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2003 J. Phys.: Condens. Matter 15 4605

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Antiferromagnetism and low-temperature specific heat of β -Mn_{1-x}Ir_x alloys

M Miyakawa, R Y Umetsu, K Sasao and K Fukamichi

Department of Materials Science, Graduate School of Engineering, Tohoku University,
Aoba-yama 02, Aoba-ku, Sendai 980-8579, Japan

E-mail: masa@maglab.material.tohoku.ac.jp

Received 31 January 2003, in final form 3 June 2003

Published 20 June 2003

Online at stacks.iop.org/JPhysCM/15/4605

Abstract

The site preference of Ir, the temperature dependence of magnetic susceptibility and the low-temperature specific heat of β -Mn_{1-x}Ir_x alloys have been investigated. In Mn_{1-x}Ir_x alloys, the β -phase exists below $x = 0.102$. From x-ray diffraction patterns, almost all Ir atoms are substituted at site 1 in the β -Mn lattice. The electronic specific heat coefficient γ is proportional to the Néel temperature T_N in the form $T_N^{3/4}$ in regions of low-Ir concentration. With increasing x , however, the gradual decrease in γ leads to a relative increase in γ from the straight line. Therefore, it is concluded that the magnetic state of the β -Mn_{1-x}Ir_x alloys varies from a weak itinerant-electron antiferromagnet to an intermediate antiferromagnet with increasing Ir concentration. The point to observe is that the γ versus $T_N^{3/4}$ plots give a universal line for all the β -Mn alloy systems over the entire concentration range.

1. Introduction

In β -Mn structure, the 20 atomic positions in the cubic unit are classified into two kinds of sites—i.e. site 1 of eight atoms and site 2 of 12 atoms—and β -Mn has no magnetic ordering down to 1.5 K [1–3]. By adding a small amount of impurity elements, β -Mn alloys exhibit magnetic ordering [4, 5]. In the past, the magnetic properties of β -Mn were mainly studied as weak itinerant-electron antiferromagnets because the electronic specific heat coefficient is extremely large [6] and the value of the nuclear spin lattice relaxation time $1/T_1$ satisfies a $T^{1/2}$ dependence [7, 8]. The value of $1/T_1$ at site 2 is about 20 times larger than that at site 1, implying that site 2 is more magnetic [1, 9]. In recent years, it has been pointed out that site 2 Mn forms a geometrical triangular lattice in β -Mn and the magnetic state can be highly frustrated [10, 11]. Over the last decade the β -Mn_{1-x}Al_x alloy system in which the substitutional atoms almost exclusively occupy site 2 has been studied extensively. This has suggested that the magnetic state changes from a spin-liquid state to a spin-glass-like state

with increasing Al concentration [10–14]. On the other hand, more recent data on β -Mn alloy systems in which the substitutional atoms almost exclusively occupy site 1 have been reported [15–18]. We have reported that the platinum group elements such as Ru and Os are almost exclusively substituted at site 1 in β -Mn. The magnetic properties of itinerant-electron systems such as β -Mn_{1-x}Ru_x [15] and β -Mn_{1-x}Os_x [16–18] alloys have been explained by considering spin fluctuations [19]. It has also been pointed out that the antiferromagnetic state in β -Mn_{1-x}Ru_x and β -Mn_{1-x}Os_x alloy systems varies from a weak itinerant-electron state to an intermediate state with increasing substitutional elements.

We now pay attention to the substitution of Ir, which belongs to the platinum group, like Ru and Os. For β -Mn_{1-x}Ir_x alloys, it has been reported that the β -phase coexists with the γ -phase at $x = 0.06$, and then the β -phase exists below $x = 0.06$ in the quenched state from 1033 K [20]. On the other hand, γ -Mn_{1-x}Ir_x alloys have been investigated intensively for application to spin-valve layers in giant magneto-resistance and tunnelling magneto-resistance heads [21–25]. Furthermore, the relationship between the spin structure and the lattice distortions of γ -Mn_{1-x}Ir_x alloys has been discussed experimentally and theoretically [26, 27]. Accordingly, it is important to investigate the magnetic properties in the vicinity of the limit of concentration of the γ -phase.

