Magnetic Field Induced Sign Reversal of the Anomalous Hall Effect in a Pyrochlore Ferromagnet $Nd_2Mo_2O_7$: Evidence for a Spin Chirality Mechanism

Y. Taguchi,[1,2] T. Sasaki,[1] S. Awaji,[1] Y. Iwasa,[1] T. Tayama,[3] T. Sakakibara,[3] S. Iguchi,[2] T. Ito,[4] and Y. Tokura[2,4,5]

[1]*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

[2]*Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan*

[3]*Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan*

[4]*Correlated Electron Research Center (CERC), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8562, Japan*

[5]*Spin Superstructure Project (SSS), ERATO, Japan Science and Technology Corporation (JST), Tsukuba 305-8562, Japan*

(Received 24 January 2003; published 25 June 2003)

The anisotropic Hall effect and magnetization have been investigated for single crystals of $Nd_2Mo_2O_7$ under high magnetic fields up to 27 T and at low temperatures down to 50 mK. The magnetization curves indicate that the Nd moments with strong Ising anisotropy are coupled antiferromagnetically with the Mo spins and show field-induced flipping on the respective sites. The Hall resistivity changes its sign with increasing field applied along the [111] direction, while it monotonously approaches zero with the field applied along the [100] or [110] direction. This behavior is in accord with the prediction by the Berry phase theory and is interpreted in terms of the field-induced reversal of spin chirality on the pyrochlore lattice.

DOI: 10.1103/PhysRevLett.90.257202 PACS numbers: 75.47.–m, 75.30.Mb, 75.50.Cc

In strongly correlated electron systems, a spin state strongly affects the charge dynamics. For example, colossal magnetoresistance phenomena in manganites can be viewed as a consequence of the modified magnitude of the transfer integral by the spin configuration [1]. The phase of the transfer integral should also be modified by the nontrivial spin texture and could produce the gauge flux [2]. Recently, this phase (termed Berry phase) has been argued to be responsible for anomalous Hall effect (AHE) or transverse conductivity observed for several ferromagnetic transition-metal oxides [3–12]. Weak coupling regime has also been examined very recently [13]. According to the Berry phase theories (BPT), the conduction electron feels a fictitious magnetic field produced by the noncoplanar spin configuration or spin chirality, and the transverse conductivity appears. On the other hand, there is some criticism [14,15] against the interpretation of the AHE in pyrochlore-type $Nd_2Mo_2O_7$ (NMO) in terms of the BPT. In this Letter, we present concrete evidence that the AHE in the pyrochlore ferromagnet does arise from this spin chirality.

The pyrochlore ($A_2B_2O_7$) lattice is composed of two sublattices of A site and B site, which are structurally identical but are displaced by half a lattice constant from each other. In each sublattice, the ions locate at the vertex of corner-sharing tetrahedra, as depicted in the inset of Fig. 1(a). The single crystals of NMO used in this study were grown by the floating-zone method in Ar atmosphere. The measurements of Hall resistivity (ρ_H) were done by using the conventional method with dc excitation at 1.6 K up to 27 T. We eliminated the longitudinal voltage drop by reversing the field direction. A vibrating-sample magnetometer was used for the magnetization measurements at 1.7 K up to 23 T. The magnetic field was provided by a hybrid magnet at High Field Laboratory, IMR, Tohoku University. Besides this high field study, we have also measured the magnetization down to 50 mK and up to 12 T. In this low temperature study, we used a capacitive Faraday-force magnetometer in a dilution refrigerator [16].

In Fig. 1(a), we show the temperature variation of the magnetization at $H = 0.5$ T for the NMO and $Gd_2Mo_2O_7$ (GMO) crystals, whose Curie temperatures (T_C) are 89 and 45 K, respectively. The Nd moments align antiparallel to the Mo $4d$ spins at T_C and begin to grow rapidly around ≈ 40 K, resulting in a reduction of the total magnetization at low temperatures. A thick bar indicates the Mo spin moment ($\approx 1.4\mu_B$/Mo) at low temperature which was deduced from the analysis of magnetization data and neutron diffraction data [8]. In the GMO, the Gd $4f$ moment couples ferromagnetically with the Mo $4d$ spin. This is evidenced by the large total magnetization at low temperatures which well exceeds the expected value for Mo spin alone ($\approx 1.4\mu_B$/Mo).

