Spontaneous Hall Effect in the Spin Liquid Phase of Pr$_2$Ir$_2$O$_7$

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Abstract. An electric current flowing through a conductor in a magnetic field produces a transverse voltage drop known as the Hall effect. In the absence of the field, this effect also appears in ferromagnets in a plane normal to its spontaneous magnetization vector owing to the spin-orbit coupling. Generally, it may also detect a nontrivial order parameter breaking the time-reversal symmetry on a macroscopic scale, for example, scalar spin chirality. Here, we present our recent results in the study of the frustrated magnetism and Hall transport of the metallic magnet Pr$_2$Ir$_2$O$_7$. Strikingly, a spontaneous Hall effect is observed in the absence of both an external magnetic field and conventional magnetic long-range order. This strongly suggests the existence of a chiral spin liquid, a spin-liquid phase breaking the time-reversal symmetry. Both our measurements indicate that spin-ice correlations in the liquid phase lead to a non-coplanar spin texture forming a uniform but hidden order parameter: the spin chirality.

1. Introduction

Time-reversal symmetry (TRS) is one of the basic concepts in the broad fields of science. In condensed-matter physics, many prototypical TRS-broken states can be found in magnetic materials where electron spins $S$ form various ordering patterns. Once the magnetic long-range order (LRO) sets in at low temperatures, the state loses the invariance under the time-reversal operation $S \rightarrow -S$. For magnetic materials, however, the source of the spontaneously broken TRS is not restricted to the LRO of spins. Since any product of odd number of spins is also odd under the time-reversal, it may in principle become a hidden but primary order parameter for the TRS breaking. The simplest one except the spin magnetic dipole moment is given by the scalar spin chirality [1], the solid angle subtended by three neighboring spins, $\kappa_{ijk} = S_i \cdot S_j \times S_k$. Despite extensive researches over decades, however, no clear answer has been made experimentally on the intriguing possibility of the TRS breaking in the absence of conventional magnetic LRO until quite recently [2].

When the TRS is broken macroscopically, the anomalous Hall effect (AHE) can be a useful probe for the detection of such hidden TRS breaking in metallic magnets. The AHE

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Figure 1. Two classes of the anomalous Hall effect in metallic magnets. (a) In a magnetic long range ordered state, the uniform spin magnetization $M_z$ and the fictitious magnetic field $b_z$ due to the spin-orbit interaction produce the anomalous Hall effect even at zero magnetic field $B_z = 0$. (b) In a spin-disordered state without uniform magnetization $M_z = 0$, the anomalous Hall effect may appear at $B_z = 0$ when the time-reversal symmetry is macroscopically broken with the finite fictitious magnetic field $b_z$ produced by the uniform spin chirality. is a fundamental transport phenomenon where the electric current generates the transverse voltage drop in the normal plane to the spontaneous magnetization $\mathbf{M}$ in ferromagnets [3] (Fig. 1a). This effect has attracted revived interest because of its topological and dissipationless character [4, 5, 6, 7, 8, 9, 10], and its potential application in spintronics [11]. In particular, it has been shown [8, 9, 10] that the intrinsic mechanism [4] of the AHE can capture the dominant part in moderately dirty metals. This intrinsic AHE can be understood in terms of the adiabatic motion of the Bloch electrons under the electric field $\mathbf{E}$ [6], which acquire a quantum geometrical Berry phase [12] in the wavevector ($\mathbf{k}$) space because of the spin-orbit interaction and the spin magnetization. This phase acts as a fictitious magnetic field $\mathbf{b}_k$ in the $\mathbf{k}$ space and bends the orbital motion of electrons as in the case of the Lorentz force due to the real magnetic field $\mathbf{B}$. Thus, it causes the AHE characterized by the finite Hall conductivity $\sigma_H$ at $B = 0$.