To understand the magnetic state in the β -Mn_{1-x}Ir_x alloys, it is meaningful to investigate the electronic specific heat coefficient, which is reflected by the spin fluctuations. In this paper, we have carried out x-ray diffraction, magnetic susceptibility and specific heat measurements of the β -Mn_{1-x}Ir_x alloys. From x-ray diffraction, the site preference of Ir atoms has been determined, while the magnetic and electrical results have been discussed within the framework of the self-consistent renormalization (SCR) theory.

2. Experimental details

The specimens were prepared by arc-melting and quenched into ice water from 1123 K after annealing for 24 h. The phase obtained was identified by x-ray powder diffraction patterns with Cu K α radiation. The alloy compositions were deduced from the density, which was derived from the lattice constants. The temperature dependence of magnetic susceptibility and the low-temperature specific heat were measured using a superconducting quantum interference device magnetometer and a relaxation method, respectively.

3. Results and discussion

Figure 1 shows the room-temperature x-ray diffraction patterns of Mn_{1-x}Ir_x alloys, together with the calculated pattern of β -Mn. Below $x = 0.102$, all the peaks are identified as a β -phase. The mixed state of the β - and γ -phases is observed at $x = 0.121$. In a previous study, it was reported that the β -phase exists below $x = 0.06$ in the quenched state from 1033 K [20]. However, the β -phase in the quenched state from 1123 K exists in a wider range up to $x = 0.102$. In the β -Mn_{1-x}Ir_x alloys, the peaks shift to the lower-angle side with increasing x , indicating an increase in the lattice constant. To check the site preference of Ir atoms in the β -Mn lattice, an x-ray diffraction pattern for $x = 0.102$ and two kinds of calculated patterns are shown in figure 2. For the calculations, it was assumed that all Ir atoms occupy site 1 (a) or site 2 (b) in the β -Mn lattice. Taking the intensities of the 110 and 111 peaks into consideration, the diffraction pattern for $x = 0.102$ tells us that almost all Ir atoms occupy site 1, in analogy with Ru [15] and Os [16].

The temperature dependence of magnetic susceptibility, $\chi(T)$, of the β -Mn_{1-x}Ir_x alloys is shown in figure 3. With increasing x , $\chi(T)$ decreases and a small peak associated the

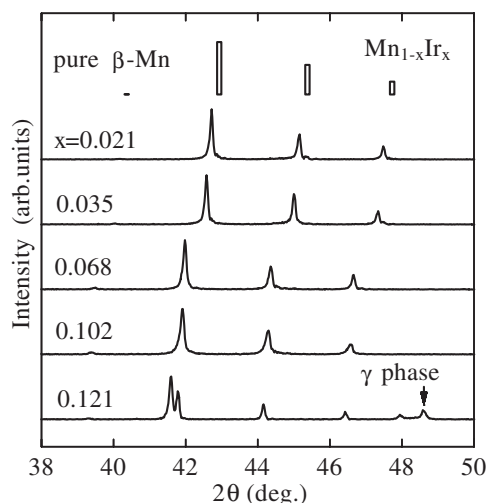


Figure 1. The room-temperature x-ray diffraction patterns of $\text{Mn}_{1-x}\text{Ir}_x$ alloys and the calculated pattern of β -Mn.

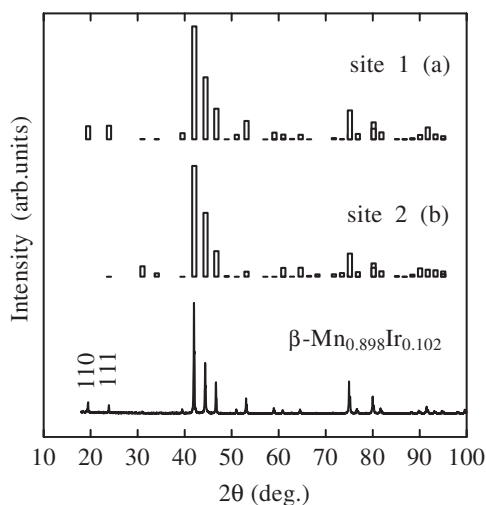


Figure 2. The x-ray diffraction pattern of $x = 0.102$ and two kinds of calculated patterns. In the calculations, all Ir atoms are assumed to occupy only (a) site 1 or (b) site 2 in the β -Mn lattice.

