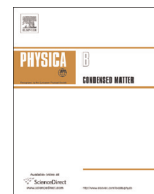




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Chemical short range order and magnetic correction in liquid manganese–gallium zero alloy



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ABSTRACT

The $\text{Mn}_{66}\text{Ga}_{34}$ alloy at this particular composition is known to be zero alloy in which the linear combination of the two neutron scattering lengths weighted by the atomic compositions vanish. Thus for this specific concentration, the effect of the partial structure factors S_{NN} and S_{NC} is cancelled by a weighted term, which value is zero. Then the measured total structure factor $S(q)$ gives directly the concentration–concentration structure factor $S_{CC}(q)$. We present here the first experimental results of neutron diffraction on the $\text{Mn}_{66}\text{Ga}_{34}$ “null matrix alloy” at 1050 °C. The main peak of the experimental $S_{CC}(q)$ gives a strong evidence of a hetero-atomic chemical order in this coordinated alloy. This order also appears in real space radial distribution function which is calculated by the Fourier transform of the structure factor. The degree of hetero-coordination is discussed together with other manganese–polyvalent alloys. However manganese also shows abnormal magnetic scattering in the alloy structure factor which must be corrected. This correction gives an experimental information on the mean effective spin of manganese in this liquid alloy. We present the first critical theoretical calculations of the magnetic correction factor in Mn–Ga zero-alloy based on our accurate experimental measurements of $S_{CC}(q)$.

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1. Introduction

The study of manganese based alloys presents a great interest, because Mn is Polyvalent transition metal which has the Fermi energy in the d-band. This may lead to complex alloy energy band corresponding to surprising properties such as the sign of thermoelectric power. Such transition metal presents magnetic properties due to the unpaired electron spin in the d-band. The magnetism can be modified when mixed with a polyvalent metal such as Ga bringing more electrons in the d-band. From the experimental point of view, manganese is the less well known than other transition metals. It has the lowest melting point of all transition metals which makes measurements at the liquid state are possible. Manganese neutron scattering length is negative and it permits to access the chemical order and enhances the ordering potential. Manganese presents also to a magnetic scattering during the neutron diffraction experiments, which are considered as a parasite effect, which provides complementary information on liquid

alloy magnetism.

In the Mn–Ga binary system, there are many intermetallic compounds and especially some of them with ordered fct structures are known to exhibit a ferromagnetic properties with high coercivity [1,2]. Nowadays, the study of (Mn–Ga)-based ternary alloy is extremely important because of the strong ferromagnetic ternary alloy properties, such as Ni_2MnGa [3,4], Mn_2NiGa [5] and Fe_2MnGa [6]. These alloys are showing thermoelastic–martensitic transformation which have been receiving much attention as promising candidates for magnetic refrigerant materials using the magnetocaloric effect and for high temperatures magnetic shape memory for new class of magnetic actuators and sensor materials which uses magnetic field-induced strain.

Liquid metal can be mixed in all proportions (it is impossible for solid alloy). Consequently, it is possible to realize specific composition such that corresponding to that of the zero alloy and unreachable for solid alloy. We measured the neutron scattering length (NSL) of manganese and gallium as well as the total structure factor $S(q)$ at a gallium concentration, $c_2 = 34.38$ at%, where manganese concentration, $c_1 = 1 - c_2$. According to Bhatia–Thornton [7] statistical mechanical approach $S(q)$ is directly proportional to the number–number, number–concentration and

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concentration–concentration partial structure factors $S_{NN}(q)$, $S_{NC}(q)$ and $S_{CC}(q)$, respectively, with complicated matrix relation, but for particular composition the so called "null matrix composition", Ji-Chen et al. [8], the simplified relation holds:

$$S(q) = \frac{S_{CC}(q)}{c_1 c_2} \quad (1)$$

which has also been used by Ruppertsberg et al. [9] for the Li–Na alloy. In the Mn–Ga alloy, the zero alloy composition is found to be 66 at% Mn and 34 at% Ga.

In the present work, we measured the structure factor of $\text{Mn}_{66}\text{Ga}_{34}$ at 1050 °C on the two axis spectrometer 7C2 built on the hot source of the LLB¹'s Orphée reactor at Saclay. In Section 2, we present the relations between the alloy total structure factor and the partial structure factors and the relations between partial structure factors $S_{ij}(q)$ and partial pair correlation functions $g_{ij}(r)$. We deduce pertinent functions like the Radial concentration Correlation Function (RCF) [10] and the degree of hetero-coordination on the first neighbours shell given by the Warren chemical short-range order (CSRO) parameter [11]. The experimental set-up, the standard corrections and the manganese magnetic scattering correction method are described in Section 3. In Section 4, the experimental results are presented and discussed.

2. Theoretical formalism

The total structure factor $S(q)$ of an alloy is related to three sets of partial structure factors $S_{ij}(q)$ by Ashcroft–Langreth [12], by Faber–Ziman [13] $a_{ij}(q)$ and the Bhatia–Thornton [7] partial structure factors, $S_{NN}(q)$, $S_{NC}(q)$ and $S_{CC}(q)$.

