Spin quantum well-like behavior in single-crystal $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$

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Rare earth orthoferrites (RFeO_3 , $\text{R} = \text{Y, Gd, Sm, Nd}$) have recently attracted much attention due to their novel magnetic and electrical properties such as magneto-electrics, magneto-dielectrics, and magneto-optics [1–3]. The complex interactions between the two sublattices of R^{3+} and Fe^{3+} in RFeO_3 provide the feasibility of controlling spins in these solids, which makes them promising candidates for the spintronics [4]. A major challenge is to develop a kind of material in which favorable properties such as stability, tunability, and non-volatility [5] can be combined. In this paper, we prepared a high-quality single-crystal of $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ and investigated its magnetic properties from 2 K to room temperature. Different from other RFeO_3 orthoferrites ($\text{R} = \text{rare earth}$, such as Gd [6], La [7], Er [8], Sm [9,10], and Tm [11]), the weak-ferromagnetic (wFM) moments of the single-crystal of $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ are discrete and have only two states: the same in magnitude but in opposite direction along the ferromagnetic axis, thus being not able to rotate randomly. The AFM coupling between Gd and Fe sublattices is broken in $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$, which leads to a spin-flip. The moment of the Gd sublattice behaves in the same direction with the moment of Fe sublattice until the external magnetic field is large enough. On the other hand, different from common materials whose coercivity often varies with the temperature [12–18], the coercivity in $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ is nearly constant from 50 to 350 K, avoiding being influenced by temperature in the application. These features give $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ great potential

as a convenient and reliable material for information storage.

The single-phase sample of $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ was prepared by a solid-state method in a floating zone furnace (see Supplementary information). Fig. 1a illustrates the crystallographic structure of $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ and the magnetic structure of Fe atoms above the anti-ferromagnetic (AFM) ordering temperature of Gd. The X-ray intensity profiles of the powdered single crystal (Fig. 1b) verify the successful preparation of $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ with high crystalline quality (Fig. S1). All the peaks in the diffraction pattern can be indexed well with the typical perovskite structure having a monoclinic group of $Pnma$ [19]. No visible structure phase transition is observed in the temperature range of 5–300 K (Fig. S2).

In our work, all the magnetic measurements were performed along the b -axis of the crystal (Fig. S1). $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ was first cooled to 2 K under a magnetic field of 0 Oe in the zero-field-cooled (ZFC) measurement and 300 Oe in the field-cooled (FC) measurement. The magnetization curves were then obtained under a measuring magnetic field of 50 Oe. The similar shapes and data values of ZFC and FC curves (Fig. 2a) indicate that $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ is not sensitive to the initial cooling process, different from other reported ABO_3 orthoferrites [6,10,20]. The spin ordering temperature (Néel temperature (T_N)) of the Gd sublattice is 3 K, while the Fe sublattice shows an AFM ordering ($T_N^{\text{Fe}} = 695$ K) with a wFM of $0.06 \mu_B$ per formula unit (f.u.) (Fig. 2b), likely due

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Figure 1 (a) Orthorhombic atomic structure and the magnetic structure of Fe atoms in $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$. The arrows represent the directions of magnetic moments. (b) X-ray diffraction (XRD) patterns of the single-crystal $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ and powdered single-crystal at room temperature.

Figure 2 (a) Temperature dependence of magnetization after ZFC and FC in $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ after demagnetization. (b) The hysteresis loop at 300 K. The inset shows the magnified view of the hysteresis loop where points A and B represent the original status when the external magnetic field is zero. (c, d) Temperature dependence of magnetization measured under different applied magnetic fields of (c) 0, -100, -200, -300 Oe and (d) -400 Oe after the positive magnetization.

to the Dzyaloshinskii-Moriya (DM) interaction [21].

