Sequential Cobalt Magnetization Collapse in ErCo2: Beyond the Limits of Itinerant Electron Metamagnetism

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The itinerant electron metamagnetism (IEM) is an essential physical concept, describing magnetic properties of rare earth – transition metal (R-TM) intermetallics, demonstrating technologically important giant magnetoresistance and magnetocaloric effects. It considers an appearance of TM magnetization induced by spontaneous magnetization of surrounding R atoms, which provides significant response of the magnetic and transport properties on variation of external parameters (temperature, pressure, magnetic field) due to strong coupling between magnetic sublattices. The RCo2 compounds were generally considered as model systems for understanding of basic properties of IEM intermetallics. However, microscopic nature of magnetic properties still remains unclear. In our experimental and theoretical study of ErCo2 in a wide range of thermodynamic parameters a sequential collapse of cobalt sublattice magnetization in the background of nearly unchanged Er sublattice magnetization was revealed. The uncoupled magnetizations behavior challenges the IEM concept applicability and evidences more complex nature of magnetism in ErCo2 and related RCo2 systems.
The good candidate to explore the microscopic nature of Co magnetization formation in RCO_2 compounds and check the limits of the applicability of IEM concept is ErCo_2, where the exchange interactions are moderate, as evidenced by the Curie temperature, \( T_c \approx 35 \text{ K} \). Several pressure studies were previously performed on ErCo_2 and Er_{1-x}Y_xCo_2 compounds using different experimental techniques. A decrease of Curie temperature with a rate \( dT_c/dP = -8 \text{ K/GPa} \) has been established in resistivity measurements on ErCo_2 up to \( P = 2 \text{ GPa} \) and it was attributed to pressure induced destabilization of the itinerant d system. In subsequent resistivity studies over extended pressure range, it was found that \( T_c \) stops to diminish and remains nearly constant at pressures above 4 GPa. The X-ray adsorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) studies evidenced a progressive reduction of Co magnetic moment as the applied pressure increases, but it is not canceled for \( P \leq 4.2 \text{ GPa} \). A remarkable reduction of the 3d spin moments upon compression was observed in ErCo_2 in the magnetic Compton scattering experiments at pressures up to 1.84 GPa. An asymmetry in \( T_c \) – related ac susceptibility peak has been also observed under pressure and it was attributed to decoupling of the magnetic ordering of the Er and Co sublattices.

In the studies of pseudobinary Er_{1-x}Y_xCo_2 compounds Hauser et al. evidenced that the metamagnetic behavior of itinerant d sub-system vanishes for \( x > 0.4 \). Two separate magnetic ordering temperatures were detected by means of specific heat, thermal expansion and electrical resistivity measurements. In contrast, using XCMD technique Chaboy et al. reported that both Er and Co magnetic sublattices order at the same temperature for \( x \leq 0.6 \). In particular, no decoupling of the magnetic ordering for both Er and Co sublattices has been shown for the compound with \( x = 0.4 \). The neutron diffraction study on \( \text{Er}_{0.57}\text{Y}_{0.43}\text{Co}_2 \) compound, at \( P \leq 0.6 \text{ GPa} \), evidenced the same Curie temperatures for both Er and Co sublattices, which decrease with a rate \( dT_c/dP = -4 \text{ K/GPa} \).

The above studies reported rather contradictory results. Also the collapse of the cobalt moment, as already mentioned, was suggested by indirect techniques as XAS, XMCD, magnetic Compton scattering, magnetic susceptibility or resistivity studies. In order to obtain more detailed and reliable information on magnetic properties of ErCo_2 at microscopic level, we have performed neutron diffraction study of crystal and magnetic structure of ErCo_2 compound in a wide range of thermodynamic parameters, high pressure 0–4 GPa and temperature 10–290 K, supported by extensive ab-initio theoretical calculations. While application of external high pressure allows to tune internal magnetic field value by reduction of interatomic distances, neutron diffraction measurements provide a possibility of simultaneous determination of both crystal and magnetic structure features in detail. We found that magnetizations of R and Co sublattices become uncoupled under pressure and the ordered Co magnetic moments collapsing in the background of nearly unchanged ordered Er magnetic moments.

