Optical diode effect at telecom wavelengths in a polar magnet

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Abstract

Magnetoelectric multiferroics such as rare earth manganites host nonreciprocal behavior driven by low symmetry, spin-orbit coupling, and toroidal moments, although less has been done to explore whether lanthanides like Er^{3+} might extend functionality into the hard infrared for optical communications purposes. In this work, we reveal nonreciprocity in the f-manifold crystal field excitations of h-$Lu_{0.9}Er_{0.1}MnO_3$. In addition to contrast in the highest fields, we demonstrate nonreciprocity at technologically-relevant energy scales - specifically in the E-, S-, and C-bands of the telecom wavelength range - and at low magnetic fields and room temperature. In fact, the low field behavior is consistent with possible altermagnetism. These findings advance the overall understanding of localized excitations in rare earth-containing systems and pave the way for entirely new types of telecom applications.

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INTRODUCTION

Nonreciprocal directional dichroism is a type of asymmetric light absorption that depends upon propagation direction.[1,2] It is a defining feature of materials that simultaneously break spatial inversion and time-reversal symmetries.[3,4] Although discovered in magnetoelectrics including CuB_2O_4,[5–8] $FeZnMo_3O_8$,[9] $LiNiPO_4$,[10] $LiCoPO_4$,[11] $Nd_2Ti_2O_7$,[12] $Pb(TiO)Cu_4(PO_4)_4$,[13] and Ni_3TeO_6,[14–16] nonrecprocity is under-explored in rare earth-containing systems. This is because f-manifold crystal field excitations (which are both spin- and parity-forbidden) have been presumed not to host large nonreciprocal effects even though mixing of electric and magnetic dipoles at noncentrosymmetric rare earth sites can generate significant magnetoelectric coupling.[1,17] Properties arising from Er^{3+} are especially relevant to amplifiers, isolators, modulators, and rectifiers at optical communications wavelengths.[18–27] The prospect of integrating additional functionality in the form of nonreciprocity to the rare earth excitations that power these telecommunications technologies is therefore both challenging and potentially transformative. Rather than testing drawn quartz fibers sprinkled with powdered Er,[22,23] we employed h-$Lu_{0.9}Er_{0.1}MnO_3$ as a platform for examining whether rare earth f-manifold excitations have the potential to host nonreciprocal behavior in the hard infrared. This system sports a dillute ensemble of Er^{3+} ions within a $P6_3mc$ matrix that combines spontaneous polarization along c (arising from improper ferroelectricity involving the Mn centers) with antiferromagnetism due to Mn^{3+} ordering ($T_N \approx 80$ K) and a rare earth related transition near 30 K [Fig. 1**a**].[28,29] In fact, h-$Lu_{0.9}Er_{0.1}MnO_3$ is likely an altermagnet due to antisymmetric spin splitting in the $6'mm'$ magnetic ground state.[30] Our work is enabled by the development of monopolar domain single crystals which grow in a characteristic canopy-like shape [Fig. 1**b**]. Such a material - if functioning as a secure communications element - should host higher fidelity and lower loss than glass fibers with Er randomly distributed throughout.

The symmetry requirements for toroidal nonreciprocity in h-$Lu_{0.9}Er_{0.1}MnO_3$ dictate that polarization, magnetic field, and light propagation direction must be mutually orthogonal.[1,4,16] We therefore performed magneto-optical spectroscopy in this fashion. Strikingly, these measurements reveal strong nonreciprocal behavior in the f-manifold crystal field excitations of Er^{3+} that persists not only at high fields but also at modest magnetic fields and

even up to room temperature. At 1525 nm, we find a contrast of 3.2% at 1.2 T and 296 K. These findings challenge the conventional wisdom about localized excitations, opening the door to entirely new types of nonreciprocal behavior and applications.

