dependence of each contribution to $\chi$ to be the same, they are considered individually. The first task, § 2, is to fix the end points, that is, the pure metallic values. $\chi_i$ is obtained from a standard Slater-type expression (Slater 1930, Angus 1932). $\chi_d$ is obtained by extending a previous calculation for solid metals (Lai et al 1978, to be referred to as I) to the liquid metal case. In the hope of minimising calculational errors and to circumvent a complex calculation for $\chi_a$, equation (1.1) is used with the calculated $\chi_i$ and $\chi_d$ and with the observed $\chi$ to infer
values for $\chi_s$. Although individual errors from $\chi_l$ and $\chi_d$ may now be contained in $\chi_s$, all the values should be reasonable and any subsequent characteristic concentration dependences should be manifested satisfactorily. The second task is to determine the concentration dependence of $\chi$ for the alloy systems. This is done in § 3 for the NFE like binary alloys, K–Rb, Na–K and Na–Cs. The expressions for $\chi_l$ and $\chi_d$ given in § 2 are modified and calculated directly. The features of the expression for $\chi_s$ given by the Landau theory of a Fermi liquid are used to obtain the concentration dependence of $\chi_s$.

Since the modifications appearing in § 3 are not sufficient for Li–Pb alloys at all concentrations, they are dealt with as a special case in § 4.

Finally, the results are collected together and discussed in § 5. The procedure suggested throughout permits a description of $\chi$ for some liquid binary alloys with a dramatically varying $\chi$ as well as those with a smoothly varying $\chi$.

2. Magnetic susceptibilities of pure liquid metals

The first step in determining the concentration dependence of the three contributions to the total magnetic susceptibilities of the liquid alloys in question is to fix their end-point values, that is, their values in the pure components. Since the major computational work herein is related to the diamagnetic susceptibilities of the valence electrons it is presented first. Afterwards, the results for the ionic core contributions and for the valence electron paramagnetic contributions are given.

2.1. Calculation of the valence electron diamagnetic susceptibilities

In I a formulation in the NFE approximation, using a non-local pseudopotential, was presented for the total valence electron magnetic susceptibilities of both solid and liquid pure metals and for the corresponding diamagnetic susceptibilities of solid pure metals. This formulation was also applied in I, with considerable success, to the calculation of the above quantities for the alkali metals and for a number of simple liquid metals. In addition, the approximations used in I were re-examined (Wang et al 1980b) and deemed to be quite reasonable. Therefore, in order to obtain $\chi_d$ for the pure liquid metals we simply apply the formulation in I, suitably modified for the liquid case.

The general expressions for $\chi_d$ are given in § 2.3 and § 2.4 of I. $\chi_d$ is written as

$$\chi_d = \chi_{df}(1 + \delta_d + \delta_c) + \Delta \chi_{ae}$$

(2.1)

where $\chi_{df}(=-\chi_l/2)$ is $\chi_d$ in the free-electron theory and, hence, $\chi_l$ is the Landau–Pauli free-electron value. $\delta_d$ arises from the electron–ion interaction and is

$$\delta_d = -\frac{1}{8\mu_0^2} \sum_q F(q, k_F) \left[ \frac{b_0^2}{2} \ln \left| 1 + \frac{b_0^2}{1 - b_0^2} \right| + \frac{b_0}{b_0 - 1} \right. + \left. \frac{2(q_x^2 + q_y^2)}{q^2} \left( \frac{b_0^2(5 - 3b_0)}{8(b_0 - 1)^2} + \frac{3b_0^2}{16} \ln \left| 1 + \frac{b_0^2}{1 - b_0^2} \right| \right) \right]$$

(2.2)

where the magnetic field, $B$, is taken in the $z$ direction, $\mu_0(=k_0^2/2)$ is the chemical potential, $\mu$, in the free-electron theory, $b_0 = 4\mu_0/E(q)$ with $E(q) = q^2/2$ and $F(q, k_F)$ describes the scattering between plane-wave states due to the total electron–ion pseudopotential in the system and is, for one-component systems,

$$F(q, k_F) = S^*(q)S(q)w_{-q}(k_F + q)w_q(k_F).$$

(2.3)
Here \( S(q) = N^{-1} \sum \exp(-i \mathbf{q} \cdot \mathbf{R}_i) \) in which \( N \) and \( \mathbf{R}_i \) are, respectively, the total number of the ions and the position vector of the \( \lambda \)th ion in the system under consideration. \( w_{\mathbf{q}}(k_F) \) is the on-the-Fermi-surface form factor with electronic exchange and correlation effects included. \( \delta_c \) arises from the non-local part of the electron–ion interaction and is

\[
\delta_c = \frac{\partial w_0(k_F)}{\partial \mu} \left( \frac{1}{3} + \frac{1}{16\mu_0} \sum_q F(q, k_F)b_0^{1/2} \ln \frac{1 + b_0^{1/2}}{1 - b_0^{1/2}} \right) \\
- \frac{1}{8\mu_0} \sum_q \left( \frac{\partial}{\partial \mu} F(q, k_F) \right) \left( 1 + \frac{b_0 - 1}{2b_0^{1/2}} \ln \frac{1 + b_0^{1/2}}{1 - b_0^{1/2}} \right). 
\]

We note that the derivative of a form factor with respect to \( \mu \) can be evaluated as in I as

\[
\frac{\partial w_{\mathbf{q}}(k_F)}{\partial \mu} = \left. \frac{\partial w_{\mathbf{q}}(k)}{\partial k} \left( \frac{\partial}{\partial \mu} \right)_{k=k_F} \right| \frac{\partial k}{\partial \mu}
\]

with \( \mu = k^2/2m^* \) and \( m^* \) being the electronic effective mass which may be taken to be the electronic density of states effective mass at the Fermi surface, as in electron transport coefficient calculations. (See Lai et al (1981) and Lai and Wang (1982) for the justification of using \( m^* \) as the electronic density of states effective mass for disordered metallic systems.) Finally, \( \Delta \chi_{\text{ex}} \) in equation (2.1) comes from conduction-electron exchange-correlation effects and has been investigated by a number of authors (Pines 1955, Kanazawa 1957); it can be obtained straightforwardly within the effective mass approximation using a formula derived by Kanazawa (1957). It is worth noting that, since the radius parameter, \( r_s \), lies between 2 and 6 for the systems of interest, this formula by Kanazawa is probably more appropriate than the one used by Dupree and Seymour (1970) in their calculation of \( \chi_d \), because the one used by them is valid only for \( r_s \leq 1 \).

