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The spin chirality induced anomalous Hall effect in pyrochlore ferromagnets

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Abstract

Temperature and magnetic field dependences of the anomalous Hall effect have been investigated for single crystals of Mo-based ferromagnets with pyrochlore structure. The Hall resistivity of $Nd_2Mo_2O_7$ compound shows unconventional temperature dependence whereas $Gd_2Mo_2O_7$ exhibits rather normal behaviour. The Berry phase model can explain the difference well; it is attributed to the difference in nature of the anisotropy of the rare-earth moment and the resultant Mo spin state. The Hall resistivity of $Nd_2Mo_2O_7$ changes its sign with increasing field applied along the [111] direction, while it monotonically approaches zero with the field applied along the [100] or [110] direction. This behaviour is also in accord with the prediction of the Berry phase theory.

Materials with pyrochlore structure ($A_2B_2O_7$) have recently attracted much interest because of the rich variety of intriguing phenomena that they exhibit, such as spin ice behaviour [1], spin glass behaviour [2], spin liquid behaviour [3] superconductivity [4, 5], and ferromagnetism [6–8]. Among these phenomena, the lack of long range ordering in the former three cases is closely related to the unique structure and resultant 'geometrical frustration'. The pyrochlore lattice is composed of two sublattices of A- and B-sites, which are structurally identical but are displaced by half a lattice constant from each other. In each sublattice, the ions locate at the vertices of corner-sharing tetrahedra, as shown schematically in figure 1(a). The projection onto the (111) plane is the Kagome lattice, and one B ion is located at the centre of a hexagon formed by six A ions (and vice versa). This unique connectivity among the magnetic sites prevents, in many cases, the long range magnetic ordering. On the other hand, there are some pyrochlore materials that exhibit ferromagnetic ground states, for example vanadate [6],

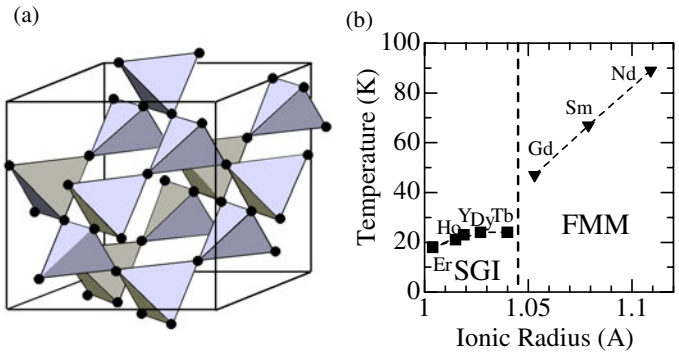


Figure 1. (a) A schematic diagram of the A-site (or B-site) sublattice of the pyrochlore structure. (b) The electronic phase diagram of pyrochlore-type molybdate where spin freezing temperatures (closed squares) and Curie temperatures (closed triangles) are plotted against the ionic radius of the A-site ion. FMM and SGI denote ferromagnetic metal and spin glass insulator, respectively.
(This figure is in colour only in the electronic version)

molybdate [7], and manganite [8]. Even in these ferromagnetic cases, complex and non-trivial spin structure is anticipated when the A-site and the B-site are both magnetic.

Among the many pyrochlore-type materials, the molybdate system is of particular interest because it shows a transition from a spin glass insulator to a ferromagnetic metal with change of the A-site ion [7]. Figure 1(b) shows the electronic phase diagram [9] plotted against the ionic radius of the A-site species. Similar phase diagrams have also been obtained by several other groups [7, 10]. It has been indicated by an optical study [9] that the electron correlation effect is of substantial importance in the metallic phase. In such a correlated metallic state, a spin state strongly affects the charge dynamics. The most dramatic manifestation of such interplay between spin state and charge dynamics is the so-called colossal magnetoresistance phenomenon in manganites [11], in which the longitudinal conductivity strongly depends on the Mn t_{2g} core spin configuration (together with additional charge/orbital ordering instabilities and/or Jahn–Teller interaction). This effect is viewed as a consequence of the magnitude of the transfer integral being modified by the spin configuration [12]. The phase of the transfer integral should also be modified by the non-trivial spin texture and could produce a gauge flux [13]. Such a gauge flux acts as a fictitious magnetic field and affects the charge dynamics in the same way as a real magnetic field does. Recently, the anomalous Hall effect or transverse conductivity has been discussed in terms of this phase (termed the Berry phase) in several ferromagnetic transition-metal oxides, for example perovskite-type manganites [14–16], molybdates with pyrochlore structure [18–22], and rutile-type CrO_2 [23]. The weak coupling regime has also been examined very recently in connection with the canonical spin glass system [24]. According to the Berry phase theories, the conduction electron feels a fictitious magnetic field produced by the non-coplanar spin configuration or spin chirality, and transverse conductivity appears. On the other hand, there are some arguments [25, 26] against the interpretation of the anomalous Hall effect in $Nd_2Mo_2O_7$ in terms of the Berry phase theory. Here, we present recent experimental evidence that the anomalous Hall effect in the pyrochlore ferromagnet does arise from the spin chirality.

