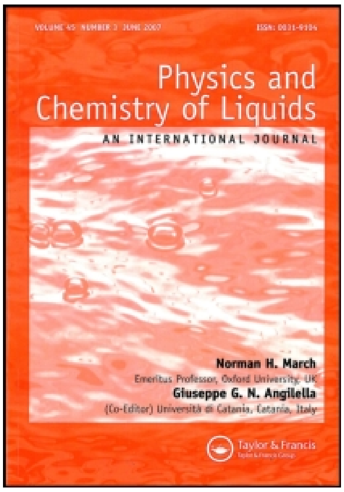

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THE ELECTRICAL RESISTIVITY OF LIQUID GERMANIUM-NICKEL ALLOYS

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The electrical resistivity of the liquid $Ni_x\ Ge_{(1-x)}$ system has been measured from pure Germanium to 77 at. % nickel. The resistivity shows a maximum for nickel rich alloys and a minimum for the resistivity temperature coefficient. The resistivity is interpreted and discussed with the *t*-matrix formulation using hard-sphere structure factors.

KEY WORDS: Nearly free electron theory, *t*-matrix, resistivity, liquid alloys, germanium, nickel.

I INTRODUCTION

It is largely agreed that electrical transport in simple liquid metals has been well resolved from a theoretical point of view. In these liquid alloys the transport properties are well described by the nearly-free electron theory with Ziman's formula[1] in the case of pure metals and with that of Faber and Ziman[2] for alloys. With transition metals one uses the extended Faber-Ziman formula with the *t* matrix formalism. After recalling the basic formula in Section 2 and the experimental method in Section 3, we present and discuss our new experimental results for the resistivities of the Ge-Ni system and compare them with different calculated values.

II THEORY

Ziman[1] has shown that the electrical resistivity of a pure liquid metal can be computed using the expression:

$$\rho = \frac{3\pi m^2 \Omega_0}{4e^2 \hbar^3 k_f^6} \int_0^{2k_f} a(q) v(q)^2 q^3 \, dq \tag{1}$$

where $a(q)$ is the static structure factor, $v(q)$ the pseudo (or model)-potential form factor, Ω_o the atomic volume and k_f the Fermi wave vector. Other symbols have their customary meaning. This formula can be extended to binary alloys[2] by replacing the product $a(q)\ v(q)^2$ by:

$$[v_1(q)]^2[c(1-c)+c^2a_{11}(q)]+[v_2(q)]^2[c(1-c)+(1-c)^2a_{22}(q)] \\ +2v_1(q)\cdot v_2(q)\cdot c\cdot(1-c)\cdot[a_{12}(q)-1]$$

here c is the atomic fraction of constituent 1, the $a_{ij}(q)$ are the Faber-Ziman[3] partial structure factors describing a mixture of randomly distributed hard spheres with different diameters, $v_i(q)$ are the ith pseudopotential form factors *in the alloy*.

With the noble, transition and rare earth metals we use the scattering approach replacing the model potential form factor $[v_i(q)]^2$ by a t matrix $[t_i(q)]^2$ of specie i in the alloy, expressed in term of phase shifts[4]. The different contributions have been grouped in 4 terms:

$$c^2a_{11}(q)|t_1(q)|^2+(1-c)^2a_{22}(q)|t_2(q)|^2+c(1-c)a_{12}(q)[t_1(q)t_2^*(q)+t_1^*(q)t_2(q)] \\ +c(1-c)\{|t_1(q)|^2+|t_2(q)|^2-0.5[t_1(q)t_2^*(q)+t_1^*(q)t_2(q)]\}$$

We have calculated the Faber-Ziman structure factors $a_{ij}(q)$ using Ashcroft-Langreth[5] (A.L.) analytical hard sphere partial structure factors. We chose, as parameters, two hard sphere diameters for the two species and held them constant with concentration but not with temperature. These hard sphere diameters have been obtained from the pure metal data. At each temperature, the hard sphere diameters are deduced from the experimental densities of the pure metals, compiled by Crawley[6] and from the packing fractions given by the Waseda empirical law[7a] where the parameters A_i and B_i have been taken from Waseda's book[7b]. The packing fractions and the hard sphere diameters are:

$$\eta_i(T)=A_i\exp(-B_iT) \quad \text{and} \quad \sigma_i^3(T)=[6\eta_i(T)\Omega_0(T)/\pi]$$

In the alloy, we have taken into account the modification of the mean atomic volume $\Omega_0(T)$ which is the normalization of the form factor and of the Fermi wavevector k_f:

$$k_f^3=(3\pi^2Z(c)/\Omega_0(T,c))$$

where $Z(c)$ and $\Omega_0(T,c)$ are respectively the mean valence and the mean atomic volume of the alloy obtained by a linear interpolation of the pure metal values. However it was not possible to take into account the energy dependence of the phase shifts.

