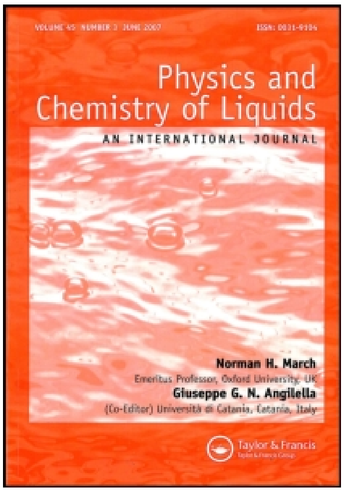

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MAGNETIC ANOMALIES IN LIQUID GOLD ALLOYS

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The anomalous variation of the magnetic susceptibility in liquid alloys of gold with polyvalent metals is discussed in terms of electron concentration and conventional pseudopotential theory. The characteristic minima of the susceptibility are shifted systematically towards the Au-rich side if the valency of the alloying element is increased, but considering the average electron concentration they all seem to occur at a critical electron to atom ratio of $e/a \approx 1.6$. They are obviously a manifestation of the electronic stabilization which gives rise to the formation of electron compounds in the critical range of composition.

Theoretical calculations based on simple model potentials reproduce the typical shape of the observed susceptibility curves and also their systematic variations due to the change in valency. The results indicate that the susceptibility minima are primarily due to interactions between the ionic cores and the electrons which increase the diamagnetic contribution of the conduction electrons to the electronic susceptibility.

Keywords: Magnetic susceptibility; electronic properties; liquid gold alloys

1. INTRODUCTION

In the past, considerable interest has been focused on the electronic transport properties of liquid alloys. As the number of papers on liquid metals and alloys increased, unexpected results have become apparent. The pioneering work of Endo [1] revealed a close relationship between the variation of the magnetic susceptibility and the appearance of compounds in the solid state. In a series of careful investigations focused on the anomalous properties of liquid copper alloys Takeuchi and coworkers [2] found strong evidence of a non-random arrangement of the constituent atoms. It seemed that electronic anomalies – characteristic maxima in the resistivity associated with negative temperature coefficients, or minima in the magnetic susceptibility – coincide with the pronounced deviations of the thermodynamic properties from a simple mixing behaviour, both as a result of chemical short range order prevailing at particular compositions. This is in fact a feature common to strongly interacting systems, but it does not necessarily apply to alloy systems with weak chemical interactions such as most of the noble metal – polyvalent metal systems. Based on experimental results on transport properties and thermodynamic quantities in liquid gold alloys, it has been concluded that the anomalies of different physical properties may appear in the same system with different intensities and even at different compositions [3–5].

Owing to the concepts based on theory of Faber and Ziman [6, 7], the electrical resistivities of liquid metals are, at least in a qualitative manner, well understood. Extrema in the resistivities, or negative temperature coefficients may come out from the theory, without the implication of compound formation or chemical short range order. The magnetic susceptibilities, on the other hand, are less understood, although practicable theoretical concepts are available [8, 9].

In the systems Au-In, Au-Ge and Au-Sb the magnetic susceptibility was found to pass through characteristic minima located on the Au-rich side. A tentative interpretation was given by assuming the formation of compounds or chemical complexes of plausible stoichiometry [3–5]. Inasmuch as such a treatment has a rather formal character, its physical relevance seems doubtful. The present paper is an attempt i) to analyse the experimental results in terms of a critical electron concentration and ii) to see whether the minima and their systematic variations with the valency of the alloying element can be reproduced by pseudopotential theory.