The conversion from the general case to the liquid case is given in § 3.2 of I as well as the conversion from sums to integrals. Following I, \( \delta_d \) and \( \delta_c \) can be converted to

\[
\delta_d = -\frac{3Z}{32\mu_0} \int_0^{\infty} dp \ F_i(p k_F, k_F) \left( 2(p + 2p^{-1}) \ln \frac{p + 2}{p - 2} - \frac{8}{3} \frac{3p^4 - 22p^2 + 24}{(4 - p^2)^2} \right)
\]

and

\[
\delta_c = \frac{\partial w_0(k_F)}{\partial \mu} \left( \frac{1}{3} + \frac{3Z}{16\mu_0} \int_0^{\infty} dp \ F_i(p k_F, k_F)p \ln \frac{p + 2}{p - 2} \right) \\
- \frac{3Z}{16\mu_0} \int_0^{\infty} dp \left( \frac{\partial}{\partial \mu} F_i(p k_F, k_F) \right) \left( p^2 + \frac{1}{4} p(4 - p^2) \ln \frac{p + 2}{p - 2} \right)
\]

where

\[
F_i(p k_F, k_F) = a(p k_F)w_{-p_F}(k_F + p k_F)w_{p_F}(k_F).
\]

Here \( a(p k_F) \) is the liquid structure factor. Note that in \( \delta_c \) the partial derivative with respect to \( \mu \) operates only on the form factor parts of \( F_i \) and that the difficulty arising from the singularity in the integral in equations (2.6) or (2.7) can be overcome as in I.

Numerical values for \( \chi_\lambda \) are obtained by using (i) the form factors from Wang et al (1980a), (ii) the on-the-Fermi-surface approximation for angles between wavevectors in \( w_{\mathbf{q}}(k_F) \) as in I and (iii) the input data listed in table 1. Here \( \eta \) is the packing density required for the \( a(q) \) from the Percus–Yevick hard-sphere theory and determined, as in Wang and Lai (1980a, to be referred to as II), using the same pseudopotentials as used for the form.
Table 1. Values for $\Omega_0$, the atomic volume in atomic units, $\eta$, the packing density, $m^*$, the electronic density of states effective mass in atomic units, $r_s$, the electronic radius parameter, and $\chi$, the Landau–Pauli free electron susceptibility in units of $10^{-6}$ cm$^3$ mol$^{-1}$, for the pure liquid metals considered herein.

<table>
<thead>
<tr>
<th>Metal</th>
<th>$T$ (°C)</th>
<th>$\Omega_0$</th>
<th>$\eta$</th>
<th>$m^*$</th>
<th>$r_s$</th>
<th>$\chi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li</td>
<td>800</td>
<td>170.2</td>
<td>0.332</td>
<td>1.32</td>
<td>3.44</td>
<td>7.62</td>
</tr>
<tr>
<td>Na</td>
<td>100</td>
<td>277.7</td>
<td>0.481</td>
<td>1.06</td>
<td>4.05</td>
<td>10.57</td>
</tr>
<tr>
<td>K</td>
<td>65</td>
<td>528.9</td>
<td>0.481</td>
<td>1.02</td>
<td>5.02</td>
<td>16.24</td>
</tr>
<tr>
<td>Rb</td>
<td>40</td>
<td>646.8</td>
<td>0.480</td>
<td>0.96</td>
<td>5.36</td>
<td>18.57</td>
</tr>
<tr>
<td>Cs</td>
<td>30</td>
<td>830.1</td>
<td>0.483</td>
<td>0.93</td>
<td>5.83</td>
<td>21.93</td>
</tr>
<tr>
<td>In</td>
<td>156</td>
<td>154.3</td>
<td>0.483</td>
<td>0.89</td>
<td>2.31</td>
<td>10.30</td>
</tr>
<tr>
<td>Pb</td>
<td>800</td>
<td>231.3</td>
<td>0.272</td>
<td>0.84</td>
<td>2.40</td>
<td>14.85</td>
</tr>
</tbody>
</table>

2.2. Ionic and paramagnetic susceptibilities

The ionic susceptibilities, $\chi_1$, are diamagnetic, arising from the ion cores, which are taken to be fixed throughout this work. We use the values, see table 3, given by Slater's (1930) method as tabulated by Angus (1932).

As noted earlier, direct calculations of either the total $\chi$ or $\chi_s$, e.g. I, do not yield precise values. Since the primary purpose of this work is to describe the concentration dependence of the total $\chi$, we attempt to circumvent inherent errors in calculations of $\chi_s$ by using the calculated $\chi_d$ and $\chi_1$ in equation (1.1) to infer $\chi_s$ from the observed values of $\chi$. These results are given in table 3 along with other values of $\chi_s$, either inferred differently (Dupree and Seymour 1970, Perdew and Wilkins 1973) or observed (see table 3 for references).
Table 3: Values of $\chi_d$ and $x_s/x_d$, determined using the presently calculated values of $\chi_d$ (table 2), theoretical $\chi_t$ from Angus (1932) and the experimental total $\chi$ (from (i) Hackstein et al (1980) for Li, (ii) Collings (1965) for the other alkali metals, (iii) Dupree and Seymour (1970) for In and (iv) Hackstein et al (1980) for Pb). The $\chi$ are in units of $10^{-6}$ cm$^3$ mol$^{-1}$. For the purpose of comparison, the values deduced theoretically for $(x_s/x_d)$ by Dupree and Seymour (1970) and those by Perdew and Wilkins (1973) are given. Also, the experimental $(x_s/x_d)$ in the liquid case, denoted by $(x_s/x_d)_{exp,L}$ is given and compared with that in the solid case, denoted by $(x_s/x_d)_{exp,S}$.

