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# ELECTRICAL RESISTIVITIY OF LIQUID Ag-Au ALLOY *

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ABSTRACT

Calculations of the dependence of the electrical resistivity in liquid Ag-Au binary alloy on composition are reported. The structure of the binary alloy is described as a hard-sphere system. A one-parameter local pseudopotential, which incorporates s-d hybridization effects phenomenologically, is employed in the resistivity calculation. A reasonable agreement with experimental trend is observed.

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## I. INTRODUCTION

The pseudopotential theory has proved to be a powerful, theoretical tool in the study of electronic properties of simple liquid metals. Some difficulties are expected to arise when the method is applied to the noble, transition and rare earth metals. In the noble metals Ag and Au, for example, narrow d-bands are filled and lie few ev below the Fermi energy. The experimental studies suggest that d-bands are not seriously affected by melting i.e. by liquid disorder. The pseudopotentials are energy dependent and non-local. One may thus expect that the use of local pseudopotentials in the liquid noble metals will not give reasonable results. But a number of studies with local pseudopotentials have already proved to be successful [1]. It is interesting to further test this simple approach in the case of alloys as a first order approximation.

Relatively few calculations exist for liquid noble metal alloys. The main reason may be the non-availability of experimentally determined partial structure factors. In this paper, we study the electrical resistivity of the liquid Ag-Au alloy.

The plan of the paper is as follows. In Sec.II, we present a brief discussion of the model pseudopotential and partial structure factors we have used in the diffraction theory of Faber and Ziman [2] for electrical resistivity of liquid binary alloys. In Sec.III, we present our results on the composition dependence of electrical resistivity along with a brief summary of our conclusions.

## II. THE ELECTRICAL RESISTIVITY OF NOBLE METAL BINARY ALLOYS

The well-known Faber-Ziman formula for the resistivity of a liquid binary alloy modified by Ashcroft and Langreth [3];

$$\rho = \frac{4\pi^3\hbar Z^*}{e^2 k_F}\int_0^1 dy\, y^3\left\{x V_2^2(y) S_{22}(y) + 2\left[x(1-x)\right]^{1/2} V_1(y) S_{12}(y) V_2(y) + (1-x) V_1^2(y) S_{11}(y)\right\}, \qquad (1)$$

where

$y = q/2k_F$, $x$ = concentration of species 2, $Z^*$ = effective valence,

requires a knowledge of electron-ion pseudopotentials $V_i$ and partial structure factors $S_{ij}$. In the following, we give a brief discussion of each of these factors.

A. Model Pseudopotentials

The simplest of the commonly used model pseudopotentials in the study ofelectronic properties of liquid metals is the Ashcroft empty-core potential,

$$V_i(y) = -\frac{\lambda^2 \cos(S_i y)}{\lambda^2 + \lambda^2 f(y)} \quad , \tag{2}$$

where $S_i = 2k_F R^i_{core}$, $\lambda^2 = (\pi a_0 k_F)^{-1}$ and $f(y)$ is the Linhard dielectric function. In their study of the electrical resistivity Ashcroft and Langreth have chosen $R_{core}$ to fit the resistivity of the molten metal at both ends of the concentration. A reasonable agreement with experiment is claimed.

In this study, the model pseudopotential recently introduced by Idrees et al. [1] is employed. The potential has the form,

$$V^b(r) = \begin{cases} \dfrac{Ze^2(R_m^2 - R_w r)}{R_w^2 (R_w - R_m)} & 0 < r < R_m \\ -2\dfrac{Ze^2}{R_w} & R_m < r < R_w \\ -\dfrac{Ze^2}{r} & R_w < r \quad . \end{cases} \tag{3}$$

