variable $\theta=k_z a$. Let

$$I=\frac{1}{\pi}\int_0^\pi d\theta\,\sin\theta\varphi_B(\theta)=-\frac{1}{\pi}\int_0^\pi d\theta\,\frac{\partial}{\partial\theta}(\cos\theta)\varphi_B(\theta)$$

$$=-\frac{1}{\pi}\cos\theta\varphi_B(\theta)\Big|_0^\pi+\frac{1}{\pi}\int_0^\pi d\theta\,\cos\theta\frac{\partial\varphi_B}{\partial\theta}$$

$$1+\int_0^\pi\frac{d\theta}{\pi}\frac{\cos\theta(1-\gamma^2)}{2\gamma\cos\theta-(1+\gamma^2)}$$

$$=-1+\frac{1-\gamma^2}{2\gamma}-\frac{1-\gamma^4}{2\pi\gamma}\int_0^\pi\frac{d\theta}{(1+\gamma^2)-2\gamma\cos\theta}.$$

The remaining integral may be obtained from standard integral tables:

$$\int_0^\pi\frac{d\theta}{1+\gamma^2-2\gamma\cos\theta}=\frac{\pi}{1-\gamma^2}.$$

Hence

$$\frac{1}{\pi}\int_0^\pi d\theta\,\sin\theta\varphi_B(\theta)=-(1+\gamma),$$

and we obtain

$$\mathcal{J}=-(B/\gamma)[1+(2\gamma/\pi)(\varphi_1-\tfrac{3}{2}\pi)]. \tag{A3}$$

PHYSICAL REVIEW B VOLUME 1, NUMBER 1 1 JANUARY 1970

Properties of the Ni-Ir Alloy System

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(Received 25 August 1969)

Measurements of the lattice parameter, electronic specific heat, magnetic susceptibility, and Debye temperature are presented in the Ni-Ir alloy system from 0 to 100 at.% Ir. This system forms an uninterrupted series of solid solutions over the entire concentration range. No ordering nor decomposition could be observed after annealing for several days at temperatures between 500 and 1100°C. The system develops a sharp peak in the electronic specific heat versus concentration at 85 at.% Ni, a concentration slightly higher than the critical concentration determined magnetically (81 at.% Ni). A weak anomaly was found also in the T^3 term of the low-temperature specific heat around the same concentration. Generally, no low-temperature anomaly, such as was predicted by the early paramagnon theories, was detected down to 1.5°K. The susceptibility was measured on the paramagnetic side only up to 79 at.% Ni, where a spontaneous moment develops at low temperature.

I. INTRODUCTION

Among the transition-metal binary alloys the Ni-Ir system is one of the few cases which has not yet been explored. Nevertheless, this system presents some interesting aspects related to the questions of paramagnons.[1,2] In this respect, its behavior differs remarkably from the isoelectronic system Ni-Rh which we investigated earlier.[3] We shall discuss this difference briefly after presenting structural and electronic data of the Ni-Ir system.

II. RESULTS

A. Lattice Parameters

Prior to our investigation of the electronic properties, the phase diagram of the Ni-Ir system had to be established. Using powder x-ray diffraction, we found that the system exhibits complete solid solubility over the entire range of concentration. Figure 1 shows the lattice parameter a of the fcc lattice and the molar volume V_M as a function of the Ir concentration, both showing a positive deviation from linearity. This behavior is identical with Ni-Rh,[4] except that the difference in lattice constants between the end members is even larger in Ni-Ir. The formation of complete solid solutions is not self-evident. This can be demonstrated by considering the corresponding Rh alloy systems. While Rh forms complete solid solutions with Ni,[4] Pd,[5,6] and Cu,[4,7] at least at high temperatures, Ir shows decomposition in alloying with Pd [6] and partial immiscibility with Cu [8] in the solid state. Special attention was therefore paid to the possible occurrence of ordered phases or decomposition, but no indication of either

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[1] N. F. Berk and J. R. Schrieffer, Phys. Rev. Letters **17**, 433 (1966).

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could be detected in the x-ray films. Furthermore, the smooth variation of the electronic properties such as susceptibility, electronic specific heat, and Debye temperature confirm that Ni and Ir indeed form random solid solutions similar to Ni and Rh.[4]

B. Magnetic Susceptibility

The low-temperature susceptibility values are shown in Fig. 2, together with the electronic specific heat and the Debye temperatures. At 1.4°K the system develops a spontaneous moment at concentrations higher than 78 at.% Ni, which could not be removed by annealing for up to 14 days. The magnetization curve showed a small saturation field and linear behavior between 3 and 15 kOe. No magnetic measurements were done on the ferromagnetic side as that range has been covered by Crangle and Parsons.[9] Their Curie points extrapolate linearly to zero at about 81 at.% Ni. For measurements of the susceptibility and specific heat, we refer the reader to Ref. 3.