**Results**

**Experimental results.** The magnetization isotherms, measured for ErCo_2 compound, are shown in fig. 1. They demonstrate a presence of the first order magnetic transition at \( T_c \approx 35 \text{ K} \), as previously reported. The neutron diffraction patterns of ErCo_2, measured below the Curie temperature (fig. 2), evidenced the rhombohedrally distorted crystal structure of \( R3m \) symmetry. There are two inequivalent positions for Co atoms, 9e and 3b, and one position, 6c for Er atoms in this structure, illustrated in fig. 3 (inset). The obtained structural parameters and ordered magnetic moments of Er and Co atoms at selected pressures and temperatures are presented in Table 1. The \( a \) and \( c \) lattice parameters decrease nearly linearly under pressure with compressibility coefficients \( k_a = 0.0051(9) \) and \( k_c = 0.0076(2) \) GPa\(^{-1} \) at 10 K. In the analysis of the magnetic contributions to neutron diffraction patterns at high pressures, we considered equal values of Co ordered magnetic moments located at positions 9e and 3b.

![Figure 1](https://www.nature.com/scientificreports)

**Figure 1** | Magnetization isotherms for ErCo_2 measured in external magnetic fields up to 12 T for selected temperatures in 4–250 K range.

![Figure 2](https://www.nature.com/scientificreports)

**Figure 2** | (a) Neutron diffraction patterns of ErCo_2 compound, measured at selected pressures and temperatures and processed by the Rietveld method. Experimental points and calculated profiles are shown. Ticks below represent calculated positions of the structural peaks. The characteristic peaks with the most intense magnetic contribution are marked as “FM”. (b) The parts of neutron diffraction patterns measured at \( P = 2.1 \text{ GPa} \) and \( T = 10 \text{ K} \), and calculated profiles for fixed Er magnetic moment \( M_{\text{Er}} = 9.0 \mu_B \) and two values of Co magnetic moments \( M_{\text{Co}} = -0.7 \mu_B \) and 0.0 \mu_B. The noticeable changes in difference curves demonstrate possibility of accurate determination of Co moments using neutron diffraction technique.
contribution to intensities of (1 0 1)/(2 1 0) and (1 1 3)/(2 1 3) peaks located at d-spacings \( d_{hkl} \) = 4.13, 2.53 and 2.16 Å, respectively, the magnetization of Co sublattice contributes mostly to the (1 0 1)/(-1 1 1) peak only. As a result, the changes in magnetization of Co sublattice can be determined precisely with respect to magnetization of Er sublattice, as illustrated by model calculations in fig. 2b.

The experimentally determined temperature dependences of the erbium and cobalt magnetic moments at different pressures are given in fig. 3 and their ferrimagnetic arrangement is illustrated in fig.3 (inset). The obtained magnetization per formula unit, \( M_{\text{tot}} \approx 7.7 \ \mu_B \), is close to one observed in magnetization data at low temperatures (fig. 1). The Curie temperature of erbium sublattice magnetization is little influenced by pressure, \( dT_{\text{Cur}}(\text{Er})/dP \approx 0.3 \ \text{K GPa}^{-1} \) and very close to that of the whole ErCo₂ compound at ambient pressure, \( T_c \approx 36 \ \text{K} \) (fig. 3, inset). The Er ordered magnetic moments at \( T = 10 \ \text{K} \) also remain nearly unchanged under pressure. In contrast, a collapse of the cobalt sublattice magnetizations at temperatures \( T_{\text{Cur}}(\text{Co}) \), which decrease with a rate \( dT_{\text{Cur}}(\text{Co})/dP \approx -(3.45 \pm 0.3) \ \text{K GPa}^{-1} \), was evidenced (fig. 3). The ordered cobalt magnetic moments determined at 10 K as well as the extrapolated values at \( T = 0 \) decrease with a pressure rate \( dM_{\text{Co}}/dP \approx -0.1 \ \mu_B \text{GPa}^{-1} \), similar to that in relevant RCo₂ compounds (R = Tb, Ho)\(^{12,15}\). The relation \( dT_{\text{Cur}}(\text{Co})/dM_{\text{Co}} = 34.5 \pm 3 \ \text{K GPa}^{-1} \), resulting from the data above, describes well the experimentally determined \( T_{\text{Cur}}(\text{Co}) \) values – Table 2. These findings provide clear evidence of the magnetization collapse of Co sublattice under pressure at temperatures \( T_{\text{Cur}}(\text{Co}) < T_c(\text{Er}) \) and therefore, uncoupled behavior of Co and Er magnetic sublattices under pressure. The obtained results also imply that previously reported pressure dependences of the Curie temperatures\(^{12,16,17,23} \) for ErCo₂ should be related to the collapse of cobalt sublattice magnetization only, characterized by \( T_{\text{Cur}}(\text{Co}) \) and not to the total magnetization of the whole compound as reported.

