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Magnetic hyperfine fields on ^{140}Ce probes substituting for the rare earth in RCo_2 laves phases

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Abstract

The magnetic hyperfine fields (MHF) acting on ^{140}Ce probes substituting for the rare-earth sites in RCo_2 Laves phases (R = Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm) were determined by perturbed angular correlation spectroscopy. The MHF scales very well with the Co magnetic moment throughout the series as already observed with ^{181}Ta, and to a lesser extent, with ^{111}Cd probes. It is shown that the orbital contribution to the MHF is about one order of magnitude smaller than in the free Ce^{+3} ion. The decrease was attributed to the de-localization of the Ce 4f electrons.

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In rare-earth–transition-metal binary alloys the magnetic properties are determined by the interaction of the magnetic moments of the rare earth (R) 4f electrons with the itinerant 3d electrons of the transition metals. In the RCo_2 Laves phases, R being a rare earth, the density of states at the Fermi level is not sufficient to satisfy the Stoner criterion and then the 3d magnetization is reached by the f–d exchange mechanism in which the 4f spins polarize the R 5d band which is hybridized with the Co 3d band. The 3d spins are antiparallel to the 4f spins and the 3d polarization increases monotonically with the 4f spin of the rare earth throughout the RCo_2 series [1].

The measurement of magnetic hyperfine fields (MHF) with suitable probes provides useful information on some microscopic properties of the electronic structure of magnetic materials. In the present investigation one reports on the results of perturbed angular correlation (PAC) measurements of MHF acting on the ^{140}Ce probes substituting for the R in the RCo_2 series of compounds. Since the ^{140}Ce probe possesses an open 4f electronic shell, an orbital contribution to the MHF is expected besides the contribution due to conduction electron polarization (CEP) and the core polarization (CP), usually seen with non-rare earth probes. By comparing the ^{140}Ce MHF values with those obtained previously with ^{181}Ta [1] and ^{111}Cd [2], one can estimate the orbital contribution to the MHF and thus infer on the nature of the 4f states of the R ions in RCo_2 compounds. These compounds are especially suited for this purpose because as stated above, the 3d–5d polarization varies as a function of the 4f spin state and then a decomposition between the spin and orbital effects on the MHF may, in principle, be performed.

The RCo_2 samples were prepared by repeatedly melting the constituent elements along with very small amounts (less than 0.1%) of the radioactive ^{140}La isotope produced by irradiating natural La with neutrons in the IEA-R1 reactor. The samples were annealed under ultra pure argon for 48 h at 700°C and analyzed by X-ray diffraction. The PAC measurements were carried out with a conventional fast-slow coincidence set-up with four conical BaF_2 detectors. The gamma cascade of 329–487 keV in ^{140}Ce populated from the β^- decay of ^{140}La and with an intermediate level of spin $I = 4^+$ at 2083 keV ($T_{1/2} = 3.45$ ns) in ^{140}Ce was used to measure the MHF at ^{140}Ce. A detailed

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description of the method can be found in Ref. [3]. The measurements were taken at temperatures between 4.2 to 10 K, depending on the sample. A unique MHF is observed with narrow distribution of the Larmor frequencies, except for $GdCo_2$ and $SmCo_2$, giving a good indication that the probes substitute for the R, since, in Co sites, more than one MHF component is found by symmetry arguments [4].

The results are shown in Fig. 1 where the MHF acting on ^{140}Ce is plotted against the R 4f-spin projection, given by $(g-1)J$, along with the corresponding data for ^{181}Ta and ^{111}Cd taken from Ref. [1].

