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Magnetic hyperfine field at Hf site in $Hf(Fe_{1-x}Co_x)_2$ and $Hf(Fe_{1-x}Co_x)_2H_y$ at low Co concentration measured by TDPAC

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Abstract

The magnetic hyperfine field (mhf) on ^{181}Ta at Hf site was measured for the pseudobinary alloys $Hf(Fe_{1-x}Co_x)_2$ and $Hf(Fe_{1-x}Co_x)_2H_y$ with $0 \leqslant x \leqslant 0.4$ and $y \sim 0.15$. The results show three different values for the mhf in the range $0.01 \leqslant x \leqslant 0.1$ which we believe are associated with the following Hf nearest neighbour configurations: (12 Fe), (11 Fe, 1 Co) and (10 Fe, 2 Co). Relative magnitudes of the mhf for each configuration show that a short-range magnetic interaction is predominant in these compounds.

Keywords: Laves-phase compounds; Hyperfine fields; Nuclear methods

The stoichiometric AB_2 cubic Laves compounds are well suited for the study of the magnetic hyperfine field behaviour because of their cubic symmetry and well-defined positions of the atoms in the lattice. The Laves-phase compound $HfFe_2$ is a ferromagnet with $T_C \sim 600$°C, whereas $HfCo_2$ is a Pauli paramagnet. The pseudobinary compounds $Hf(Fe_{1-x}Co_x)_2$ crystallize in the cubic C15 Laves phase showing a spontaneous magnetization which decreases with increasing x and presents a transition from ferromagnetism to paramagnetism at $x \sim 0.7$, where a sharp collapse of the magnetic moment occurs [1, 2].

At low Co concentration, different Hf nearest-neighbour configurations can simultaneously be present in several sublattices ranging from undisturbed neighbourhood (12 Fe nearest-neighbour atoms) to configurations where one, two or more Co replace Fe atoms. The measurement of the magnetic hyperfine field at the non-magnetic Hf site in these situations is, therefore, an ideal way to investigate microscopically the collapse of the ferromagnetic order.

In the present work, the magnetic hyperfine field has been investigated at Hf site in pseudobinary alloys $Hf(Fe_{1-x}Co_x)_2$ and $Hf(Fe_{1-x}Co_x)_2H_{0.15}$ in the range of $0 \leqslant x \leqslant 0.4$ by TDPAC measurements.

The alloys were prepared by arc melting the constituent elements under argon with about 0.1% of the Hf atoms being substituted with radioactive ^{181}Hf. The resulting alloys were annealed for 24 h at 800°C. X-ray diffraction analysis showed the presence of one phase only. The hydrogenation of the samples was carried out in a standard Sievert-type facility. The final hydrogen concentration was determined by weighting the samples after hydrogenation in a very precise balance.

The TDPAC method is based on the hyperfine interaction of nuclear moments with magnetic field (H_{hf}) or electric field gradient (EFG). Since the EFG is vanishingly small due to the cubic symmetry of the charge distribution around a particular site in the cubic Laves phase, only the magnetic interaction is expected. Further details about the TDPAC method and the way the measurements were made is described in Ref. [3].

The TDPAC measurements were carried out with a conventional fast–slow coincidence set-up using a spectrometer with four BaF_2 detectors. The measurements were performed at 77 K in all cases and for the samples without Hydrogen, measurements were also made above 650 K in order to verify the cubic symmetry of the samples.

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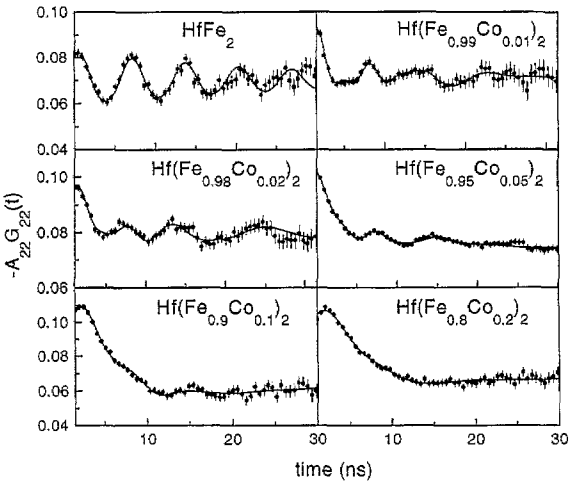

Fig. 1. TDPAC spectra for some of the alloys $Hf(Fe_{1-x}Co_x)_2$.

Results of the least-squares fitting of the experimental data to an appropriate function [3] for the magnetic interaction in a polycrystalline sample for some of the compounds are shown in Fig. 1. It can be seen that for $x = 0$ there is a unique field (H_1) indicating that the ^{181}Ta nuclear probes see only one nearest-neighbour Fe configuration that is 12 Fe atoms surrounding the probe. For $0.01 \leqslant x \leqslant 0.10$ there are three fields which we believe are due to the co-existence of the three different configurations corresponding to (a) Hf nuclei in undisturbed sites with only Fe nearest-neighbours (H_1), (b) Hf sites with 11 Fe and 1 Co nearest neighbour (H_2) and (c) the Hf nuclei where two or more Co atoms replace some of the 12 nearest neighbour Fe atoms (H_3). At still higher Co concentration, $x > 0.1$ only two magnetic fields are observed as the probability of the existence of totally unperturbed Hf nearest-neighbour configuration becomes very small.