In figure 2(a), we show the temperature variation of the magnetization at $H = 0.5$ T and the neutron scattering intensity of (200) peak for the $Nd_2Mo_2O_7$ crystal, whose Curie temperature was determined from ac susceptibility measurements to be 89 K. The Nd moments align

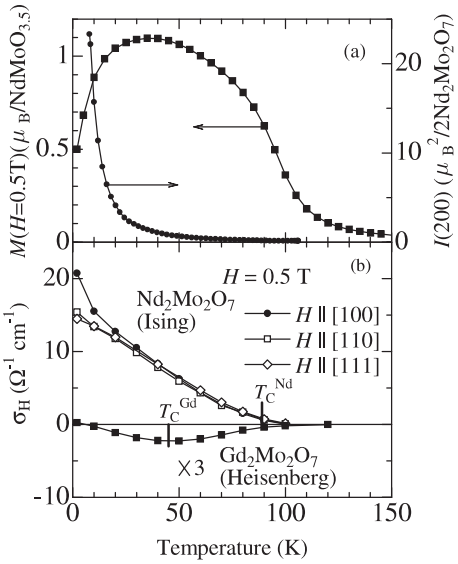


Figure 2. (a) The temperature dependence of the magnetization at $H = 0.5$ T and the background-subtracted and properly normalized neutron scattering intensity I of the (200) reflection, which represents the ordering of the transverse component. (b) The temperature variation of the Hall conductivity for $Nd_2Mo_2O_7$ and $Gd_2Mo_2O_7$. The data for the Gd compound are multiplied by 3.

antiparallel to the Mo 4d spins at the Curie temperature and begin to grow rapidly around a crossover temperature (≈40 K), resulting in a reduction of the total magnetization at low temperatures. At around this crossover temperature, the neutron scattering intensity of the (200) reflection begins to grow. The Nd moments and the Mo spins form an 'umbrella structure' [19] due to the strong ⟨111⟩ Ising anisotropy of the Nd moment and the antiferromagnetic interaction between the Nd moment and the Mo spin. The (200) reflection represents the ordering of the transverse component of the umbrella structure. The Mo spin moment at low temperatures was deduced to be approximately 1.4 μ_B/Mo from analysis of the magnetization data and neutron diffraction data [19].

We show in figure 2(b) the Hall conductivity $\sigma_{xy} = \rho_H/(\rho_{xx}^2 + \rho_H^2)$ at $H = 0.5$ T for $Nd_2Mo_2O_7$ and $Gd_2Mo_2O_7$. The most remarkable feature is that the Hall conductivity of $Nd_2Mo_2O_7$ for each direction continuously increases down to 2 K, and finally saturates only below 2 K (not shown here). Such a behaviour is totally different from the conventional behaviours of the anomalous Hall effect [27, 28]. By contrast, the Gd compound shows a conventional temperature dependence: the absolute value of the Hall conductivity takes a maximum near the Curie temperature (=45 K), and decreases toward low temperatures. Furthermore, the absolute value itself is much smaller than that for the Nd compound. It seems that the conventional theories can hardly explain this remarkable difference between two materials with very similar longitudinal transport properties since they are 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 Berry phase theory can naturally account for the difference. $Nd^{3+}(4f^3)$ has Ising-like anisotropy with a ⟨111⟩ axis while $Gd^{3+}(4f^7)$, without orbital angular momentum, has a Heisenberg-like one [29]. 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 why the unconventional behaviour of the Hall conductivity is observed for $Nd_2Mo_2O_7$