2. METHOD

The total magnetic susceptibility of metals χ_{tot} may be considered as superposition of a diamagnetic contribution arising from the ionic cores χ_{ion}, and an electronic contribution χ_{el} according to

$$\chi_{\text{tot}} = \chi_{\text{ion}} + \chi_{\text{el}} \qquad (1)$$

Since the underlying diamagnetism is assumed to be a characteristic property of the ionic cores under consideration (i.e., Au^{+}, In^{3+}, Ge^{4+}, Sb^{5+}), the analysis will be concentrated on the electronic part of the magnetic susceptibility which is obtained by subtracting the diamagnetic core contribution from the total susceptibility. As the values given in the literature for the same ionic species are very divergent, the choice of values is essential to the absolute values of χ_{el}. The electronic susceptibilities of the pure elements listed in Table I refer to the ionic core susceptibilities given by Hurd and Coodin [10], and alternatively to those compiled by Selwood [11]. The divergency between the two sets of χ_{el} amounts to about $4 \cdot 10^{-6}$ cm^3 mol^{-1} in the case of Au, $1 \cdot 10^{-6}$ cm^3 mol^{-1} for In and Ge, and $2 \cdot 10^{-6}$ cm^3 mol^{-1} for Sb, which adds to the experimental inaccuracy of $\pm 2 \cdot 10^{-6}$ cm^3 mol^{-1}. For practical reasons, in this paper all experimental data given for χ_{el} relate to the ionic susceptibilities quoted by Hurd and Coodin [10]. This choice is certainly debatable, but the analysis will be focused on the deviations from the additive law, i.e., rather on the shape of the experimental susceptibility vs composition curve.

For an assembly of free non-interacting electrons theory yields a simple expression which relates χ_{el} to the molar volume V_m and the number of valence electrons per atom Z.

$$\chi_{el}^{\circ} = \frac{2\mu_B^2 m_e N_A^{1/3}}{h^3 (3\pi^2)^{2/3}} \cdot V_m^{2/3} \cdot Z^{1/3} \tag{2}$$

As regards a more elaborate theoretical interpretation the electronic susceptibility of the conduction electrons χ_{el} has to be split into a

TABLE I Magnetic susceptibilities of liquid Au, In, Ge, and Sb: χ_{tot} total experimental susceptibility, χ_{ion} ion core susceptibility, χ_{el} electronic susceptibility (experimental), χ_{el}° free electron susceptibility (calculated)

	Magnetic Susceptibility in 10^{-6} cm^3 mol^{-1}				
	χ_{tot}	$\chi_{ion}^{1)}$	$\chi_{ion}^{2)}$	$\chi_{el}^{1)}$	$\chi_{el}^{2)}$	χ_{el}°
Au	−31.0	−36.17	−40.0	5.17	9.00	6.26
In	−7.6	−19.69	−19.0	12.09	11.40	12.43
Ge	4.9	−8.21	−7.0	13.11	11.90	11.10
Sb	−1.3	−15.92	−14.0	14.62	12.70	16.14

1), 2) The values are based on the ionic core susceptibilities of Au^{+}, In^{3+}, Ge^{4+} and Sb^{5+} taken from Ref. [10], and Ref. [11], respectively.

paramagnetic spin susceptibility term χ_{spin} and a diamagnetic contribution χ_{dia} which strongly depend on the interactions between the particles of such a simple model system. The appropriate relations can be given in terms of the free electron expressions $\chi^{\circ}_{\text{spin}}$ and $\chi^{\circ}_{\text{dia}}$ according to

$$\chi_{\text{spin}} = \chi^{\circ}_{\text{spin}} \cdot (I + \Delta_p) \cdot \delta^{\text{ec}}_p \tag{3}$$

$$\chi_{\text{dia}} = \chi^{\circ}_{\text{dia}} \cdot (I + \Delta_d) \cdot \delta^{\text{ec}}_d \tag{4}$$

The quantities Δ_p and Δ_d represent the corrections for the interaction between the ionic cores and the conduction electrons, while δ^{ec}_p and δ^{ec}_d take account of exchange and correlation effects between the electrons. For a nearly free electron system, Timbie and While [8] have derived expressions which relate Δ_p and Δ_d to integrals of the form:

$$\Delta_p = -\frac{3 \cdot Z}{32 \cdot E_f^2} \cdot \int_0^{\infty} a(q) \cdot \nu^2(q) \cdot G_p(q) \cdot dq \tag{5}$$

$$\Delta_d = -\frac{3 \cdot Z}{32 \cdot E_f^2} \cdot \int_0^{\infty} a(q) \cdot \nu^2(q) \cdot G_d(q) \cdot dq \tag{6}$$

