^{59}Co NMR in $ErCo_3$

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Abstract

NMR measurements of ^{59}Co in ferrimagnetic $ErCo_3$ have been carried out between 4.2 and 120 K and in external fields up to 7 T. The zero field powder spectrum at 4.2 K shows three resolved lines at 23, 65, and 96 MHz. In a field along the c-axis the two lower frequencies decrease with increasing field at 10 MHz/T, while the upper line increases smoothly at the same rate. This shows that the orientation of the moments on the part of the Co sites is inconsistent with the results of the neutron diffraction. The temperature dependence of the upper line gives strong indication that the Co-moment of this sublattice changes discontinuously to a smaller value above the metamagnetic transition temperature at 100 K.

Key words: NMR; ferrimagnet; rare earth transition-metal compound; itinerant electron metamagnetism

The RCo_3 compounds crystallize in the rhombohedral $PuNi_3$-type structure with two nonequivalent crystallographic sites for rare earth (R), with R_I on 3a and R_{II} on 6c positions, and three Co-sites, with Co_I on 3b, Co_{II} on 6c and Co_{III} on 18h positions [1]. Among the 1:3 compounds, YCo_3 is by far most intensively studied. These studies revealed that YCo_3 is a very weak itinerant ferromagnet (T_c =300 K). In this compound two field-induced metamagnetic transitions were observed at 60 and 82 T [2]. In the case that R is a magnetic rare earth element the magnetism in such an intermetallic compound is strongly modified due to the interaction of the localized 4f-electrons and the itinerant d-electron system. Owing to the strong molecular field the net value of the d-sublattice magnetization is modified. In a previous study of the magnetic structure of $ErCo_3$ by neutron diffraction an antiparallel moment arrangement along the c-axis was found at 4.2K (the Er and Co sublattices are orientated in an opposite direction along the c-axis) [3] . Recently, the phase transition associated with the temperature induced change in the Co magnetic state was found [4,5]. In order to clarify the nature of $ErCo_3$ microscopically, including the anomaly at 100 K, measurements of Co NMR were carried out.

The polycrystalline $ErCo_3$ sample material was prepared in a high-frequency induction furnace under argon atmosphere in a water-cooled copper boat. For some RCo_3 compounds a wide homogeneity region has been found, the stoichiometry varying between 1:2.88 and 1:3. For the sample used in this investigation the 1:3 stoichiometry was chosen. The $ErCo_3$ ingot was subsequently annealed at 850°C for 24 hours. The NMR spectra were measured by frequency and magnetic field sweeps in the temperature range from 4.2 to 120 K using a homemade phase-coherent pulsed spectrometer.

Three resonance lines of ^{59}Co NMR were found at

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about 23, 65, and 95 MHz at 4.2 K from the measurement of frequency sweep, as can be seen in Fig. 1. Comparison with the NMR line shapes observed by Itoh et al. [6] in $Y_{1-x}Nd_xCo_3$ and $Y_{1-x}Gd_xCo_3$ would suggest that the comparatively narrow lines at 95 and 23 MHz correspond to Co_{III}(18h), the broad one at 65 MHz to Co_I(3b). Co_{II}(6c) might produce another around 180 MHz, above the upper frequency limit of our spectrometer.

Experiments in external fields show that all three frequencies change at a rate of 10.01 MHz/T, the gyromagnetic ratio of the ^{59}Co isotope. This proves that all resonances are due to Co, and that all Co moments are aligned collinear to the applied field. This is expected for an easy axis magnetic powder where the grains will align along the field. This orientation was further confirmed by measurements of a powder which was oriented in an external field and fixed in paraffin at room temperature for the low temperature experiments. There is no indication of any field induced magnetic transition below 7 T at low temperature. Fig. 2 shows the temperature dependence of 95 MHz resonance line. The metamagnetic transition near 100 K reported by neutron diffractions [5] is observed in the resonance. The intensities of this line decreases with increasing temperatures below 100 K and the resonance frequencies move down corresponding to decreasing moments at 18h sites. The 23 MHz resonance (not shown) indicates another transition at lower temperature by a similar behavior near 50 K.

With increasing the applied magnetic field the Co-resonance frequencies change in two opposite ways - the frequency of the 23 MHz- and 65 MHz-sites decreases, on the other hand, the line at 95 MHz increases. This proves that the Co moments belonging to the 23 and 65 MHz lines are parallel to the overall magnetic moment (to the external field), but the Co moments at the 95 MHz-site are anti-parallel to the overall moment.

The inconsistency of this assignment with the results of neutron diffraction might be resolved as follows. Sites of Co and R (rare earth) are cubic and hexagonal for both elements in $ErCo_3$. The easy axes in cubic site of R are one of the (001) directions along the c-axis (in the pseudo cubic positions), and the ones in hexagonal site are the (111) of the cubic site corresponding to the c-axis. Thus there exists frustration of exchange interactions between Co and R, especially for Co_{III}(18h). An evidence of the fluctuation can be found in the split lines of Co_{III}(18h) in $NdCo_3$ NMR [6]. When the moments form domains in two stable states NMR detects the internal fields in both the states, however, neutron scattering will observe only the averaged moment on the site. The macroscopic magnetization will be difficult to describe in this interpretation, depending on the correct relative line intensities. Therefore, further investigations of ^{59}Co NMR in $ErCo_3$, including spin-

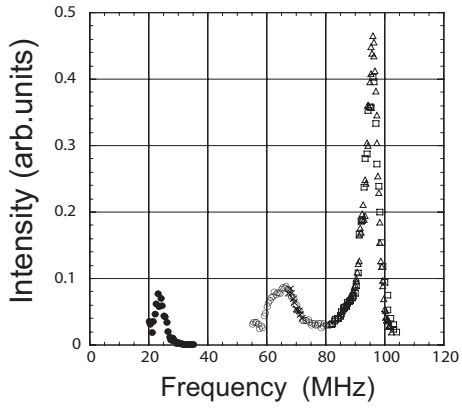

Fig. 1. Frequency-sweep (open symbols) and field-sweep (crosses) ^{59}Co NMR spectra in $ErCo_3$ taken at 4.2 K in zero external field. We confirmed that there is no resonance peak in the frequency range between 30 and 55 MHz. Each resonance intensity was divided by the measured frequency.

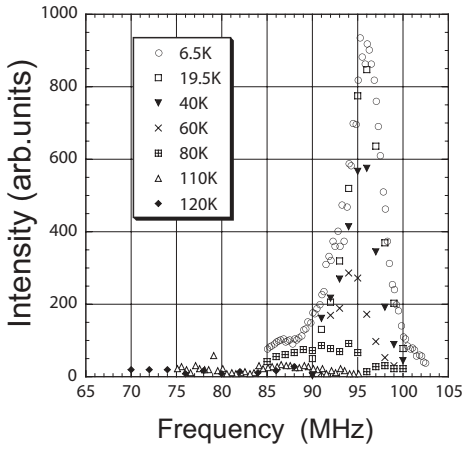

Fig. 2. Temperature dependence of Frequency-swept ^{59}Co 95 MHz-resonance line in $ErCo_3$ in zero external field. Each resonance intensity was multiplied by the measured temperature.

relaxation time T_1 and sin-spin relaxation time T_2, will be needed and are now in progress.

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