So that, it is repulsive in the region from $r = 0$ to $r = R_m^2/R_w = R_m$ (core radius), attractive and constant in the region $r = R_m$ to $r = R_w$ $(=(3\Omega/4\pi)^{1/3}$, the Wigner-Seitz radius) and purely Coulombic beyond. The attractive and constant part is claimed to give an improved account of the s-d mixing which has been introduced phenomenologically. It is important to note that



this is a one-parameter potential. The Fourier transform of Eq.(3) is given by

$$V^b(q) = \frac{4\pi}{\Omega q^3}\Big[(AR_m^2 + B - AR_wR_m)(\sin qR_m - qR_m \cos qR_m) - B(\sin qR_w - qR_w \cos qR_w) - \frac{AR_w}{q}(qR_m \sin qR_m + 2(\cos qR_m - 1)) - Ze^2 q \cos qR_w\Big] \quad .$$

where

$$A = Ze^2/R_w^2(R_w - R_m)$$
$$B = 2Ze^2/R_w \quad . \tag{4}$$

The parameter $R_m$ is fixed by fitting the theoretical factor Eq.(4) to the experimental one or to a better calculation. The electron-ion pseudopotential to be used in Eq.(1) is the screened one. We have used both Shaw [4] and Vashista-Singwi [5] dielectric functions without any significant difference in the final result. This may be due to the fact that in the diffraction theory of Faber and Ziman for liquid metal alloys, the electrical resistivity is mainly determined by the structure factors in the region of wave number $q$ less than but close to the value $2k_F$. And the role of conduction electron screening is becoming less important and almost equivalent in this region according to the prescriptions mentioned above.

B. Partial Structure Factors

For the partial structure factors, the hard sphere solution of the Percus-Yevick equation is used. The analytic expressions for these are given by Ashcroft and Langreth [6]. (Their original paper contains a number of typographical errors which are corrected in a following Errata.) We give the correct expressions in the Appendix for the sake of completeness.

## III. NUMERICAL RESULTS AND CONCLUSIONS

The input parameters and the model pseudopotentials appropriate for Ag and Au are taken from Ref.[1]. Liquid metal properties like electrical resistivities, thermoelectric power and effective ion-ion interactions for liquid Ag and liquid Au, based upon these pseudopotentials are discussed at some length in their paper. The results seem to be somewhat better that those of other investigations. Thus, our work can be considered as an extra test of the proposed pseudopotentials.

We have checked the accuracy of the hard sphere structure factors by comparing them with the experimental results for pure liquid Ag and Au at both ends of the concentrations range. The experimental values are taken from Y. Waseda [7]. The results seem to be reasonable as shown in Figs.1 and 2. The partial structure factors are shown in Fig.3.

The concentration dependence of the calculated electrical resistivity of liquid Ag-Au system is shown in Fig.4. The experimental trend is generated well. In intermediate concentrations the model pseudopotential and hard-sphere structure factors used seem to be overestimating the resistivity. To get a better numerical agreement with experimental resistivity values (taken from Roll and Motz [8] as noted by Howe and Enderby [9]) one is forced to use hard sphere diameters which tend to result in structure factors that are in disagreement with experimental ones.

In our calculations, we have made use of the fact that the atomic volume and the Fermi wave number of the alloy remain practically constant across the whole concentrations range [9]. Thus, the atomic volume of the alloy is linearly interpolated between the values of the pure components. The Fermi wave number is calculated with this atomic volume. The packing fraction is taken to be 0.45.

Our results confirm the generalization that the resistivity of binary alloys is a "smooth" function of concentration at constant temperature. The addition of Au in Ag increases the resistivity in an expected manner. And the use of local pseudopotentials even in the case of noble metals seems to be reasonable. But it is still interesting to see what role any non-locality of the pseudopotential will play in binary alloys. Work along these lines is in progress.



## APPENDIX

The analytical expressions for the partial structure factors $S_{ij}(y)$ obtained by Ashcroft and Langreth in the hard-sphere Percus-Yevick model are as follows:

$$S_{11}(y) = \left[1 - n_2 C_{22}(y)\right] / D(y)$$

$$S_{22}(y) = \left[1 - n_1 C_{11}(y)\right] / D(y)$$

$$S_{12}(y) = (n_1 n_2)^{1/2} C_{12}(y) / D(y)$$

$$D(y) = \left[1 - n_1 C_{11}(y)\right]\left[1 - n_2 C_{22}(y)\right] - n_1 n_2 C_{12}^2(y) \ ,$$