Néel temperature smears away and then an inflection becomes clear at $x = 0.102$, similar to $\chi(T)$ of β -Mn $_{1-x}$ Ru $_x$ [15] and β -Mn $_{1-x}$ Os $_x$ [16–18] alloys. This suggests that the magnetic states of the β -Mn $_{1-x}$ Ir $_x$ alloys are itinerant-electron antiferromagnets, in analogy with β -Mn $_{1-x}$ Ru $_x$ [15] and β -Mn $_{1-x}$ Os $_x$ [16–18] alloys, and the small peaks and the inflection points correspond to the Néel temperature T_N .

Figures 4(a) and (b) show the concentration dependence of the room-temperature lattice constant and the Néel temperature T_N of the β -Mn $_{1-x}$ Ir $_x$ alloys, respectively, together with those of β -Mn $_{1-x}$ Ru $_x$ [15] and β -Mn $_{1-x}$ Os $_x$ [17] alloys. Comparing the results of the β -Mn $_{1-x}$ Ir $_x$ alloys with those of the others, the lattice constant and the Néel temperature T_N of the β -Mn $_{1-x}$ Ir $_x$ alloys increase more significantly than those of β -Mn $_{1-x}$ Ru $_x$ and β -Mn $_{1-x}$ Os $_x$.

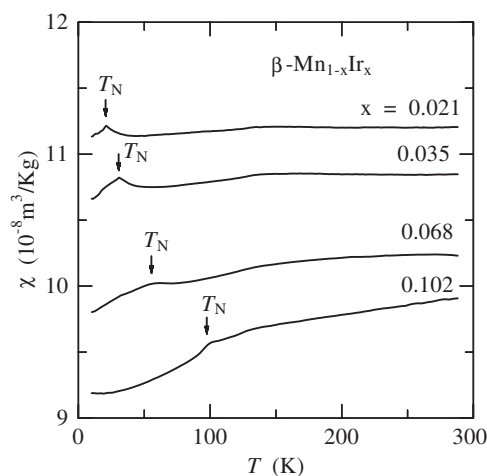


Figure 3. The temperature dependence of the magnetic susceptibility, χ , of $\beta\text{-Mn}_{1-x}\text{Ir}_x$ alloys. The arrows indicate the Néel temperature T_N .

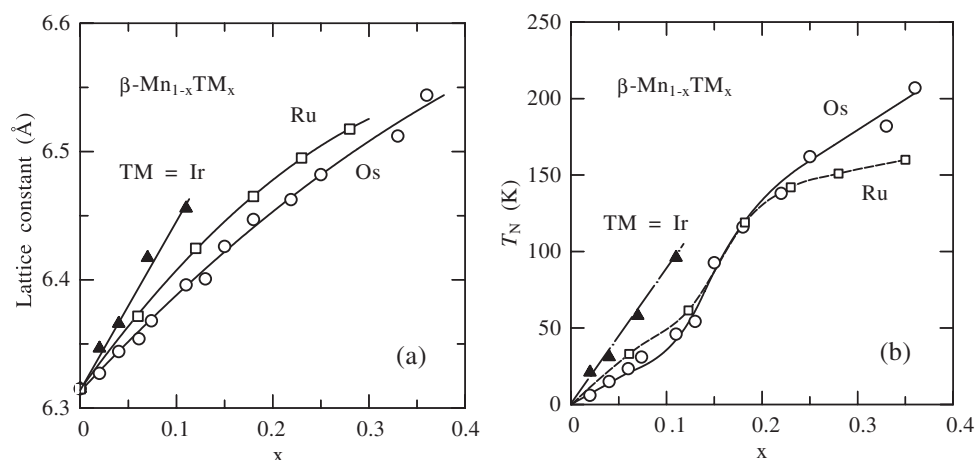


Figure 4. The concentration dependence of the room-temperature lattice constant (a) and that of the Néel temperature (b) of the $\beta\text{-Mn}_{1-x}\text{Ir}_x$ alloys, together with the data of $\beta\text{-Mn}_{1-x}\text{TM}_x$ (TM = Ru [15] and Os [17]) alloys in which Ru and Os atoms preferentially occupy site 1.