The gradual collapse of cobalt magnetization, at temperatures \( T_{\text{Cur}}(\text{Co}) \) can be analysed in correlation with temperature dependence of the exchange field, \( H_{\text{ex}} \) acting on cobalt atoms \( (H_{\text{ex}} = 2J_{\text{CoCo}}M_{\text{Co}} + J_{\text{CoEr}}M_{\text{Er}}) \). Starting from a two sublattices molecular field model\(^{1,10} \), the \( J_{ij} \) (i,j = Er, Co) parameters describing the exchange interactions inside and between magnetic sublattices were evaluated using a fitting program\(^{20} \) of the magnetizations as determined by neutron diffraction. The values \( J_{\text{CoCo}} \approx 95 \) and \( J_{\text{CoEr}} \approx -5 \) were obtained. In case of heavy rare-earth RCo₂ compounds, the \( J_{\text{CoCo}} \) values are linearly dependent on De Gennes factor, similar as their R5d band polarizations, as expected for a 4f-5d-3d exchange interaction model\(^{20} \). The thermal variations of exchange fields were computed assuming a Brillouin type dependence of cobalt magnetization and using the experimentally determined \( M_{\text{Co}} \) values as illustrated in Fig. 3. At temperatures \( T_{\text{Cur}}(\text{Co}) \), where the cobalt magnetization collapses, the exchange field was \( H_{\text{ex}} = 74 \pm 3 \ \text{K} \). This value corresponds to the critical field necessary for supporting the cobalt ordered moment and it is nearly the same as that involved in inducing a cobalt ordered moment in LuCo₂ by external field\(^{b} \), or by internal field in pseudobinary RCo₂ compounds\(^{20,24} \). Conversely, assuming that the collapse of cobalt moment takes place at \( H_{\text{ex}} = 74 \ \text{T} \), then \( T_{\text{Cur}}(\text{Co}) \) values close to those experimentally determined were obtained. – Table 2.

**Theoretical calculations.** The pressure evolution of cobalt moments at \( T = 0 \ \text{K} \), was theoretically analysed in correlation with modification of the band structures – fig. 4. The computed cobalt moments at 3b sites are by \( \approx 0.05 \ \mu_B \) higher than those at 9e

| Table 1 | Structural parameters and ordered magnetic moments of Er and Co atoms in ErCo₂ at selected pressures and temperatures. In the ambient temperature cubic \( \text{Fd}3\text{m} \) structure Er atoms locate at sites \( 8(a) \) (0.125, 0.125, 0.125) and Co atoms locate at sites \( 16(d) \) (0.5, 0.5, 0.0) and 9(e) (0.5, 0.0, 0.0). In the low temperature rhombohedral \( \text{R}3\text{m} \) structure Er atoms locate at sites \( 6(c) \) (0, 0, 0) and Co atoms locate at sites \( 3(b) \) (0, 0, 0.5) and 9(e) (0.5, 0.0, 0.0). The reliability \( R_p \) and \( R_{\text{wp}} \) factors values are also given.