The Hall conductivity $\sigma_{xy} = \rho_H/(\rho_{xx}^2 + \rho_H^2)$ at $H = 0.5$ T for the each sample is displayed in Fig. 1(b). As already reported [8], the Hall conductivity of NMO for each direction continuously increases down to 2 K, and finally saturates only below 2 K (not shown here). Such a behavior is distinct from the conventional behaviors of the AHE [17]. By contrast, the GMO shows a conventional temperature dependence as the ferromagnetic metal: The absolute value of the Hall conductivity takes maximum near T_C (= 45 K), and decreases toward low temperature. Furthermore, the absolute value itself is much smaller than that of the NMO. This remarkable difference between the two materials with very similar longitudinal transport properties can hardly be reconciled with the

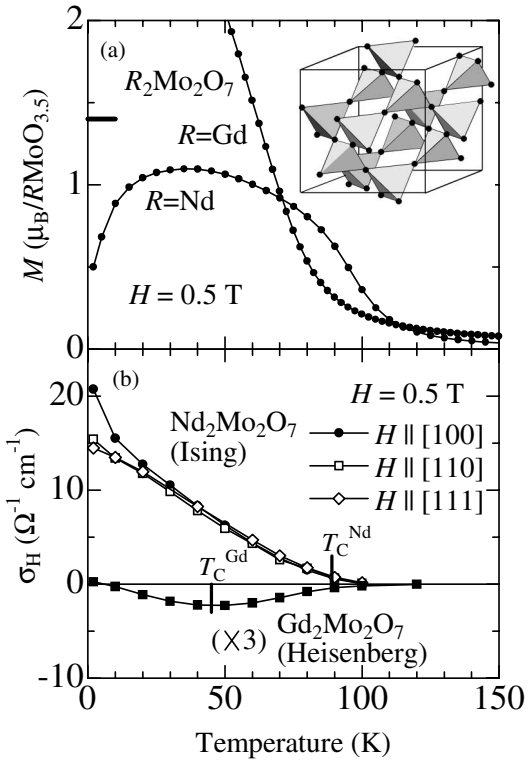

FIG. 1. (a) Temperature dependence of magnetization for $R_2Mo_2O_7$ crystal with R = Nd and Gd. A thick bar indicates the estimated value of Mo spin moment at low temperatures. (b) Temperature variation of the Hall conductivity for $Nd_2Mo_2O_7$ and $Gd_2Mo_2O_7$. The data for Gd compound are multiplied by 3. The inset of (a) shows the schematic figure of the Mo sublattice of the pyrochlore structure.

conventional theories based upon the spin-orbit interaction of the conduction electrons which would be least affected by the rare-earth species. On the other hand, the difference is naturally understood in terms of the BPT. The Nd^{3+} ($4f^3$) is of Ising-like anisotropy with $\langle 111 \rangle$ axis while Gd^{3+} ($4f^7$) without orbital angular momentum is of Heisenberg-like one [18]. Therefore, the spin chirality is produced in the Mo spin system via the f-d interaction only in the case of the Nd compound. This is the reason why the unconventional behavior of Hall conductivity is observed for NMO and also for $Sm_2Mo_2O_7$ with the Ising anisotropy of the Sm moment [6,9], but not for GMO.