RESULTS AND DISCUSSION

Er^{3+} crystal field excitations in the hard infrared

Figure 1**c,d** displays the near infrared absorption of h-$Lu_{0.9}Er_{0.1}MnO_3$ as a function of temperature. This particular wavelength range focuses on the Er^{3+} f-manifold crystal field excitations in the E-, S-, and C-bands of the telecom range. These excitations are well-known to be sharp and highly localized. The clusters of peaks between 1440 and 1540 nm can be assigned as ${}^4I_{15/2} \rightarrow {}^4I_{13/2}$. This set of excitations is well-studied in Er-containing oxides, chalcogenides, and silica glasses[20–25,31] because the excitations are responsible for the substantial gain near 1550 nm in Er-doped fiber amplifiers for long-range optical communication. h-$Lu_{0.9}Er_{0.1}MnO_3$ contains two unique Er^{3+} centers, each with seven primary ${}^4I_{15/2} \rightarrow {}^4I_{13/2}$ excitations at base temperature, with additional features arising from temperature-induced population effects. The two Er^{3+} sites and the combination of both ab-plane and c-oriented excitations add to the complexity of the 4 K data. The impact of population effects along with the Er^{3+}-related and Mn^{3+} magnetic ordering transition is evident in the contour plot as well.[30,32]

Nonreciprocity in f-manifold crystal field excitations

In order to explore how low symmetry and spin-orbit coupling impact the properties of f-manifold crystal field excitations, we measured the magneto-optical response of h-$Lu_{0.9}Er_{0.1}MnO_3$ at base and room temperatures and calculated the nonreciprocity which is defined as $\Delta\alpha_{\mathrm{NDD}} = \alpha(+H,+k)$ - $\alpha(-H,+k)$. Figure 2 summarizes these results in the form of contour plots. As anticipated, the excitations shift linearly in magnetic field with different slopes given by the respective g factors. We find features that cross and merge as well as excitations that host avoided crossings. The latter defines critical fields at 6 and 30 T, in line with the magnetization of h-$ErMnO_3$[30,32] as well as our own measurements of h-

$Lu_{0.9}Er_{0.1}MnO_3$ [Fig. S6, Supplementary Information]. The key point is that nonreciprocity is observed at both low fields and high temperatures and is sensitive to the development of different field-induced magnetic states.

Figure 3 summarizes the dichroic spectra of h-$Lu_{0.9}Er_{0.1}MnO_3$. These data were taken in the toroidal configuration with $\pm H$ which is symmetrically equivalent to $\pm k$ [Fig. S2, Supplementary Information].[33] As a reminder, toroidal dichroism occurs when light propagation is along the toroidal moment T, ($\vec{k} \parallel T = P \times M$, where P and M are the electric polarization and magnetic moment).[3,34–38] We polished and mounted our crystal consistent with this configuration [Fig. S1, Supplementary Information].

Figure 3**a** displays the nonreciprocal directional dichroism of h-$Lu_{0.9}Er_{0.1}MnO_3$ at 5.5 K, well below the Er^{3+}-related transition and magnetic ordering temperature of Mn^{3+}. The dichroic response is unexpectedly large (18.8% at 1516 nm and 55 T) for such a highly localized set of excitations. These results unequivocally confirm that f-manifold crystal field excitations in a magnetoelectric material can host nonreciprocity. The effect is sharp and clear, with a very systematic response as a function of applied magnetic field. Even more remarkably, we find that this system does not need the highest fields to reveal functionality. $\Delta\alpha_{\mathrm{NDD}}$ is approximately 10.4% at 1525 nm and 1.2 T. That such a well-defined nonreciprocal effect can be observed in the telecom wavelength range allows us to conceive of a number of unique opportunities.

To test the impact of magnetic ordering on the dichroic effect, we performed similar experiments at elevated temperatures. Figure 3**b,c** displays the nonreciprocal directional dichroism of h-$Lu_{0.9}Er_{0.1}MnO_3$ at 31 and 186 K, respectively. There is no rare earth ordering at 31 K,[30,32] although the Mn^{3+} centers are still magnetically ordered ($T_{\mathrm{N}} \approx 80$ K). Even so, nonreciprocity is surprisingly strong. Increasing temperature to 186 K eliminates magnetic ordering involving the Mn^{3+} sites as well. The consequence is a substantial decrease in the size of the dichroic signal. The overall shape changes due to population effects as well. Clearly, Er$\cdots$Mn interactions are important - but not essential - for this process.