To investigate the evolution of magnetization, we kept the direction of the applied magnetic field B downward and referred the magnitudes of fields to the absolute value (Fig. 2c and d). The corresponding directions of applied magnetic field B , Fe moments and Gd moments have been illustrated with arrows in the diagram of Fig. 2c and d. The $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ crystal was then magnetized un-

der a large magnetic field so that the magnetization was fixed at the magnetic state of A or B shown in Fig. 2b. We also obtained M - T curves in the FC cooling (FCC) processes when the magnetic fields were applied either antiparallel to the b -axis in Fig. 2c or parallel to the b -axis in Fig. S3. The negative and positive signs herein represent the magnetic moments in the downward and upward directions, respectively.

Specifically, the sample with a positive remanence value showed upward magnetic moments, staying at point A before all the measurements were performed. As shown in Fig. 2c and d, in the temperature range of 50–300 K, the magnetizations keep nearly flat, which results from the Fe sublattice. Thus, the positive sign indicates the upward direction of the total magnetic moments of the Fe sublattice. As the temperature decreases below 50 K, the magnetization changes dramatically, likely resulting from the magnetic ordering of Gd moments [22,23]. Based on the effective field model, the $M(T)$ curves under different magnetic fields (Fig. 2c and d) below 50 K could be divided into three types. The effective field B_{eff} applied to Gd atoms can be described as

$$B_{\text{eff}} = B_{\text{Fe}} + B_{\text{ex}}, \quad (1)$$

where B_{Fe} and B_{ex} are the effective field from the Fe sublattice and the external magnetic field, respectively. The estimated value of B_{Fe} from GdFeO_3 is about ~ 310 Oe [6,24].

(1) When the absolute value of the B_{ex} is smaller than 100 Oe, the magnetization increases with the decrease of the temperature. In this case, the B_{Fe} that is relatively larger than B_{ex} plays a more important role in the magnetic moments of the Gd sublattice. Thus, the magnetic moments of the Gd sublattice are induced in the same upward direction with the Fe sublattice, resulting in the sudden rise in the $M(T)$ curve at about 30 K.

(2) When the value of B_{ex} reaches 200 Oe, the magnetization exhibits the first decrease at 50 K to a minimum, and then a sharp increase at $T = 5.2$ K. The B_{ex} here becomes a major part of B_{eff} and cannot be neglected. As the temperature decreases from 50 K to lower, the Gd atoms are partially affected by the external field with a downward direction and produce downward magnetic moments, leading to the lessening of the upward total

magnetic moment and the decline of $M(T)$ curve. At 5.2 K, more Gd atoms become magnetic-ordered and produce upward magnetic moments, in the same direction with the Fe sublattice and opposite to the applied field, leading to a sharp increase of magnetization.

(3) When the value of B_{ex} becomes 400 Oe, the external field is strong enough to overcome the B_{Fe} , leading to the reverse to a large negative value (Fig. 2d). Thus, due to the strong effect of B_{ex} to the Gd sublattice at temperatures below 50 K, the magnetic moment decreases first. Since the total magnetic anisotropy field is lower than the external field, it indicates that both moments of the Fe and Gd sublattices are magnetized along the applied field below 17 K, where the total magnetic moments suddenly turn from a positive to a negative value. This is a distinctive spin-flip behavior which is not found in pure GdFeO_3 [6,25].

The hysteresis loops under different temperatures were also measured with the measuring field applied along b -direction. As shown in Fig. 3a, each hysteresis loop contains a linear curve in a broad range of measuring fields and a small loop whose saturated magnetization is much smaller than the linear part. Notably, the magnetization of the loop switches sharply. This implies that the $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ is mainly AFM, but the canted ferromagnetic component survives due to the DM interaction. The extracted coercivity is plotted in Fig. 3b, which clearly shows that the coercivity is almost a constant above 50 K and declines when the temperature decreases below 50 K. It is commonly accepted that the coercivity usually decreases with the increasing temperature, with very few exceptions. It is unique to have a type of material with a constant coercivity within a wide temperature range, whose stability is desired for the technical application. Based on the structure of $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$, the Hamiltonian is as follows:

Figure 3 (a) Hysteresis Loops measured along b -direction. (b) Coercivity as a function of temperature.

