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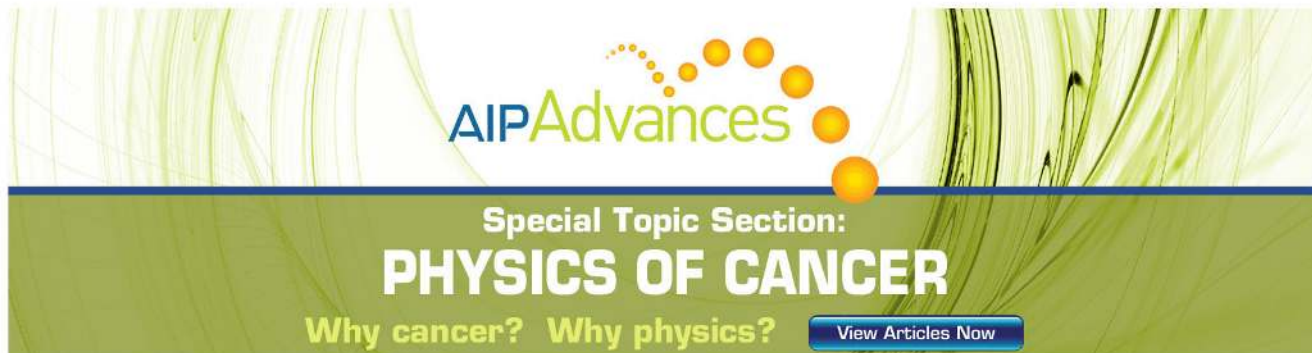
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Magnetic switching and magnetic transitions in ErCo₂ probed by radio-frequency transverse susceptibility

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ErCo₂ represents a typical example of magnetism of itinerant electron systems and metamagnetic processes and has been the subject of extensive research. We present here the first study of radio-frequency transverse susceptibility (TS) of bulk ErCo₂ using a self-resonant tunnel-diode oscillator technique. TS measurements reveal the collective magnetic switching of the Er moments at temperatures below the ferrimagnetic transition temperature, $T_c \sim 32$ K, and the existence of Co nanoclusters with short-range correlations at $T_c < T < T_f$ (T_f denoted as the flipping temperature). The difference in the magnetic configuration between the ferrimagnetic, parimagnetic, and paramagnetic states, as well as the change from the paramagnetic to parimagnetic regime upon varying dc magnetic fields are also probed by TS experiments. These findings are discussed in the context of our previous investigations using other different techniques which provide further insights into the magnetism and the so-called parimagnetism phenomenon in ErCo₂. © 2011 American Institute of Physics. [doi:10.1063/1.3553936]

Laves-phase ferrimagnetic compound ErCo₂ represents a typical example of magnetism of itinerant electron systems and metamagnetic processes and has been the subject of extensive research.¹ Its magnetism in the ordered state is dominated by the magnetic moments of the Er sublattice ($8.8 \mu_B$ /atom), whereas Co has a low magnetic moment in the paramagnetic phase of about $0.2 \mu_B$ which increases up to $1 \mu_B$ at low temperatures upon cooling through the ferrimagnetic ordering temperature, $T_c \sim 32$ K.² Recently, a new magnetic configuration has been identified as *parimagnetism* in ErCo₂ by means of x-ray magnetic circular dichroism, small angle neutron scattering (SANS), and ac magnetic susceptibility.³ This configuration is present at $T_c < T < T_f$ (T_f —a certain flipping temperature), where the net Co magnetic moment is aligned antiparallel to the Er one and the applied field. Moreover, the formation of magnetic nanoclusters (0.8 nm) in the parimagnetic range has been attributed to correlated Co moments, giving rise to a Griffiths-like phase in ErCo₂.⁴ At $T > T_f$, the Co moments are, on average, oriented parallel to the applied magnetic field and to the Er sublattice, as expected in a standard paramagnetic phase. Interestingly, a magnetic phase diagram of this compound (Fig. 1) shows three different magnetic configurations corresponding to the ferrimagnetic, parimagnetic, and paramagnetic regimes.

To study the spin dynamics of ErCo₂ in these three regions, radio-frequency (RF) transverse susceptibility (TS) measurements were performed using a very sensitive, self-resonant tunnel-diode oscillator (TDO) technique. The

ErCo₂ sample was fabricated following the same procedure as described in previous works.^{3–5} The obtained results provide physical insights toward a complete understanding of the magnetic switching of the Er and Co sublattices and the so-called parimagnetism phenomenon in ErCo₂.