The momentum transfer vector k is expressed in units of the Fermi vector k_f according to $q = k/k_f$, Z denotes the valency of the polyvalent metal, E_f the Fermi energy, $a(q)$ the structure factor, and $v(q)$ the pseudopotential from factor of the liquid metal. $G_p(q)$ and $G_d(q)$ are functions given as

$$G_p = 2q \cdot \ln\left|\frac{q+2}{q-2}\right| - \frac{8q^2}{q^2 - 4} \tag{7}$$

$$G_d = \frac{2}{q} \cdot (q^2 + 2) \cdot \ln\left|\frac{q+2}{q-2}\right| - \frac{8}{3} \cdot \frac{4 - 22q^2 + 3q^4}{(q^2 - 4)^2} \tag{8}$$

The corrections due to electron – electron interactions (δ^{ec}_p and δ^{ec}_d) were deduced from the electron sphere radius r_s as suggested by Wilk *et al.*

[12], and Kanazawa and Matsudaira [13]. It has been pointed out that the expression given for δ_d^{ec} might be inappropriate for metallic densities [8, 14], but its contribution to the principal form of the susceptibility vs composition curve is small.

The analytic form of the corrections adopted in this paper, the procedure of computing the integrals in view of the divergencies of $G_p(q)$ and $G_d(q)$ at $q=2$ as well as the adaptation of the formalism to the case of binary alloys are basically those of Dupree and Sholl [9]. Their calculations were based on the semiempirical model potential proposed by Borchi and DeGennaro [14] for liquid Au, empty core potentials for the polyvalent elements [15], and the structure factors of an hard sphere assembly [16]. Despite the simplifications their calculations were successful in reproducing the trends in certain noble metal – polyvalent metal systems. In this paper the form factors given by Heine, Abarenkov and Animalu [17, 18] were adopted for the polyvalent metals, as already practised by Timbie and White [8]. On account of the characteristic shoulder on the first maximum of the structure factor, a mixture of hard spheres interacting via an attractive square-well [19] was assumed to be representative of the liquid structure. The choice of input parameters has some arbitrary character, however, this paper is not aimed at fundamental aspects of theory, it is primarily concerned with physical trends of the systems.

The volumes of the pure liquid metals were taken from the compilation of Crawley [20], those of the alloys were assumed to be additive. In general, the effect of mixing on the volume of weakly interacting metallic systems is of the order of 2–3%, hence its influence on the principal shape of the susceptibility vs composition curve is expected to be rather small. The theoretical treatment will be focused on the systems Au-In, Au-Ge, and Au-Sb. The general reference temperature throughout the paper was chosen as 1300 K.

3. RESULTS AND DISCUSSION

The experimental electronic susceptibilities of the pure liquid elements (Au, In, Ge, Sb) are compared in Table I with the values predicted for free non-interacting electrons (Eq. (2)). In view of the high degree of incertainty of around $3-6\cdot10^{-6}$ cm^3 mol^{-1} the agreement seems to be acceptable, irrespective of the choice of core susceptibility. However, considering the results of the binary alloys (Figs. 1 – 3) it becomes apparent that neither the particular variation of the electronic susceptibility with the composition (i.e., the appearance of characteristic minima at certain concentrations) nor the systematic changes within the series from Au-In to Au-Ge and Au-Sb can be accounted for by such a simple expression. Eventual effects brought about by a non-ideal variation of the volume would be of about one order of magnitude smaller than the observed anomaly.

In a series of preceding papers [3 – 5] it has been attempted to assign the thermodynamic and the electronic anomalies to the simultaneous formation of two different types of compounds with the stoichiome-

