Nuclear Gamma Resonance Study of the Ir—Fe and Ir—Ni Alloy Systems

R. L. MÖSSBAUER, M. LENGSFELD, W. VON LIERES, W. POTZEL, P. TESCHNER and F. E. WAGNER

Physik-Department, Technische Universität München, Munich, Germany

G. KAINDL

Lawrence Radiation Laboratory, University of California, Berkeley, California

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Dedicated to Prof. Dr. H. Maier-Leibnitz on the occasion of his 60th birthday

The Ir-Fe and Ir-Ni alloy systems were studied over the whole composition range by means of the nuclear resonance absorption of the 73 keV γ-rays of ^{193}Ir and of the 14.4 keV γ-rays of ^{57}Fe. The magnetic hyperfine field at the Ir-nuclei in Ir-Ni alloys decreases approximately linearly with the Ir concentration from -460 kOe at 4.2 K in very dilute alloys to zero at about 20 at.-% Ir. This behaviour is paralleled by the decrease of the magnetic moment per Ni atom as determined from bulk magnetization measurements. The hyperfine fields at both Ir and Fe were measured for the ferromagnetic bcc phase of the Ir-Fe system. They turned out to be virtually independent of concentration with values of about -1400 kOe and -330 kOe, respectively. Linewidths increasing with the Ir concentration indicate a distribution of hyperfine fields. The fcc phase of the Ir-Fe system has been found to be paramagnetic at 4.2 K throughout the range of its existence. The dependence of the hyperfine fields on concentration is discussed in terms of a rigid 3d-band model combined with local shielding. A discussion of the concentration dependence of the ^{193}Ir and ^{57}Fe isomer shifts has to take into account lattice expansion as well as band repopulation effects.

1. Introduction

The understanding of the magnetic behaviour of 3d solutes in 3d solvents as represented by the Slater-Pauling curve is fairly well advanced [1]. The present work was undertaken in an effort to improve the knowledge about 5d solutes in 3d solvents. We report a systematic study of solid solutions of iridium in iron and nickel covering the entire concentration range.

The nuclear resonance absorption of the 73.2 keV γ-rays of ^{193}Ir has been used to study the properties of the Ir-Ni and Ir-Fe alloy systems. The lifetime of the 73 keV level is sufficiently long [2] for well-resolved magnetic hyperfine patterns to be observed [3–6], from which the hyperfine fields can be obtained with high accuracy. For the Ir-Fe alloys supplementary information on the iron hyperfine fields was obtained from the nuclear resonance absorption of the 14.4 keV γ-rays of ^{57}Fe. Information on the s-electron density at the nuclei of both Ir and Fe is obtained from the isomer shifts of the 73.2 keV and 14.4 keV γ-rays.

Magnetic susceptibility measurements performed at 4.2 K on all ferromagnetic alloy samples yielded the ordered magnetic moment per Fe or Ni atom, respectively, while x-ray patterns taken from all samples gave information on the phase constitution and lattice constants of the alloys.

The Ir-Ni alloy system has only recently been investigated extensively by BUCHER et al. [7], who studied the crystal structure, specific heat, and magnetic susceptibility. According to these authors iridium and nickel form a continuous series of face-centered cubic solid solutions. No ordered phase could be detected. Ferromagnetism has been observed [8] for Ni-rich alloys, with both the Curie temperature and the ordered magnetic moment decreasing with the Ir concentration and extrapolating to zero at about 20 at.% Ir.

Reprint requests to Prof. Dr. R. L. MÖSSBAUER, Physik-Department E 15 der Technischen Universität München, *D-8000 München 2*, Arcisstraße 21.

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Ir and Fe likewise form a continuous series of solid solutions. The phase diagram has been established by Fallot[9,10]. For Ir concentrations below about 20 at.% the fcc γ-phase is stable at elevated temperatures only, whereas the stable low-temperature modification is of the bcc α-iron type. The Curie temperature of the bcc α-phase stays virtually constant up to 5 at.% Ir[9], where the transition temperature begins to fall below the Curie point. On the other hand, the fcc phase remains paramagnetic down to 4.2 K for all compositions.

2. Experimental Details

The nuclear gamma resonance absorption of the 73.2 keV γ-rays of ^{193}Ir was observed with both the source and the absorbers cooled to 4.2 K in a liquid-He cryostat. Sources of ^{193}Os ($T_{1/2}=31$ h) in Os metal were produced by neutron irradiation of 25 mg quantities of enriched ^{192}Os of 98.7% isotopic abundance in a thermal flux of $2\cdot10^{13}$ n/s·cm^2. Due to the non-zero electric field gradient at the nuclear sites in the hexagonal osmium lattice the emission line of such sources is an unresolved quadrupole doublet. Since the magnitude of the quadrupole splitting is, however, well known[5], it can be taken into account in the evaluation of the data and has hardly any influence on the reliability of the results obtained for the hyperfine parameters of the absorbers. The 73.2 keV γ-rays were detected by a Ge(Li) diode and stored in a multichannel analyzer operated in the multiscaler mode and synchronized to the sinusoidal motion of the source.

The transmission experiments with the 14.4 keV γ-rays of ^{57}Fe were performed with the source of ^{57}Co in Pd and the absorbers at room temperature. During these experiments the velocity calibration was monitored by a second ^{57}CoPd source mounted on the velocity drive and an absorber of pure metallic iron. The γ-rays were detected by two argon-methane proportional counters, and both transmission spectra were stored into the 1024 channel memory of one multichannel analyzer by means of an electronic interface described elsewhere[11].