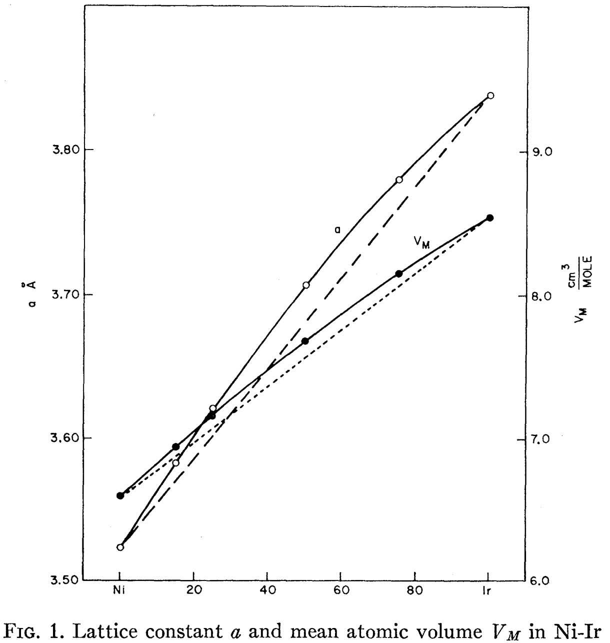

FIG. 1. Lattice constant a and mean atomic volume V_M in Ni-Ir alloys as a function of Ir concentration.

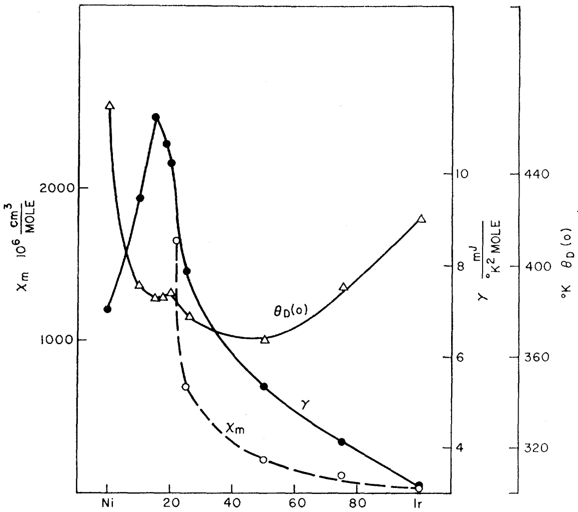

FIG. 2. Molar susceptibility χ_m, electronic specific heat γ and Debye temperature $\theta_D(0)$ in Ni-Ir alloys as a function of Ir concentration.

C. Electronic Specific Heat

The variation of the electronic specific heat γ as a function of Ir concentration is shown in Fig. 2. The most striking feature is the sharp peak occurring at 85 at.% Ni. According to our current understanding of the electronic specific heat in transition metals, this behavior can be understood qualitatively in a simple way. From band-structure calculations, a peak in the bare density of states $N_0(\epsilon_F)$ is expected close to a number of 10 e/at. in the fcc phase.[10] Therefore, dissolving Ni in Ir will raise the electronic specific heat. Simultaneously, the exchange interaction J will also rise, and at a critical concentration, the product of $N_0(\epsilon_F)$ and J will reach unity at low temperatures, leading to split spin-up and spin-down bands and hence to ferromagnetism. It is interesting to ask the question how the splitting of the bands at a slightly over critical concentration will affect the further variation of the density of states as a function of concentration. It is a simple matter to show that the behavior is determined in the first approximation by the sign of $\partial^2 N/\partial\epsilon^2|_{\epsilon=\epsilon_F}$. This means that the peak in the bare density of states does not necessarily need to occur at the critical concentration for ferromagnetism (which is actually the case in Ni-Ir). It can be said, however, that the bare density of states is expected to drop if we go towards the more rich Ni side because one is already close to the density-of-states peak at the critical concentration. Qualitatively at least, one would expect a behavior of the bare electronic specific heat as shown in Fig. 2. According to recent theories,[1,2,11] however, the shape of the peak should be modified by spin fluctuations or paramagnons, occurring at low temperatures around the critical ferromagnetic concentration. The theory in its simplest form predicts a logarithmic enhancement of the bare electronic specific heat[11]:

$$\gamma=\gamma_0\left[1+\tfrac{9}{2}\mu\ln\left(1+\frac{1}{3(1-\mu)}\right)\right], \qquad (1)$$

where μ is given by

$$\mu=N_0(\epsilon_F)J, \qquad (2)$$

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[9] J. Crangle and D. Parsons, Proc. Roy. Soc. (London) **A255**, 509 (1960).