Nonreciprocity at room temperature

Inspired by the possibility that a well-ordered magnetic state is not required for the development of nonreciprocity in f-manifold crystal field excitations, we measured the magneto-optical properties of h-$Lu_{0.9}Er_{0.1}MnO_3$ at room temperature. As a reminder, nonreciprocal directional dichroism requires magnetoelectric coupling. This is because the magnetoelectric susceptibility $\chi^{me}_{\alpha\beta}$ is proportional to $\langle 0|P_\alpha|n\rangle\langle n|M_\beta|0\rangle$, so both the polarization and magnetization matrix elements must be non-zero.[1]

Figure 3**d** displays the dichroic response of h-$Lu_{0.9}Er_{0.1}MnO_3$ at room temperature. Strikingly, the effect is not quenched due to a lack of long-range magnetic order in the Mn framework. This is because the toroidal configuration does not require magnetic order since $\vec{k} \parallel T = P \times M$.[3,34,36–39] Examination reveals that $\Delta\alpha_{\mathrm{NDD}}$ is still as high as 13.7% at 1516 nm and full field, although the exact magnitude varies depending upon the excitation, and the overall size of the spectral features are much more field sensitive than before. Not only is the signal larger in higher fields, but the highest fields are needed to obtain well-formed peak shapes. This is likely because larger fields are required to overcome thermal fluctuations, align Er moments, and break time-reversal symmetry in the paramagnetic phase above the ordering temperature. Even at 296 K, h-$Lu_{0.9}Er_{0.1}MnO_3$ continues to host nonreciprocal directional dichroism at very modest fields. $\Delta\alpha_{\mathrm{NDD}}$ at 1.2 T, for instance, is still distinct [inset, Fig. 3**d**]. These findings demonstrate that it is indeed possible to realize room temperature nonreciprocity under low field conditions in the telecom range.

Breaking time reversal symmetry in this system

Thus far, we have seen that at low temperatures, where Mn is antiferromagnetically ordered, Er$\cdots$Mn interactions break time-reversal symmetry to enable nonreciprocity in the f-manifold excitations. We speculate that, in the presence of magnetic order, the Mn-induced local exchange fields on the Er moments cooperate with the external field for an enhanced dichroic response. This provides a way to understand the dramatic spectral changes across the Mn ordering temperature. Below the ordering temperature, h-$Lu_{0.9}Er_{0.1}MnO_3$ likely exhibits B_2-type symmetry, corresponding to a $6'mm'$ magnetic point group, suggesting the potential for symmetric and antisymmetric spin splitting (S/A-type altermagnetism).[40] This

may provide a possible explanation for the substantial enhancement of the nonreciprocal signal below T_N [Fig. S4, Supplementary Information]. At higher temperatures, where Mn is no longer ordered, the applied field acts on the Er moments directly to break time reversal symmetry, the result generating a smaller, yet still appreciable nonreciprocal signature - even at room temperature. These effects are well within the limits of our sensitivity. Due to the low energy scales for f-orbital excitations, small external perturbations lead to a remarkably large nonreciprocal response, 3.2% at 1525 nm and 1.2 T, even at room temperature.

To summarize, we report the discovery of nonreciprocity in the rare earth crystal field excitations of an Er^{3+}-containing oxide across the telecom wavelength range. The effect is surprisingly strong - a finding that we attribute to the ability of the applied field to rotate Er^{3+} moments. At the same time, there is a polar environment around the noncentrosymmetric rare earth ions leading to the formation of net toroidal moments even in the presence of very small magnetic fields. While there is some sensitivity to magnetic ordering likely due to altermagnetic character below T_N, the toroidal configuration generally supports low loss nonreciprocity in monopolar crystals of h-$Lu_{0.9}Er_{0.1}MnO_3$ at readily accessible fields and temperatures - including room temperature. These findings open the door to the development of structure-property relations as well as low power devices in this unique application space.

METHODS

Single crystal growth: h-$Lu_{0.9}Er_{0.1}MnO_3$ powder was synthesized by sintering a stoichiometric mixture of Lu_2O_3, Er_2O_3, and Mn_2O_3 powders at 1350 °C for 30 hours, with two intermediate grindings. For single crystal growth, the synthesized powder was mixed with Bi_2O_3 flux in a molar ratio of 1:10. The mixture was placed in a platinum crucible, heated to 1100 °C for 10 hours, then cooled at a rate of 2 °C per hour to 800 °C, followed by rapid cooling at 100 °C per hour to room temperature. The residual flux was removed using dilute hydrochloric acid to isolate h-$Lu_{0.9}Er_{0.1}MnO_3$ crystals. Compared to previously reported flux growth methods that yielded multi-ferroelectric-domain crystals, the approach used here incorporates a higher ratio of Bi_2O_3 flux. As a result, crystallization occurred at the surface rather than at the bottom of the crucible. This surface crystallization effectively poled the

crystals into a single ferroelectric domain, as confirmed by optical microscope images of the chemically etched surfaces. The crystals were polished to reveal the c-axis (since the polar c-axis is normal to the large natural growth face) and to obtain a proper optical density for our experiments. In this case, the thickness was 230 μm. This configuration is shown in Fig. S1, Supplementary Information.