III EXPERIMENTAL METHOD

Resistivity measurements were performed by the four probes method using a quartz cell fitted with tungsten electrodes. This kind of cell has several advantages. The first one is the possibility of filling the capillary from the bottom in order to avoid the presence of bubbles by pressing the liquid metal in the secondary storage tank. Bubbles can be detected by the resistivity change which occurs when a variation of pressure of about 0.25 bar is applied over the sample. Indeed their sizes are modified and this effect is traduced by a different voltage drop. Bubbles can be eliminated either by clearing the capillary again or by increasing the pressure of argon. The second advantage is the possibility of changing the composition of the alloy during the experiment. The whole arrangement is heated under vacuum until the metals are melted. An absolute pressure of argon of 1 to 3 bars is then applied over the liquid sample and pushes the liquid alloy into the capillary tube. Full experimental details are described by Gasser[8] (thesis 1982). A stable constant current is furnished by a General Resistance DIAL DAS 86 generator. The voltage drop is measured with a 1 μV resolution 120000 points Hewlett Packard 3490 voltmeter. Thermoelectric e.m.f. are eliminated by inverting the current. The geometrical constant of the cell has been carefully calibrated with triple distilled mercury which has been afterwards eliminated

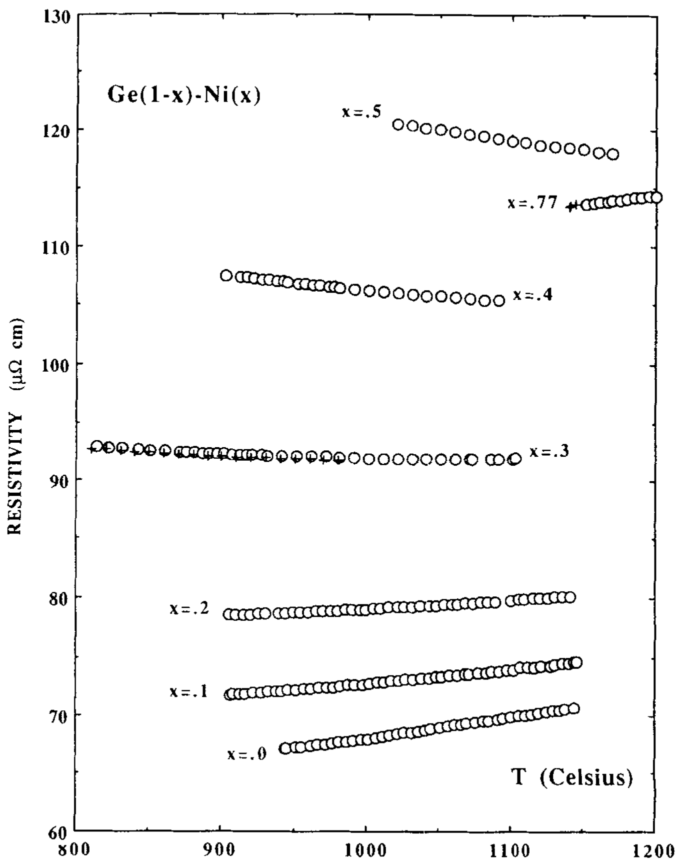

Figure 1 Electrical resistivity of liquid nickel-germanium alloys versus temperature.

by distillation. The accuracy of the electrical resistivity is estimated to 0.4%, that of the composition of the alloy to 0.3 at % and that of the temperature to 0.3%. The main error arises from the knowledge of the composition in the capillary which is estimated to 0.5 atomic %. Pure Germanium (Ge) and Nickel (Ni) have been used (99.999% purity).

IV EXPERIMENTAL RESULTS AND DISCUSSION

The electrical resistivity of Ni_x-Ge_{lx} alloys has ever been measured earlier by Güntherodt and Künzi[11]. Our experimental measurements are reported as a function of temperature in Figure 1 and as a function of concentration at 1000°C in Figure 2. The general shape of the curve is similar to Güntherodt and Künzi values. However important differences occur (about 15 $\mu\Omega$ cm in the middle of the phase diagram) from a quantitative point of view. The temperature coefficient is plotted in Figure 3. Here also, the agreement is only qualitative, not quantitative.