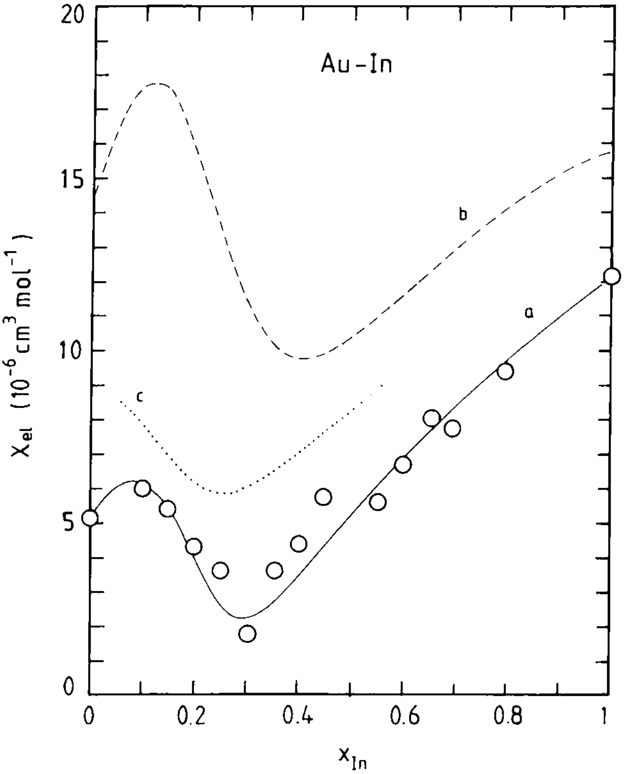

FIGURE 1 Experimental (a) and calculated (b,c) electronic suceptibility of liquid Au-In alloys at 1300 K.

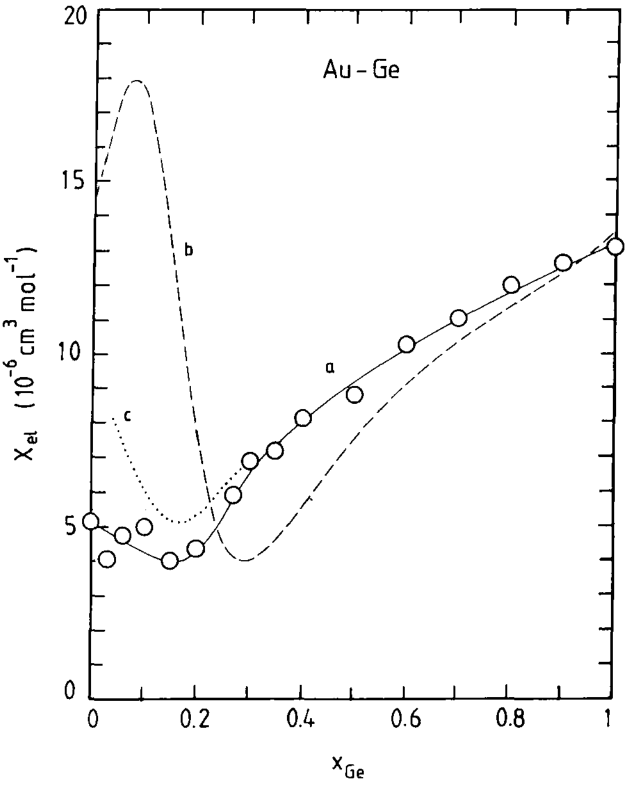

FIGURE 2 Experimental (a) and calculated (b, c) electronic suceptibility of liquid Au-Ge alloys at 1300 K.

tries AuIn and $AuIn_3$, AuGe and $AuGe_4$, and AuSb and $AuSb_5$ which reduce the abundance of free electrons and thus – according to Eq. (2) – also the electronic susceptibility. Despite the success in describing different quantities with the same model, this interpretation has to be considered of tentative character without real physical background.

The analysis in terms of electron concentration, on the other hand, indicates that the minima obviously occur at the same critical electron the atom ratio $e/a \approx 1.6$ electrons per atom. As a matter of fact, the minima in Au-In, Au-Ge and Au-Sb occur at $x_{In} \approx 0.30$, $x_{Ge} \approx 0.18$ and $x_{Sb} \approx 0.16$ which imply electron to atom ratios of 1.60, 1.54, and 1.64, respectively, Sodek *et al.* [21] reported for Au-Ge a pronounced minimum at $x_{Ge} \approx 0.2$ ($e/a \approx 1.6$), while Vatolin *et al.* [22] observed it