where $y = q\,\sigma_2$, $\sigma_2$ is the hard-sphere diameter of the larger sphere and $n_i$ are number densities of the ith component. The total packing fractions $\eta$, the ratio of the hard-sphere diameters $\alpha$ and the concentration $x$ are defined as

$$\eta = n_1 + n_2 = \pi\, n_1\, \sigma_1^3/6 + \pi\, n_2\, \sigma_2^3/6$$

$$\alpha = \sigma_1/\sigma_2 \qquad (0 \leq \alpha \leq 1)$$

$$x = n_2/(n_1 + n_2) \ .$$

The direct correlation functions $C_{ij}(y)$ can be expressed in terms of parameters $\eta$, $\alpha$ and $x$ and are given by the following expressions.

$$-n_i C_{11}(y) = \frac{24\eta_1}{\alpha^3 y^3}\left\{ a_1\left(\sin\alpha y - \alpha y \cos\alpha y\right) + \frac{\beta_1}{\alpha y}\left[2\alpha y \sin\alpha y - (\alpha^2 y^2 - 2)\cos\alpha y\right] + \frac{\gamma_1}{\alpha^3 y^3}\left[(4\alpha^3 y^3 - 24\alpha y)\sin\alpha y - (\alpha^4 y^4 - 12\alpha^2 y^2 + 24)\cos\alpha y + 24\right]\right\}$$

To obtain $-n_2\, C_{22}(y)$, simply replace $\eta_1$ by $\eta_2$, $\alpha_1$ by $\alpha_2$, $\alpha y$ by $y$, $\beta_1$ by $\beta_2$ and $\gamma_1$ by $\alpha^{-3}\gamma_1$.

$$-(n_1 n_2)^{1/2} C_{12}(y) = 3(1-\alpha)^3 \eta\, x^{1/2}(1-x)^{1/2} \frac{a_1}{x+(1-x)\alpha^3} \frac{\sin y_\lambda - y_\lambda \cos y_\lambda}{y_\lambda^3}$$

$$+24\eta \frac{x^{1/2}(1-x)^{1/2}\alpha^3}{x+(1-x)\alpha^3}\left[\frac{\sin y_\lambda}{y_1^4}\left\{\beta_{12}\left[2y_1\cos y_1+(y_1^2-2)\sin y_1\right]\right.\right.$$

$$+\frac{\gamma_{12}}{y_1}\left[(3y_1^2-6)\cos y_1+(y_1^3-6y_1)\sin y_1+6\right]$$

$$\left.+\frac{\gamma_1}{y_1^2}\left[(4y_1^3-24y_1)\cos y_1+(y_1^4-12y_1^2+24)\sin y_1\right]\right\}$$

$$+\frac{\cos y_\lambda}{y_1^4}\left\{\beta_{12}\left[2y_1\sin y_1-(y_1^2-2)\cos y_1-2\right]\right.$$

$$+\frac{\gamma_{12}}{y_1}\left[(3y_1^2-6)\sin y_1-(y_1^3-6y_1)\cos y_1\right]$$

$$\left.+\frac{\gamma_1}{y_1^2}\left[(4y_1^3-24y_1)\sin y_1+(y_1^4-12y_1^2+24)\cos y_1+24\right]\right\}$$

$$+\frac{a_1}{y_1}\left\{\cos y_\lambda\left(\frac{\sin y_1-y_1\cos y_1}{y_1^2}+\frac{1-\alpha}{2\alpha}\,\frac{1-\cos y_1}{y_1}\right)\right.$$

$$\left.\left.+\sin y_\lambda\left(\frac{\cos y_1+y_1\sin y_1-1}{y_1^2}+\frac{1-\alpha}{2\alpha}\,\frac{\sin y_1}{y_1}\right)\right\}\right]$$

where

$$y_1=\alpha y\ ,\quad y_\lambda=\tfrac{1}{2}\,y(1-\alpha)\ ,\quad a_1=\frac{\partial}{\partial\eta_1}(\beta\beta')\ ,\quad a_2=\alpha^{-3}\frac{\partial}{\partial\eta_2}(\beta\beta')\ ,$$