<table>
<thead>
<tr>
<th>Metal</th>
<th>$\chi_t$</th>
<th>$\chi_{exp}$</th>
<th>$x_s$</th>
<th>$(x_s/x_d)$</th>
<th>$(x_s/x_d)_{DS}$</th>
<th>$(x_s/x_d)_{PW}$</th>
<th>$(x_s/x_d)_{exp,L}$</th>
<th>$(x_s/x_d)_{exp,S}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li</td>
<td>0.7</td>
<td>27.5</td>
<td>31.30</td>
<td>2.74</td>
<td>2.04</td>
<td>—</td>
<td>2.67 ± 0.13</td>
<td>2.84 ± 0.1</td>
</tr>
<tr>
<td>Na</td>
<td>3.7</td>
<td>15.0</td>
<td>24.05</td>
<td>1.52</td>
<td>1.65</td>
<td>1.73</td>
<td>1.57 ± 0.08</td>
<td>1.59 ± 0.05</td>
</tr>
<tr>
<td>K</td>
<td>13.1</td>
<td>20.0</td>
<td>41.75</td>
<td>1.71</td>
<td>1.72</td>
<td>1.57</td>
<td>1.68 ± 0.10</td>
<td>1.70 ± 0.006</td>
</tr>
<tr>
<td>Rb</td>
<td>24.1</td>
<td>17.6</td>
<td>52.14</td>
<td>1.87</td>
<td>1.94</td>
<td>1.58</td>
<td>1.71 ± 0.16</td>
<td>1.72 ± 0.01</td>
</tr>
<tr>
<td>Cs</td>
<td>37.2</td>
<td>28.2</td>
<td>78.04</td>
<td>2.37</td>
<td>1.90</td>
<td>—</td>
<td>2.05 ± 0.20</td>
<td>2.17 ± 0.06</td>
</tr>
<tr>
<td>In</td>
<td>28.1</td>
<td>−9.6</td>
<td>23.73</td>
<td>1.54</td>
<td>1.57</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Pb</td>
<td>33.8</td>
<td>−13.0</td>
<td>30.96</td>
<td>1.39</td>
<td>1.26</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>

† From (i) Schumacher and Slichter (1956) for Li and (ii) Perdew and Wilkins (1973) for others.
‡ From (i) Flesner and Schultz (1976) for Li and (ii) Knecht (1975) for the other alkalis.

Here $x_d$ is the free-electron value. The indications are that this procedure will be reasonably successful for the lighter alkalis but less so for the heavier metals.

At this point, all of the end point values have been determined for each component of $\chi$ and we now turn our attention to the alloys.

3. Magnetic susceptibilities of metallic-like liquid alloys

In this section the concentration dependence of each contribution to $\chi$ is determined for a number of metallic-like binary alloys. These are alloys that closely resemble the pure metallic components, the charge-transfer effects are small and the NFE approximation is valid.

3.1. Calculation of the valence electron diamagnetic susceptibilities

Since the valence electrons in a metallic-like binary alloy are not appreciably different from those in a pure metal we use exactly similar methods to calculate $\chi_d$ in both cases. Thus we need only modify the expressions given in § 2.1.

The scattering between plane waves is still given by the matrix elements of the total pseudopotential and hence we make the substitutions

$$S(q)w_i(\mathbf{k}_F) \Rightarrow \sum_l S_l(q)w_{i,q}(\mathbf{k}_F)$$

and

$$F(q, \mathbf{k}_F) \Rightarrow \sum_l \sum_j S_l^*(q)S_j(q)w_{i-q}(\mathbf{k}_F + \mathbf{q})w_{j,q}(\mathbf{k}_F).$$

Here $S_l(q) = \langle 1/N \rangle \sum_i \exp(-i\mathbf{q} \cdot \mathbf{R}_i)$, where $N$ is the total number of ions in the alloy and
$R_{ij}$ gives the positions of each $i$th-type ion in the alloy; $w_{j,\mathbf{q}}(k_F)$ is the form factor for the component $j$ of the binary alloy (Wang et al 1980a).

Of course, the above expressions apply to a single configuration of the system. To proceed further, we follow the literature (Ashcroft and Langreth 1967, Hafner 1977) to take the average over all possible configurations in the liquid state and write the substitution (3.2) in the form

$$F_i(p_{kF}, k_F) = \sum_{i,j} (C_i C_j)^{1/2} a_{ij}(p_{kF}) w_{i, -p_{kF}}(k_F + p_{kF}) w_{j, p_{kF}}(k_F).$$

(3.3)

In this case

$$Z = \sum_i C_i Z_i.$$  (3.4)

Here $C_i$ is the concentration of the $i$th component and $a_{ij}(q)$ is the Ashcroft–Langreth-type partial structure factor.

The calculation of $\chi_d$ for the alloy now proceeds exactly as in the pure metal. The input data for the alloys are listed in table 4. $\eta_i$ are the partial packing densities required for the $a_{ij}(q)$. These packing densities and the average atomic volumes were determined as described in II. The values of $m^*$ were obtained in the same way as in the above pure metallic case. $\delta_e$ and $\delta_d$ for the alloys are obtained by substituting equations (3.3) and (3.4) into equations (2.6) and (2.7) and performing the integrations. $\chi_d$ is then given by equation (2.1) with $\chi_{df}$ and $\Delta \chi_{dc}$ being obtained as in the pure metals.

The numerical results for K–Rb, Na–K and Na–Cs alloys are noted in table 5. The results have very similar features, in general, to the pure metallic case. In addition, one sees that the concentration dependence of $\chi_d/\chi_{df}$ deviates more from a linear relation as one goes from a system with small charge transfer, e.g. Na–K, to one with a larger charge transfer, e.g. Na–Cs (see II for a discussion of the charge transfer). Note that, although it is included in table 5 for convenience, Li–Pb is discussed in § 4 as a special case.