alloys with an increase in the substitutional elements. The d-electron number in Ir is larger than that in Ru and Os. In $\beta\text{-Mn}$ alloys substituted by the 3d transition metal, it has been pointed out that the magnetic ordering is influenced by the increases of the lattice constant and the d-electron number [5]. In the present study, this trend was confirmed from figures 4(a) and (b). Figure 5 shows the room-temperature lattice constant versus the Néel temperature T_N of $\beta\text{-Mn}$ alloys in which the substitutional elements mainly occupy site 1. The present $\beta\text{-Mn}_{1-x}\text{Ir}_x$ alloys as well as $\beta\text{-Mn}_{1-x}\text{Ru}_x$ [15] and $\beta\text{-Mn}_{1-x}\text{Os}_x$ [16, 17] alloys show the same relationship between the lattice constant and the Néel temperature in low-concentration ranges, though the data of $\beta\text{-Mn}_{1-x}\text{Co}_x$ alloys [28] is much different from those alloys. In high-concentration ranges, $\beta\text{-Mn}_{1-x}\text{Ru}_x$, $\beta\text{-Mn}_{1-x}\text{Os}_x$ and $\beta\text{-Mn}_{1-x}\text{Ir}_x$ alloys exhibit a different slope, compared with that in low concentrations. It has been well known that magnetic properties of Mn alloy

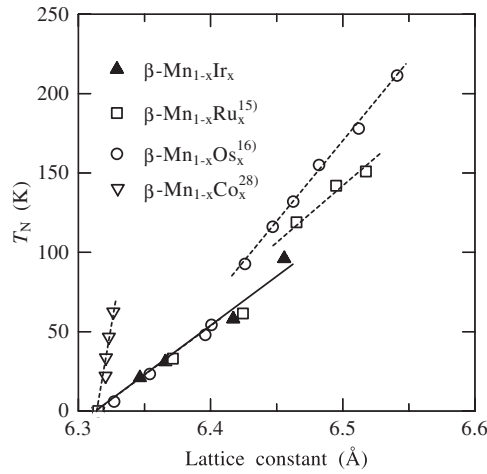


Figure 5. The Néel temperature T_N versus the room-temperature lattice constant plot of the β -Mn alloys in which the additional elements preferentially occupy site 1.

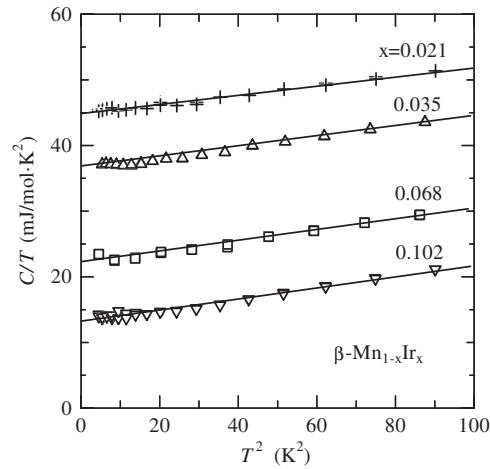


Figure 6. Temperature versus the specific heat plots, in the form of C/T versus T^2 , for the β -Mn $_{1-x}$ Ir $_x$ alloys.

systems are very sensitive to the Mn–Mn distance [29]. Strictly speaking, β -Mn alloys in which the substitutional elements occupy site 1 do not follow this relation, as seen from figure 5, suggesting that they are less in the localized state than in the itinerant-electron state.