For the preparation of the alloys iridium metal of 99.9% purity from Degussa AG, Hanau, and Fe metal of 99.998% as well as Ni metal of 99.999% purity, both supplied by Koch-Light Ltd., Colnbroke, England, were used. The samples weighing several grams each were melted in an argon-arc furnace, the buttons being turned over and remelted several times to improve the homogeneity. The weight loss of the samples during melting and remelting was less than 1% of the sample weight.

From the Ir-Ni alloy ingots, which covered the composition range between 1.5 at.-% and 73.5 at.-% Ir, powder samples were filed and enclosed in lucite containers for use as resonance absorbers.

From the Ir-Fe alloys, the composition of which ranged from 0.7 at.-% to 72.4 at.-% Ir, foils of appropriate thickness for the ^{57}Fe measurements were produced by coldrolling. Absorbers of suitable thickness for the measurements on ^{193}Ir were obtained by stacking several layers of these foils. Additionally, Ir-Fe alloy absorbers for the ^{193}Ir measurements were prepared by filing like in the case of Ir-Ni alloys. Since the filings contained iron contaminations from the file, they were not used for the ^{57}Fe measurements.

From part of each of the alloy ingots, as well as from pure Fe and Ni, small cylinders of 3 mm diameter and a length of 5 mm were turned on a lathe. These were used for measurements of the bulk magnetization at 4.2 K by an induction method[12] in fields up to 50 kOe produced by a superconducting solenoid. From the saturation magnetization of the ferromagnetic samples the magnetic moment per Fe or Ni atom, respectively, was derived. For the pure Fe and Ni samples used to calibrate the apparatus magnetic moments of 2.22 μ_B and 0.61 μ_B per atom[13] were assumed. The magnetic moments per nickel and iron atom, respectively, which are given in Tables 1 and 2 were calculated on the assumption that the Ir atoms in both alloy system behave like magnetic vacancies and carry no magnetic moment.

3. Results

The results of the nuclear resonance absorption and magnetization measurements are compiled in Tables 1 – 3, and some of the transmission spectra are reproduced in Figs. 1 – 3.

The 73.2 keV transition in ^{193}Ir, proceeding between the first excited state with spin $1/2^+$ and the $3/2^+$ groundstate, is of mixed E2/M1 character. Since the excited state g-factor is much larger[4] than that of the groundstate, the magnetic hyperfine patterns consist of two well-separated sets of four lines each. The groundstate splitting is resolved in the Ir-Fe but not in the Ir-Ni alloys. The least-squares fits of 16 Lorentzian lines to the measured Zeeman patterns allowed for the quadrupole splitting in the Os metal source, the magnitude of which was always kept constant at the known[5] value of $eQV_{zz}/4=(0.24\pm0.01)$ mm/s. The relative intensities of the eight lines in the absorber could be

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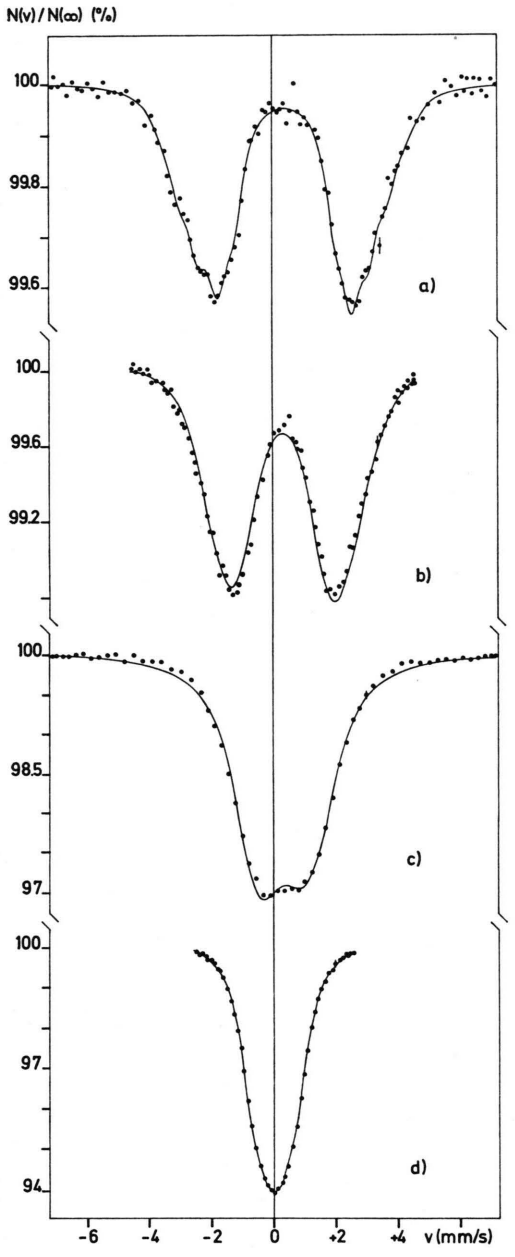

Fig. 1. Nuclear resonance spectra of the 73.2 keV γ-rays of ^{193}Ir measured at 4.2 K with an Os metal source and Ir-Ni alloy absorbers containing 1.5 at.-% (a), 7.1 at.-% (b), 14.1 at.-% (c), and 41.7 at.-% (d) iridium.

