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IMPURITIES IN ANTIFERROMAGNETS AT LATTICE SITES OF ZERO EXCHANGE FIELD

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Résumé. — La réapparition graduelle de l'aimantation macroscopique est observée dans des alliages Fe-Rh-Ir quand la température est réduite bien au-dessous du point de transition AF-F. Ce phénomène peut être expliqué en supposant qu'une petite fraction des atomes de fer occupe, comme « impureté », les sites Rh avec un champ d'échange nul en moyenne. Il est qualitativement démontré que les fluctuations du champ d'échange sont transmises à une longue distance par les impuretés magnétiques, donc une interaction effective entre les impuretés est attendue. Par conséquent, les moments Fe autour de ces « impuretés » sont inclinés produisant l'aimantation macroscopique observée dans les expériences.

Abstract. — A gradual reappearance of the macroscopic magnetization is observed in some Fe-Rh-Ir alloys on lowering the temperature well below the AF-F transition point. The phenomenon is explained by assuming a small fraction of iron atoms to occupy as « impurities » the Rh sites with zero average exchange field. The fluctuations of the exchange field are transmitted — as shown qualitatively — by the magnetic impurities to a fairly long distance and thus an effective interaction is expected to occur between the impurities leading to the canting of the Fe moments around the « impurities » which produces the observed net macroscopic magnetization.

The magnetic properties of the nearly equi-atomic Rh-Fe alloys have been intensively investigated during the last ten years [1]. The alloys are of CsCl-type ordered structure with G-type antiferromagnetism below the AF-F transition temperature T_i . Both experimental [2] and theoretical [3] evidences show that the Rh atoms have no magnetic moment in the AF state, while a fairly large magnetic moment $\sim 1 \mu_B$ appears in the F state on the Rh atoms as a result of the polarizing effect of the Fe neighbours.

If the Rh atoms are partially substituted by a third type of atoms and the substituent is a 3 d element, then T_i decreases radically and in most cases the F state remains stable in the whole temperature range until 0 °K. On the other hand, if the substituent is Ir or Pt, T_i increases, that is, the AF state is stabilized by this third component.

In the following we describe and tentatively interpret a new phenomenon in $\text{FeRh}_{1-x}\text{Ir}_x$ ($0 \leq x \leq 0,1$) alloys, namely, a gradual reappearance of the macroscopic magnetization on lowering the temperature well below T_i .

Above and near the AF-F transition the shape of the magnetization versus temperature curves and the other data reflect the tendencies known already from previous measurements [4]. However, well below the transition temperature several surprising anomalies were observed, not reported earlier.

The anomalies in the AF state can be explained by the presence of magnetic impurities at the lattice sites of zero average exchange field, i. e. in our case at the Rh sites. The aim of this paper is to show how such impurities affect the magnetic properties of the AF state.

Figure 1 shows the magnetization versus magnetic field curves for the $\text{FeRh}_{0,9}\text{Ir}_{0,1}$ sample at various temperatures below T_i . The straight line obtained just below T_i becomes gradually curved with decreasing temperature. This behaviour is characteristic for the onset of a ferromagnetic phase and it is a very remarkable fact that the field necessary to saturate this

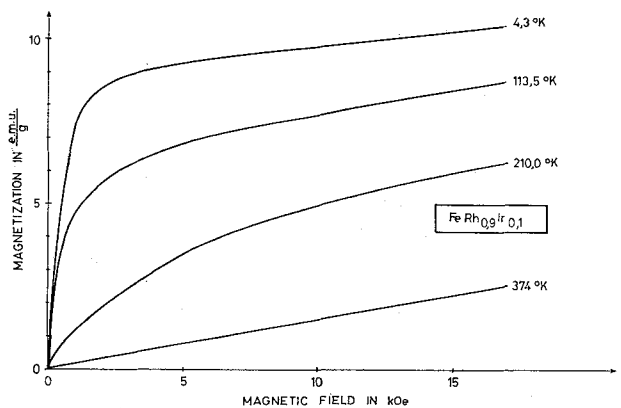


FIG. 1. — Magnetization curves for the sample $\text{FeRh}_{0,9}\text{Ir}_{0,1}$. The temperature of measurement is indicated on each curve.

phase decreases with decreasing temperature, while the « saturation » susceptibility remains essentially the same at each temperature. A « spontaneous » magnetization can be defined by extrapolating the saturation value to $H = 0$ along the straight line corresponding to the « saturation » susceptibility.