The TDO system consists of a self-resonant circuit with a LC tank, operating at a resonant frequency of around 20 MHz.⁶ The sample ingot was inserted into a gel cap that snugly fits into the inductive coil, which is integrated into the physical property measurement system (PPMS) from quantum design such that the perturbing RF magnetic field inside the coil ($\sim 10^{-3}$ T) is oriented perpendicular to the superconducting magnet. In this experiment, the measured quantity is the shift in the resonant frequency of the circuit as the static field and the temperature are varied. Since the change in frequency of the circuit is a direct consequence of the change in inductance as the sample is magnetized, the quantity Δf is directly proportional to $\Delta\chi_T$. In the present study, we have considered the quantity $\Delta\chi_T/\chi_T\% = [\chi_T(H) - \chi_T^{\text{sat}}] * 100/\chi_T^{\text{sat}}$, where χ_T^{sat} is the transverse susceptibility at the saturating field $\mu_0 H^{\text{sat}}$. It has been theoretically shown that a TS scan peaks at the anisotropy fields ($\pm H_K$) and at the switching field (H_S) for a unipolar sweep of the dc magnetic field.⁷ This TS technique has been used by us over the years with great success to study the anisotropic magnetic properties in a variety of systems from multilayered thin films⁸ to single crystals⁹ and nanoparticles.¹⁰ A careful analysis of TS profiles (their shape and magnitude) has also revealed several interesting features, such as phase coexistence and magnetic clusters that are present in doped manganites⁹ and cobaltites.¹¹

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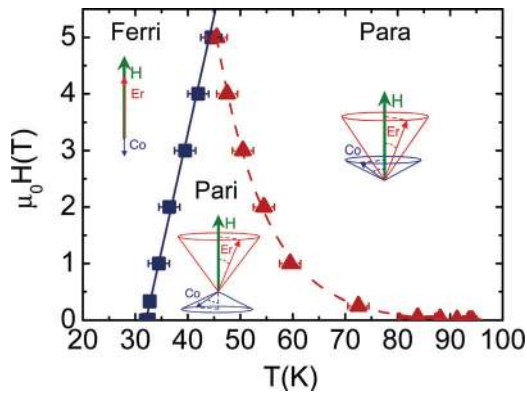


FIG. 1. (Color online) Magnetic phase diagram of the ErCo_2 compound.³ (■) denotes the first-order ferrimagnetic transition (T_c) and (▲) indicates the flipping temperature (T_f).

Since the spin dynamics of ErCo_2 depends on the initial configuration of the magnetic moments, in the present study TS measurements were performed by saturating the sample to $\mu_0 H^{\text{sat}} = 1$ T and 5 T over a temperature range $10 \text{ K} < T < 100 \text{ K}$. Figure 2 shows the bipolar TS scans for $\mu_0 H^{\text{sat}} = 1$ T and 5 T for three different temperatures ($T = 20, 50,$ and 90 K) representing the magnetic behaviors in the corresponding ferrimagnetic, parimagnetic, and paramagnetic ranges. It can be seen in Figs. 2(a) and 2(d) that in the ferrimagnetic region (e.g., $T = 20 \text{ K}$), the switching peaks are very sharp and the peak location (H_S) remains almost unchanged with the saturating field, $\mu_0 H^{\text{sat}}$. Evolution of H_S with temperature is shown in Fig. 3(a) for measurements with $\mu_0 H^{\text{sat}} = 1$ T. We have found that at $T = 20 \text{ K}$ the H_S is about $9.5 \times 10^{-2} \text{ T}$, which remain almost constant in the ferrimagnetic region until T reaches $T_c \sim 32 \text{ K}$. There is a sharp increase in H_S at T_c (0.135 T for $\mu_0 H^{\text{sat}} = 1$ T and 0.13 T for

$\mu_0 H^{\text{sat}} = 5 \text{ T}$). This increase is likely associated with the magnetostructural phase transition at T_c in ErCo_2 , similar to the case reported in $\text{Pr}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ where a magnetocrystalline anisotropy transition is driven by a structural transition.¹¹ These observations reveal a collective switching of the Er moments at low temperatures below T_c . This is fully consistent with the previous study on RCo_2 compounds ($\text{R} = \text{rare earth}$) that indicated that the magnetic response in the ordered state was dominated by the rare earth and almost temperature independent below T_c .³ At $T_c < T < 55 \text{ K}$ ($\sim T_f$), a strong decrease in H_S has been observed; however, finite values of H_S are still detected, suggesting the existence of magnetic clusters in the parimagnetic region. For $T > T_f$, the disappearance of H_S is expected as the ErCo_2 system is now in the paramagnetic regime.

Furthermore, we note that there is a distinguishable difference in the shape of the TS curves taken at $T = 20, 50,$ and 90 K (Fig. 2). Such a difference is expected as ErCo_2 exhibits three different magnetic configurations in the ferrimagnetic, parimagnetic, and paramagnetic regimes, respectively. A strong variation in the shape of the TS curves upon varying saturation fields ($\mu_0 H^{\text{sat}} = 1 \text{ T}$ versus $\mu_0 H^{\text{sat}} = 5 \text{ T}$) is also noted. In addition, the magnitude of the $\Delta\chi_T/\chi_T\%$ ratio at $\mu_0 H_{\text{dc}} = 0 \text{ T}$ (which we defined as η) has been found to remarkably vary with temperature in the ferrimagnetic, parimagnetic, and paramagnetic regimes.