$$\beta\beta'=\left\{(\eta_1+\alpha^3\eta_2)(1+\eta+\eta^2)-3\eta_1\eta_2(1-\alpha)^2\left[1+\eta_1+\alpha(1+\eta_2)\right]\right\}(1-\eta)^{-3}\ ,$$



$$\beta_1=-6\left[\eta_1 g_{11}^2+\tfrac{1}{4}\eta_2(1+\alpha)^2\alpha\, g_{12}^2\right]\ ,$$

$$\beta_2=-6\left[\eta_2 g_{22}^2+\tfrac{1}{4}\eta_1\alpha^{-3}(1+\alpha)^2 g_{12}^2\right]\ ,$$

$$g_{11}=\left[(1+\tfrac{1}{2}\eta)+\tfrac{3}{2}\eta_2(\alpha-1)\right](1-\eta)^{-2}\ ,$$

$$g_{22}=\left[(1+\tfrac{1}{2}\eta)+\tfrac{3}{2}\eta_1\left(\tfrac{1}{\alpha}-1\right)\right](1-\eta)^{-2}\ ,$$

$$g_{12}=\left[(1+\tfrac{1}{2}\eta)+\tfrac{3}{2}\frac{1-\alpha}{1+\alpha}(\eta_1-\eta_2)\right](1-\eta)^{-2}\ ,$$

$$\gamma_1=\eta_1 a_1+\alpha^3\eta_2 a_2\ ,$$

$$\gamma_{12}=2\gamma_1\left(\frac{1-\alpha}{\alpha}\right)\ ,$$

$$\beta_{12}=-3\alpha(1+\alpha)\left(\alpha^{-2}\eta_1 g_{11}+\eta_2 g_{22}\right) g_{12}\ .$$

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## FIGURE CAPTIONS

Fig.1 Comparison of theoretical and experimental structure factors for iquid Ag.

Fig.2 Comparison of theoretical and experimental structure factors for liquid Au.

Fig.3 Calculated structure factors $S_{ij}(y)$ for the combinatuons $n = 0.45$, $x = 0.50$ and $\alpha = 0.95$.

Fig.4 The resistivity of liquid Ag-Au. • Present results.
x Experimental values from Roll and Motz (Ref.8).