One of the important predictions by the BPT is that the sign of ρ_H changes when a strong magnetic field is applied along the [111] direction, whereas it does not when the field is applied along the [100] or [110] direction. To make a more precise statement, we define the fictitious magnetic field that penetrates a single Mo tetrahedron $\vec{b}_{Mo}$ as a vector sum:

$$\vec{b}_{Mo} = \sum_{\langle i,j,k \rangle} (\vec{S}_i \cdot \vec{S}_j \times \vec{S}_k)\vec{n}_{ijk},$$

where $\vec{n}_{ijk}$ is a normal vector (with unit length) of a triangle formed by site i, j, and k. When the field is along the [100] or [110] direction, a relation that $\vec{b}_{Mo} \cdot \vec{H} < 0$ always holds, whereas $\vec{b}_{Mo} \cdot \vec{H}$ changes sign when strong field is applied along the [111] direction. Therefore, the ρ_H arising from the fictitious field changes sign when the field direction is [111].

Schematic configurations of the Nd moments and Mo spins are shown in Fig. 2. Figure 2(a) shows a case where a weak field (e.g., H = 0.5 T) is applied along the [100] direction. The Mo spins are almost ferromagnetically aligned along the field direction. The Nd moments feels a downward effective field due to the antiferromagnetic interaction with the Mo spins. Thus, the Nd moments, which are subject to strong single-ion anisotropy, form the "2 in, 2 out" configuration. Then, in turn, Mo spins are tilted from the direction of net magnetization by the antiferromagnetic interaction with Nd moments. In the pyrochlore structure, Mo site 1 locates at the center of a hexagon formed by two Nd 2′, two Nd 3′, and two Nd 4′. Taking into account the antiferromagnetic interaction

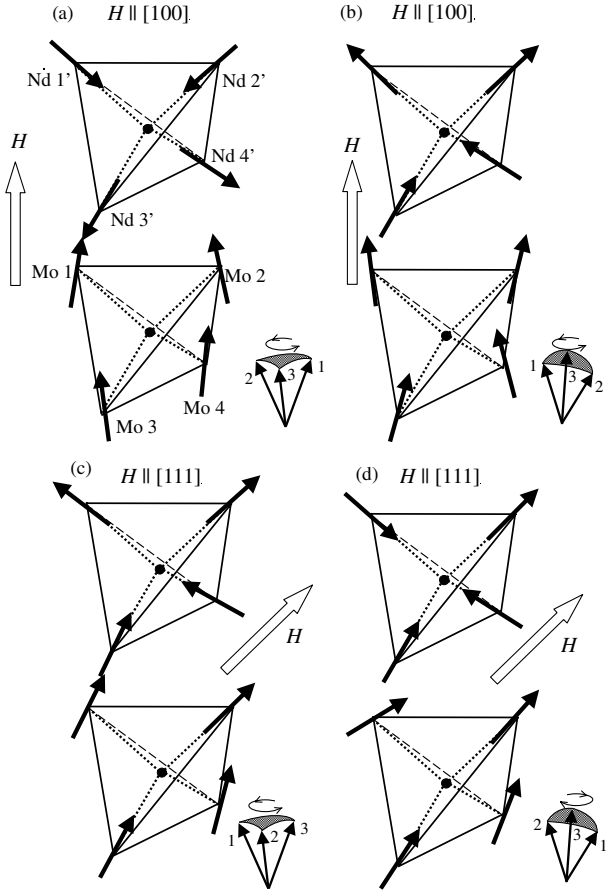

FIG. 2. Schematic figures of spin configurations for Nd and Mo tetrahedra. In each panel, upper and lower tetrahedra correspond to Nd and Mo tetrahedra, respectively. The relationship among the Mo spin 1, 2, and 3 is schematically shown at the right-hand side of each Mo tetrahedron. Open arrows depict directions of applied magnetic field. In (a) and (b), and (c) and (d), magnetic field is applied along the [100] and [111] directions, respectively. See text for details.