To better illustrate these intriguing features, Fig. 3(b) displays the temperature dependences of η and FWHM (the full width at half maximum of a TS curve) for the case of $\mu_0 H^{\text{sat}} = 1 \text{ T}$. The temperature dependence of the correlation length (ξ) as determined from SANS experiments is also included in Fig. 2(c) for discussion.³ It can be observed in Fig. 3(b) that η remains almost unchanged in the ferrimagnetic region ($T < T_c$), increases sharply at T_c , and then

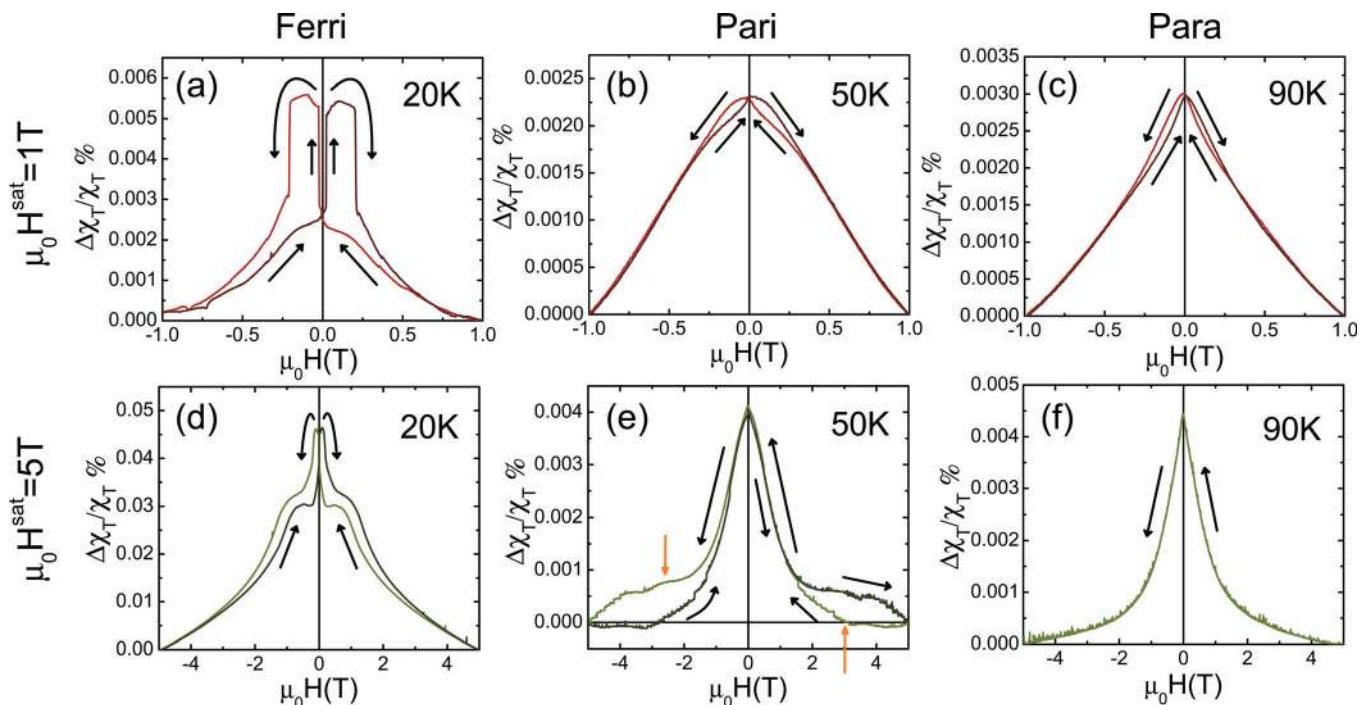


FIG. 2. (Color online) Plot of relevant TS bipolar scans of ErCo_2 . (a), (b), and (c) measured for $\mu_0 H^{\text{sat}} = 1$ T at 20, 50, and 90 K, respectively. (d), (e), and (f) measured at the same temperatures for $\mu_0 H^{\text{sat}} = 5$ T. The black arrows indicate the direction of the field sweeping.