### 3.2. Valence electron paramagnetic susceptibilities

In order to determine $\chi_s$ in alloys we use the features of the expression for $\chi_s$ as given by the Landau theory of a Fermi liquid. Thus

$$\chi_s = \chi_{df} m^*(1 + B_0)^{-1}$$  (3.5)

### Table 4. Values for (i) the atomic volume $\Omega_0$, (ii) the partial packing density $\eta_i$, (iii) the electronic density of states effective mass $m^*$, (iv) the electronic radius parameter $r_s$, and (v) $\chi_{df}$ for the binary alloys of simple metals (see table 1 for units).

<table>
<thead>
<tr>
<th>A–B</th>
<th>$C_B$</th>
<th>$T$(°C)</th>
<th>$\Omega_0$</th>
<th>$\eta_A$</th>
<th>$\eta_B$</th>
<th>$m^*$</th>
<th>$r_s$</th>
<th>$\chi_{df}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>K–Rb</td>
<td>0.8</td>
<td>100</td>
<td>619.3</td>
<td>0.085</td>
<td>0.380</td>
<td>0.97</td>
<td>5.29</td>
<td>18.04</td>
</tr>
<tr>
<td>Na–K</td>
<td>0.4</td>
<td>100</td>
<td>378.3</td>
<td>0.222</td>
<td>0.255</td>
<td>1.04</td>
<td>4.49</td>
<td>12.99</td>
</tr>
<tr>
<td>Na–K</td>
<td>0.7</td>
<td>100</td>
<td>455.6</td>
<td>0.091</td>
<td>0.382</td>
<td>1.03</td>
<td>4.77</td>
<td>14.70</td>
</tr>
<tr>
<td>Na–Cs</td>
<td>0.3</td>
<td>100</td>
<td>443.1</td>
<td>0.216</td>
<td>0.264</td>
<td>1.05</td>
<td>4.75</td>
<td>14.43</td>
</tr>
<tr>
<td>Na–Cs</td>
<td>0.5</td>
<td>100</td>
<td>557.0</td>
<td>0.125</td>
<td>0.346</td>
<td>1.03</td>
<td>5.10</td>
<td>16.81</td>
</tr>
<tr>
<td>Na–Cs</td>
<td>0.7</td>
<td>100</td>
<td>649.5</td>
<td>0.064</td>
<td>0.399</td>
<td>1.01</td>
<td>5.37</td>
<td>18.62</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>0.4</td>
<td>800</td>
<td>183.6</td>
<td>0.165</td>
<td>0.106</td>
<td>0.96</td>
<td>2.71</td>
<td>10.43</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>0.6</td>
<td>800</td>
<td>201.8</td>
<td>0.096</td>
<td>0.158</td>
<td>0.92</td>
<td>2.58</td>
<td>12.04</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>0.8</td>
<td>800</td>
<td>217.0</td>
<td>0.042</td>
<td>0.213</td>
<td>0.88</td>
<td>2.48</td>
<td>13.48</td>
</tr>
</tbody>
</table>
Magnetic susceptibilities of liquid binary alloys

Table 5. Theoretical values for $\delta_d$, $\delta_c$, various $\chi_a$ and the electron exchange correlation correction, $\Delta\chi_{ec}$, for the binary alloys of the simple liquid metals considered herein.

<table>
<thead>
<tr>
<th>A-B</th>
<th>C_B</th>
<th>$-\chi_{dr}$</th>
<th>$\delta_d$</th>
<th>$\delta_c$</th>
<th>$-\chi_d^{(0)}$</th>
<th>$-\chi_c^{(0)}$</th>
<th>$-\chi_{em}$</th>
<th>$-\Delta\chi_{ec}$</th>
<th>$\chi_a/\chi_{dr}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>K-Rb</td>
<td>0.8</td>
<td>9.02</td>
<td>0.024</td>
<td>0.006</td>
<td>9.29</td>
<td>9.30</td>
<td>0.81</td>
<td>1.12</td>
<td></td>
</tr>
<tr>
<td>Na-K</td>
<td>0.4</td>
<td>6.50</td>
<td>-0.046</td>
<td>-0.013</td>
<td>6.12</td>
<td>6.25</td>
<td>0.46</td>
<td>1.01</td>
<td></td>
</tr>
<tr>
<td>Na-K</td>
<td>0.7</td>
<td>7.35</td>
<td>-0.037</td>
<td>-0.006</td>
<td>7.03</td>
<td>7.14</td>
<td>0.51</td>
<td>1.03</td>
<td></td>
</tr>
<tr>
<td>Na-Cs</td>
<td>0.3</td>
<td>7.22</td>
<td>0.040</td>
<td>-0.060</td>
<td>7.08</td>
<td>6.88</td>
<td>0.51</td>
<td>1.05</td>
<td></td>
</tr>
<tr>
<td>Na-Cs</td>
<td>0.5</td>
<td>8.41</td>
<td>0.036</td>
<td>-0.046</td>
<td>8.33</td>
<td>8.17</td>
<td>0.67</td>
<td>1.07</td>
<td></td>
</tr>
<tr>
<td>Na-Cs</td>
<td>0.7</td>
<td>9.31</td>
<td>0.020</td>
<td>-0.025</td>
<td>9.26</td>
<td>9.22</td>
<td>0.84</td>
<td>1.08</td>
<td></td>
</tr>
<tr>
<td>Li-Pb</td>
<td>0.4</td>
<td>5.22</td>
<td>0.020</td>
<td>0.047</td>
<td>5.57</td>
<td>5.44</td>
<td>0.16</td>
<td>1.10</td>
<td></td>
</tr>
<tr>
<td>Li-Pb</td>
<td>0.6</td>
<td>6.02</td>
<td>0.140</td>
<td>0.097</td>
<td>7.45</td>
<td>6.54</td>
<td>0.18</td>
<td>1.27</td>
<td></td>
</tr>
<tr>
<td>Li-Pb</td>
<td>0.8</td>
<td>6.74</td>
<td>0.180</td>
<td>0.119</td>
<td>8.76</td>
<td>7.66</td>
<td>0.13</td>
<td>1.32</td>
<td></td>
</tr>
</tbody>
</table>

(Rice 1968, Knecht 1975) where $\chi_{dr}$ is the free-electron value, $m^*$ is the electronic density of states effective mass occurring in previous expressions and $B_0$ is a Legendre coefficient of the Landau scattering function. Although a number of approximations are available for $B_0$ (see Kushida et al 1976 for an excellent review) no single approximation works equally well for a given set of simple metals. Thus we first infer the dependence of $B_0$ on $m^*$ and $r_s$ from the $\chi_a$ as determined above for the pure metals. These results are then used to infer the values of $B_0$ corresponding to the values of $m^*$ and $r_s$ appropriate for the alloys and hence the concentration dependence of $\chi_a$ in the alloys.