The specific heat, in the form of C/T versus T^2 plots for the β -Mn $_{1-x}$ Ir $_x$ alloys, is given in figure 6. Since all the plots are well fitted by a straight line below 10 K, the electronic specific heat coefficient γ is obtained by a linear extrapolation. The value of γ consists of two terms: the band term γ_{band} and the spin fluctuation term γ_{sf} , which is given by the following expression within the framework of the SCR theory [30]:

$$\gamma = \gamma_{band} + \gamma_{sf} = \frac{\pi^2 k_B^2}{3} D(\varepsilon_F) + \frac{3N_0}{4T_0} \ln\left(1 + \frac{1}{K_0^2}\right) \quad (1)$$

where k_B is the Boltzmann constant, $D(\varepsilon_F)$ is the density of states at the Fermi level ε_F , N_0 is the number of magnetic atoms, T_0 corresponds to the energy scale of spin fluctuations,

and $1/K_0$ is proportional to the magnetic correlation length, respectively. When the effect of spin fluctuations is significant at low Ir concentrations with a low Néel temperature, the long-wavelength mode would become dominant and hence the magnetic correlation length would become extensively large. As a result, $1/K_0$ becomes large and γ_{sf} brings about a remarkable contribution to the value of γ . Taking into account the band term, which is estimated to be $8 \text{ mJ mol}^{-1} \text{ K}^{-2}$ [31], the value of γ_{sf} is extremely large in the range of low Ir concentration and rapidly decreases with increasing Ir concentration. As a result, the value of γ comes close to γ_{band} , as seen from the figure.

It has been pointed out that the spin fluctuation term, γ_{sf} , of the electronic specific heat coefficient γ is proportional to $T_N^{3/4}$ in weak itinerant-electron antiferromagnets [32]. The value of γ_{sf} is expressed by the following expressions from the SCR theory [32]:

$$\gamma_{sf}(\alpha) - \gamma_{HF}(\alpha) = \gamma_{sf}(1) - \gamma_{HF}(1) - W_A(\alpha - 1)^{1/2} \quad (2)$$

$$\alpha = 2I\chi_S \quad (3)$$

where γ_{HF} is the term obtained by the Hartree–Fock approximation, W_A is the constant associated with the band structure, and I and χ_S are the exchange interaction and the staggered susceptibility, respectively. The magnetic state on the verge of antiferromagnetism is given by $\gamma_{sf}(1)$ and $\gamma_{HF}(1)$ in equation (2). The value of α given in equation (3) represents the condition for the on-set of antiferromagnetism. Depending on the value of α , i.e. below 1 or above 1, the magnetic state becomes paramagnetic or antiferromagnetic, respectively. When the components of spin fluctuations with wavevectors are close to the antiferromagnetic vector Q , T_N is related to the following relation [33]:

$$T_N \propto (\alpha - 1)^{2/3}. \quad (4)$$

The Hartree–Fock term in equation (2) is almost independent of the alloy concentration, and hence the relation between γ_{sf} and T_N is given by the following expression in weak itinerant-electron antiferromagnets:

$$\gamma_{sf} = A - BT_N^{3/4} \quad (5)$$

where A and B are the constants. The relation between the Néel temperature, T_N , and the electronic specific heat coefficient, γ , of the $\beta\text{-Mn}_{1-x}\text{Ir}_x$ alloys is shown in figure 7, together with that of $\beta\text{-Mn}_{1-x}\text{Fe}_x$ [6], $\beta\text{-Mn}_{1-x}\text{Co}_x$ [6], $\beta\text{-Mn}_{1-x}\text{Ni}_x$ [6], $\beta\text{-Mn}_{1-x}\text{Ru}_x$ [15], and $\beta\text{-Mn}_{1-x}\text{Os}_x$ [17] alloys, in which the substitutional elements preferentially occupy site 1. The plots in the form of $\gamma - T_N^{3/4}$ are consistent with equation (5) by assuming that the value of γ_{band} is insensitive to the Ir concentration. The Ir concentration of four specimens is indicated in the same figure. The value of γ is proportional to $T_N^{3/4}$ below $x = 0.035$ and, in the range of high γ values, all the data are plotted on the same straight line regardless of the kind of the substitutional elements. However, with increasing substitutional elements, or with increasing T_N , the gradual reduction of γ leads to a relative increase of γ from the straight line, hence the alloys are no longer weak itinerant-electron antiferromagnets, and eventually γ becomes close to γ_{band} . Therefore, it is concluded that the magnetic state in the $\beta\text{-Mn}_{1-x}\text{Ir}_x$ alloys varies from the weak itinerant-electron antiferromagnetic to the intermediate antiferromagnetic state with increasing Ir concentration. Moreover, what is important in figure 7 is that the curve exhibits universality for all the $\beta\text{-Mn}$ alloy systems, indicating that the magnitude of the contribution to the spin fluctuations is insensitive to the kinds of additives by scaling the Néel temperature.