between Mo and Nd, we can infer that the transverse component of the Mo spin 1 points parallel to that of the Nd moment 1′. Thus, the Mo spin system acquires the same "tilting habit" as the Nd moment system. In this particular spin configuration, $\vec{b}_{\mathrm{Mo}} \cdot \vec{H}$ is negative. Figure 2(b) depicts the situation where all the Nd moments are reversed toward the field direction by a strong enough field (e.g., $H = 5$ T). Similar to the case of Fig. 2(a), the anisotropy of the Nd moment system is transmitted to the Mo spin system. The Mo spin configuration in Fig. 2(b) is obtained from the configuration in Fig. 2(a) by operating a rotation around the field direction by 180°. Importantly, the spin chirality is invariant under the global spin rotation; thus, the fictitious magnetic field that penetrates the Mo tetrahedron points to the same direction as in Fig. 2(a). Therefore, the ρ_{H} will not change sign in terms of the Berry phase model, even when all the Nd moments are reversed by the field along the [100] direction. The situation is analogous when the field is applied along the [110] direction; the 2 in, 2 out tilting habit for the transverse Mo moments is similar to the case of $\vec{H} \parallel [100]$, and hence there will be no change in sign of the Mo spin chirality and hence in sign of the Hall effect as well.

Figure 2(c) represents a case where the field is applied along the [111] direction. Here, the field is already strong enough to reverse the averaged Nd moment, but not yet so strong (e.g., $H = 5$ T) as to make all the spins satisfied in terms of Zeeman energy. This state also corresponds to the 2 in, 2 out structure. Figure 2(d) shows a spin configuration where a magnetic field (e.g., $H = 12$ T) applied along the [111] direction forces all the Nd spins to align so as to satisfy the Zeeman energy. In this case, the Nd tetrahedron consists of the "3 in, 1 out" (or "1 in, 3 out") configuration. Then, the Mo spin system again acquires the same tilting habit as that in the Nd moment system, namely, the 3 in, 1 out structure, where $\vec{b}_{\mathrm{Mo}} \cdot \vec{H}$ is positive. This means that the fictitious field operating on conduction electrons changes its sign, and hence the sign of the ρ_{H} will be reversed.

In Fig. 3(a), we show the magnetization curve for fields along the [100] and [110] direction at 1.7 K and 50 (or 70) mK. The estimated value of the Mo 4*d* moment in this temperature range is $\approx 1.4\mu_B$ [8], and the *averaged* Nd moment should vanish at the field of $H_0 \approx 3$ T. This consideration is consistent with recent results of neutron diffraction in a magnetic field [15,19]. For the respective field directions, the magnetization curves are almost identical at 1.7 K and 50 (or 70) mK. Saturation moments above 10 T are in accord with the estimated values (indicated by horizontal thick bars) under the following assumption: We assumed that the longitudinal component of the Mo spin moment is field independent (because of the small tilting angle) and that the Nd moment is of strong Ising anisotropy with its effective moment and Ising axis being $g_{\mathrm{eff}}J \approx 2.3$ and the $\langle 111 \rangle$ direction, respectively. Smooth nature of magnetization curves below 10 T even at 70 (or 50) mK suggests that the Nd moments flip incoherently from site to site, not in a cooperative and metamagnetic manner. Therefore, the local spin structure, and hence the local spin chirality, is different from the *averaged* spin structure during the magnetization process.

In Fig. 3(b), the ρ_{H} is plotted as a function of magnetic field applied along the [100] or [110] direction. For both field directions, the ρ_{H} decreases monotonously and approaches zero. At the magnetic field of H_0 (≈ 3 T), both of the longitudinal and transverse components of the *averaged* Nd moment vanish. Then, the transverse component of *averaged* Mo moment also vanishes. However, as noted above, each Nd moment does not vanish at all even at H_0, and hence each Mo moment does have a transverse component, namely, local spin chirality. Therefore, finite ρ_{H} appears at H_0. What should be compared with the experimental result is the *averaged* fictitious field generated from the local Mo spin chirality ($= \langle \vec{S}_1 \cdot \vec{S}_2 \times \vec{S}_3 \rangle$), but not the fictitious field calculated from such an *averaged* spin strucuture ($= \langle \vec{S}_1 \rangle \cdot \langle \vec{S}_2 \rangle \times \langle \vec{S}_3 \rangle$) [15] as determined from the neutron diffraction experiment. At high magnetic fields, the Mo spins are aligned along the field direction. Therefore,