The condition for observing the AHE at $B = 0$ is the macroscopically broken TRS [13], which ensures a nonzero average $\mathbf{b}$ of the Berry phase over the occupied Bloch states. It does not necessarily require a finite ferromagnetic (FM) spin alignment (Fig. 1a), but a noncoplanar spatial distribution of spins. While unconventional scenarios directly relying on a noncoplanar spin texture with the uniform scalar spin chirality have been addressed experimentally so far for ferromagnets and for spin glasses [3, 14, 15, 16, 17], the macroscopically broken TRS and a resultant nonzero $\sigma_H$ in the absence of a uniform spin magnetization have also been proposed theoretically; for example, in antiferromagnetic (AF) states and spin-liquid states with the scalar spin chirality [18] (Fig. 1b). In this exotic example of a chiral spin-liquid [1], the uniform scalar spin chirality shows the LRO, but the spin magnetic moment does not. Experimentally, however, the AHE at zero field has never been observed in the absence of the uniform spin magnetization associated with the ferromagnetism [3] or the spin freezing [19, 15].

Here, we discuss our recent results in the study of the frustrated magnetism and Hall transport of the metallic magnet Pr$_2$Ir$_2$O$_7$, which has revealed a macroscopically TRS-broken spin-liquid
state [2]. The spontaneous Hall effect is found in the geometrically frustrated Kondo lattice Pr$_2$Ir$_2$O$_7$ [20] even above its spin freezing temperature $T_f$ $\sim$ 0.3 K. A clear hysteresis is observed in the Hall conductivity around zero field below the onset temperature $\theta$ $\sim$ 1.5 K, whereas that in the magnetization curve appears only below $T_f$ within an experimental accuracy. Namely, a large anomalous Hall conductivity $\sigma_{\text{H}}$ is found even at zero field where the magnetization practically vanishes, in sharp contrast to the conventional AHE in ferromagnets. This indicates an emergence of a hidden order at $T = \theta$ that macroscopically breaks the TRS without invoking a magnetic LRO. The phenomenon may be understood in terms of a formation of the uniform spin chirality out of “2-in, 2-out” configurations of localized magnetic moments of Pr$^{3+}$ ions in an analogy to spin-ice systems [21].

2. Experimental

Single crystals of Pr$_2$Ir$_2$O$_7$ were grown by a flux method [20, 22]. Energy dispersive x-ray analysis found that the ratio between Pr and Ir is 1:1.03 and stoichiometric within experimental errors. The four-circle x-ray diffraction measurements confirmed the pyrochlore lattice structure of the crystals. The transport properties and magnetization were measured at low temperatures down to 0.03 K and under magnetic fields up to 7 T.

The field sweep measurements were performed with a small fixed rate of 1 Oe/s at each temperature $T \leq 0.5$ K, while at $T \geq 0.7$ K the field was fixed at each point for $\sim$ 2 min. before the measurement. We also performed the measurements by using both procedures above at 0.5 K and confirmed that basically the same results can be obtained by the different field sweep methods. The field dependence was measured at each temperature after the same zero field cooling procedure. Namely, we first increased the temperature above 2 K before each measurement, and then cooled down the sample to a target temperature under zero field.

The transport measurements were made by using a conventional four-probe method with a current path along the [110] direction. For the transverse Hall resistivity measurements, the longitudinal voltage drop was eliminated by reversing the field direction. For the field sweep measurement in Fig. 2c, the field was first increased from zero up to 7 T, and then a hysteresis loop was measured between 7 T and −7 T. Only the results in the positive field region are presented in Fig. 2c. The virgin curve between 0 T and 7 T is not shown because of the uncertainty in determining the longitudinal voltage drop.

The magnetization above and below 2 K was measured by the conventional SQUID magnetometer and by the Faraday method using a dilution refrigerator [23], respectively. The nonlinear susceptibility was obtained by fitting $M$ vs. $B$ curves to $M = \chi_1 B + \chi_3 B^3/3!$ at each temperature. For the analysis of the results at $T \leq T_f$, we added a constant term $M_0$ for the frozen component. The ac susceptibility measurements were performed by using the mutual inductance method with ac fields of $B_{ac} = 5 \mu$T with frequency $f = 16$ Hz.