We note that, experimentally (table 3) and theoretically (Wang et al 1980b), $\chi_a/\chi_{dr}$ is almost temperature independent and hence the major temperature dependence resides in $\chi_{dr}$. It follows that $B_0$ will depend primarily on $m^*$ and $r_s$ for a given alloy system. In order to determine this dependence $B_0$ has been evaluated for the pure metals at (i) liquid temperatures where $m^*$ is given in table 1 and (ii) at low temperatures where $m^*=m_b$, the band effective mass, the effects of the electron–phonon interaction being negligible. The corresponding values of $B_0$ are summarised in table 6. Included also are the theoretical values (Wilk et al 1979) for Li, Na and K metals at low temperature.

By inspection one sees that $B_0$ has a complex dependence on $m^*$ and $r_s$, contrary to the results of Rice (1968), which depend only upon $r_s$. We also notice that the values of $B_0$ appear to lie in two distinct ranges of $r_s$ values, $r_s \leq 3.5$ and $r_s \geq 3.5$ such that in each range $B_0$ depends primarily on $m^*$ but with different functional relations. Specifically, for

Table 6. Values of the Legendre coefficient of the Landau scattering function, $B_0$, for (i) $m^*$ given in table 1 and (ii) $m^*$ equal to $m_b$, as defined in text for seven pure simple metals.

<table>
<thead>
<tr>
<th>Metal</th>
<th>Pb</th>
<th>In</th>
<th>Li</th>
<th>Na</th>
<th>K</th>
<th>Rb</th>
<th>Cs</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\chi_a/\chi_{dr}$</td>
<td>1.39</td>
<td>1.54</td>
<td>2.74</td>
<td>1.52</td>
<td>1.71</td>
<td>1.87</td>
<td>2.37</td>
</tr>
<tr>
<td>$r_s$</td>
<td>2.40</td>
<td>2.31</td>
<td>3.44</td>
<td>4.05</td>
<td>5.02</td>
<td>5.36</td>
<td>5.83</td>
</tr>
<tr>
<td>$m^*$</td>
<td>0.84</td>
<td>0.89</td>
<td>1.32</td>
<td>1.06</td>
<td>1.02</td>
<td>0.96</td>
<td>0.93</td>
</tr>
<tr>
<td>$-B_0(r_s, m^*)$</td>
<td>0.40</td>
<td>0.42</td>
<td>0.52</td>
<td>0.30</td>
<td>0.41</td>
<td>0.49</td>
<td>0.61</td>
</tr>
<tr>
<td>$m^*=m_b$†</td>
<td>0.90</td>
<td>0.91</td>
<td>1.61</td>
<td>1.05</td>
<td>1.10</td>
<td>1.11</td>
<td>1.27</td>
</tr>
<tr>
<td>$-B_0(r_s, m^*)$</td>
<td>0.35</td>
<td>0.41</td>
<td>0.41</td>
<td>0.31</td>
<td>0.36</td>
<td>0.41</td>
<td>0.46</td>
</tr>
<tr>
<td>$-B_{0, \text{wcy}}$‡</td>
<td>—</td>
<td>—</td>
<td>0.46</td>
<td>0.38</td>
<td>0.36</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>

† From Ashcroft and Lawrence (1968) for In, Pb and from So et al (1977) for the alkali metals.
Complementing the above with the fact that \( m^* \) and \( r_s \) vary more or less linearly with concentration in metallic-like binary alloys we anticipate that it will be reasonable to apply a linear relation to obtain \( B_0 \) in these alloys. Accordingly, we set \( B_0 = \sum C_i B_{0i} \) for the alloy, \( B_{0i} \) being the pure metallic values. Finally, equation (3.5) yields \( \chi_s \). The results of this calculation are summarised in table 7.

### 3.3. Ionic and total magnetic susceptibilities

Since the ion cores are assumed to be independent of the environment, the concentration dependence of \( \chi_i \) in the alloy will be given by \( \chi_i = \sum C_j \chi_{ij} \), where \( \chi_{ij} \) is the \( \chi_i \) for the \( j \)th component.

Now \( \chi_d \), \( \chi_s \) and \( \chi_i \), as just calculated, are combined to give \( \chi \) in the alloy and hence its concentration dependence. The corresponding results, i.e. \( \chi_i, \chi_s = \chi_{dt} + \chi_{st} \) (the total valence electron magnetic susceptibility) and \( \chi \) are displayed in figures 1(a) and (b) for Na–K and Na–Cs, respectively. However, before analysing these results the Li–Pb case will be discussed.

### 4. Magnetic susceptibility of Li–Pb alloys

The works of Ruppersberg and Egger (1975) and of Bhatia and Ratti (1976) on neutron scattering in the Li–Pb alloys indicate that short-range structure exists in these alloys. Thus a Li–Pb alloy containing a significant amount of Pb appears to be a ternary-like mixture containing primarily Li₄Pb molecular units plus separated Pb⁺⁺ and Li⁺ ions. Hence, at first sight, the formulation applied in § 3 is inappropriate for Li–Pb alloys. In addition, in the vicinity of \( C_p = 0.20 \) the alloy appears to undergo a phase transition to a non-metallic phase. Nevertheless, we attempt a description of Li–Pb alloys using the concept of a charge transfer to modify the procedure presented in § 3.