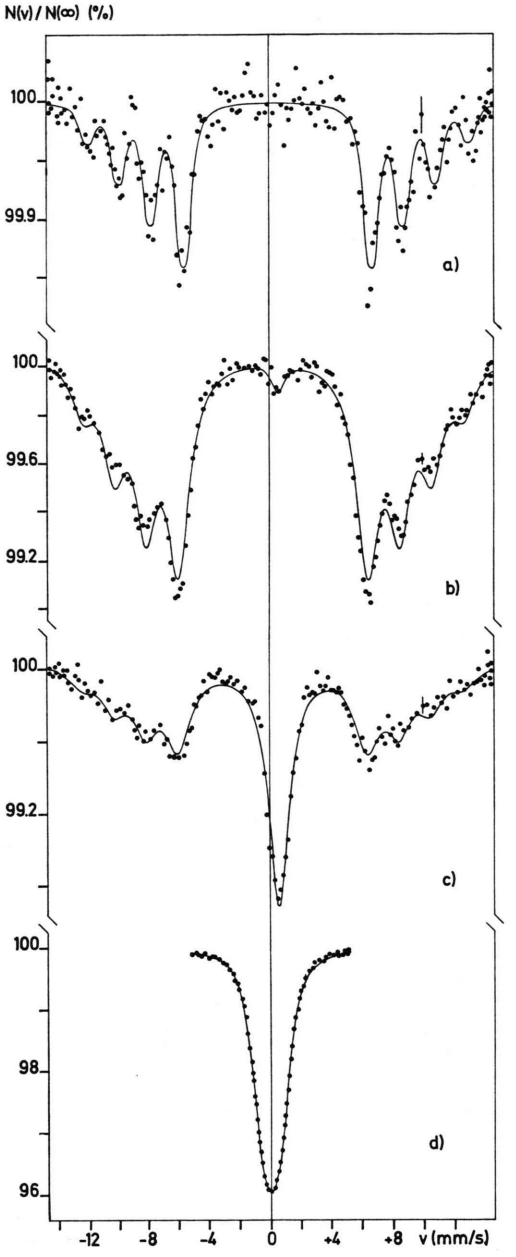

Fig. 2. Nuclear resonance spectra of the 73.2 keV γ-rays of ^{193}Ir measured at 4.2 K with an Os metal source and Ir-Fe alloy absorbers containing 0.7 at.-% (a), 13.5 at.-% (b) 16.2 at.-% (c), and 53.6 at.-% (d) iridium. The shown spectra were all taken with filed samples as absorbers.

treated as free parameters in the Ir-Fe alloys. In the case of the Ir-Ni alloys, where the resolution is rather poor, they had to be kept constant at their theoretical values[14] resulting from the known E2/M1 mixing ratio $\delta^2 = 0.311 \pm 0.006$[4].

An electric quadrupole interaction of the $3/2^+$ ground-state with an axially symmetric field gradient was allowed for in all alloy absorbers. In

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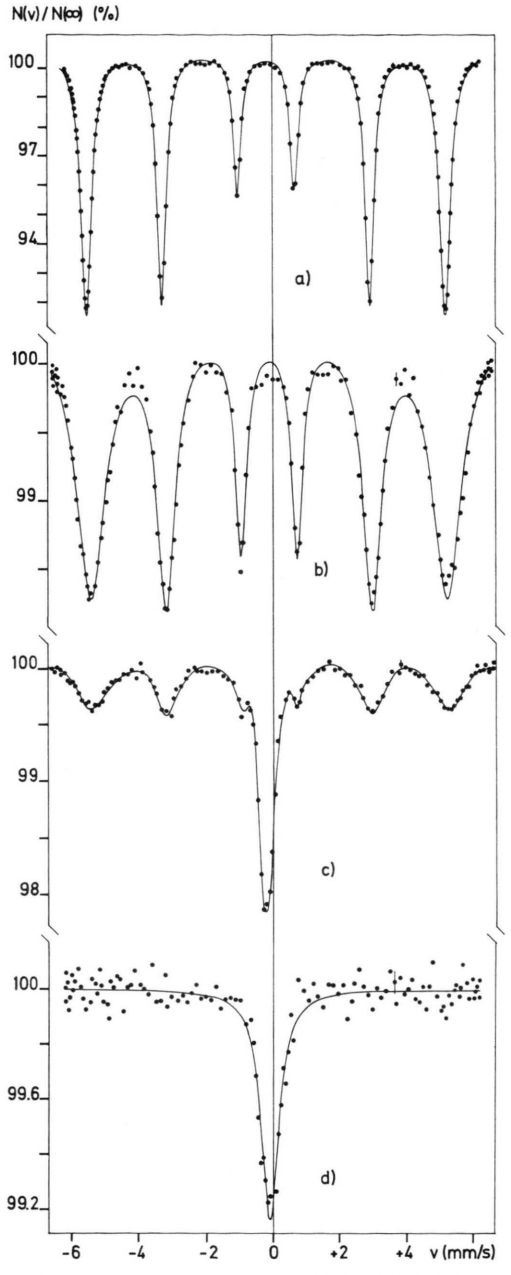

Fig. 3. Nuclear resonance spectra of the 14.4 keV γ-rays of ^{57}Fe measured at room temperature with a ^{57}CoPd source and absorbers containing 0.7 at.-% (a), 16.2 at.-% (b), 22.5 at.-% (c), and 53.6 at.-% (d) iridium. All spectra were taken with rolled foils as absorbers.

the ferromagnetic phases the symmetry axis of the electric field gradient was assumed to be parallel to the magnetic hyperfine field. Actually one expects the local field gradients to depend on the individual environment of the iridium atoms, and the $eQV_{zz}/4$ values given in Tables 1 and 2 have to be regarded as average values only. Whenever the magnetic hyperfine interaction of the $3/2^+$ state is large compared to the electric quadrupole coupling, only the projection of the electric gradient tensor on the direction of the hyperfine field is seen in the nuclear resonance spectra. For a random relative orientation of both, the average value of $eQV_{zz}/4$ may become zero, the net effect on the spectra being some line broadening. The obvious absence of this effect in the paramagnetic alloys may explain the abrupt increase of the average quadrupole splitting $eQV_{zz}/4$ on the breakdown of ferromagnetism as displayed by the data of Tables 1 and 2.