3. Experimental Results and Discussion

3.1. Spin Ice Correlation in the Spin Liquid Phase

The pyrochlore iridate Pr$_2$Ir$_2$O$_7$ is a cubic system consisting of Pr and Ir corner-sharing tetrahedra that are displaced by half the lattice constant. The Pr$^{3+}$ ions provide localized (111) Ising-like 4f magnetic moments pointing inwards or outwards to the center of the Pr tetrahedron (Fig. 2a) with the effective size of $p_{\text{eff}}$ $\sim$ 3.0 $\mu$B/Pr, while Ir 5d conduction electrons are weakly correlated and Pauli-paramagnetic with an appreciable spin-orbit coupling [20]. The localized moments interact with the conduction electrons through the AF Kondo coupling $J_{fd}$. Strong geometrical frustration [24] is apparent as the magnetic moments do not exhibit any LRO, but only a freezing behavior below $T_f$ $\sim$ 0.3 K, two orders of magnitude lower than the AF Weiss temperature $T^* \sim -20$ K [20].
Figure 2. Magnetic and crystal structure, and field dependence of the magnetization and Hall conductivity of Pr$_2$Ir$_2$O$_7$ along the [111] field direction [2]. (a) The pyrochlore lattice is alternating stacking of kagome and triangular layers along the [111] direction. Under zero field, the Pr moments most likely form the “2-in, 2-out” configuration as denoted by three red arrows and one blue arrow in each tetrahedron. Application of the field $B$ along the [111] direction flips the blue moments and stabilizes the “3-in, 1-out” (1-in, 3-out) configuration formed by four red arrows. (b) Field dependence of the magnetization $M$ at 0.06 and 0.5 K (left axis) and its derivative $dM/dB$ at 0.06 K (right axis). (c) Field dependence of the Hall conductivity $\sigma_H$ at 0.06 K. Dashed line represents the metamagnetic transition field $B_c \sim 2.3$ T. Inset: Low field (b) $M$ and (c) $\sigma_H$ at fixed temperatures. During the measurements, the field was continuously swept with the rate of 1 Oe/s at temperatures $T \leq 0.5$ K, while it was fixed at each measurement field point at $T \geq 0.7$ K. The arrows indicate up and down field sweep sequences. (d) Field dependence of the magnetization $M(B)$ for fields along the [100], [110], and [111] directions at 0.1 K. Inset: Hysteresis in the magnetization $M(B)$ at the metamagnetic transition for fields along the [111] direction at 0.03 K.
Before discussing the results of the Hall transport measurements, we clarify low-temperature magnetic properties of Pr$_2$Ir$_2$O$_7$. First, we have performed the field-sweep measurements of the magnetization under the field along the [111] direction. Figure 2b represents the magnetization curve $M(B)$ obtained at 0.06 K and 0.5 K. In contrast with $M(B)$ at 0.5 K, a small but clear step-like feature is found at 0.06 K. This anomaly is also clearly observed as a kink in the field derivative (Fig. 2b). On further cooling down to 0.03 K, around the step-like feature there appears a hysteresis between the up and down sweep sequences, indicating a first-order metamagnetic transition (Fig. 2d inset). We also performed similar measurements under fields along [100] and [110] directions. However, we observed neither an anomaly nor a hysteresis, but a smooth increase of the magnetization (Fig. 2d).