Figure 2 shows this « spontaneous » contribution for three samples representing three typical magnetic behaviours. Curve A corresponds to the case of the pure antiferromagnetic state which is stable in the whole temperature range below T_i , while curve B illustrates that the pure antiferromagnetic state appearing only just below the AF-F transition temperature is followed by a gradual onset of a ferromagnetic-like phase. Curve C shows the ferromagnetic phase to be present in the whole temperature range below T_i . The composition of the samples is indicated in figure 2, but no correlation between the low temperature behaviour and the composition could be found. The magnetic properties of the samples were found to be insensitive to a prolonged heat treatment at 1 200 °C.

The unusually large susceptibility in the AF state

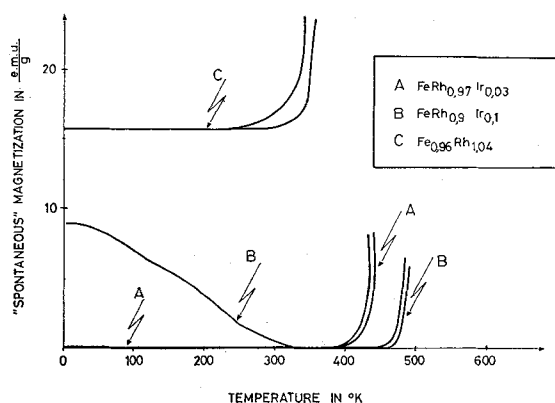


FIG. 2. — The «spontaneous» magnetization as a function of temperature, for various samples.

and other evidences found by Pál [5] indicate that there exists a strong intrasublattice (ferromagnetic) interaction in this material which defines the temperature of magnetic ordering, and a weak intersublattice coupling which is responsible for the antiferromagnetism below T_i . Because of the weakness of this second interaction an external magnetic field turns the sublattice magnetizations easily into the field direction. Just the same thing happens locally, if a magnetic impurity is placed in a Rh site. Since the impurity occupies a lattice point of zero average exchange field, its spin direction should follow the fluctuations of this field but since the coupling between the impurity and its nearest Fe neighbours is ferromagnetic, a local region of fluctuating canted iron moments should appear around the impurity. The canting is transmitted to rather distant iron atoms by the strong intrasublattice interaction when the intersublattice coupling is weak. The linear dimensions of the perturbed region defined by the ratio of these two interactions might be rather large (up to 5-10 atomic distances).

If two regions overlap, an effective interaction between the (perhaps very distant) impurities is expected to occur. This will define an ordering temperature T_0 for the iron moments, which depends on the range of the local canting and on the concentration of the impurities. Since the range of the effective interaction is large, a very small concentration of impurities is needed only to produce a noticeable effect. Below the ordering temperature the impurity spins are essentially parallel to each other resulting in a spatially and temporally fluctuating canting angle for the iron moments. If the average canting angle is large, it may even destroy the stability of the AF state. This fact may account for the effect of the 3d elements in Fe-Rh alloys as observed by Walter [6].

The three types of magnetic behaviour indicated in figure 2 can be interpreted by the different values of the ordering temperature T_0 . (Curve A corresponds to the case of $T_0 = 0$, while B and C to those of $T_0 < T_i$ and $T_0 > T_i$, respectively.)

The appearance of Fe atoms in Rh sites may be due to a small deviation from the exact stoichiometric composition, but mainly to a natural «disorder» which is probably connected with lattice defects. However, it is not quite clear, what governs the amount of this «disorder» iron in the Rh sites. The role of Ir seems to be nothing else than to strengthen the intersublattice interaction resulting in a definite decrease in the ordering temperature of the impurity moments. The sensitivity of the Fe-Rh alloys to the atomic disorder is well demonstrated by the experiments of Lommel and al. [7]. In alloys disordered by cold work the heat treatment re-establishes first the CsCl-type structure and then the complete AF-F transition. The difference in the time behaviour may be the consequence of a small residual iron disorder not observable by X-ray analysis. The large differences of virtually «identical» Fe-Rh samples may also reflect the variation of the iron disorder from sample to sample.

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