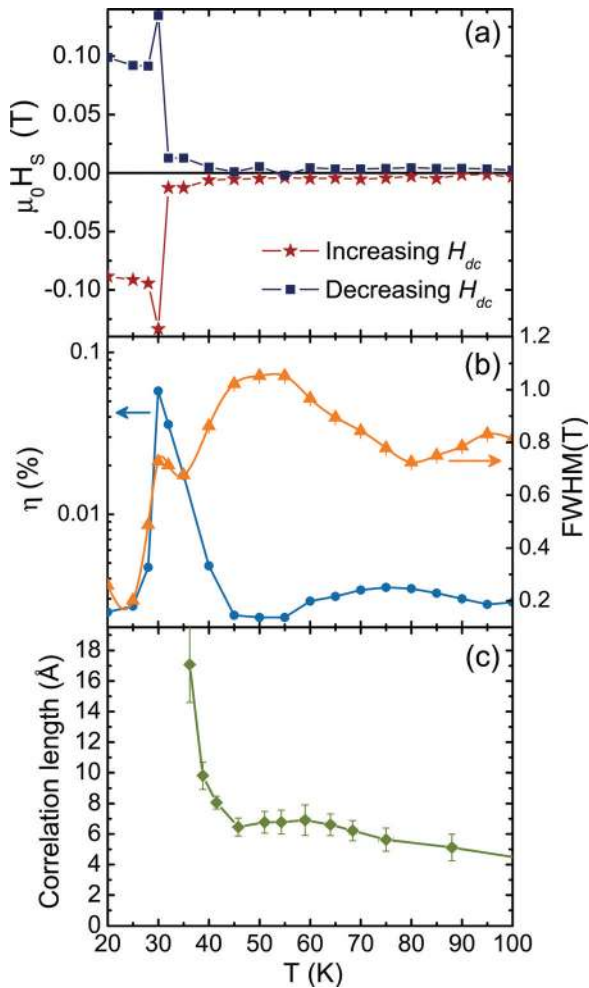


FIG. 3. (Color online) (a) Switching field H_S (■ and *) identified from TS measurements with $\mu_0 H^{\text{sat}} = 1$ T in ErCo_2 . (b) Temperature dependence of η (●) and FWHM (▲) of the TS profiles for $\mu_0 H^{\text{sat}} = 1$ T. (c) Correlation length ξ obtained from SANS at (◆).³

decreases but not to zero in the paramagnetic region ($T_c < T < T_f$). This trend is similar to the temperature dependence of H_S [Fig. 3(a)], which once again indicates that the magnetic response of ErCo_2 in the ferrimagnetic region is almost temperature independent, and that the magnetic clusters are present in the paramagnetic region. We note that the strong decrease of η just above T_c can be associated with the drop in ξ . For $T > T_f$, η increases again with increasing temperature, reaches a maximum at 75 K, and finally decreases at higher temperatures. This can be reconciled with the fact that the net Co moment changes in sign from “negative” to “positive” at $T_f \sim 55$ K and gradually increases with temperature until $T \sim 75$ K, as we have found in previous studies on ErCo_2 .^{2,3} The temperature dependence of η can be correlated with that of FWHM. In the ferrimagnetic range the FWHM sharply increases and peaks at $T_c \sim 32$ K. However, in the paramagnetic regime the increase of FWHM corresponds to the decrease of η . The maximum of FWHM

and the minimum of η at 55 K clearly set a transition from the paramagnetic state to the paramagnetic one. In the paramagnetic region the inverse relationship is also found, with the minimum of FWHM corresponding to the maximum of η at 75 K. In this temperature range the correlation length gradually decreases with increasing temperature.

A final note is the change in the sign of TS from “negative” to “positive” at 50 K [Fig. 2(e)] as the magnetic field is swept from $\mu_0 H^{\text{sat}} = 5$ T to 0 (or from $\mu_0 H^{\text{sat}} = -5$ T to 0) after the sample is saturated to $\mu_0 H^{\text{sat}} = 5$ T. We recall from the magnetic phase diagram (Fig. 1) that at 50 K and for $\mu_0 H^{\text{sat}} > 3$ T the ErCo_2 sample exhibits a paramagnetic characteristic. In this case, the net Co moment tends to align along the direction of the Er moment and the applied magnetic field. However, for $\mu_0 H^{\text{sat}} < 3$ T the ErCo_2 sample lies in the paramagnetic state, where the net Co moment tends to align antiparallel to the direction of the Er moment and the applied magnetic field. Therefore, as the magnetic field is swept from $\mu_0 H^{\text{sat}} = 5$ T to 0 the change in the sign of the TS profile from negative to positive at $\mu_0 H^{\text{sat}} \sim 3$ T is expected.

In summary, our study based on TS measurements on ErCo_2 is coherent with all the previous results from magnetic and transport studies^{3,12} confirming the existence of short-range correlations at $T_c < T < T_f$ and the power of the technique for studying magnetism at the nanoscale. A remarkable evidence of these correlations was given by probing the existence of a finite H_S in the paramagnetic region in ErCo_2 .

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