The fact that the metamagnetic transition occurs only for $B \parallel [111]$ provides an evidence for the formation of the “2-in, 2-out” configurations of (111) Ising moments on each tetrahedron. In general, four Ising moments on a tetrahedron form two distinct configurations, depending on the sign of the nearest-neighbor interaction: “all-in, all-out” and “2-in, 2-out” for AF and FM interactions, respectively. The “all-in, all-out” state has locally $M = 0$. Hence, to induce a finite $M$, a metamagnetic transition is expected for fields along all three directions. However, this is not the case in our experiment. In contrast, for “2-in, 2-out”, the transition occurs only along the [111] direction as observed in our case as well as in the spin-ice compound Dy$_2$Ti$_2$O$_7$ [25], and stabilizes the “3-in, 1-out” state in high fields (Fig. 2a).

Further consistency with the “2-in 2-out” correlation can be found for the values of the magnetization $M_c$ and the magnetic field $B_c$ at the metamagnetic transition. In spin-ice systems, the magnetization just below the metamagnetic transition is known to reach $M_c = g_J J/3$ in the kagome-ice state, which can be viewed as the stacking sequence of the “2-in, 2-out” kagome-lattice layer and the fully polarized triangular-lattice layer along the [111] direction [21] (Fig. 2a). Using the Landé factor $g_J$ for Pr$^{3+}$ and $J$ obtained from $p_{\text{eff}} = g_J \sqrt{J(J+1)}$, $M_c$ is estimated to be 0.9 $\mu_B$/Pr, which is close to our observation of $M_c \sim 0.8 \mu_B$/Pr. On the other hand, the effective nearest-neighbor FM coupling $J_{\text{ff}}^{\text{eff}}$ in the spin-ice systems can be estimated from $B_c$ as $J_{\text{ff}}^{\text{eff}} \approx (1/3) g_J J \mu_B B_c / k_B$. In our case, the local “2-in 2-out” correlation inferred from the observed magnetization curves also indicates a FM coupling between nearest-neighbor 4f moment pairs. The observed $B_c = 2.3$ T indicates $J_{\text{ff}}^{\text{eff}} \sim 1.4$ K which is close to the peak temperature ($\sim 2$ K) of the magnetic specific heat $C_\text{m}$ [20] as in spin-ice systems [21]. Furthermore, the nonlinear magnetic susceptibility $\chi_3$ exhibits a steep negative increase below $\theta$ and saturate to a large negative value (not shown) [2], consistent with the FM correlation among 4f magnetic moments. Therefore, we conclude that at $T \leq J_{\text{ff}}^{\text{eff}}$, the “2-in, 2-out” configuration appears with the highest probability. However, there is a sharp contrast to the spin-ice case where the spin freezing occurs at the same temperature scale, i.e., $T_1 \sim J_{\text{ff}}^{\text{eff}}$ [21], while the present material remains in a spin-liquid state down to $T_1 \sim 0.3$ K even below the temperature $\theta \sim J_{\text{ff}}^{\text{eff}}$.

Two possible sources of the FM interaction of the order of $J_{\text{ff}}^{\text{eff}} \sim 1.4$ K can be considered. Here, the magnetic dipole interaction cannot be the origin because it is an order of magnitude smaller for Pr$^{3+}$ ($\sim 0.1$ K) than for Dy$^{3+}$ ($\sim 1$ K) of the dipolar spin-ice system Dy$_2$Ti$_2$O$_7$ [21]. Instead, one possibility is the superexchange coupling between Pr 4f moments through the superexchange path mediated by 2$p$ orbitals at the oxygen O(1) site [26]. The significant deviation from 180$^\circ$ of the bond angle for the Pr-O-Pr path alters the sign of the superexchange and should make it ferromagnetic. Another possibility is the RKKY interaction mediated by conduction electrons via the Kondo coupling. Indeed, the RKKY interaction should be FM between the nearest-neighbor pairs, according to the analyses based on the simple Fermi gas model using the carrier concentration $\sim 20$ %/Pr found by the Hall effect measurements [27]. A first-principles calculation also predicts a single Fermi surface with a carrier concentration $\sim 20$ %/Pr [2]. A recent Monte-Carlo simulation [28] has indicated that this RKKY interaction with
Figure 3. (a) Temperature dependence of the Hall conductivity $\sigma_H$ (left axis) and the dc susceptibility $\chi$ (right axis) under a magnetic field of $B = 0.05$ T along the [111] direction [2]. Both the zero-field cooled (ZFC) and field-cooled (FC) results are plotted. Vertical dashed lines denote $\theta \sim 1.5$ K and $T_f \sim 0.3$ K, respectively. (b) Temperature dependence of the remnant Hall conductivity $\sigma_H(B = 0)$ (left axis) and remnant magnetization $M(B = 0)$ (right axis) at zero field, obtained after a field sweep down from 7 T in the hysteresis loop measurements [2]. Inset: Temperature dependence of the ac-susceptibility obtained under zero field with the ac-field of $5\mu$T with frequency $f = 16$ Hz. The peak indicates (possibly partial) freezing of Pr moments below $T_f \sim 0.3$ K [20]. (c) Temperature dependence of the longitudinal conductivity $\sigma$ under $B = 0.05$ T along the [111] direction. No hysteresis is found between the results obtained in the ZFC and FC sequences.
the experimentally obtained $k_F$ also explains the AF Weiss temperature in agreement with our experiment, while it predicts a magnetic dipole order for the ground state.