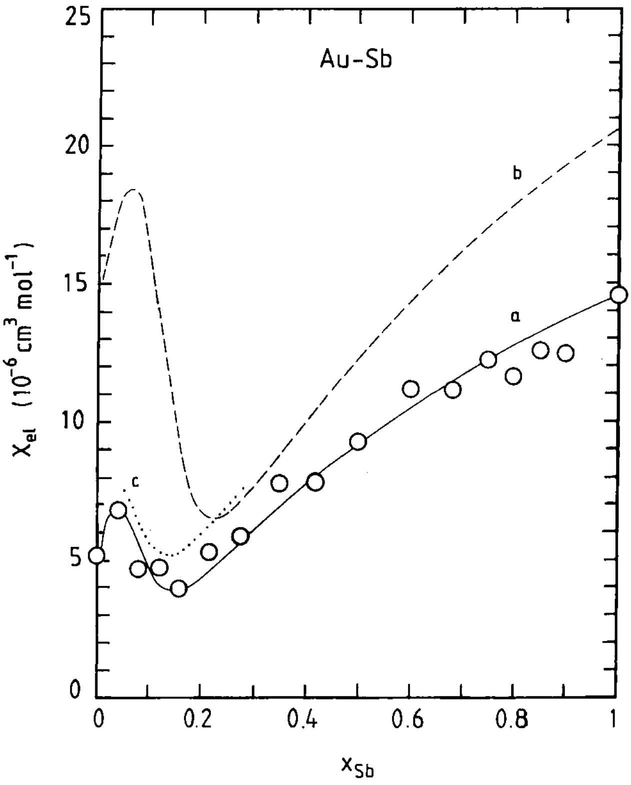

FIGURE 3 Experimental (a) and calculated (b, c) electronic suceptibility of liquid Au-Sb alloys at 1300 K.

around the eutectic composition of the system ($x_{Ge} \approx 0.25$ or $e/a \approx 1.75$).

This argumentation is supported by the results reported for the systems Au-Sn, Au-Ga, Au-Zn, Au-Cd, and possibly also Au-Te. The minimum observed in liquid Au-Sn at $x_{Sn} \leq 0.2$ indicates $e/a \leq 1.6$ and has already been treated by pseudopotential theory [9]. Yatsenko *et al.* [23] observed a likewise minimum in Au-Ga around $x_{Ga} \approx 0.25$ pointing to an electron to atom ratio of $e/a \approx 1.5$. Tsuchiya and Tamaki [24] reported broad minima for liquid Au-Zn ($x_{Zn} \approx 0.55$) and liquid Au-Cd ($x_{Ca} \approx 0.5$) giving evidence of a critical electron concentration of about $e/a \approx 1.5$. The obvious correlation between the concentration where the minima occur and the formal valency (group number) of the polyvalent element is illustrated in Figure 4 which also includes a data point for liquid Au-Te [25]. The full line shown in the figure was calculatd with an average value of $e/a \approx 1.56$. The results reported for Au-Te do not show a clear minimum, but the weak anomaly around $x_{\mathrm{Te}} \approx 0.2$ might suggest $e/a \approx 2$. There are strong indications that the number of valence electrons in liquid Te increases gradually with the temperature. Hall effect and the magnetic susceptibility data point rather to a formal valency of about 3.5 at the given reference temperature which places the valence electron concentration for $x_{\mathrm{Te}} \approx 0.2$ at $e/a \approx 1.5$ [26]. The point given in Figure 4 refers to the hypothetical valency of 3.5 instead of the formal valency