These relations are given in Waseda's book [14]. In the present work, we present the link between these two sets of partial structure factors and facilitate the use of these formalisms. The total structure factor $S(q)$ is expressed as a function of the Faber–Ziman partial structure factors $a_{ij}(q)$, with $i, j = 1, 2$:

$$S(q) = 1 + \frac{c_1^2 b_1^2 (a_{11}(q) - 1) + 2c_1 c_2 b_1 b_2 (a_{12}(q) - 1) + c_2^2 b_2^2 (a_{22}(q) - 1)}{c_1 b_1^2 + c_2 b_2^2}, \quad (2)$$

where b_1, b_2 are the neutron scattering lengths of each metal. The partial structure factors $a_{ij}(q)$ are connected to the partial pair correlation functions $g_{ij}(r)$ via the standard statistical mechanics relation:

$$g_{ij}(r) - 1 = h_{ij}(r) = \frac{1}{2\pi^2 \rho_0 r} \int_0^\infty q (a_{ij}(q) - 1) \sin(qr) dq, \quad (3)$$

where ρ_0 is the average number density. Here, q is wave vector or the so called the Fourier space coordinate. The total structure factor is also related to the Bhatia–Thornton partial structure factors $S_{NN}(q)$, $S_{NC}(q)$ and $S_{CC}(q)$ by:

$$S(q) = \frac{(c_1 b_1 + c_2 b_2)^2 S_{NN}(q) + 2(c_1 b_1 + c_2 b_2)(b_1 - b_2) S_{NC}(q) + (b_1 - b_2)^2 S_{CC}(q)}{c_1 b_1^2 + c_2 b_2^2}. \quad (4)$$

When the quantity $(c_1 b_1 + c_2 b_2)$ is set equals to zero, it can be shown that $S(q) = S_{CC}(q)/(c_1 c_2)$. The difference between the structure factor $S_{CC}(q)$ and the product $c_1 c_2$ is considered the measure of the nature of the chemical order, homo or hetero-coordination. It can be written in terms of the Faber Ziman partial structure factors:

$$S_{CC}(q) = c_1 c_2 (1 + c_1 c_2 (a_{11}(q) + a_{22}(q) - 2a_{12}(q))). \quad (5)$$

In inverse Fourier transform of $S_{CC}(q)$ is simply given as:

$$\frac{1}{2\pi^2 \rho_0 r c_1 c_2} \int_0^\infty \left(\frac{S_{CC}(q)}{c_1 c_2} - 1 \right) q \sin(qr) dq = g_{11}(r) + g_{22}(r) - 2g_{12}(r) = \frac{g_{CC}(r)}{(c_1 c_2)^2}. \quad (6)$$

The quantity $g_{11}(r) + g_{22}(r) - 2g_{12}(r)$ is linked to the difference between the homo-coordination, characterized by $g_{11}(r) + g_{22}(r)$, and the hetero-coordination, characterized by $2g_{12}(r)$ indicates the nature of the chemical order, in the first nearest neighbours shell.

On the other hand, Ruppertsberg and Egger [10] introduced the Radial concentration Correlation Function "RCF" defined as:

$$RCF(r) = 4\pi r^2 \rho_{CC}(r) = \frac{2r}{\pi} \int_0^\infty q \left(\frac{S_{CC}(q)}{c_1 c_2} - 1 \right) \sin(qr) dq. \quad (7)$$

The quantity $\rho_{CC}(r)$ can be expressed in terms of partial radial densities $\rho_{ij}(r)$ [10] as:

$$\rho_{CC}(r) = c_2 (\rho_{11}(r) + \rho_{12}(r)) + c_1 (\rho_{22}(r) + \rho_{21}(r)) - \frac{\rho_{21}(r)}{c_1}. \quad (8)$$

Where $\rho_{ij}(r) = c_j \rho_0 g_{ij}(r)$. If we use the simplest model pair potential, the hard sphere model characterized with hard sphere diameters σ_{ij} ,

$$V_{hs}(r) = \begin{cases} \infty & r \leq \sigma_{ij} \\ 0 & r > \sigma_{ij} \end{cases} \quad (9)$$

and assuming nearly equal hard sphere diameters $\sigma_{11} \cong \sigma_{22} \cong \sigma_{12}$, then $S_{NC}(q)$ strictly vanishes and the arrangement of atoms of species 1 and atoms of species 2 around an atom 1 (given by $\rho_{11}(r) + \rho_{12}(r)$) is the same as around an atom 2 (given by $\rho_{22}(r) + \rho_{21}(r)$). Then we have:

$$\rho_{CC}(r) = \left(\bar{\rho}(r) - \frac{\rho_{21}(r)}{c_1} \right)_{S_{NC}=0} \quad (10)$$

with average density $\bar{\rho}(r) = \rho_{11}(r) + \rho_{12}(r) = \rho_{22}(r) + \rho_{21}(r)$. For a real system with weak attractive potentials, one can consider this equation as a first approximation. The quantity $\rho_{CC}(r)$ vanishes for a random distribution because we have $\rho_{21}(r) = c_1 \bar{\rho}(r)$ and has a negative sign for a hetero-coordinated distribution with $\rho_{21}(r) > c_1 \bar{\rho}(r)$.

By integrating the RCF function in the real space, it may be possible to calculate the Warren [11] short-range order (WSRO) parameter α_p via the relation,

$$\alpha_p = \frac{1}{Z_p} \left(\int_{\text{on the } p^{\text{th}} \text{ shell}} RCF(r) dr \right)_{S_{NC}=0}, \quad (11)$$

where Z_p is in the atoms number of species 1 or 2 in the p^{th} shell.

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