The variation of the measured hyperfine field (H_n) as well as the site population (P_n) corresponding to different Hf nearest-neighbour configurations are plotted in Fig. 2 together with the width distribution of the hyperfine fields (δ_n) for some of the alloys. While the fractional population (P_1) corresponding to unperturbed configuration drops rather dramatically from 100% at $x = 0$ to 8% at $x = 0.1$ and then goes to almost zero for $x > 0.1$, the value (P_2) for the configuration (11 Fe, 1 Co) increases rapidly, goes through a maximum of 47% for $x = 0.05$ and then declines to 30% and stabilizes at this value for $x \geqslant 0.1$. The population (P_3) of the sites (10 Fe, 2 Co) also increase with increasing Co concentration, attaining a maximum value of about 60% at $x = 0.1$, and then increases only slightly for $x > 0.1$. These results are not quite consistent with the simple statistical calculations of the probabilities for various Hf near-neighbour

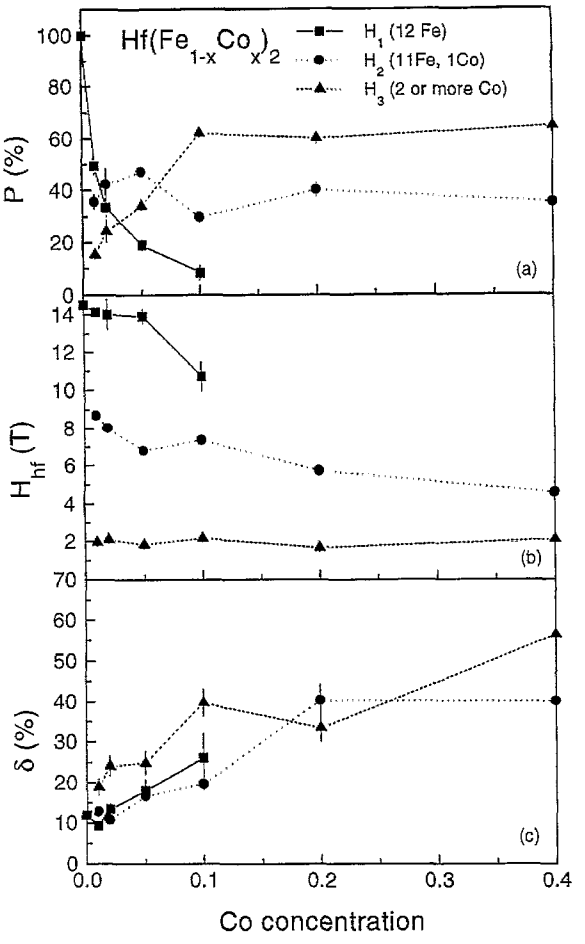

Fig. 2. Site population P (a), hyperfine field H_{hf} (b) and field distribution width δ (c) as a function of Co concentration.

configurations. Complex chemical order effects probably play rather important role in determining the near-neighbour configurations and may be responsible for this disagreement. A small amount of local disorder in these compounds, shown by the presence of quadrupole interactions, is discussed later.

The hyperfine field component H_1 declines slowly between $x = 0$ and 0.05, and rather dramatically from $x = 0.05$ to $x = 0.1$. This component is not observed any more for $x > 0.1$. The field H_2 also decreases with increasing Co concentration. On the other hand, the field H_3 remains practically unchanged in the studied range of Co concentrations. This behaviour indicates that long-range interactions play important role in determining the magnetic order in these compounds. At the same time, the fact that the observed field H_1 is much higher than both H_2 and H_3 indicates that the local environment around the probe plays a dominant role in the formation

of the magnetic moment and may involve some short-range interactions like hybridization of d orbitals. The distribution of hyperfine fields was found to increase with increasing Co concentration in all field components (see Fig. 2). This may indicate instability in the Fe–Co magnetic moments which are probably very sensitive to the local environment leading to wide distribution of hyperfine fields.

The TDPAC measurements carried out on samples above 650 K showed very small quadrupole interaction implying an almost cubic symmetry. The small quadrupole frequency was, however, observed to increase almost linearly with the Co concentration indicating some distortion of the local charge distribution around the probe as more Co atoms substitute Fe in the compound $HfFe_2$.

The maximum quantity of hydrogen that could be absorbed by the samples at 1 atm was found to be quite small, corresponding to a composition $Hf(Fe_{1-x}Co_x)_2H_{0.15}$, and was practically independent of the Co concentration. The TDPAC measurements on hydrogenated alloys revealed very similar hyperfine field behaviour as observed for the non-hydrogenated alloys.

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