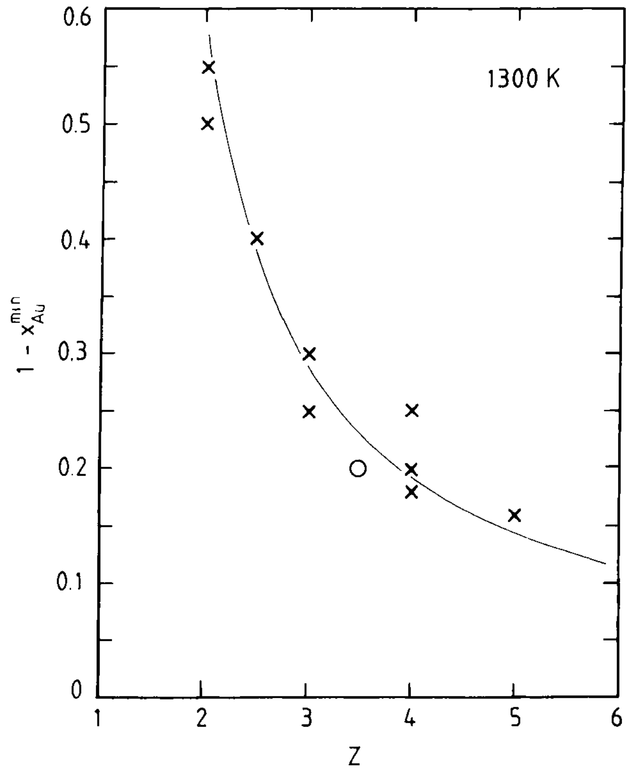

FIGURE 4 Correlation between the position of the minima in the magnetic susceptibility and the valency of the alloying element in liquid Au-based alloys at 1300 K (x, o experimental values, - calculated with the average electron to atom ratio of 1.56.

of 6. In addition to the binary systems, Figure 6 also includes the result of recent measurements in the ternary section $Au_x(In_{0.5}Zn_{0.5})_{1-x}$ where according to an average formal valency of 2.5 a flat minimum occurs around $x_{Au} \approx 0.6$ suggesting $e/a \approx 1.6$ [27].

The position of the minima $x_{Au}^{min}$and the corresponding electron concentrations e/a are listed in Table II, together with some information on the phases appearing in that critical range of composition [28]. It is a remarkable but also a plausible coincidence, that in all systems the susceptibility minima occur at those compositions where electron compounds with electron to atom ratios of 3:2 or 21:13 are formed in solid state. They appear as stable phases in the systems Au-Zn, Au-Cd, Au-In, and Au-Sn, and as metastable phases

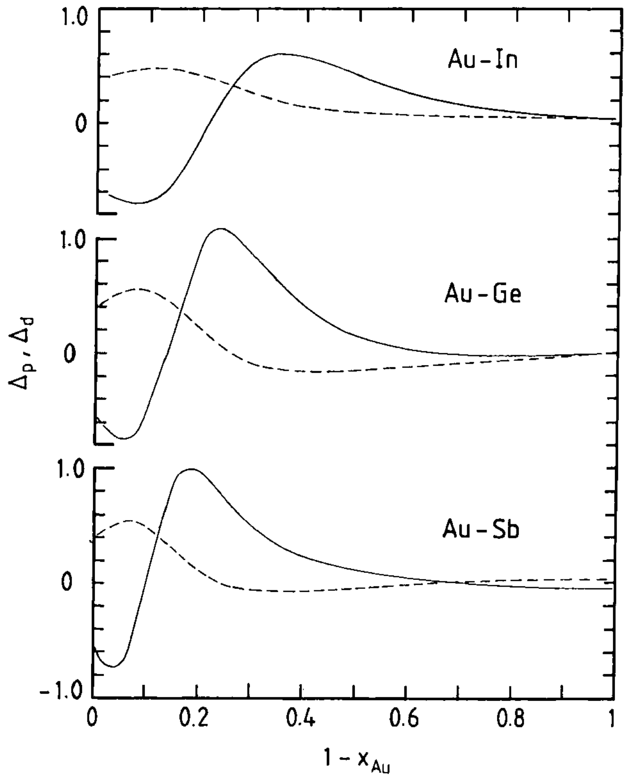

FIGURE 5 Pseudopotential corrections to the magnetic susceptibility of liquid Au-In, Au-Ge and Au-Sb alloys at 1300 K (- - - Δ_p, —Δ_d).