Now, we must recognise that neutron diffraction gives virtually no information about the electronic structure of a system. For this we turn to available electronic properties. Nguyen and Enderby (1977) measured the electronic transport coefficients for the Li–Pb

<table>
<thead>
<tr>
<th>A–B</th>
<th>( C_B )</th>
<th>( r_s )</th>
<th>( m^* )</th>
<th>( -B_0(r_s, m^*) )</th>
<th>( \chi_s/\chi_d )</th>
<th>( \chi_s/\chi_{st} )</th>
<th>( \chi_s )</th>
</tr>
</thead>
<tbody>
<tr>
<td>K–Rb</td>
<td>0.8</td>
<td>5.29</td>
<td>0.97</td>
<td>0.47</td>
<td>1.83</td>
<td>27.06</td>
<td>49.52</td>
</tr>
<tr>
<td>Na–K</td>
<td>0.4</td>
<td>4.49</td>
<td>1.04</td>
<td>0.34</td>
<td>1.58</td>
<td>19.49</td>
<td>30.79</td>
</tr>
<tr>
<td>Na–K</td>
<td>0.7</td>
<td>4.77</td>
<td>1.03</td>
<td>0.37</td>
<td>1.63</td>
<td>22.05</td>
<td>35.94</td>
</tr>
<tr>
<td>Na–Cs</td>
<td>0.3</td>
<td>4.73</td>
<td>1.05</td>
<td>0.39</td>
<td>1.72</td>
<td>21.65</td>
<td>37.24</td>
</tr>
<tr>
<td>Na–Cs</td>
<td>0.5</td>
<td>5.10</td>
<td>1.03</td>
<td>0.46</td>
<td>1.91</td>
<td>25.22</td>
<td>48.17</td>
</tr>
<tr>
<td>Na–Cs</td>
<td>0.7</td>
<td>5.37</td>
<td>1.01</td>
<td>0.52</td>
<td>2.10</td>
<td>27.93</td>
<td>58.65</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>0.0</td>
<td>3.44</td>
<td>1.32</td>
<td>0.52</td>
<td>2.74</td>
<td>11.43</td>
<td>31.32</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>0.4</td>
<td>2.71</td>
<td>0.96</td>
<td>0.47</td>
<td>1.81</td>
<td>15.65</td>
<td>28.33</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>0.6</td>
<td>2.58</td>
<td>0.92</td>
<td>0.45</td>
<td>1.67</td>
<td>18.06</td>
<td>30.16</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>0.8</td>
<td>2.48</td>
<td>0.88</td>
<td>0.42</td>
<td>1.52</td>
<td>20.22</td>
<td>30.73</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>1.0</td>
<td>2.40</td>
<td>0.84</td>
<td>0.40</td>
<td>1.39</td>
<td>22.28</td>
<td>30.97</td>
</tr>
<tr>
<td>Li–Pb</td>
<td>0.2</td>
<td>2.96</td>
<td>0.88</td>
<td>0.45</td>
<td>1.60</td>
<td>11.91</td>
<td>19.06</td>
</tr>
</tbody>
</table>
Magnetic susceptibilities of liquid binary alloys

Figure 1. Various theoretical susceptibilities and $f$ as functions of concentration for alkali-metal binary alloys at or just above their liquidus temperatures.

system and reported results (see figure 2 for the thermoelectric power, $Q$) that are simple metallic-like at all concentrations except for a small range about $C_{Pb} = 0.2$. Having formulated (Wang et al. 1980a) the theory for the transport coefficients of the liquid binary alloys of simple metals using the NFE type approach, Wang and Lai (1980b, to be referred to as III) examined the extent to which this formulation could be used with the Li-Pb like alloys. First, they used the usual NFE approach in which all of the valence electrons are treated as conduction electrons and found results for $Q$ in reasonable agreement with observation only for $C_{Pb}$ greater than 0.4 and less than 0.1 (see figure 2). This result is

Figure 2. Theoretical and experimental thermoelectric power, $Q(C_{Pb})$, for Li–Pb alloys.
consistent with recent conclusions (Lai and Wang 1982) on the applicability of the usual NFE approach to liquid binary alloys. Second, in order to reproduce the observed main features of the concentration dependence of \( Q \) for \( C_{\text{Pb}} \) between 0.1 and 0.4, they introduced a modified NFE approach. In this case, some fraction of the valence electrons are considered to be bound on the more electronegative ion (the Pb\(^{4+}\) in Li–Pb alloys) from the charge-transfer point of view and only the remaining valence electrons are treated as conduction electrons; that is, as carriers in a NFE approach. In this connection we note that since the electrical resistivity, \( \rho = m^* (nr)^{-1} \), depends on the product of the carrier density, \( n \), and the scattering time, \( \tau \), one can have a significant increase in \( \rho \) and still remain in the weak scattering limit (NFE limit) if there is a correspondingly significant decrease in \( n \), for example, caused by a partial localisation of the valence electrons on the ions (II). Now the average electron charge, \( \nu \), needed to be localised around a Pb ion in order to fit \( Q \) at \( C_{\text{Pb}} = 0.2 \) was found (III) to be 0.664. Of course, the ability of a particular model to fit a single property is not a sufficient test of that model. Hence the modified NFE approach was used to interpret the anomalous temperature dependence of \( \rho \), i.e. \( d\rho (C_{\text{Pb}})/dT \), in Li–Pb alloys recently (Lai and Wang 1981). Using \( \nu = 0.664 \) results in a calculated value of the above derivative at \( C_{\text{Pb}} = 0.2 \) of \( -0.347 \mu \Omega \text{ cm deg}^{-1} \). The corresponding experimental value is \( -0.514 \mu \Omega \text{ cm deg}^{-1} \). In order to obtain reasonable agreement with this experimental value one requires \( \nu \approx 1 \). Now having two values of \( \nu \), \( Q \) at \( C_{\text{Pb}} = 0.2 \) was recalculated for \( \nu = 1 \) and the result is shown by the full triangle in figure 2. The theoretical \( Q(C_{\text{Pb}}) \) curve has the general character of the experimental curve and because of the sensitivity of \( Q \) to the details of the form factors and the liquid structure factors at \( q = 2k_F \) we consider the agreement with \( \nu = 1 \) quite reasonable (cf theoretical and experimental \( Q \) for Li in figure 2). Thus we favour the value \( \nu = 1 \). The picture emerging from above is that for electronic dynamic properties the modified NFE approach with binary-type liquid structure factors applies to Li–Pb alloys and differs significantly from the usual NFE approach for \( 0.1 < C_{\text{Pb}} < 0.4 \), because in this range of \( C_{\text{Pb}} \) \( \nu \) is too large to allow all the valence electrons to be treated as nearly free. At this point it is worth noting that the modified NFE approach gives a very poor result for \( \rho \) of the Li\(_{12}\)Bi\(_{25}\) alloy, which is much more non-metallic than the Li\(_{10}\)Pb\(_{20}\) alloy.