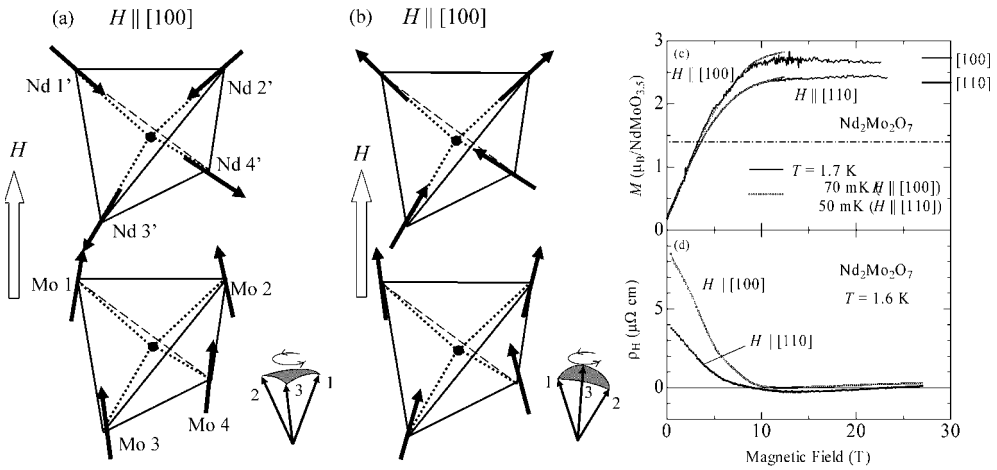


Figure 3. (a), (b) Schematic diagrams of spin configurations for Nd and Mo tetrahedra when the field is applied along the [100] direction. In each panel, upper and lower tetrahedra correspond to Nd and Mo tetrahedra, respectively. The relationship among the Mo spins 1, 2, and 3 is shown schematically on the right-hand side of each Mo tetrahedron. Open arrows depict the directions of the applied magnetic field. See the text for details. (c) Magnetization curves for the applied field along the [100] and [110] directions. The horizontal dot–dashed line represents the contribution from the Mo spin moment. Thick horizontal bars indicate the expected values of the saturation moment for each field direction under the assumption of strong Ising anisotropy of Nd moments. See the text for details. (d) $\rho_{\rm H}$ is plotted as a function of the magnetic field which is applied along the [100] or [110] direction.

and also for $Sm_2Mo_2O_7$ with the Ising anisotropy of the Sm moment [17, 20], but not for $Gd_2Mo_2O_7$.

The Berry phase theory has an important prediction that the sign of the Hall resistivity 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] directions. To make a more precise statement, we define the fictitious magnetic field that penetrates a single Mo tetrahedron, $\vec{b}_{\rm Mo}$, as a vector sum:

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

where $\vec{n}_{ijk}$ is a normal vector (with unit length) of a triangle formed by sites i, j, and k. The relation $\vec{b}_{\rm Mo} \cdot \vec{H} < 0$ always holds when the applied field is along the [100] or [110] directions. On the other hand, $\vec{b}_{\rm Mo} \cdot \vec{H}$ changes its sign when a strong field is applied along the [111] direction. Thus, the Hall resistivity that arises from the fictitious field changes sign when the field is applied along the [111] direction.

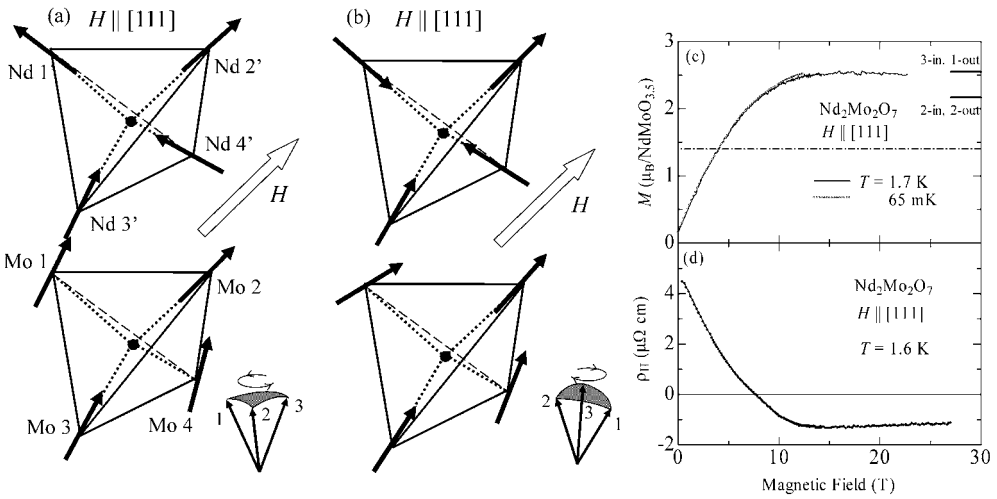
In figures 3(a) and (b), we display schematic configurations of the Nd moments and Mo spins, where the magnetic field is applied along the [100] direction. In both panels, the upper tetrahedron represents the Nd tetrahedron and the lower one corresponds to the Mo tetrahedron. Figure 3(a) shows a case 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 centre of a hexagon formed by two Nd 2′, two Nd 3′, and two Nd 4′. Taking into account the antiferromagnetic interaction 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, $\vec{b}_{\rm Mo} \cdot \vec{H}$ is negative. Figure 3(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). Similarly to the case of figure 3(a), the anisotropy of the Nd moment system is transmitted to the Mo spin system. The Mo spin configuration in figure 3(b) is obtained from the configuration in figure 3(a) by effecting 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 in the same direction as in figure 3(a). Therefore, the Hall resistivity will not change sign in the Berry phase model, even when all the Nd moments are reversed by the field applied 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.