The magnetic hyperfine fields H_i given in Tables 1 and 2 were derived from the experimental values of the excited state Zeeman splitting $g_{1/2}\,\mu_N H_i$ with $g_{1/2} = 1.007 \pm 0.003$, the g-factor determined by nuclear gamma resonance in an external magnetic field [15, 16]. About 7% larger values for the hyperfine fields would be obtained from the groundstate splitting $g_{3/2}\,\mu_N H_i$ together with the NMR value [17] $g_{3/2} = 0.10589 \pm 0.00006$ for its g-factor. This discrepancy is due to the large hyperfine anomaly [18, 19] between the $1/2^+$ and the $3/2^+$ state of ^{193}Ir [5, 6, 15], and renders the interpretation of magnetic data in terms of effective magnetic fields somewhat uncertain. Our definition of H_i has been prompted by nuclear model arguments, according to which the uncommonly large hyperfine anomaly is due to the properties of the $3/2^+$ groundstate of ^{193}Ir [16], as wel as by the fact that $g_{1/2}\,\mu_N H_i$ is the parameter which actually is determined by our least squares fits, particularly in cases of poor resolution or statistics, where the ratio of splitting factors was kept constant at $G = g_{1/2}/g_{3/2} = 8.874 \pm 0.013$ [4, 6] for the Ir-Fe and at $G = 8.87 \pm 0.05$ [6] for the Ir-Ni alloys.

Good fits to the Zeeman patterns of the 14.4 keV γ-rays of ^{57}Fe were obtained with a superposition of six Lorentzian lines, if allowance was made for the

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Table 1. Summary of results obtained for Ir-Ni alloys at 4.2 K: Magnetic hyperfine fields H_i at the Ir nuclei, isomer shifts S of the 73.2 keV γ-rays with respect to Ir metal, mean values of the electric quadrupole coupling parameter $e\,Q\,V_{zz}/4$ of the groundstate of ^{193}Ir, full linewidths W at half maximum and magnetic moments μ per Ni atom. The sign of $e\,Q\,V_{zz}/4$ could only be determined for absorbers with non-zero hyperfine field H_i. For the samples containing 1.5 at.-% and 5.1 at.-% Ir the magnetic moment μ per Ni atom was not measured.

Ir concentration [at.-%]	Absorber thickness [mg Ir/cm^2]	H_i [kOe]	S [mm/s]	$\overline{eQV_{zz}/4}$ [mm/s]	W [mm/s]	μ [μ_B]
1.5	18	-429 ± 5	$+0.90 \pm 0.01$	-0.03 ± 0.01	0.78 ± 0.04	—
3.3	45	-399 ± 4	$+0.92 \pm 0.01$	-0.02 ± 0.01	0.90 ± 0.04	0.58 ± 0.03
5.1	85	-354 ± 5	$+0.91 \pm 0.01$	$+0.00 \pm 0.01$	1.02 ± 0.04	—
7.1	85	-314 ± 4	$+0.90 \pm 0.01$	-0.02 ± 0.01	1.16 ± 0.02	0.51 ± 0.03
14.1	150	-172 ± 5	$+0.85 \pm 0.01$	$+0.01 \pm 0.01$	1.56 ± 0.02	0.23 ± 0.03
20.0	160	-66 ± 12	$+0.81 \pm 0.01$	$+0.09 \pm 0.03$	0.98 ± 0.02	0.05 ± 0.03
23.5	115	—	$+0.73 \pm 0.01$	0.19 ± 0.02	1.36 ± 0.06	—
31.5	200	—	$+0.64 \pm 0.01$	0.42 ± 0.01	1.04 ± 0.02	—
41.7	230	—	$+0.55 \pm 0.01$	0.44 ± 0.01	1.00 ± 0.02	—
55.1	350	—	$+0.41 \pm 0.01$	0.48 ± 0.01	1.18 ± 0.02	—
73.5	100	—	$+0.26 \pm 0.01$	0.37 ± 0.02	0.96 ± 0.06	—
100.0	100	—	0	0	0.89 ± 0.02	—

Table 2. Summary of results obtained for Ir-Fe alloys at 4.2 K: Magnetic hyperfine fields H_i at the Ir nuclei, isomer shifts S of the 73.2 keV γ-rays with respect to Ir metal, mean values of the electric quadrupole coupling parameter $e\,Q\,V_{zz}/4$ of the groundstate of ^{193}Ir, full linewidths W at half maximum and average magnetic moments μ per Fe atom. The sign of $e\,Q\,V_{zz}/4$ could only be determined for cases with non-zero hyperfine field H_i.