$$\begin{aligned}
 H = & \sum_{\langle ij \rangle} J_{\text{Fe}} S_i \cdot S_j + \sum_{\langle ij \rangle} J_{\text{Gd}} S_i \cdot S_j \\
 & + \sum_{\langle ij \rangle} J_{\text{GdFe}} S_i \cdot S_j + \sum_{\langle ij \rangle} D \cdot S_i \times S_j \\
 & + \sum_{\langle i \rangle} K_2 S_{iz}^2 + \sum_{\langle i \rangle} B_z S_{iz},
 \end{aligned} \quad (2)$$

where the first and the second terms denote the exchange interaction of the Fe sublattice and Gd sublattice, respectively. The third term is the coupling between Fe and Gd sublattices. The DM interaction from the Gd sublattice has been added as the fourth one. The last two terms come from the ionic anisotropy and external magnetic field.

The Gd sublattice is paramagnetic in the high-temperature region. Thus, the high-temperature magnetic properties only stem from the Fe sublattice. By down-folding the mean effect of the Gd sublattice from the external field of Fe lattice, the Hamiltonian becomes

$$\begin{aligned}
 H \approx & \sum_{\langle ij \rangle} \tilde{J}_{\text{Fe}} S_i \cdot S_j + \sum_{\langle ij \rangle} D \cdot S_i \times S_j \\
 & + \sum_{\langle i \rangle} K_2 S_{iz}^2 + \sum_{\langle i \rangle} \tilde{B}_z S_{iz}.
 \end{aligned} \quad (3)$$

Some typical representative values of the parameters are selected without loss of generality. $\tilde{J}_{\text{Fe}}$, D , K_2 and $\tilde{B}_z$ are selected as 1 a.u., 0.05 a.u., (0, 0, 0.05) a.u., and 0 respectively. The calculated total energy vs. spatial rotating angle of two Fe spins is plotted in Fig. 4a. The energy surface reveals a minimum when the spin directions of two Fe atoms are antiparallel. However, due to the DM interaction, the location of the energy minimum is not exactly at the position where two spin directions are antiparallel. Thus the ground state is not entirely AFM since the magnetic moments are canted along the b -axis, resulting in the wFM ordering. Considering the term of anisotropy, there is a barrier between two minima. A field large enough can overcome the barrier and switch wFM

components, which leads to the distinctive spin-flip. Therefore, the magnetic states can be treated as two-fold quantum wells, where the magnetization is an analogy to a trapped quasiparticle. The barrier can be estimated by multiplying the saturated magnetic moment and magnetic field. The atomic doping can be used to adjust the barrier since the atomic doping could change the spin-orbital coupling between Fe atoms. In Fig. 4b, the magnetic moments are trapped in either state of quantum wells. Hence, the coercivity and magnetic moment remain almost constant in the high-temperature region. In the low-temperature region with decreasing temperature, the magnetic ordering of Gd sublattices appears, yielding an effect on the total anisotropy field. Thus, the coercivity, the value of the external reverse field, reduces abruptly as the temperature decreases below 50 K.

Based on the above phenomena, materials with similar states may be suitable for fabricating devices that work at room temperature, especially for non-volatile memory that retains information when the power is off. To estimate the potential application, the magnetic domains were measured by a magnetic force microscope (MFM) under different measuring fields at a temperature of 3.5 K (Fig. 5). There were clear changes in the magnetic domains when the measuring field was applied. For instance, the small measuring field of 100 Oe can affect the domains without a hitch. The written pattern can be also read without destroying when a small field was applied to the magnetic probe of MFM.

Notably, the induced magnetic moment of the Gd sublattice has the same direction with the Fe sublattice, different from the usual AFM coupling between the rare-earth atom and Fe atom in RFO₃ compounds [1,6,10,26–28]. In a similar material of GdFeO₃, Das *et al.* [6] reported that the exchange interaction between the Gd and

Figure 4 Illustration of the magnetic quantum wells in Gd_{0.75}La_{0.25}FeO₃. (a) Calculated energy surface. θ_1 and θ_2 are the relative spatial angles of two Fe atoms from the z -axis in space. The color represents the energy E . (b) Illustration of the magnetic quantum wells and the corresponding magnetic configurations of two Fe atoms.