We now speculate that the distinguishing feature between these two systems is the lifetime of the molecular units, being much longer in the Li\(_{15}\)Bi\(_{25}\) system than the \( 10^{-12} \) s (Soltwisch et al 1980) in the Li\(_{10}\)Pb\(_{20}\) system. We further anticipate that this lifetime becomes considerably shorter in an externally applied field due to the perturbation that this field produces on the states of the valence electrons being responsible for the binding of a molecular unit. Thus, in an external field, the number of molecular units decreases significantly and the ionic structure becomes very dynamic with the continual creation and annihilation of molecular units. Hence, even in the scattering time of the nearly free carriers (about \( 10^{-14} \) s) the effective scattering potentials seen by the carriers are those of the Li and Pb ions in the modified NFE approach. This is not the case in Li\(_{15}\)Bi\(_{25}\). We suggest that this is the reason why the modified NFE approach will work for the Li–Pb type of alloys.

We now look to see where the above conjecture leads, when applied to the calculation of the electronic susceptibilities in the Li–Pb alloys. To this end, as in obtaining the theoretical \( Q(C_{\text{Pb}}) \) curve in figure 2, we apply the NFE approach to the Li–Pb alloys for \( C_{\text{Pb}} \geq 0.4 \) and the modified NFE approach to the Li–Pb alloy at \( C_{\text{Pb}} = 0.2 \). Now the expressions for \( \chi_d \) using the usual NFE approximation in § 3.1 can be applied directly to the alloys with \( C_{\text{Pb}} = 0.4, 0.6 \) and \( 0.8 \), the results being given in table 5. For the application of the modified NFE approach to the calculation of \( \chi_d \) for the Li–Pb alloy at \( C_{\text{Pb}} = 0.2 \), we
follow III and consider this alloy to consist of (i) a set of Li ions with \( Z_{Li} = 1 \) unchanged, (ii) a set of Pb ions with \( Z_{Pb} = 4 - \nu \) (\( \nu = 1 \) from above) (here the localised charge on a Pb ion is treated as a point charge, this being similar to the treatment of a depletion charge in OPW type pseudopotential theory) and (iii) only \( n_e(1-f) \) nearly free electrons rather than the usually used value (the valence electron density \( n_e \)). Here, \( f \) is associated with the valence electron charge transferred to each electronegative ion by the equation

\[
\nu = f(C_A Z_A + C_B Z_B)C_B^{-1}
\]  

(4.1)

where \( C_A \) and \( C_B \) are the concentrations of the electropositive and electronegative ions, respectively, and \( Z_A \) and \( Z_B \) their corresponding valences. Because \( f=0 \) for the pure metals for which \( \nu = 0 \), \( f \) may be interpreted as the fraction per valence electron localised and is 0.125 for the Li\(_{80}\)Pb\(_{20}\) alloy. The corresponding total diamagnetic susceptibility due to all of the valence electrons may be written as

\[
\chi_d = \chi_{d1} + \chi_{d2}.
\]  

(4.2)

\( \chi_{d1} \) is the contribution from the \( n_e(1-f) \) nearly free electrons and is calculated using the methods of § 3.1. \( \chi_{d2} \) is the contribution due to the \( \nu \) electrons localised on each Pb ion, being on average in the 6s like atomic state, and is estimated using the Slater-type method (Slater 1930, Angus 1932), which appears more reliable than other methods (Myers 1952, Dupree and Seymour 1970). We obtain \( \chi_{d2} = -1.52 \times 10^{-6} \text{ cm}^3 \text{ mol}^{-1} \). The results and input data are listed in table 8.

For \( \chi_i \), we assume, for simplicity, that the ion cores in the Li–Pb alloys are unaffected by the changes in the valence electron structure. Hence, \( \chi_i \) can be obtained as discussed in § 3.3, the results being displayed in figure 3.

Next, we note that the Li–Pb alloys are NFE-like and \( r_s \) is small in the concentration range \( C_{Pb} \geq 0.4 \). Hence the methods of § 3.2 apply to the calculation of \( \chi_i \) here and using the values in table 7 yield the results for \( \chi_s \) in this table.

The linear procedure, § 3.2, of estimating \( B_0(r_s, \mu^*) \) and hence \( \chi_s/\chi_{mf} \) does not apply to the \( n_e(1-0.125) \) nearly free electrons in the Li–Pb alloy at \( C_{Pb} = 0.2 \) because the calculated \( \mu^* \) at this point, being 0.88 au, differs significantly from the linearly interpolated value of 1.224 au. However, since the corresponding \( r_s = 2.96 \) we conclude that the functional dependence of \( B_0 \) on \( r_s \) and \( \mu^* \) lies in the range of \( r_s \leq 3.5 \) in the pure metallic case (§ 3.2) and is about the same as that shown in table 7 for Li, Pb and their metallic binary alloys. Using this table, if the \( r_s \) dependence is ignored we would take \( B_0 \approx -0.42 \) at \( \mu^* = 0.88 \) au and if the \( \mu^* \) dependence is ignored we would take \( B_0 \approx -0.48 \) at \( r_s = 2.96 \). Notice that a decreasing magnitude of \( B_0 \) with decreasing \( r_s \) is consistent with previous work (Rice 1968, Dupree and Geldart 1971). Taking both \( \mu^* \) and \( r_s \) dependence together implies \( B_0 \approx -0.45 \) for Li–Pb at \( C_{Pb} = 0.2 \) for the nearly free conduction electrons. Now \( \chi_s/\chi_{mf} = 1.60 \) which is only 10% greater than the well-known first-principles calculation value of 1.45 (Dupree and Geldart 1971). We note that for this partial localisation model,