Ir concentration [at.-%]	Absorber thickness [mg Ir/cm^2]	H_i [kOe]	S [mm/s]	$\overline{eQV_{zz}/4}$ [mm/s]	W [mm/s]	μ [μ_B]
0.7	10	-1405 ± 8	1.03 ± 0.02	-0.03 ± 0.02	0.78 ± 0.06	2.36
1.5	19	-1399 ± 8	0.99 ± 0.02	-0.01 ± 0.01	0.84 ± 0.06	2.41
3.1	40	-1413 ± 7	1.00 ± 0.01	-0.00 ± 0.01	1.28 ± 0.06	2.55
4.9	52	-1404 ± 7	0.94 ± 0.01	-0.03 ± 0.02	1.10 ± 0.06	2.51
6.8	50	-1411 ± 8	0.88 ± 0.02	-0.03 ± 0.02	1.18 ± 0.06	2.70
8.8	80	-1418 ± 8	0.84 ± 0.02	-0.02 ± 0.02	1.28 ± 0.06	2.70
11.0	130	-1420 ± 7	0.81 ± 0.01	-0.01 ± 0.01	1.42 ± 0.05	2.70
13.5[a,c]	110	-1435 ± 7	0.75 ± 0.01	-0.02 ± 0.01	1.66 ± 0.04	2.66
		—	1.12 ± 0.08	0.40 ± 0.20	0.94 ± 0.18	
13.5[b]	35	-1417 ± 7	0.77 ± 0.01	-0.01 ± 0.01	1.62 ± 0.02	—
16.2[a,c]	100	-1440 ± 7	0.72 ± 0.03	$+0.01 \pm 0.02$	1.98 ± 0.10	2.14
		—	1.09 ± 0.02	0.37 ± 0.02	0.96 ± 0.06	
16.2[b]	35	-1421 ± 7	0.71 ± 0.02	-0.01 ± 0.01	1.74 ± 0.06	—
22.5[a,c]	90	-1346 ± 30	0.74 ± 0.01	-0.13 ± 0.10	1.52 ± 0.32	0.90
		—	1.10 ± 0.01	0.74 ± 0.04	1.06 ± 0.06	
22.5[b,c]	38	-1401 ± 14	0.76 ± 0.05	0.02 ± 0.02	1.74 ± 0.12	—
		—	1.06 ± 0.01	0.78 ± 0.03	0.88 ± 0.04	
30.3	120	—	0.89 ± 0.01	0.43 ± 0.01	0.80 ± 0.06	—
40.4	140	—	0.77 ± 0.03	0.45 ± 0.01	1.04 ± 0.06	—
53.6	80	—	0.60 ± 0.02	0.43 ± 0.01	1.16 ± 0.04	—
72.4	90	—	0.27 ± 0.02	0.50 ± 0.01	1.46 ± 0.04	—
100.0	100	—	0	0	0.89 ± 0.02	—

[a] Results from spectra measured with filed samples as absorbers.
[b] Results from spectra measured with rolled foils as absorbers. For these the magnetic moments were not measured.
[c] The samples consisted of a mixture of the ferromagnetic α-phase and the paramagnetic γ-phase. The results in the upper line refer to the former, those in the lower line to the latter. The values for the magnetic moments μ are averaged over both phases.

broadening of particularly the outer lines in the alloys with all but the lowest Ir concentrations. The results obtained for the widths of the outermost (W_1), intermediate (W_2) and inner pairs (W_3) of lines are given in Table 3 together with the average hyperfine field at the iron nuclei. A single line was fitted to the ^{57}Fe spectra of the paramagnetic γ-phase alloys.

Table 3. Summary of results obtained from the nuclear gamma resonance absorption of the 14.4 keV γ-rays of ^{57}Fe at room temperature: Magnetic hyperfine fields H_i at the Fe nuclei, isomer shifts S with respect to the source of ^{57}Co in Pd, and full widths at half maximum of the outermost (W_1), intermediate (W_2) and inner (W_3) pairs of lines. The width of the single absorption peak observed for the paramagnetic γ-phase is given in the column of W_1.

Ir concentration [at.-%]	Absorber thickness [mg Ir/cm^2]	H_i [kOe]	S [mm/s]	W_1 [mm/s]	W_2 [mm/s]	W_3 [mm/s]
0.7	16	-330 ± 1	-0.185 ± 0.001	0.386 ± 0.002	0.322 ± 0.002	0.288 ± 0.002
1.5	15	-331 ± 1	-0.180 ± 0.001	0.410 ± 0.002	0.334 ± 0.002	0.282 ± 0.002
3.1	11	-331 ± 1	-0.162 ± 0.001	0.580 ± 0.002	0.420 ± 0.002	0.338 ± 0.004
4.9	7	-329 ± 1	-0.164 ± 0.001	0.470 ± 0.004	0.358 ± 0.004	0.292 ± 0.002
6.8	13	-332 ± 1	-0.153 ± 0.001	0.636 ± 0.004	0.458 ± 0.006	0.326 ± 0.006
8.8	8	-331 ± 1	-0.141 ± 0.001	0.816 ± 0.008	0.534 ± 0.008	0.402 ± 0.008
11.0	8	-332 ± 1	-0.129 ± 0.001	0.844 ± 0.006	0.564 ± 0.006	0.402 ± 0.006
13.5	8	-330 ± 1	-0.105 ± 0.001	0.938 ± 0.006	0.594 ± 0.006	0.424 ± 0.006
16.2	10	-328 ± 1	-0.111 ± 0.002	1.034 ± 0.014	0.658 ± 0.010	0.402 ± 0.010
22.5[a]	7	-331 ± 5	-0.111 ± 0.012	1.300 ± 0.056	0.642 ± 0.034	0.243 ± 0.042
		—	-0.197 ± 0.003	0.974 ± 0.018	—	—
30.3	5	—	-0.147 ± 0.009	0.630 ± 0.028	—	—
40.4	4	—	-0.111 ± 0.011	0.604 ± 0.034	—	—
53.6	4	—	-0.095 ± 0.008	0.620 ± 0.024	—	—
72.4	1	—	-0.052 ± 0.020	0.750 ± 0.050	—	—

[a] The sample consisted of a mixture of the ferromagnetic α-phase and the paramagnetic γ-phase. The results in the upper line refer to the former, those in the lower line to the latter.

The signs of the hyperfine fields do not follow directly from the nuclear gamma resonance data reported here. However, in pure iron the field is known [20] to be negative. The fields at Ir in Fe and Ni were, in the course of the present work, shown to be equally negative by measurements of the change of the effective hyperfine field with the magnitude of an externally applied magnetic field.