In figure 3(c), we show the field dependence of the magnetization for fields applied along the [100] and [110] directions at 1.7 K and 50 (or 70) mK. The estimated value of the Mo 4d moment in this temperature range is $\approx$1.4 $\mu_{\rm B}$ [19], 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 [26, 30]. For the respective field directions, the magnetization curves are almost identical at 1.7 K and 50 (or 70) mK. Above 10 T, the magnetization nearly saturates, and these saturation moments are roughly in accord with the estimated values (indicated by horizontal thick bars) under the following assumption: that the longitudinal component of the Mo spin moment is field independent (because of the small tilting angle) and that the Nd moment has strong Ising anisotropy with its effective moment and Ising axis being $g_{\rm eff} J \approx 2.3$ and the $\langle 111 \rangle$ direction, respectively. The smooth nature of the 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 figure 3(d), the Hall resistivity is plotted as a function of the magnetic field, which is applied along the [100] or [110] direction. For both field directions, the Hall resistivity decreases monotonically, and approaches zero. At the magnetic field of H_0 ($\approx$3 T), both the longitudinal and transverse components of the *averaged* Nd moment vanish. Then, the transverse component of the *averaged* Mo moment also vanishes. However, as noted above, each Nd moment does not vanish at all even at H_0, and hence each Mo moment does have a transverse component, namely, the local spin chirality. Therefore, a finite Hall resistivity appears at H_0. What should be compared with the experimental result is the *averaged* fictitious field generated from the local Mo spin chirality ($=\langle \vec{S}_1 \cdot \vec{S}_2 \times \vec{S}_3 \rangle$), and not the fictitious field calculated from such an *averaged* spin structure ($=\langle \vec{S}_1 \rangle \cdot \langle \vec{S}_2 \rangle \times \langle \vec{S}_3 \rangle$) [26] as determined from the neutron diffraction experiment. At high magnetic fields, the Mo spins are aligned along the field direction. Therefore, the tilting angle of the Mo spin becomes smaller and smaller as the field is increased. This reduction of the tilting angle results in a decrease of the Hall resistivity.

Figure 4(a) 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 4(b) shows a spin configuration where a



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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 requirement. 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 in Nd moment system, namely, the '3 in, 1 out' structure, where $\vec{b}_{\rm Mo} \cdot \vec{H}$ is positive. This means that the fictitious field operating on conduction electrons changes its sign, and hence the sign of the Hall resistivity will be reversed.

In figure 4(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 for this field direction also 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 is clearly larger than the expected value for the '2 in, 2 out' configuration. The magnetization process below about 10 T is gradual for this field direction also. Therefore, the spin configuration of the Mo tetrahedra is a random mixture of the '2 in, 2 out' and '3 in, 1 out' structures with the ratio changing as the field is increased[6], and finally becomes the '3 in, 1 out' configuration for all tetrahedra above 13–15 T.

The Hall resistivity for the applied field along [111] is displayed in figure 4(d). Clearly, it changes sign at 7.5 T. With the magnetic field increasing from zero, the number of tetrahedra with the '3 in, 1 out' structure increases. In the high field limit, all the tetrahedra adopt 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 it is parallel in the

[6] 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 [31, 32]. However, the presence of the Mo channel still obscures the transient feature even at 65 mK.

case of '3 in, 1 out', as mentioned above. Therefore, in the course of the field increasing from zero to a strong enough field to make all the tetrahedra adopt the '3 in, 1 out' configuration, the internal fictitious field cancels out, becoming zero in the bulk limit. This field corresponds to 7.5 T, where the Hall resistivity vanishes. With further increasing field, the tetrahedra with the '3 in, 1 out' configuration become dominant, and the fictitious field for the conduction electron changes sign. This results in sign reversal of the Hall resistivity, as observed.

In summary, we have clarified the unconventional behaviours of the anomalous Hall effect in the ferromagnet $Nd_2Mo_2O_7$ with the pyrochlore structure. The temperature dependence of the Hall conductivity of $Nd_2Mo_2O_7$ is totally different from the prediction of existing theories, but well understood in terms of the Berry phase theory. The magnetization curve shows little temperature dependence below 2 K and the values of the saturation moments are in accord with the assumption that the Nd moment has Ising anisotropy with the magnitude and Ising axis being $\approx 2.3\ \mu_B$ and along the $\langle 111 \rangle$ direction, respectively. The Hall resistivity changes sign when the field is applied along the [111] direction, but does not do this when the field is applied along the [100] or [110] direction. This fact is consistent with the prediction of the Berry phase theory, evidencing the spin chirality mechanism of the anomalous Hall effect for $Nd_2Mo_2O_7$ and related compounds.

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