| \( P \) (GPa) | \( T \) (K) | \( 0 \) | \( 2.1 \) | \( 4.1 \) |
|---|---|---|---|---|
| \( T \) (K) | 290 | 10 | 290 | 10 | 290 | 10 |
| \( \alpha \) (Å) | 7.1115(6) | 5.0495(4) | 7.0717(3) | 5.0419(7) | 7.0492(7) | 5.0337(7) |
| \( c \) (Å) | - | 12.307(7) | - | 12.124(1) | - | 12.027(8) |
| \( z \) | - | 0.1238(5) | - | 0.1241(1) | - | 0.1221(1) |
| \( B_{\text{Mo}} \) (Å²) | 0.42(6) | 0.20(6) | 0.5(6) | 0.17(8) | 0.29(6) | 0.16(8) |
| \( M_{\text{Co}} \) (μB) | 3 | 9.60(8) | 9.0(1) | 8.8(1) | - | - |
| \( C_{\text{Mo}} \) (Å²) | 0.43(7) | 0.30(7) | 0.41(7) | 0.28(8) | 0.38(6) | 0.26(8) |
| \( M_{\text{Er}} \) (μB) | - | -0.95(5) | - | -0.70(7) | - | -0.50(7) |
| \( R_p \) | 4.72 | 4.93 | 5.38 | 5.45 | 5.73 | 5.93 |
| \( R_{\text{wp}} \) | 5.21 | 5.42 | 6.03 | 6.18 | 6.28 | 6.42 |
Table 2 | The Curie temperatures of cobalt sublattice magnetization

| Method                                      | $T_c$(Co) [K] |
|---------------------------------------------|---------------|
| Experimental                               | $P = 0$ GPa   | $P = 1.1$ GPa | $P = 2.1$ GPa | $P = 4.1$ GPa |
| $T_c$(Co) = 34.5 MCo                       | 34.9 ± 0.4    | 28.6 ± 0.3    | 24.8 ± 0.4    | 21.1 ± 0.3    |
| From critical value of exchange field, $H_{ex}$ = 74 T | 32 ± 2        | 30.5 ± 0.7    | 25 ± 0.8      | 22.0 ± 0.5    |

Figure 4 | Total and partial densities of states of ErCo$_2$ at pressures $P = 0$ GPa ($v/v_0 = 1$) (a), 4.1 GPa ($v/v_0 = 0.97$) (b) and $v/v_0 = 0.90$ (c) (right part).

In the left part the energy bands of ErCo$_2$ decorated with orthogonal orbital character are shown. The location of the centers of the corresponding “fat” bands are only indicated. The contribution of Co3d and Er5d ($d_{x^2-y^2}$, $d_{z^2}$, $d_{xy}$, $d_{yz}$, $d_{zx}$), $p$ ($p_x$, $p_y$, $p_z$) and $s$ orbitals to the “fat” bands were analysed as well as their evolution with pressure, relative volume, respectively on the energy scale. For some directions of the reciprocal lattice peculiar orbitals have dominant contributions, as mentioned in text.
Discussion

The origin of Co magnetization collapse in ErCo$_2$ is different from relevant intermetallic YCo$_5$, where Co electronic configuration changes from high-to low spin state and accompanied by an isomorphic lattice collapse under pressure.\textsuperscript{6,2} No anomalies in lattice parameters were detected in ErCo$_2$ at temperatures T$_c$(Co). This suggests that magnetovolume coupling is controlled mainly by erbium sublattice. The collapse of cobalt moments at T = 0 K, as effect of pressure, is well correlated with evolution of exchange splitting, $\Delta E_{\text{ex}}$ of their 3d band. There is a linear dependence of $M_{\text{Co}}$ on $\Delta E_{\text{ex}}$, in agreement with previous work.\textsuperscript{8,10} No changes in populations of the two Co3d subbands can be shown as effect of pressure.

In addition to itinerant metamagnetism model,\textsuperscript{2,5} a model of induced magnetism has been also proposed, correlated with exchange field $H_{\text{ex}}$ of external or internal nature, acting on cobalt atoms.\textsuperscript{15,11} In the framework of this model, the cobalt magnetic moment is weakly dependent on the exchange field below critical value $H_{\text{ex}}$ and it jumps to significantly higher value on approaching $H_{\text{ex}}$. Upon further increase of the exchange field in the $H_{\text{ex}} > H_{\text{ex}}$ region a linear variation of cobalt moment followed by its saturation is expected. The cobalt moment has been correlated with the variation of the exchange splitting of their 3d band. Analysing the band structures of ErCo$_5$ at different pressures, relative volume, respectively, there is a shift of spin up and spin-down sub-bands, respectively in opposite directions. The exchange splitting is thus diminished simultaneously with the decrease of cobalt moment. As showed previously\textsuperscript{8,10,11}, there is a linear dependence of cobalt moment with the exchange splitting. No change in the population of the Co sub-bands is evidenced by band structure calculations, as required in the collective electron metamagnetism model. In addition, the cobalt moment is stable only in the presence of high external or exchange field. In all studies, particularly by neutron diffraction, a degree of localization of cobalt moment can be shown. Even in case of exchange enhanced LuCo$_{5}$ paramagnet, at 100 K, in field of 5.72 T the density on Co atoms has a form factor which is similar to that of 3d electrons in Co metal.\textsuperscript{12} Localized features for cobalt magnetism were reported also by NMR or other experiments.\textsuperscript{13} These data show a more complex magnetic behavior than suggested by the model of collective electron metamagnetism. A better correlation with their 3d band exchange splitting, as function of the pressure is evidenced for ErCo$_2$, as expected in the induced magnetism model.