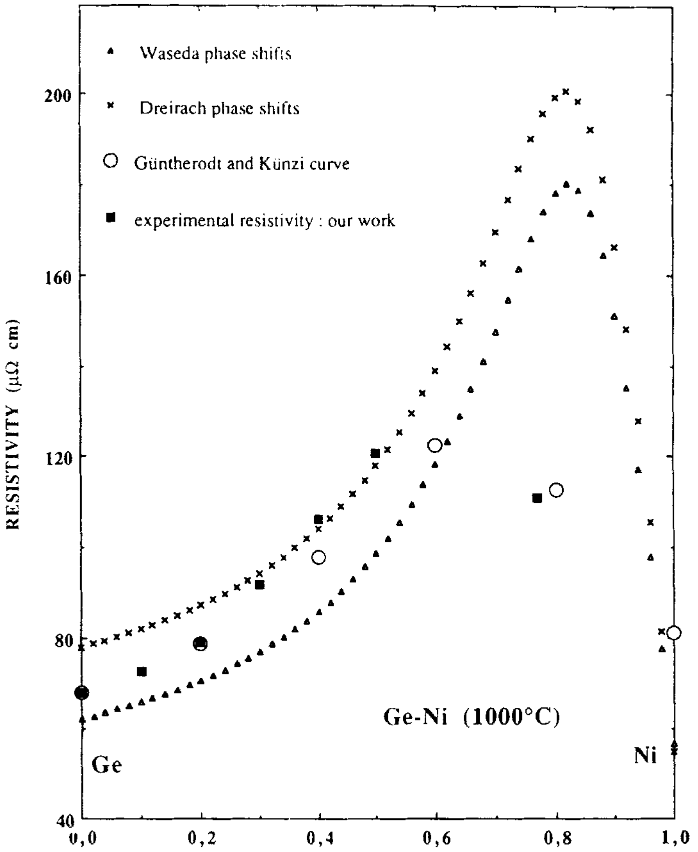

Figure 2 Resistivity of liquid nickel-germanium alloys versus nickel concentration.

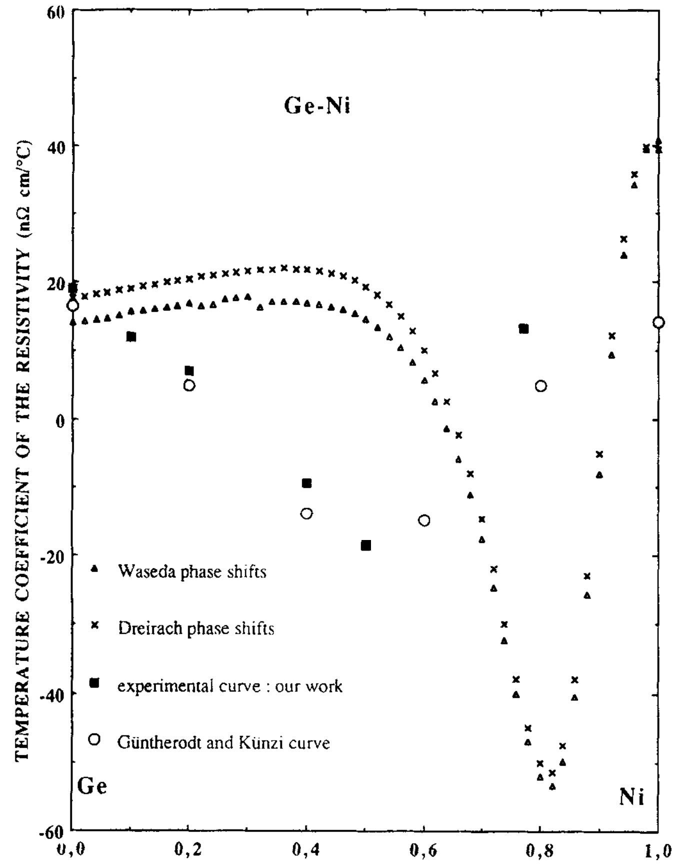

Figure 3 Temperature coefficient of the resistivity of liquid nickel-germanium alloys.

We have compared our experimental values to those obtained with the extended Faber-Ziman formula (Dreirach *et al.*[4]) We used Waseda's phase shifts and Dreirach for Germanium and Nickel. The results of our calculations are represented on Figure 2 and 3. For these alloys, there is evidence of a resistivity maximum in 50–77 at % Ni concentration range. The temperature coefficient becomes negative for Ni concentration greater than 30 at %. After passing through a minimum between 50–77 at % Ni concentration range, it becomes again positive. The maximum of ρ and the minimum of $d\rho/dT$ lie at approximately the same concentration. The behaviour of the Ni-Ge system shows a great similarity with that of Mn-Sn alloys studied by Gasser[8]. The resistivity curve is qualitatively represented by our calculations which gives a maximum for the resistivity versus concentration curve and a minimum for the temperature coefficient versus concentration curve. Nevertheless, these extrema are located near the middle of the phase diagram. The difference between experimental and calculated resistivities can be explained by the fact that the phase shifts determined for the pure metals cannot be used for alloys. The Fermi energy for pure Nickel is different from the pure Germanium one. The important energy dependence of the phase shifts, (especially the resonant η_2 phase shift of Nickel), can bring a justification to that difference.

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