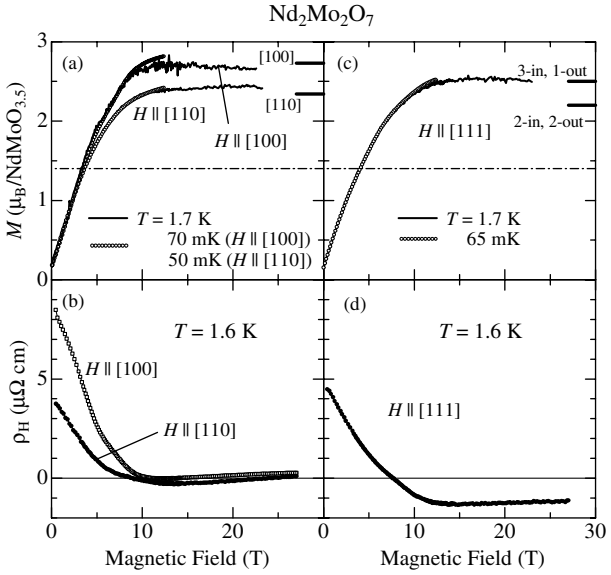

FIG. 3. Magnetization curves for the applied field (a) along the [100] and [110] directions, and (c) along the [111] direction. ρ_{H} is plotted as a function of the magnetic field which is applied (b) along the [100] or [110] direction, and (d) along the [111] direction. In (a) and (c), the horizontal dot-dashed line represents the contribution from the Mo spin moment. Thick horizontal bars in (a) indicate the expected values of the saturation moment for each field direction under the assumption of strong Ising anisotropy of Nd moments. See text for details. In (c), thick horizontal bars indicate the expected values of magnetization when the magnetic configurations of all the Nd tetrahedra are "3 in, 1 out" and "2 in, 2 out," respectively.

the tilting angle of the Mo spin becomes smaller and smaller as the field is increased. This reduction of the tilting angle results in the decrease of ρ_H.

In Fig. 3(c), we plot the magnetization curve for the field applied along the [111] direction at 1.7 K and 65 mK. Surprisingly, the magnetization process is temperature independent also for this field direction at these low temperatures. The saturation moment at high fields almost coincides with the expected value for the 3 in, 1 out structure under the same assumption as above, but apparently larger than the expected value for the 2 in, 2 out configuration. The magnetization process below about 10 T is gradual also for this field direction. Therefore, the spin configuration of the Mo tetrahedra is a mixture of the 2 in, 2 out and the 3 in, 1 out structure with the ratio changing as the field is increased [20], and finally becomes the 3 in, 1 out configuration for all the tetrahedra above 13–15 T.

The ρ_H for the [111] field direction is shown in Fig. 3(d). Clearly, it changes sign at 7.5 T. With increasing magnetic field from zero, the number of tetrahedron with the 3 in, 1 out structure increases. In the high field limit, all the tetrahedra become the 3 in, 1 out configuration as evidenced by the magnetization curve. The local fictitious magnetic field is antiparallel to the local magnetization in the case of 2 in, 2 out while parallel in the case of 3 in, 1 out, as mentioned above. Therefore, in the course of increasing field from zero to strong enough field to transform all the tetrahedra to the 3 in, 1 out configuration, the internal fictitious field cancels out to be zero in the bulk limit. This field corresponds to 7.5 T, where the ρ_H vanishes. With further increasing field, the number of the tetrahedra with the 3 in, 1 out configuration becomes dominant, and the fictitious field for the conduction electron changes sign. This results in the sign reversal of the ρ_H, as observed.

The Nd moments preserve Ising anisotropy even at the highest field in this magnetization measurement ($= 23$ T). Therefore, the Mo spins are still tilted at this high field region regardless of the applied field direction, although the tilting angle is much smaller than the zero field value. This small tilting angle produces finite ρ_H. The ρ_H at high fields for the three field directions suggest that the contribution arising from conventional spin-orbit interaction, which does not depend on the field direction, must be negative. Then, it is likely that both contributions add (with negative sign) for the [111] direction, and almost cancel out for the [100] and [110] directions.