Figure 5 (a) Hysteresis loop at 3.5 K. (b) Magnetic domains related to the hysteresis loop at 3.5 K. The blue and red denote magnetic moments along up and down directions, respectively.

Fe moments produced an effective magnetic field at the Gd^{3+} site with the opposite direction to the canted Fe^{3+} moment. We also tested GdFeO_3 with the same method as described above (see Figs S4 and S5), and the performance is the same as Das's report. As there are only 25% non-magnetic La atoms doping in the GdFeO_3 and no structural phase transition, such a vast change in Gd-Fe coupling is unexpected. We thus propose that the paramagnetic La atoms replace the magnetic Gd atoms in a cell unit of $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$, leading to the destruction of the coupling between the Gd and Fe sublattices. At low temperatures, $\sum_{\langle ij \rangle} J_{\text{Gd}} S_i \cdot S_j$ and $\sum_{\langle ij \rangle} J_{\text{GdFe}} S_i \cdot S_j$ are smaller than usual and depend on the way that La atoms replace the Gd sublattice. Therefore, the binary quantum well will still exist at low temperatures without the coupling between Gd and Fe sublattices. When there is no AFM coupling, the total magnetic moments of $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ are easier to reverse than that of GdFeO_3 .

In summary, $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ is synthesized as a perfect single crystal with uniqueness in coercivity temperature coefficient, magnetization process, and spin-flip. The observed phenomena can be explained qualitatively by a Hamiltonian model based on the spin quantum well-like energy surface that can trap magnetic moments in a certain direction. $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ is easier to switch in the binary quantum well, being suitable for spintronic devices. Furthermore, the doping of La destroys the AFM coupling in Gd and Fe sublattices and promotes the influence of Fe sublattice-derived effective magnetic field on the Gd atoms.

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Author contributions Wen X and Yang J conceived the idea. Fang Y, Cao S, Zhang X, Cheng Z prepared the materials. Song Z conducted the DFT calculations. Malik IA and Zhang J conducted the measurement of magnetic domains. Wen X performed the experiments and wrote the paper with support from Yang J, Yang W, Han J, Liu S, Du H and Hou Y contributed to the correction of the manuscript. Hou Y and Yang J revised the manuscript. All authors contributed to the general discussion.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Experimental details and supporting data are available in the online version of the paper.

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单晶 $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ 中的类自旋量子阱行为

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摘要 为满足实际应用, 自旋电子学器件需要自旋在特定方向发生自发取向, 但这通常是难以控制的. 在本文中, 我们观察了沿 b 轴的单晶 $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ 在室温下伴随着弱铁磁(wFM)成分的反铁磁(AFM)磁有序. 与普通的AFM材料不同, 这里的wFM磁矩是离散的, 类似于二维量子阱, 可以通过一个小磁场在两个状态之间切换. 量子阱中的势垒使得矫顽力在50–350 K的宽温度范围内保持稳定, 并在17 K时产生独特的自旋翻转现象. 此外, 在 RFeO_3 ($\text{R}=\text{稀土}$)中, 稀土和铁的次晶格之间普遍存在着一种AFM耦合. 而在 $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ 中, Gd与Fe次晶格之间的AFM耦合被破坏, 使得Gd被诱导出的磁矩与Fe磁矩方向不再相反, 而是表现出与Fe磁矩方向相同的诱导磁矩. 由于在 GdFeO_3 或 LaFeO_3 中不存在自旋翻转, 因此AFM耦合的破坏也被认为是材料表现出一种独特的自旋翻转现象的主要原因. 以上性质使得 $\text{Gd}_{0.75}\text{La}_{0.25}\text{FeO}_3$ 在信息存储方面具有更优良的可控性.