TABLE II Some experimental data of Au-based alloys: $x_{\mathrm{Au}}^{\mathrm{min}}$concentration of the susceptibility minima in the liquid alloys, *e*/*a* electron to atom ratio at $x_{\mathrm{Au}}^{\mathrm{min}}$, and some information on the solid phases appearing in the systems (notation of phase, Pearson symbol and prototype of structure, range of composition in terms of *e*/*a*)

System	$x_{\mathrm{Au}}^{\mathrm{min}}$	*e*/*a*	*Phases*	*Structure*	*Composition*
Au-Zn	0.55	1.55	β'-AuZn	$cP2$(CsCl-type)	$1.38 \leq e/a \leq 1.57$
			γ-AuZn	$cI52$(Cu_5Zn_8-type)	$1.63 \leq e/a \leq 1.86$
Au-Cd	0.50	1.50	β-AuCd	$cP2$(CsCl-type)	$1.43 \leq e/a \leq 1.57$
			δ-AuCd	$cI52$(Cu_5Zn_8-type)	$1.48 \leq e/a \leq 1.61$
Au-In	0.70	1.60	γ-AuIn	$cI52$(Cu_5Zn_8-type)	$1.58 \leq e/a \leq 1.63$
			ζ-AuIn	$hP2$(Mg-type)	$1.26 \leq e/a \leq 1.46$
Au-Ge	0.82	1.54	β-AuGe(m)	$hP2$ (Mg-type)	$1.48 \leq e/a \leq 1.75$
Au-Sn	0.80	1.60	ζ-AuSn	$hP2$(Mg-type)	$1.30 \leq e/a \leq 1.56$
Au-Sb	0.84	1.64	ζ-AuSb(m)	$hP2$ (Mg-type)	$1.52 \leq e/a \leq 1.60$
Au-Te	0.80	1.50*			

(m) metastable phase.

* corrected valency of 3.5 according to Ref. [26].

TABLE III Electronic susceptibilities and corrections to the free electron susceptibilities of liquid Au, In, Ge, and Sb: Calculated electronic susceptibility $\chi_{\mathrm{el}}^{\mathrm{calc}}$, pseudopotential corrections Δ_p, Δ_d, and corrections due to electron-electron interactions $\delta_p^{\mathrm{ee}}, \delta_d^{\mathrm{ee}}$

	$\chi_{\mathrm{el}}^{\mathrm{calc}}$	Δ_p	Δ_d	δ_p^{ee}	δ_d^{ee}
Au	14.46	0.394	−0.571	1.23	1.23
In	15.80	0.025	0.008	1.21	1.17
Ge	13.50	−0.013	−0.029	1.19	1.13
Sb	20.71	0.004	−0.085	1.20	1.15

in the systems Au-Ge, Au-Sb. So far, only for Au-Te metastable phases on the Au-rich side have not been reported.

In this context it is interesting to note that the anomalous transport properties of amorphous alloys seem to occur at a likewise critical electron to atom ratio of $e/a \approx 1.8$ [29].

The results of the theoretical treatment are shown in Figures 1–3 together with the experimentally deduced results. As regards the magnetic susceptibilities on an absolute scale – in particular that of pure Au – the calculated results exceed the experimental values by more than the estimated incertainty of $3-6 \cdot 10^{-6}$ cm^3 mol^{-1}. Once more it has to be emphasized that due to the use of different reference values for the core susceptibilities the electronic susceptibilities quoted in the literature are very divergent. Although the total susceptibility measured for pure Au is in excellent agreement with others [25, 30, 31], Dupree and Ford [31] claimed $17.4 \cdot 10^{-6}$ cm^3 mol^{-1} for the electronic contribution, while Borchi and DeGennaro use $13.8 \cdot 10^{-6}$ cm^3 mol^{-1} in good accordance with the theoretical value of $14.4 \cdot 10^{-6}$ cm^3 mol^{-1}. It is unclear whether the discrepancies are due to the selected model potential or the choice of core susceptibility. Nevertheless, it is remarkable that such a parameter-free treatment reproduces the essential features of the systems, i.e. the deep minima on the Au-rich side and even the initial increase of the susceptibility indicated for small additions of polyvalent metal. The positions of the minima are found to be not precisely predicted, they appear at higher concentrations of polyvalent metal, but the trends are about the same. In the particular case of Au-In the shape of the experimental susceptibility vs composition curve is found to be well reproduced, and also for Au-Sb the agreement is acceptable. The appearance of a likewise minimum in Au-Ge is evidenced by the theory, however, the extremely pronounced shape of the theoretical curve is not reflected by the experimental points. Sodeck *et al.* [21] and Vatolin *et al.* [22], on the other hand, reported in fact a much more pronounced variation of the magnetic susceptibility in Au-Ge, in better agreement with the theory. Probably, these differences reflect the experimental limitation in determining the magnetic properties of diamagnetic liquid metals.