The isomer shifts of the 73.2 keV γ-rays of ^{193}Ir (Tables 1 and 2) are all given with respect to Ir metal, which is known [5] to exhibit a shift of $-(0.540 \pm 0.004)$ mm/s with respect to an Os metal source. The ^{57}Fe isomer shifts are referred to the source of ^{57}Co in Pd.

In Figs. 4 – 6 the essential results obtained for the Ir-Ni and IrFe alloys are plotted versus the iridium concentration.

In the Ir-Ni system the hyperfine field decreases about linearly with the Ir concentration and extrapolates to zero near 20 at.% Ir. This behaviour follows that of the magnetic moment per Ni atom as given in Table 1 and by CRANGLE and PARSONS [8], whose measurements cover the region up to 6 at.% Ir only. The isomer shift decreases monotonically with the Ir concentration from $+0.90$ in dilute IrNi alloys to zero in pure Ir, with a slight change of the slope of the isomer shift versus concentration curve near 20 at.% Ir, where the collective magnetism breaks down.

In the Ir-Fe system matters are complicated by the coexistence of the α and γ phases in samples with concentrations between 11.0 at.% Ir and 22.5 at.% Ir. Since only the bcc α-phase is ferromagnetic, the fcc phase gives rise to a single absorption peak in the middle of both the ^{193}Ir and the ^{57}Fe gamma resonance spectra (cf. Figs. 2 and 3). In the ^{193}Ir spectra of absorbers consisting of alloy filings the single peak due to the γ-phase became first visible at 13.5 at.% Ir and increased in relative intensity until, at 22.5 at.% Ir, the Zeeman pattern of the α-phase had virtually disappeared. However, in the rolled absorbers which were used for measurements on both ^{193}Ir and ^{57}Fe, the pure α-phase persisted up to 16.2 at.% Ir, and the coexistence of the two phases was observed for the 22.5 at.% sample only. The $\gamma \rightarrow \alpha$-phase transformation induced by cold-rolling was confirmed by the x-ray patterns taken of both the rolled and filed samples. For all samples outside the 13.5 at.% Ir to 22.5 at.% Ir range, the rolled and filed absorbers yielded essentially the same results and only weighted averages are given in Table 2.

The magnetic moment per iron atom at first increases with the Ir content of the alloys and then, having passed through a maximum near 10 at.% Ir,

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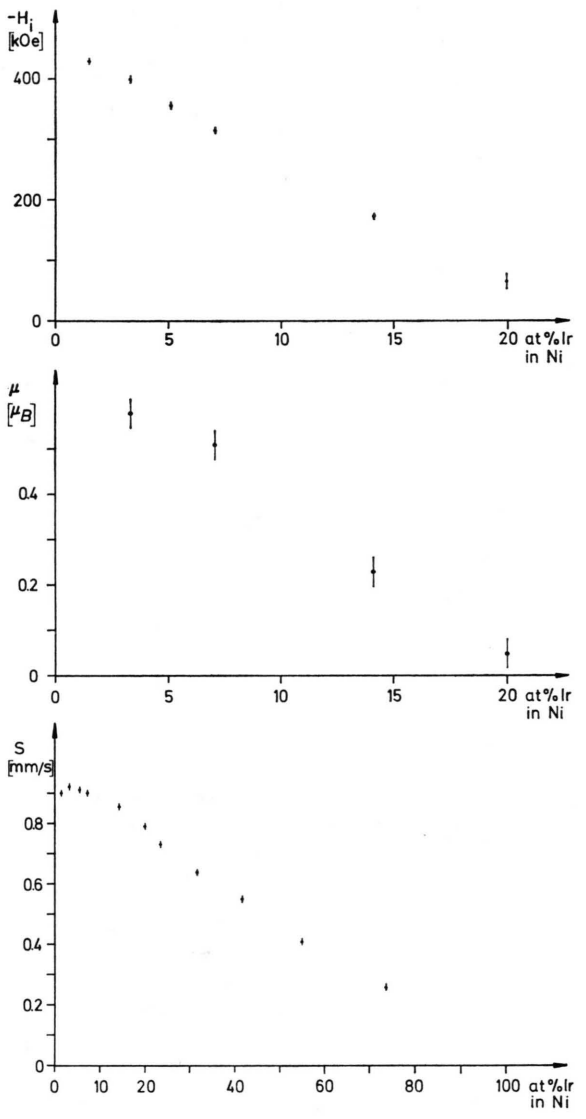

Fig. 4. Results for the Ir-Ni alloys at 4.2 K: Concentration dependence of the magnetic hyperfine field H_i at the Ir nuclei, of the magnetic moment μ per Ni atom and of the isomer shift S with respect to Ir metal.