4. Conclusion

The site preference of Ir, the temperature dependence of magnetic susceptibility, and the low-temperature specific heat of $\beta\text{-Mn}_{1-x}\text{Ir}_x$ alloys have been investigated. The substitution limit of

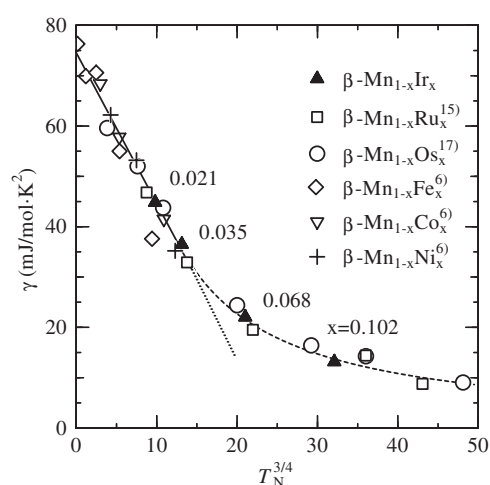


Figure 7. The relationship between the electronic specific heat coefficient, γ , and the Néel temperature, T_N , in the form of $\gamma \propto T_N^{3/4}$ for the β -Mn_{1-x}Ir_x alloys, together with that for β -Mn_{1-x}Fe_x [6] β -Mn_{1-x}Co_x [6], β -Mn_{1-x}Ni_x [6], β -Mn_{1-x}Ru_x [15], and β -Mn_{1-x}Os_x [17] alloys, in which the substitutional elements preferentially occupy site 1. Four kinds of Ir concentrations are given.

Ir and its site preference in the β -Mn lattice have been determined. The close relation between the Néel temperature T_N and the electronic specific heat coefficient γ has been discussed in terms of spin fluctuations. The main results are summarized as follows:

- From the x-ray diffraction patterns, the substitution limit of Ir in the β -phase was confirmed as $x = 0.102$ in the quenched state from 1123 K. Almost all Ir atoms occupy site 1 in the β -Mn lattice.
- Both the lattice constant and the Néel temperature of the β -Mn_{1-x}Ir_x alloys increase more significantly than those of β -Mn_{1-x}Ru_x and β -Mn_{1-x}Os_x alloys with an increase in Ir concentration.
- The relation between the electronic specific heat coefficient, γ , and the Néel temperature, in the form $T_N^{3/4}$, is held in the range of low Ir concentration, and the curve tends gradually away from a linear relation with increasing x , indicating that the magnetic state varies from the weak itinerant-electron antiferromagnetic to the intermediate antiferromagnetic state.
- A universal curve in the plots of γ versus $T_N^{3/4}$ is confirmed over the entire concentration region for all the β -Mn alloy systems in which the substitutional elements preferentially occupy site 1.

Acknowledgments

The authors wish to express their thanks to Mr K Ohchi for experimental support in this study. Two authors (M Miyakawa and R Y Umetsu) are grateful for the support of the JSPS Research Fellowships for Young Scientists. This work was supported by a Grant-in-Aid for Scientific Research (B)(2), No 13450255, from the Japan Society for the Promotion of Science.

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