As this point it should be noted that even the choice of very crude input parameters – i.e. empty core potentials for all elements with $R_c \approx R_{\rm ion}$, and hard sphere structure factors in simple form – gives a rough reproduction of the minima in the magnetic susceptibility (cf. curve *c* in Figs. 1 – 3). This illustrates that, despite the drastic difference in absolute values, the choice of parameter has only a limited influence on the principal shape of the susceptibility vs composition curves.

The corrections to the free electron susceptibilities of the pure elements are listed in Table II. The correction terms due to exchange and correlation interactions ($\delta_p^{\rm ex}$ and $\delta_d^{\rm cc}$) were found to vary gradually with the composition of the alloys, as expected from the smooth variation of r_s. The remarkable variations of the pseudopotential corrections (Δ_p and Δ_d) are shown in Figure 5. The flat maximum of Δ_p indicates a considerable enhancement of $\chi_{\rm spin}$ in the very vicinity of pure Au, which concides with the minimum of Δ_p thus giving rise to the initial increase of χ_{el} as actually indicated for Au-In and Au-Sb. The spectacular maximum of Δ_d at $e/a \approx 1.85$ results in a drastic increase of χ_{dia} as compared to the free electron value by 60% in Au-In up to about 100% in Au-Ge and Au-Sb. It is evident, that the minima apparent in the total susceptibility are primarily due to the marked enhancement of χ_{dia} due to interactions between the ionic cores and the conduction electrons.

So far, the theoretical concept applied in the present form gives a qualitative interpretation of the observed phenomena. The remaining differences in the principal shape of the curves (e.g., for Au-Ge) as well as the obvious divergency in absolute values are probably due to the assumption of a nearly-free-electron behaviour and the simplifications introduced to evaluate the theoretical expressions. It is obvious that the form factors applied in this paper are probably not adequate to describe non-simple metals like Au, and also that hard sphere structure factors – even if used in improved form – are only a crude approximation of real alloy systems.

However, judged by the uncertainty in the experimental data (i.e., the absolute values of χ_{el}, and even the shape of the experimental curves), it is worth emphasizing that the simplified theoretical treatment applied in this paper reproduces i) the principal form of the experimental susceptibility vs composition curves and i) also the systematic variations of the minima due to the increased number of valence electrons.

4. CONCLUSION

The observed minima in the magnetic susceptibility of liquid Au-alloys occur apparently around the critical electron concentration of $e/a \approx 1.6$ which is a value between classical electron to atom ratios of 3:2 and 21:13. In a convincing manner, the magnetic anomalies are accompanied by the appearance of stable (Au-Zn, Au-Cd, Au-In, Au-Sn) or metastable electron compounds (Au-Ge, Au-Sb). It can be assumed, that the minima of the magnetic susceptibility reflect an increased electronic stability of the liquid alloys in the critical concentration range which, in turn, gives rise to the appearance of electron compounds.

The theoretical treatment in terms of pseudopotential theory gives at least a qualitative interpretation of the susceptibility minima in noble metal – polyvalent metal systems. The calculations indicate that the anomalous variation of the susceptibility is primarily due to the interactions between the ionic cores and the electrons which – in the range of the compositions where the minima occur – results in a strong enhancement of the diamagnetic part of the electronic susceptibility. The structure factors and the model potentials applied in the paper are only crude approximations to real alloy systems with more or less pronounced chemical short range order, but in view of the high degree of uncertainty in the experimental susceptibilities and those of the ionic cores, they provide an acceptable representation.

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