Spectroscopic techniques: Spectroscopic measurements at zero magnetic field were performed using a Bruker Equinox 55 Fourier-transform infrared spectrometer across the infrared range from 800 - 2600 nm, more than covering the telecom wavelengths (1260 - 1625 nm) and the response of the f-manifold excitations in that vicinity. Unpolarized light was used for all experiments. The absorption coefficient was calculated as: $\alpha(\lambda) = (-1/d)\ln(\mathcal{T}(\lambda))$, where $\mathcal{T}$ is the measured normalized transmission, and d is the crystal thickness. An open-flow cryostat was used for temperature control.

Magneto-optical measurements: Magneto-optical spectroscopy was performed at cryogenic temperatures using a 65 T pulsed magnet at the National High Magnetic Field Laboratory in Los Alamos, NM. The samples were mounted in the Voigt geometry on a fiber-coupled probe. Two multi-mode optical fibers were used to deliver broadband, unpolarized, white light to the sample, and to collect the light transmitted through the sample, respectively. The collected light was dispersed in a 300 mm spectrometer, using a 600 groove/mm grating, and was detected by a 1024-pixel InGaAs array detector. We focused on the E-, S-, and C-bands of the telecom wavelength range (1460 - 1565 nm), achieving approximately 0.1 nm spectral resolution to capture changes in the sharp f-manifold excitations. To test nonreciprocal directional dichroism, the propagation direction of the light ($\pm k$) through the sample was reversed by switching the delivery and collection fibers and also by reversing the direction of the magnetic field ($\pm H$). A comparison is shown in Fig. S2, Supplementary Information. We provide an example of the raw data collected at $\pm$55 T in Fig. S3, Supplementary Information.

DATA AVAILABILITY

The datasets generated and/or analyzed during the current study are not publicly available due to active intellectual property considerations, but are available from the corresponding

authors upon reasonable request.

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AUTHOR CONTRIBUTIONS

JLM and SWC designed the study. XX grew the crystals under the supervision of SWC. KAS, SAC, and JLM performed the pulsed field magneto-optical spectroscopies. KS and YG analyzed the data with guidance from JLM. KAS and JLM developed the figures and wrote the manuscript. KAS, YG, XX, HSK, SWC, SAC, and JLM commented on the text.

COMPETING INTERESTS

Authors KAS, YG, XX., HSK, SAC, and JLM declare no competing interests. SWC serves as an editor of this journal and had no role in the peer-review or decision to publish this paper. SWC declares no competing interests.

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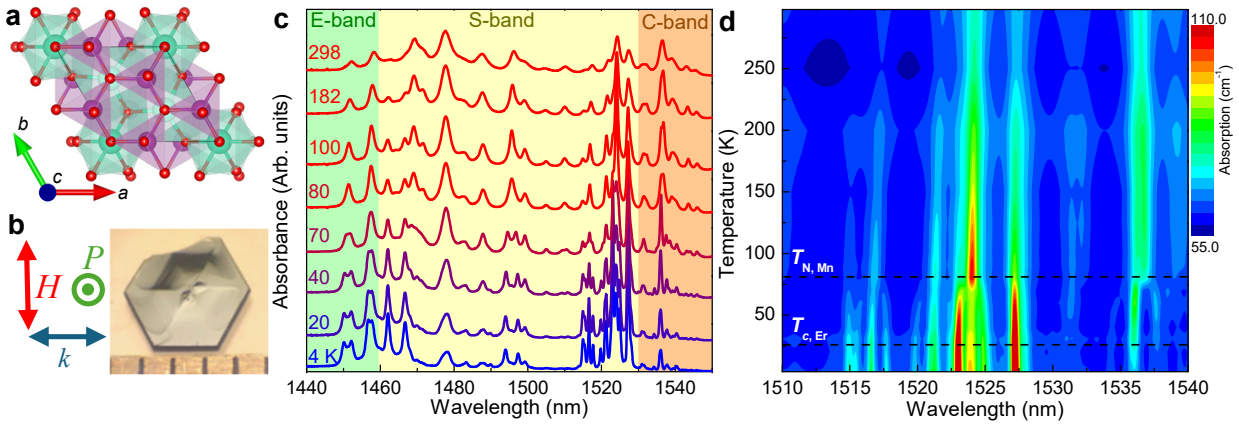