The example of ErCo$_2$ demonstrates the limits for the applicability of IEM concept for the case of RCo$_5$ and relevant R-TM intermetalics with relatively low Curie temperatures and strength of R-Co interactions. In this case, decoupling of R and TM magnetization can be observed upon variation of thermodynamic parameters. This phenomenon is mediated by values of inter-sublattice exchange interaction $J_{\text{R-TM}}$ and R 5d band polarization $M_{\text{sd}(0)}$, induced by 4f-5d local exchange and governing by DeGennes factor.\textsuperscript{12} A reduction of the cobalt magnetic moment correlating with the behavior of the 3d band exchange splitting under pressure in ErCo$_2$ is consistent with the induced magnetism model.

Methods

Experiment. The ErCo$_2$ compound has been prepared by melting the high purity elements in an induction furnace, under high purity argon atmosphere. The sample was annealed at T = 880 °C, in vacuum for one week. The electron microscopy and XRD studies, at ambient temperature, evidenced the presence of only one phase. Magnetic measurements were performed in field up to 12 T, in a large temperature range. The errors in determining the magnetizations are around 0.06 μB.

Neutron powder diffraction measurements, at θ ≤ 4.1 GPa and T ≥ 10 K, were made with DN-12 spectrometer\textsuperscript{14} at IBR-2 high-flux pulsed reactor (FNLP Dubna, Russia) using the sapphire anvil pressure cell.\textsuperscript{15} The pressure was determined by the ruby fluorescence technique with accuracy of 0.05 GPa, at each ruby chip and the pressure on the sample was obtained by averaging the values determined at different points. The data were analysed by Rietveld method using MRIA\textsuperscript{16} and Fullprof\textsuperscript{17} programs.

Theory. The ground state electronic structures and magnetic properties of ErCo$_2$ compound at various pressures, volume variations, respectively have been performed using scalar relativistic tight-binding linear muffin-tin orbital method (TB-LMTO) in atomic sphere approximation.\textsuperscript{18} The local density approximation (LSDA) has been used for the exchange and correlation energy within von Barth and Hedin parameterization.\textsuperscript{19} The valence basis consists of s-, p- and d- orbitals, whereas the Er 5s orbitals were considered as core states. The overlaps of the muffin-tin spheres

![Figure 5](image-url) | The dependence of cobalt moments at T = 0 K on relative volume change. The computed values for Co(9e) and Co(3b) moments and the experimental data extrapolated to T = 0 are given. The errors resulting from the refinements of experimental neutron diffraction patterns are ±0.10 μB.
was below 9% and the standard combined corrections (CC) terms were introduced to compensate the errors due to ASA. The band structures were computed starting from lattice parameters determined at 10 K and pressures of 0, 1.1, 2.1 and 4.1 GPa. The corresponding normalized volumes were v/v0 = 1.0, 0.989, 0.982 and 0.97, respectively. A sample with v/v0 = 0.90 has been also analysed. The minimum total energy has been obtained for a and c lattice parameters which differ from experimentally determined values by 0.6%.

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**Author contributions**

D.P.K. and E.B. prepared the main manuscript and display items. P.V. performed ab-initio calculations. E.B. performed magnetization measurements. S.E.K. and A.V.R. performed ND experiments. A.V.R. and B.N.S. made a preliminary ND data treatment. All authors reviewed the manuscript.

**Additional information**

Competing financial interests: The authors declare no competing financial interests.

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