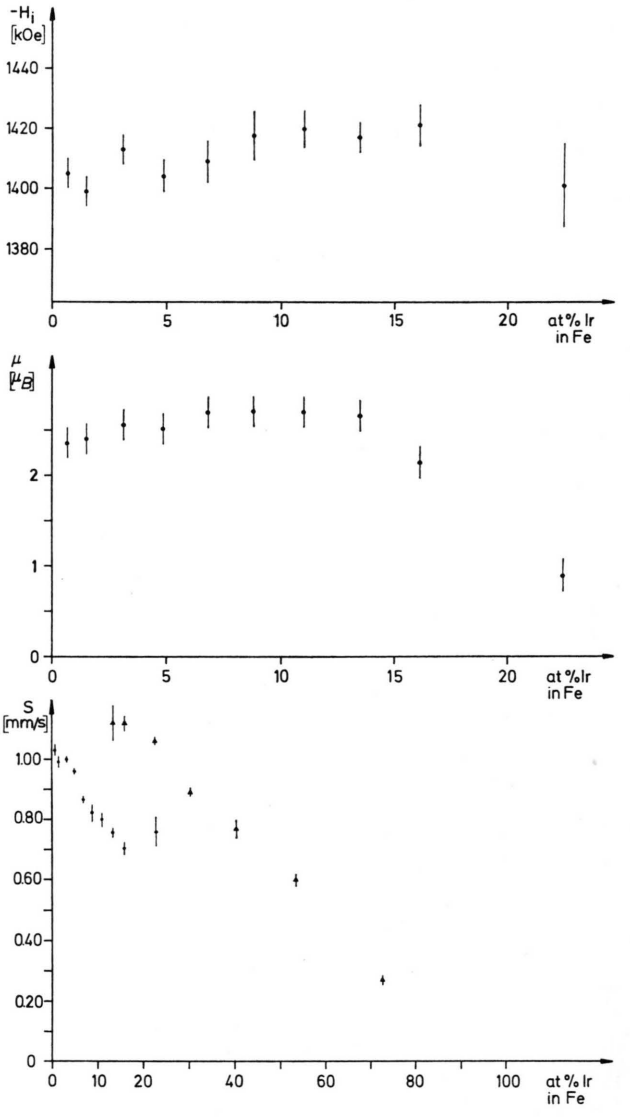

Fig. 5. Results for the Ir-Fe alloys at 4.2 K: Concentration dependence of the magnetic hyperfine field H_i at the Ir nuclei, of the magnetic moment μ per Fe atom and of the isomer shift S with respect to Ir metal. Dots refer to the bcc α-phase and triangles to the fcc γ-phase.

decreases rather rapidly (Fig. 5). This behaviour is in qualitative agreement with that observed by FALLOT [9], but the initial increase of the moment with the Ir concentration is more pronounced than in Fallot's work, the discrepancy perhaps being due to the fact that the present measurements were performed at 4.2 K and those of Fallot at room temperature. The presence of the paramagnetic γ-phase in alloys containing more than about 13 at.% Ir will result in a decrease of the average ordered moment per iron atom, as has already been pointed out by FALLOT [9]. Hence for higher Ir concentrations no definite conclusions as to the iron magnetic moment in the α-phase can be drawn from the available susceptibility data. The average hyperfine fields at both the Ir and the Fe nuclei in the α-phase alloys remain virtually constant up to 22 at.% Ir (cf. Figs. 5 and 6). At the same time

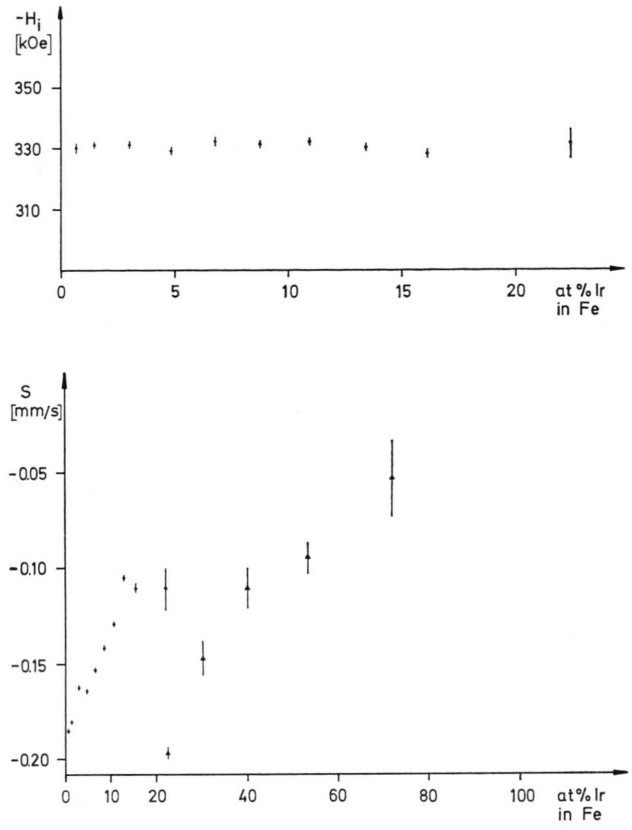

Fig. 6. Results for the Ir-Fe alloys at room temperature: Concentration dependence of the magnetic hyperfine field H_i at the Fe nuclei and of the isomer shift S with respect to the source of ^{57}Co in Pd. Dots refer to the bcc α-phase, triangles to the fcc γ-phase.

the linewidths increase considerably with the Ir concentration, which indicates the presence of a distribution of hyperfine fields. This is best seen in the ^{57}Fe spectra (Fig. 3 and Table 3), where the linewidth of the outermost pair of lines increases most rapidly.

4. Discussion

The magnetic hyperfine interactions observed at solutes of d transition elements in ferromagnetic host lattices are usually considered to originate mainly from the Fermi contact interaction due to the polarized conduction electrons and to polarization of the impurity core by unpaired d spins localized at the solute site [21]. The hyperfine fields at dilute Ir impurities in Fe, Co, and Ni are, at least approximately, proportional to the host magnetic moment [22], which suggests conduction electron polarization as their prime source. For IrFe this conjecture is borne out by the results of neutron scattering experiments [23], which yield a small value of $(0.2 \pm 0.2)\ \mu_B$ [24] for the local moment on the Ir impurities. With the Hartree-Fock value of FREEMAN et al. [25] for the core polarization field per unpaired iridium 5d spin the corresponding hyperfine field becomes $-(70 \pm 70)$ kOe, or only about 5% of the total hyperfine field at Ir in Fe. Unfortunately a similar estimate cannot be made for IrNi, where no neutron data are available. Recently an orbital contribution of $+(335 \pm 200)$ kOe to the hyperfine field at Ir in an iron matrix has been inferred from hyperfine anomaly data [15]. Similar measurements have been performed for IrNi [6]. However, ambiguities in the interpretation of such data make the magnitude of the surmised homogeneous fields at Ir in both Fe and Ni uncertain. In any case they seem to be small enough [6] to be neglected in the present context.