FIG. 1. **Structure, measurement conditions, and temperature effects in h-$Lu_{0.9}Er_{0.1}MnO_3$.** **a**, Crystal structure of h-$Lu_{0.9}Er_{0.1}MnO_3$ in the $P6_3mc$ space group.[41] The Lu/Er, Mn, and O sites are indicated by teal, magenta, and red spheres, respectively. **b**, Schematic of the measurement geometry indicating the light propagation k, applied field H, and polarization P directions (left) and optical microscope image (right) of the ab-plane of the single crystal revealing the natural canopy-like structure that is indicative of a monopolar domain sample. Our single crystals were polished to expose the c-axis (which is the direction of polarization) as well as the $\perp c$ direction. Additional information about the orientation [Fig. S1], monopolar domain character [Fig. S5], and magnetic properties [Fig. S6] is provided in the Supplementary Information. **c**, Absorption of the $^4I_{15/2} \rightarrow {}^4I_{13/2}$ rare earth crystal field excitations in h-$Lu_{0.9}Er_{0.1}MnO_3$ as a function of temperature with the corresponding telecom wavelength ranges indicated in green (E-band), yellow (S-band), and orange (C-band). The spectra are offset for clarity. **d**, Contour plot of the spectra in panel **c** with focus on the C-band features. The horizontal lines indicate Mn^{3+} antiferromagnetic ordering and the Er^{3+}-related transition.

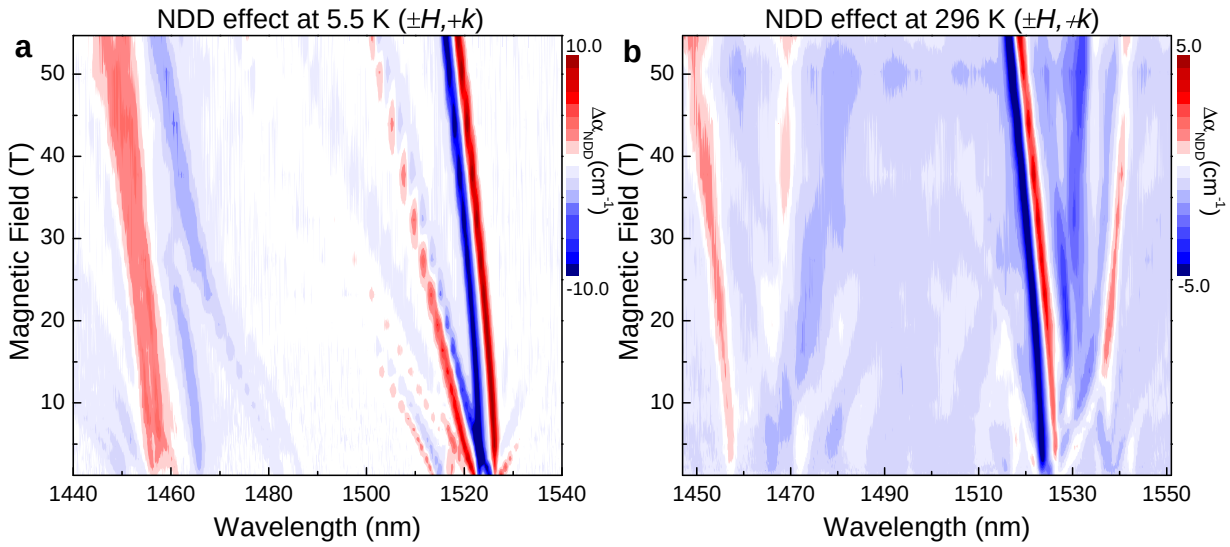

FIG. 2. **Color contour plots of dichroic effects.** Nonreciprocal directional dichroism of h-$Lu_{0.9}Er_{0.1}MnO_3$ at **a**, 5.5 K and **b**, room temperature (296 K) in a toroidal geometry in pulsed magnetic fields to 55 T and shown as contour plots. The color indicates the strength and sign of the dichroic signal. Nonreciprocal directional dichroism $\Delta\alpha_{\rm NDD}$ is calculated as $\Delta\alpha_{\rm NDD} = \alpha(+H,+k)$ - $\alpha(-H,+k)$. This quantity is the difference between field pairs.