[10] See, e.g., L. Hodges, H. Ehrenreich, and N. D. Lang, Phys. Rev. **152**, 505 (1966), and references therein.

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and is responsible for the enhancement of the spin susceptibility χ_0 according to the Stoner formula

$$\chi = \chi_0/(1-\mu)\,, \tag{3}$$

where μ is the effective exchange interaction and the subscript 0 indicates bare values. Here, we neglect further enhancement effects of γ_0 due to electron-phonon interaction or better, we assume it to be constant in the range of interest. Combining (1) and (3) one might expect a relationship

$$\gamma \sim \ln(\chi/\chi_0) \tag{4}$$

to hold approximately at the critical concentration ($\mu \lesssim 1$). Equation (4) would enable us to get an approximate idea of the enhancement factor of γ due to paramagnons. In practice, it is found that (4) is not well obeyed. This is probably due to statistical concentration fluctuations and the local nature of the Coulomb repulsion which is strongly localized at the Ni atoms. Equation (1), however, is based on a homogeneous enhancement model. But even if the latter is valid, as e.g., in the case of Pd, we know that the enhancement effects of γ are considerably over estimated by (1). Improvements are possible by refined models,[12–16] but cannot remove the over estimation entirely in the case of Pd, or in the dilute case of *Pd*:Ni.[17–21]

At low temperature, the behavior of the specific heat shows a remarkable difference compared with similar alloy systems, e.g., Ni-Rh. Generally, no low-temperature anomaly was found except in $\mathrm{Ni_{0.80}Ir_{0.20}}$ but only about 6% at 1.5°K and decreasing after a long annealing treatment. In Ni-Rh the low-temperature anomaly was attributed to an electronic term proportional to $T^3 \ln(T/T_s)$ with $T_s \approx (1-\mu)T_F$.[3] An alternative interpretation of this anomaly has been given by Hahn and Wohlfarth.[22] In their work they assumed the origin of the anomaly to be weakly ferromagnetic clusters leading to a $1/T$ anomaly in a $C/T-T$ plot. Further investigation by Triplett *et al.*,[23] in Ni-Rh alloys have verified that the $1/T$ functional dependence holds to below 65 m°K. The data also fit a $T^3 \ln(T/T_s)$ term but only over a very limited temperature range. There is also some recent theoretical support for the absence of the $T^3 \ln(T/T_s)$ term in alloys. Fulde and Luther[24] and Brinkman and Engelsberg[19] have recently investigated the effect of short mean free paths in the theory of paramagnons. They showed that the functional dependence of the low-temperature specific-heat anomaly ΔC_s changes from

$$\Delta C_s \sim T^3 \ln(T/T_s) \tag{5}$$

to a more complicated form depending on the temperature regime. At low temperatures, the specific heat is analytic but has a large-negative T^3 contribution. The specific heat then goes through an intermediate-temperature regime for temperatures greater than the spin-flip frequency $1/\tau_s$ but less than the mean collision frequency $1/\tau_0$ in which it varies as $T^{3/2}$. This term is due to diffusive modes in the dynamic susceptibility in this regime. For temperatures greater than $1/\tau$ the specific heat then returns to a $T^3 \ln T$-type behavior. The overall size of the anomaly is not changed by these effects. However, recent experiments by Hicks *et al.*[25] indicate the importance of magnetic clustering in nearly ferromagnetic alloys. The weak impurity effects considered theoretically cannot possibly describe this type situation and a suitable theory for such alloys has not yet been developed.