| Table 8. Input data for the calculation of \( \chi_{d1} \) and calculated value of \( \delta_d + \delta_e \) and of \( \chi_{d1} \) for the Li\(_{80}\)Pb\(_{20}\) alloy at \( T = 800 \text{ °C} \). Included is also \( \chi_{d1} \) plus estimated \( \chi_{d2} \), that is, \( \chi_d \) (see text for details and tables 1 and 2 for units). |
|---|---|---|---|---|---|---|---|
| \( \Omega_0 \) | \( \eta_{Li} \) | \( \eta_{Pb} \) | \( r_s \) | \( \chi_i \) | \( m^* \) | \( \delta_d + \delta_e \) | \( -\chi_{d1} \) | \( -\chi_d \) |
| 152.4 † | 0.310 | 0.078 | 2.96 | 7.94 | 0.88 | 0.15 | -4.68 | 6.20 |
| † From Ruppersberg and Egger (1975). |
as used in this work, no contribution to the paramagnetic susceptibility is expected from the spins of the localised valence electron charges on the electronegative ions in the Li$_{80}$Pb$_{20}$ alloy. This arises because the dynamic nature of the formation of molecular units in this system should maintain a zero average spin for the bound valence electrons.

Now, summing the $\chi_d$, $\chi_l$ and $\chi_s$, we obtain $\chi$ for the Li–Pb alloys, as displayed in figure 3, in which $\chi = \chi_s + \chi_d$; that is, the total valence electron magnetic susceptibility.

5. Discussions and conclusions

In order to discuss the concentration dependence of $\chi$ in relation to $f$, appearing in equation (4.1), we also displayed the values of $f$ from II in figures 1(a) and (b) and those from § 4 for Li$_{80}$Pb$_{20}$ and III for the other Li–Pb alloys in figure 3. It is interesting to note that the deviation of $\chi(C_B)$ from the linear relation, $C_A\chi_A + C_B\chi_B$, for those alloys with small $f$, figure 1, is similar to the experimental results for the Na–Cd and Na–Tl liquid alloys (Uemura et al 1978). Examination of the results in figure 3 shows that $\chi$, via $\chi_s$ (or $\chi_d$), has an anomalous behaviour only when the Li–Pb system passes from a NFE metallic-like phase into a modified NFE non-metallic like phase, as the experimental $\chi$ indicates (figure 3). This occurs when $f$ and hence the number of conduction electrons (carriers) varies strongly with concentration and is consistent with the conclusion drawn in III and

Summarising, in this work a pseudopotential procedure, originally formulated in I and re-examined in Wang et al (1980b), was used first to calculate the conduction electron diamagnetic susceptibility, $\chi_d$, for pure simple liquid metals and was then extended to enable the calculation of this same quantity in some liquid binary alloys of these metals. For the latter case we used (i) the NFE non-local pseudopotential procedure, developed in Wang et al (1980a) for binary alloys, for the metallic like binary alloys and (ii) the modified NFE model re-examined in § 4 for the non-metallic like Li$_{80}$Pb$_{20}$ alloy. The calculated results show that, for the metallic like binary alloys, (a) $\chi_d$ differs from the free-electron value, $\chi_{df}$, by from 0 to 37% and (b) the concentration dependence of $\chi_d/\chi_{df}$ goes from linear to non-linear as the charge transfer goes from smaller to larger values.

Also, the ion core diamagnetic susceptibilities from Angus (1932) were used along with the $\chi_d$ calculated to infer the conduction electron spin susceptibilities, $\chi_s$, in the pure simple liquid metals from the observed total magnetic susceptibilities. Thus, the values of the Landau scattering function, $B_0$, were deduced, using equation (3.5) for these pure metals. Next, values of $B_0$ for the metallic like binary alloys were obtained as functions of concentration using a linear interpolation. It is clear (tables 6 and 7) that $B_0$ depends not only on $r_s$ but also on $m^*$, that is, $B_0 = B_0(r_s, m^*)$. Finally, using these $B_0$ the concentration dependence of $\chi_s$ for the alloys was deduced. At present, it is difficult to determine the error inherent in $\chi_s$ when determined in this way because no experimental values are yet available. However, the functional dependences are slowly varying and hence the results should be reasonable.

The presently obtained results for the concentration dependence of the total magnetic susceptibility, $\chi$, for those binary mixtures of simple metals with small $f$ are consistent with the smoothly varying $\chi$ observed for the Na–Cd and Na–Tl liquid alloys (Uemura et al 1978). On the other hand, the observed dramatic variation of $\chi$ for Li–Pb, in which $f$ is large, is also reproduced. Thus it appears from the present study that the modified NFE procedure, as suggested in III and used recently (Lai and Wang 1981) for a determination of the electron transport coefficients in Li–Pb alloys for $0.1 \leq C_{Pb} \leq 0.4$ also provides a description for the present problem.

Acknowledgment

This work was supported in part by the Natural Sciences and Engineering Research Council of Canada.

References

Angus W R 1932 Proc. R. Soc. A 136 569–78
Hackstein K, Sotier S and Lüscher E 1980 J. Physique Coll. 41 C8 49–51
Myers W R 1952 Rev. Mod. Phys. 24 15–27
Nguyen V T and Enderby J E 1977 Phil. Mag. 35 1013–9
Slater J C 1930 Phys. Rev. 36 57–64
Soltwisch M, Quitmann D, Ruppersberg H and Suck J B 1980 J. Physique Coll. 41 C8 167–70
——— 1980b J. Physique Coll. 41 C8 535–9