In summary, we have investigated the Hall effect and magnetization for single crystals of NMO. The magnetization curve shows little temperature dependence below 2 K, and the values of saturation moments are in accord with the assumption that the Nd moment is of Ising anisotropy with the magnitude and Ising axis being $\approx 2.3\mu_B$ and the $\langle 111 \rangle$ direction, respectively. The ρ_H changes sign when the field is applied along the [111] direction, but does not when applied along the [100] or [110] direction. This fact is consistent with the prediction by the BPT, evidencing the spin-chirality mechanism of AHE for NMO and related compounds.

We thank N. Nagaosa, S. Onoda, and K. Ohgushi for enlightening discussions and T. Takenobu for help in experiments. A part of this work was done at High Field Laboratory for Superconducting Materials, IMR, Tohoku University. This work was in part supported by a Grant-In-Aid for Scientific Research Priority Area from the MEXT, Japan.

[1] P.W. Anderson and H. Hasegawa, Phys. Rev. **100**, 675 (1955).
[2] N. Nagaosa and P. A. Lee, Phys. Rev. Lett. **64**, 2450 (1990).
[3] P. Matl *et al.*, Phys. Rev. B **57**, 10 248 (1998).
[4] J. Ye *et al.*, Phys. Rev. Lett. **83**, 3737 (1999).
[5] S. H. Chun, M. B. Salamon, Y. Lyanda-Geller, P. M. Goldbart, and P. D. Han, Phys. Rev. Lett. **84**, 757 (2000).
[6] Y. Taguchi and Y. Tokura, Phys. Rev. B **60**, 10 280 (1999).
[7] K. Ohgushi, S. Murakami, and N. Nagaosa, Phys. Rev. B **62**, R6065 (2000).
[8] Y. Taguchi, Y. Oohara, H. Yoshizawa, N. Nagaosa, and Y. Tokura, Science **291**, 2573 (2001).
[9] Y. Taguchi and Y. Tokura, Europhys. Lett. **54**, 401 (2001).
[10] M. Onoda and N. Nagaosa, J. Phys. Soc. Jpn. **71**, 19 (2002).
[11] S. Onoda and N. Nagaosa, Phys. Rev. Lett. **90**, 196602 (2003).
[12] H. Yanagihara and M. B. Salamon, Phys. Rev. Lett. **89**, 187201 (2002).
[13] G. Tatara and H. Kawamura, J. Phys. Soc. Jpn. **71**, 2613 (2002).
[14] T. Kageyama *et al.*, J. Phys. Soc. Jpn. **70**, 3006 (2001).
[15] Y. Yasui *et al.*, J. Phys. Soc. Jpn. **72**, 865 (2003).
[16] T. Sakakibara, H. Mitamura, T. Tayama, and H. Amitsuka, Jpn. J. Appl. Phys. **33**, 5067 (1994).
[17] J. P. Jan and J. M. Gijsman, Physica **18**, 339 (1952); R. S. Lee and S. Legvold, Phys. Rev. **162**, 431 (1967).
[18] N. P. Raju, M. Dion, M. J. P. Gingras, T. E. Mason, and J. E. Greedan, Phys. Rev. B **59**, 14 489 (1999).
[19] Y. Oohara, H. Yoshizawa, Y. Taguchi, and Y. Tokura (unpublished results).
[20] In a typical spin-ice system, e.g., $Dy_2Ti_2O_7$, the first-order-like transition from the "2 in, 2 out" to the "3 in, 1 out" configuration is discerned below 1 K [21]. However, the presence of Mo channel in NMO still obscures the transient feature even at 65 mK.
[21] H. Fukazawa, R. G. Melko, R. Higashinaka, Y. Maeno, and M. J. P. Gingras, Phys. Rev. B **65**, 054410 (2002); Z. Hiroi, K. Matsuhira, S. Takagi, T. Tayama, and T. Sakakibara, J. Phys. Soc. Jpn. **72**, 411 (2003).