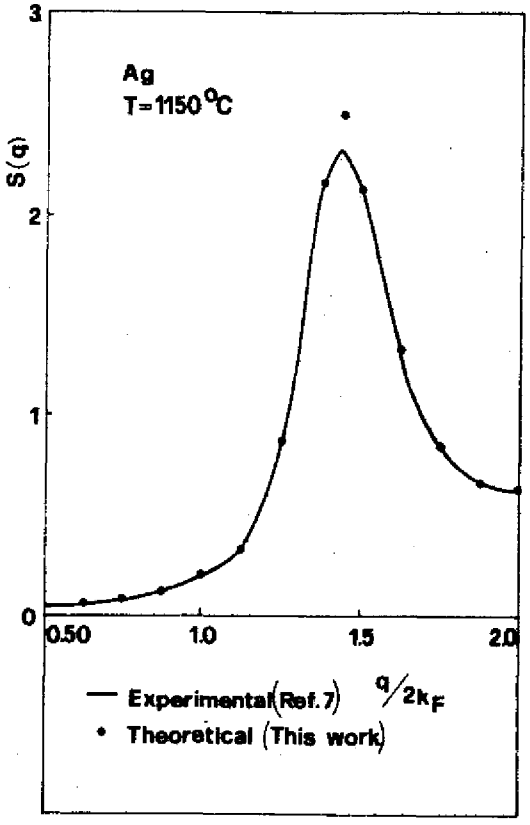


Fig. 1

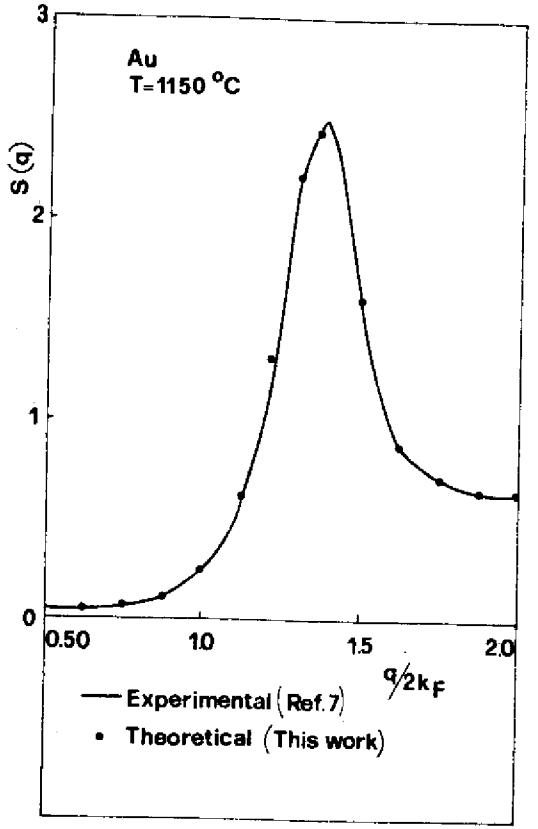
Au
T=1150 °C
S (q)
3
2
1
0
0.50
1.0
1.5
2.0
q/2k_F
Experimental (Ref. 7)
Theoretical (This work)

Fig. 2



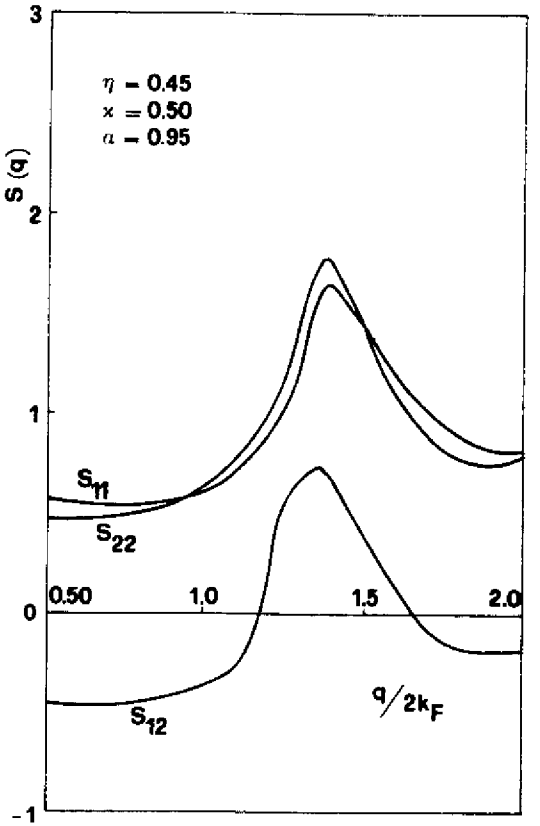
η = 0.45
x = 0.50
a = 0.95
S (q)
3
2
1
0
-1
S11
S22
S12
0.50
1.0
1.5
2.0
q/2k_F

Fig. 3

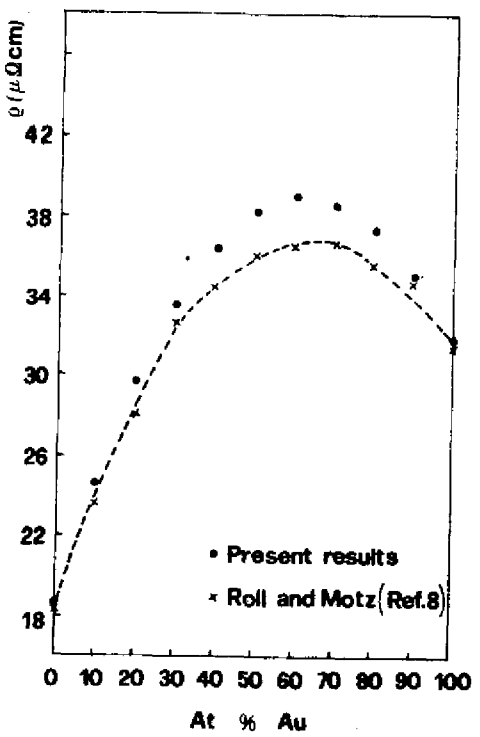


Fig. 4





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