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New magnetic configuration in paramagnetic phase of HoCo$_2$


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X-ray magnetic circular dichroism (XMCD) measurements on HoCo$_2$ reveal the inversion of Co moment at temperatures higher than the critical temperature, $T_c$, showing that the net magnetization under a field of the Ho and Co sublattices remain antiparallel even above $T_c$. The Ho moment also changes its orientation to align antiparallel to the applied field at high temperature giving rise to a new magnetic configuration in the paramagnetic regime. Transverse susceptibility (TS) and small angle neutron scattering (SANS) measurements performed above $T_c$ indicate the existence of sizable magnetic short-range correlated regions in HoCo$_2$. First principles calculations based on spin polarized local-density approximation, LSDA+U have been performed to obtain insights on the origin of the short-range correlated volume. © 2012 American Institute of Physics. [doi:10.1063/1.3672258]

INTRODUCTION

For more than 20 years, the mechanisms that define the magnetism in $RCo_2$ ($R$ = rare-earth) compounds have been intensively studied as they are an ideal starting point to understand the physical properties of other intermetallic compounds. In magnetic $RCo_2$, the exchange interaction, $J$, between $R$ 4f from a heavy rare earth and Co 3d is always negative, and one sublattice tends to align antiparallel to the magnetic moment of the other. In the paramagnetic regime, the hybridization of Co 3d and $R$ 5d is responsible for many physical properties observed in these systems. However, a new magnetic configuration within the paramagnetic phase of ErCo$_2$ was identified in 2007 as result of different experimental techniques. Within this magnetic configuration, named “parimagnetism,” the Co moments are disordered at the long-range but a net Co magnetic moment, antiparallel to the applied field and to Erbium moment, is found. At a certain temperature, denoted as flipping temperature $T_f$, well above the critical temperature, $T_c$, the Co moment changes its preferred orientation recovering the normal paramagnetic configuration. Short-range order correlations between the Co atoms have been identified within this new magnetic configuration. We have selected HoCo$_2$ compounds to investigate whether parimagnetism is present in other lanthanide-based $RCo_2$.

Polycrystalline ingots of HoCo$_2$ were prepared following the procedure described in a previous work. X-ray diffraction (XRD) pattern was recorded in a Rigaku RTO 5000RC diffractometer using Cu-Kα radiation. The Rietveld analysis shows no impurities within the 5% of accuracy of powder diffraction methods. Magnetic characterization as well as the TS measurements were performed at the Servicio de Medidas Físicas of the University of Zaragoza-CSIC. Magnetization as a function of temperature measurements show $T_c = 76$ K.

X-ray absorption (XAS) and x-ray magnetic circular dichroism (XMCD) spectra at the Co $L_{2,3}$ and Ho $M_5$ edges were measured at temperatures between 5 K and 350 K at $H = 1$ T in UE46-PGM1 beamline at the BESSY synchrotron facility. To prevent surface oxidation, samples were cleaved under high vacuum before starting the exposure to the x-rays. Figure 1 shows the XMCD spectra for both Co $L_{2,3}$ and Ho edges.
The spin dynamics of HoCo2 magnetic moments were studied by means of radio-frequency transverse susceptibility (TS). The analysis of the TS profile have previously proved the existence of magnetic clusters in ErCo2.7 The measurements in HoCo2 polycrystalline sample were performed using a self-resonant circuit oscillator based on a simple inverter cell using CMOS transistors in cross-coupled topology. Details of the technique and the circuit can be found in Refs. 7 and 8, respectively. In this particular case, we are interested in the quantity $\frac{\Delta I_T}{I_T \text{ sat}} = \frac{I_T(H) - I_T(H_s)}{I_T \text{ sat}} \times 100\%$ as a function of the applied field $H$, here $I_T$ is the transverse susceptibility at the saturating field $H_s$, which in this study has been selected as 1 T. Figure 3 shows the bipolar TS scans for selected temperatures at 10 K, 85 K, 110 K, and 150 K. The TS profile shows pronounced switching peaks in the ferromagnetic transition. However, for $T > T_c$, the peaks are narrower as a manifestation of a collective process of Ho moments at the magnetic transition. For $T > 150$ K, $H_s$ values are clearly lower but finite suggesting the occurrence of magnetic clusters in this range of temperatures. The shape of TS profiles above $T_c$ coincides with the different magnetic configurations observed by XMCD. As combination of TS measurements and our XMCD study, we propose a scheme of new magnetic configurations for HoCo2 shown in Fig. 4. The Co and Ho magnetization are arranged antiparallel below $T_c$. Above $T_c$ and up to a certain temperature $T_f$, the Co magnetization sublattice on average is aligned antiparallel to the applied field and to the Ho magnetization. Above $T_f$, the Co magnetization on average is aligned to $H$. At certain temperatures between 83 K and 92 K, the sign of the XMCD signal for Ho M5 changes above 83 K, which in the case of HoCo2 is between 83 K and 92 K. Surprisingly as the temperature is raised, the sign of the XMCD signal for Ho M5 also changes at certain temperature $T_f$. The bottom panel on the right side of Fig. 1 shows the inversion of Ho moment at 150 K. This magnetic configuration remains up to 350 K. The measurements show that the Co and Ho sublattice magnetization values are small, but the net moment is different from zero well above $T_c$ in both cases. At $T > T_f$, the moment of the Ho sublattice is aligned antiparallel to the moment of Co sublattice and Ho due to the Co–Co exchange interaction.