3.2. Spontaneous Hall Effect in the Spin Liquid Phase
Now, we present the experimental findings on the AHE and the associated macroscopic TRS breaking [2]. Figure 3a shows the temperature dependence of the Hall conductivity $\sigma_H(T) = -\rho_H J / (\rho_T^2 + \rho_{2H}^2)$ (left axis), and the dc susceptibility $\chi(T) = M / H$ (right axis) at $B = 0.05$ T along the [111] direction. Below 2 K, $\sigma_H(T)$ exhibits a strong temperature dependence and even bifurcation at $\theta \sim 1.5$ K between the zero-field cooled (ZFC) and field-cooled (FC) results, while the longitudinal conductivity $\sigma(T)$ shows no hysteresis (Fig. 2b). On the other hand, the onset temperature of the irreversibility in $\chi(T)$, $T_i \sim 0.3$ K (Fig. 3a, Fig. 3b and its inset), is significantly lower than that in $\sigma_H(T)$, $\theta \sim 1.5$ K. This hysteresis in $\sigma_H(T)$ suggests that the TRS is already broken macroscopically below $\theta$ in the spin-liquid state.

To confirm this possibility, we have performed the field-sweep measurements of $\sigma_H(B)$ with $B \parallel [111]$ at various temperatures. First, let us discuss the results obtained in the spin-frozen state below $T_i$. Figure 2c presents the results at $T = 0.06$ K. Apparently, we do not find any field range where $\sigma_H(B)$ is proportional to $M(B)$. Instead, $\sigma_H(B)$ shows the following nonmonotonous field dependence: (i) a pronounced hysteresis loop around $B = 0$ between the up-sweep $\sigma_H^{\uparrow}(B)$ and the down-sweep $\sigma_H^{\downarrow}(B)$, (ii) a peak formation at $B_p \sim 0.5$ T, (iii) a kink around the metamagnetic transition at $B = B_c \sim 2.3$ T, (iv) a sign change around 5 T. The remnant Hall conductivity $\sigma_H(B = 0)$ amounts to 10 $\Omega^{-1}$ $cm^{-1}$, which is comparable to that for the ferromagnet Nd$_2$Mo$_2$O$_7$ [14, 29]. The size of the hysteresis, i.e., $\sigma_H^{\uparrow}(B) - \sigma_H^{\downarrow}(B)$, decreases with field and vanishes at $B \sim B_c$. In contrast with the clear hysteresis observed in $\sigma_H(B)$, the magnetization $M(B)$ shows only a small remnant component $\sim 0.01\mu_B$ due to the spin freezing (Fig. 2b, inset).