The MHF curves shown in Fig. 1 scale quite well with the Co magnetic moment μ_{Co}, especially for the heavy R and for the ^{181}Ta and ^{140}Ce probes. In what follows we will restrict ourselves to this region of the plot. The data from the ^{111}Cd are well reproduced by the relation $B_{hf} = B_{CEP}(M) - B_{CEP}(R) = \chi_{3d}\mu_{3d} - \chi_{4f}(g-1)J$, with constant susceptibilities χ_{3d} and χ_{4f} , since, being a closed d shell probe, ^{111}Cd senses only the CEP form the 3d and 4f spins [1]. A similar analysis for ^{181}Ta and ^{140}Ce is not possible owing to their d polarization (dP) accompanied by the CP. The later is opposite to the dominant CEP field thus explaining why the ^{181}Ta field is smaller than the field sensed by ^{111}Cd. But if one takes the values for the outer s electron hyperfine coupling parameters [5] and combines the MHF results for ^{111}Cd and ^{181}Ta, one arrives at a value of 4.2 T due to the $(B_{CP} + B_{dP})$ at the Ta probe in the case of $DyCo_2$.

The outer shell configuration of Ta is $5d^36s^2$ while that of the Ce is $4f^15d^16s^2$ and the difference in the electronic core configuration between these two atoms is a filled 4f shell. In this way one expects that the $(B_{CP} + B_{dP})$ contribution to the MHF is very similar for the two probes owing to the opposite contribution between B_{CP} and B_{dP} and also because the polarization of Ta d^3 electrons are expected not to be too dissimilar to the polarization of Ce $4f^15d^1$ electrons, at least within a factor of 3/2. Combining these results taking into account the hyperfine coupling parameter for Ce [5] one estimates the orbital contribution to the ^{140}Ce MHF as −15.7 T, again for the $DyCo_2$ case.

It has to be taken into account that the sign of the fields were not determined and then there is the possibility that the ^{181}Ta and ^{140}Ce measured fields have opposite signs. In this case, the orbital contribution due to the $4f^1$ electrons on ^{140}Ce would be 28.9 T.

Thus the higher possible experimental value determined in this work is near one order of magnitude smaller than the known value of 192 T due to the orbital MHF of a free Ce^{+3} ion [6]. The difference is hard to be justified with possible errors made in the present approximation. Moreover, the same conclusion was already drawn before with the $CeIn_3$ compound after PAC experiments on ^{140}Ce probes and electronic structure calculation [7].

It is usually believed that the driving mechanism of the spin polarization of the hybridized $3d_{Co}$–$5d_R$ band in RCo_2 compounds, is the local 5d–4f exchange field of the rare-earth [1]. This would preserve the local nature of the R 4f state and result on a contribution to the MHF comparable to the free Ce^{+3} ion, in disagreement with the present investigation. One then concludes that in addition to this mechanism, some degree of hybridization between the 5d and 4f states may occur. In this way, the R 4f states lose the strictly local character, expand more nearly to the nearest outer 5d shell and become more affected by the crystalline field, decreasing the shielding effect observed in isolated ions. On the other hand, this feature may be exclusive to the Ce compounds, as it is known that in this case the $4f^1$ states are better treated as de-localized [8] and our measurements were also been performed with Ce. First principles electronic structure calculations and MHF measurements on other R probes instead of Ce would be helpful to confirm this hypothesis.

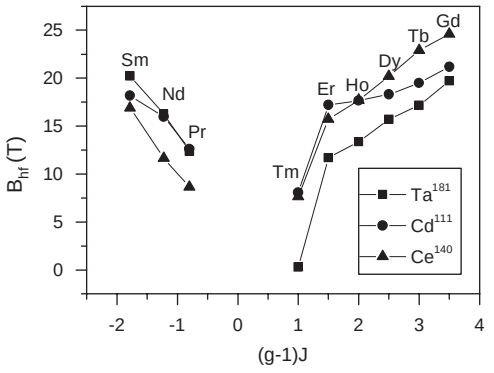

Fig. 1. Absolute values of magnetic hyperfine field B_{hf} acting on the indicated probes in RCo_2 as a function of the 4f spin projection of the rare-earth R. Ce : present work; Ta and Cd data taken from Ref. [1].

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