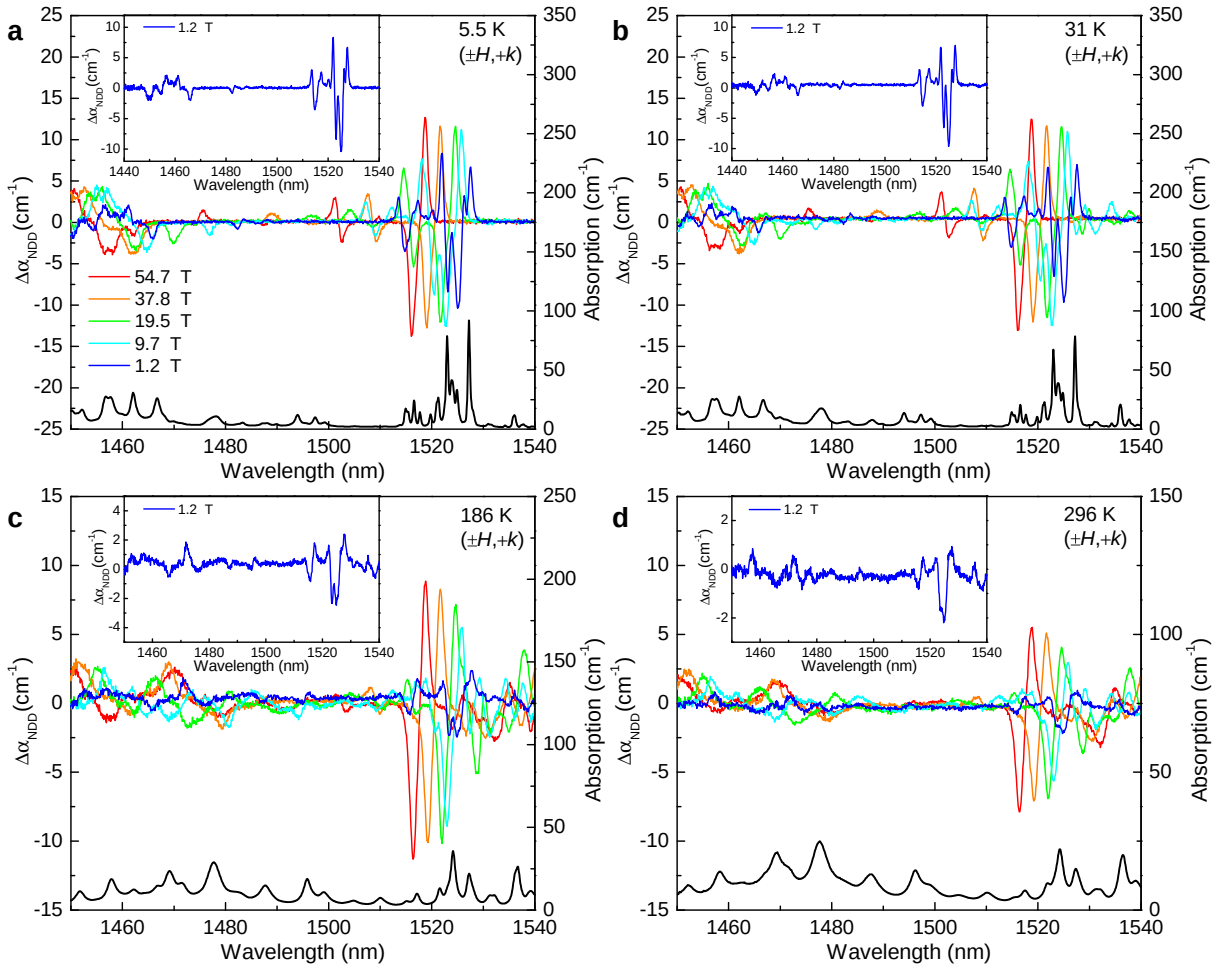

FIG. 3. **Temperature dependence of nonreciprocal directional dichroism.** Nonreciprocal directional dichroism of h-$Lu_{0.9}Er_{0.1}MnO_3$ at **a**, 5.5 K, **b**, 31 K, **c**, 186 K, and **d**, 296 K at selected fields. The absolute absorption at each temperature is at the bottom of each panel for comparison. These features are assigned as Er^{3+} $^4I_{15/2} \rightarrow {}^4I_{13/2}$ crystal field excitations. The insets show $\Delta\alpha_{\mathrm{NDD}}$ at 1.2 T.