D. Debye Temperatures

The Debye temperatures $\Theta_D(0)$ plotted in Fig. 2 were obtained from the well-known formula $\Theta_D(0) = (1944.5/\alpha)^{1/3}$ with $\alpha = d(c/T)/dT^2$ at the lowest temperatures, i.e., below about 8°K. An interesting tiny anomaly of $\Theta_D(0)$ between 10 and 30 at.% Ir can be noted. The coefficient α of the cubic term of the specific heat shows a barely detectable local maximum corresponding to a local minimum in the Debye temperatures. The maximum of α coincides with the peak in the electronic specific heat. At 20 at.% Ir corresponding about to the critical ferromagnetic concentration (determined magnetically) the Debye temperature seems to rise a little bit. The effect, however, is within the accuracy of our measurements and should not be over emphasized.

Let us for the moment assume the minimal anomaly given by the weak impurity scattering theory, namely, that there is simply a negative T^3 correction of the form given by Fulde and Luther[24] for $T < \hbar/k_B\tau_0$

$$\Delta C_s = \tfrac{2}{5}\pi k_B \left(\frac{p_s}{\hbar T_s}\right)^3 \ln\left(\frac{\pi\hbar}{4p_s l}\right) T^3\,, \tag{6}$$

where p_s is the cutoff momentum of the paramagnons

$$T_s = (2/\pi k_B) v_F p_s (1-\mu)\,, \tag{7}$$

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l is the mean free path of an electron, and v_F is the Fermi velocity. This would mean that the true Debye temperatures around the critical concentration are somewhat lower than reported here. It is very difficult to give even rough estimates of expression (6). It is tempting to draw a smooth curve in the Θ_D' between about 10 and 50 at.% Ni and attribute the "excess Debye temperature" to the negative term of (6). At the peak at 20 at.% Ir, the true Debye temperature may be at most about 10–15°K lower than the values plotted in Fig. 2. This would lead to a T^3 term due to spin fluctuations of $\simeq -4.10^{-6}$ J/°K^4 mole. If we now try to make an order-of-magnitude evaluation of (6) for some reasonable parameters, e.g., $p_s = p_F$, $T_F = 10^4$ °K, $\mu = 0.95$, and $\hbar/p_F l \sim 10^2$, we find that (6) is about three orders of magnitude too large. The origin of this discrepancy is not clear to us at the moment. It is possible that we have to assume a much smaller cutoff momentum p_s than p_F or that we have an almost identical cancellation by other terms which had to be neglected in the random-phase approximation (RPA).

A quantitative analysis of the spin-fluctuation theory in the improved version of Fulde and Luther[24] or Brinkman and Engelsberg[19] has not yet been carried out. To do this we would need elastic measurements supplementary to our calorimetric data. On the other hand, the investigation of the elastic behavior around 20 at.% Ir presents an interesting problem itself. The Geneva solid-state group Belmahi *et al.*,[26] has found an interesting correlation between the elastic and electronic properties at the strongly enhanced susceptibility peak at the end of the 4*d* band. Their measurements, as well as recent measurements in Ni-Rh,[27] suggest that we have to expect an anomaly in the Debye temperatures around 20 at.% Ir. It is generally known from transition metal alloys[28] that density-of-states peak of *d* bands are reflected in a minimum of the Debye temperatures. This softening effect by a high density of states was also nicely demonstrated in the Rh-Pd-Ag alloy system by Belmahi *et al.*,[26] and explained on the basis of an improved Fuchs model. A similar effect should occur in our Ni-Ir alloy system where the bare density-of-states peaks. The situation, however, may be much more complex than just described, since we do not know the contribution of spin waves on the ferromagnetic and paramagnetic side to the elastic properties. Our relatively smooth variation of the formally calculated Θ_D value suggest, however, that all these effects are quite small, of the order of 1–2% only.

III. CONCLUSIONS

Summarizing briefly the results of our investigation of the Ni-Ir alloy system we find the following results: (1) The system forms an uninterrupted series of solid solutions with random distribution of the components. (2) The electronic specific heat shows a sharp peak at 15 at.% Ir in Ni. We interprete the variation of γ as a variation of the bare density of states $N_0(0)$, however, enhanced around the peak by paramagnons. (3) The experimental results in Ni-Ir support the conclusion that spin-fluctuation effects in alloys are considerably smaller than predicted by any current theories and models. (4) At the critical ferromagnetic concentration a small anomaly in the T^3 term of the low-temperature specific heat is observed as a function of Ir concentration. The anomaly is due to a superposition of various effects, e.g., an anomaly in the elastic behavior as well as a negative T^3 contribution from paramagnons. All these effects, however, are very small.

ACKNOWLEDGMENT

We would like to thank Dr. T. M. Rice for his critical review of our manuscript and for his valuable comments.

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