Notably, $\sigma_H(B = 0)$ does not vanish even above $T_i \sim 0.3$ K. Although it is weakened with temperature, we still observe a hysteresis in $\sigma_H(B)$ around $B = 0$ at 0.5 and 0.7 K (Fig. 2b, inset), while that in $M(B)$ is already absent within the experimental accuracy $\sim 10^{-3}\mu_B$ (Fig. 2b, inset). Thus, $\sigma_H(T,B = 0)$ decreases on heating, and it eventually vanishes at $\theta \sim 1.5$ K, which can be clearly distinguished from $T_i$ where $M(T,B = 0)$ decreases to zero (Fig. 3b). The finite $\sigma_H$ observed in the apparent absence of $B$ and $M$ indicates that the TRS is broken spontaneously and macroscopically in the spin-liquid state, pointing to an LRO or freezing of higher degrees of freedom than spin dipole moments, for instance, the net spin chirality. There exists a close link between the macroscopic TRS-breaking and the local “2-in, 2-out” spin correlation. The onset temperature $\theta \sim 1.5$ K almost coincides with the effective FM coupling $J_{ff}^{\parallel} \sim 1.4$ K estimated from the metamagnetic transition field $B_c$. Besides, the hysteresis observed in $\sigma_H$ as a function of field disappears at $B_c$ where a large portion of “2-in, 2-out” configurations are transformed into “3-in, 1-out” (Fig. 2b and c), as we have already explained. Therefore, the macroscopically TRS-broken spin-liquid state found in $T_i \leq T \leq \theta$ should comprise the “2-in, 2-out” configurations having a net spin chirality.

3.3. Possibility of Quantum Melting of Spin Ice
In Pr$_2$Ir$_2$O$_7$, most likely the spin-ice states with a uniform chirality component become dynamic at $T \geq T_i$, forming a chiral spin-liquid that does not show a freezing of dipole moments. With this regard, the recent theory by Onoda and Tanaka has concluded an interesting possibility of quantum melting of spin ice. They focus on the superexchange path made of Pr-O-Pr bond and found that it can be represented by an effective quantum pseudospin-1/2 model [26, 30]. They found that the combination of the small size of Pr 4f moments and the trigonal crystal electrical field at Pr site enhances the quantum terms to be as large as the classical Ising term in the Hamiltonian. Using the calculation for 16 sites, they further predict that each Pr tetrahedron
may develop a spin quadrupole and spin chirality correlation at the ground state, as the results of the superposition of the “3-in, 1-out” configurations in the dominant “2-in, 2-out” states. The experimental determination of the profile of the magnetic correlations in the spin-liquid phase for $T_f \leq T \leq T_H$ is an important task for future studies.

4. Conclusion
The above results have demonstrated that a macroscopic breaking of the time reversal symmetry (TRS) can indeed occur even without a conventional magnetic order, and can be detected through a spontaneous Hall effect in the absence of external field and uniform magnetization, suggesting an unconventional anomalous Hall effect without the ferromagnetic or ferrimagnetic spin ordering. With this regard, the ac version of the anomalous Hall effect, i.e., a magneto-optical Kerr effect at zero field, may offer another useful probe of a macroscopically TRS-broken state, as actually reported in the pseudogap region of a high-temperature cuprate superconductor [31]. Future explorations searching for the spontaneous Hall effect might unveil a fertile field of exotic chiral states of matter in strongly correlated electrons.

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[13] Landau L D and Lifshitz E M 1984 Electrodynamics of Continuous Media 2nd ed (Oxford: Elsevier) the macroscopically broken $T$ means that the time-reversal operation, which inverts the spin and orbital angular momenta and the wavevector, $S \rightarrow -S$, $L \rightarrow -L$, and $k \rightarrow -k$, as well as the fictitious magnetic field $b_{\ast k} \rightarrow -b_{\ast -k}$, should not be compensated by any other symmetry operations of the crystal, e.g., translation, spatial inversion, reflection, rotation, and their combinations.
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