The origin of the paramagnetic configuration observed just above $T_c$ in ErCo2 has been explained due to the collective flipping of the short-range magnetic correlated regions formed by Co moments.5 By means of small angle neutron scattering (SANS) we aimed to observe the occurrence of short-range magnetic correlations in the paramagnetic state of HoCo2. SANS measurements were performed in D16 diffractometer at the Institute Laue Langevin. The neutron wavelength used was $\lambda = 4.54$ Å and the SANS intensity data were collected at zero applied magnetic field and at temperatures between 4 K and 300 K. From the total SANS intensity, $I_0$, only the magnetic intensity, $I_{m}$, is relevant for this study because $I_{m}$ comes from the clustering structures. At 300 K, magnetic correlations are expected to be minor. The difference between $I_0$ and the SANS intensity recorded at 300 K can be analyzed to give a good approximation of the evolution of the correlation length with temperature. Figure 2 shows the temperature dependence $I_0 - I_{300 \text{ K}}$ for selected $q$ values. The intensity increases as the temperature is reduced to reach a plateau at 140 K, which indicates the occurrence of magnetic correlated regions close to this temperature. Close to $T_c$, the curves diverge to infinity as the system approaches to the long-range order.
temperatures $T_f > T_f$, the Ho magnetization sublattice aligns antiparallel to the applied field and Co moment.

First principles calculations based on the LSDA + $U$ approximation have also been performed for HoCo$_2$ to obtain insights on the origin of the Co-short-range correlated volume. In the theoretical model, we have considered a $P1$ unit cell with eight inequivalent positions for Ho atoms and 16 inequivalent positions for Co atoms, a $U = 4.0$ eV value has been used in the calculations. Magnetic moment has been assigned to each atom in such a way that the net magnetic moment in the unit cell is 0 to simulate the paramagnetic state. The inclusion of an impurity located at an interstitial position of the cell is needed to give rise to the nucleation of Co clusters. The imbalance of electron charge density between different Co sites due to the impurity within the unit cell could give rise the formation of magnetic short range correlations between Co atoms.

In summary, our XMCD study demonstrates that Ho and Co net magnetic moments are different from zero within the paramagnetic region of HoCo$_2$. A new magnetic phase diagram proposed for HoCo$_2$ shows two paramagnetic configurations above $T_c$. The origin of these new magnetic configurations, is ascribed to the magnetic clusters detected by SANS and TS techniques. Theoretical simulations suggest that an impurity is needed for the nucleation of the magnetic correlated regions in the sample. These results also suggest that paramagnetism may be a common phenomenon among ferrimagnetic $R$Co$_2$.

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