With conduction electron polarization as the dominating source of the Ir hyperfine field the approximate proportionality between the magnetic moment per atom of the ferromagnetic host and the Ir hyperfine field (Figs. 4 and 5) is easily understood.

A strong concentration dependence of the Ni magnetic moment is known not only for Ir-Ni alloys, but also for alloys of Ni with other 4d and 5d elements [8]. Only recently a pronounced concentration dependence has been found for the hyperfine field at Ni in Ni-Pd alloys by means of nuclear gamma resonance experiments [26] on ^{61}Ni. The near linearity of the decrease of the magnetic moment and the Ir hyperfine field in the Ir-Ni system can essentially be understood in terms of a rigid band model for the host, if each solute Ir atom is assumed to donate about the same number of electrons into the Ni 3d band. The majority spin band being already full in pure Ni, the additional electrons will change the population of the minority

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band only. At about 22 at.% Ir, the concentration at which the magnetic moment extrapolates to zero, we have to assume both Ni d-bands to be filled. The fact that the Ir hyperfine field extrapolates to zero at about the same concentration seems to justify our neglect of all sources of the Ir hyperfine field other than conduction electron polarization, but definite conclusions in this respect would require more data for concentrations near 20 at.% Ir.

In the Ir-Fe system the electrons supplied by the solute may populate both half-bands. Roughly balancing population numbers can explain the fact that both hyperfine field and susceptibility depend but weakly on the Ir concentration.

A striking feature in the behaviour of the Ir-Fe system is the virtual concentration independence of the hyperfine fields at both Ir and Fe, while Fig. 5 shows that the magnetic moment per iron atom increases slightly with the Ir content of the alloys, at least up to 10 at.% Ir. Whereas the Ir hyperfine fields exhibit a slight increase with concentration (Fig. 5), the average fields at the iron nuclei remain constant to within better than 1%, but at the same time the increasing linewidth indicates a distribution of hyperfine fields. The constancy of the average field contrasts with the behaviour of iron-rich Fe-Rh alloys, where the increase of the magnetic moment with the Rh concentration is paralleled by an increasing magnitude of the iron hyperfine field [27]. In both RhFe and IrFe the nearest and next-nearest Fe neighbours to the impurities have magnetic moments which are larger by about 3% and 5% [24], respectively, than the unperturbed iron moment. The analysis of CAMPBELL and BERNAS [28] gives a phenomenological description of the observation that the Fe hyperfine field is substantially changed in the neighbourhood of impurities like Os, Re [28] and W [29], but not in the vicinity of Ir solutes. Their interpretation implies, that in the Ir case the increase of the magnitude of the iron core polarization field, which is proportional to the increase in the magnetic moment, is just cancelled by the decrease of the conduction electron polarization due to the Ir neighbour with its nearly vanishing local moment. It is felt that this concept also accounts for the virtual independence of the average iron hyperfine field on the Ir concentration. Overlapping shielding radii may account for the distribution of the fields around the average value at higher Ir concentrations.

According to the relation [30]

$$\Delta S = (2\pi/3)\, Z e^2 \Delta\langle r^2\rangle \Delta |\psi(0)|^2$$

the isomer shifts S observed for the 73.2 keV and 14.4 keV γ-rays yield information on the changes $\Delta|\psi(0)|^2$ of the s-electron densities at the nuclei. The change $\Delta\langle r^2\rangle$ of the mean square nuclear charge radius is negative [31] for the 14.4 keV transition of ^{57}Fe and positive [3, 5, 32] for the 73.2 keV transition of ^{193}Ir. Hence our isomer shift results show (Figs. 4 – 6) that the s-electron densities at the Ir nuclei in the Ir-Fe and Ir-Ni alloys and at the iron nuclei in the Ir-Fe system decrease with the Ir concentration.

A discussion of this behaviour has to take into account mere atomic volume effects as well as true changes in the electron populations at the atoms under consideration. In principle estimates of the electron density differences due to changes of the atomic volume can be obtained from the pressure dependence of the isomer shifts. Such data are available for pure bcc iron [33, 34] as well as for ^{57}Fe in various metallic hosts [35], including Ta, W, Pt, and Au but not Ir. For the 73.2 keV transition in ^{193}Ir no nuclear resonance experiments at high pressure have become known, but on gold isomer shift measurements [36] as well as self-consistent-field calculations [37] of the electron density as a function of atomic volume have been performed.

These data can be combined with the changes of the lattice constants in the Ir-Ni and Ir-Fe alloy systems as determined from our x-ray patterns. The

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resulting estimates show, that isomer shifts of the observed order of magnitude can well be due to atomic volume effects. It seems worth noting that the experimental values of the electron densities at both Ir and Fe consistently increase with decreasing volume. This is true even for the discontinuity connected with the $\alpha \rightarrow \gamma$ transition. The change of the ^{57}Fe isomer shift during this phase transition is reminiscent of a similar change observed in pure iron [38].

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It is felt that ^{193}Ir isomer shift measurements under high pressure will help to improve the understanding of the isomer shifts in Ir alloys. At present it cannot be excluded that part of the increase in the s-electron density at the Ir nuclei in Fe and Ni as compared to pure Ir metal is due to the larger value of the effective screening radius in the alloys as compared to atomic iridium 5d orbitals.

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