Anomalous Magnetothermopower in a Metallic Frustrated Antiferromagnet

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We report the temperature T and magnetic field H dependence of the thermopower S of an itinerant triangular antiferromagnet $PdCrO_2$ in high magnetic fields up to 32 T. In the paramagnetic phase, the zero-field thermopower is positive with a value typical of good metals with a high carrier density. In marked contrast to typical metals, however, S decreases rapidly with increasing magnetic field, approaching zero at the maximum field scale for $T > 70$ K. We argue here that this profound change in the thermoelectric response derives from the strong interaction of the $4d$ correlated electrons of the Pd ions with the short-range spin correlations of the Cr^{3+} spins that persist beyond the Néel ordering temperature due to the combined effects of geometrical frustration and low dimensionality.

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The interplay between itinerant electrons and even simple magnetic structures can lead to spectacular effects, the giant magnetoresistance seen in magnetic multilayers being arguably the most prominent example [1]. In geometrically frustrated magnets, complex spin textures that couple to the conduction electrons create an altogether different landscape, where short-range correlations are expected to play a major role. Moreover, since magnetism in metals can be destabilized much more readily than in insulators, magnetic frustration in metallic systems offers a rich playground to search for the emergence of novel transport phenomena. Notable recent examples include the unconventional anomalous Hall effect (AHE) observed in magnetic pyrochlores [2,3] and the suppression of thermopower in a longitudinal magnetic field in the layered Curie-Weiss metal Na_xCoO_2 [4].

Despite their obvious potential for new physics, metallic frustrated magnets have been noticeably less studied than their insulating counterparts, largely due to the fact that such materials are rare. Of particular interest are materials in which the conduction electrons and magnetic moments arise from different subsystems. In this context, the quasi-two-dimensional (quasi-2D) antiferromagnet $PdCrO_2$ [4–6] is somewhat unique. $PdCrO_2$ has a delafossite crystal structure with layers of Pd ions arranged in a triangular lattice stacked between magnetic edge-sharing CrO_6 octahedra. The latter contains Cr^{3+} ions with localized (Mott insulating) 3/2 spins which order in the 120° antiferromagnetic (AFM) structure below $T_N = 37.5$ K [7,8]. The frustration parameter f, defined as an absolute ratio of the Weiss temperature Θ_W and the ordering temperature T_N, is around 13 for $PdCrO_2$, indicating a high level of frustration [7–9]. According to band structure calculations [10], angle-resolved photoemission [10], and quantum oscillation (QO) studies [11,12], the Fermi surface (FS) of $PdCrO_2$, in the paramagnetic (PM) phase, is identical to that of the nonmagnetic analog $PdCoO_2$, and thus is derived uniquely from the $4d$ electrons on the Pd site. $PdCrO_2$ also draws special interest because it too exhibits an unconventional AHE, i.e., one that does *not* scale with its magnetization [5,11].

Here, we report the discovery of a new feature in the transport properties of $PdCrO_2$, namely, a strong magneto-thermopower (MTP) at elevated temperatures. In a transverse field, S exhibits a marked decrease which for $T > 70$ K, reaches 100% of the zero-field value at $\mu_0 H = 30$ T. The suppression is reminiscent of that first reported in Na_xCoO_2 ($x = 0.7$) [4] and attributed to a lifting of the spin degeneracy of the large spin entropy of the mobile Co^{4+} spins that gives rise to its enhanced thermopower [13]. We argue here, however, that the suppression of $S(B)$ in $PdCrO_2$ is distinct from that observed in Na_xCoO_2 and signifies instead a novel magnon drag contribution to the thermopower that persists far beyond T_N due to the highly frustrated short-range spin correlations on the Cr sublattice.

Single crystals of $PdCrO_2$ of typical dimensions $1 \times 0.4 \times 0.2$ mm^3 were grown by a flux method, as described in Refs. [11,14]. Details of our thermoelectric measurements can be found in the Supplemental Material (SM) [15]. In all measurements reported here, the magnetic field is oriented perpendicular to the thermal gradient and to the highly conducting planes.

The key finding of our study is the effect of a magnetic field on the thermoelectric response of $PdCrO_2$. This is summarized in Fig. 1 where the in-plane thermopower $S_{ab}(T)$ in zero field (solids black squares) is compared with

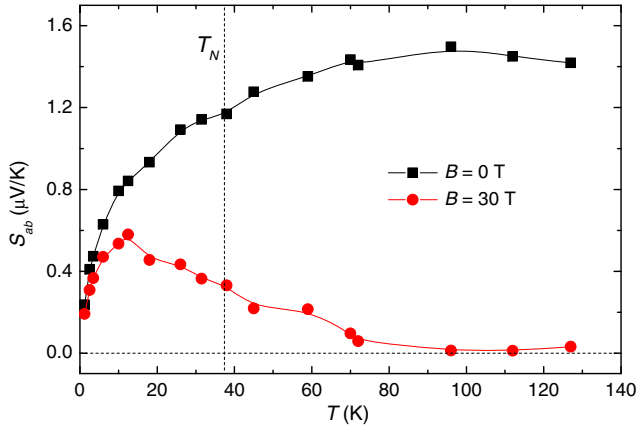

FIG. 1. Temperature dependence of the in-plane thermopower S_{ab} of $PdCrO_2$ at zero and high magnetic field $B \| c$ shows an almost complete field suppression for $T > 70$ K. The magnetic ordering transition at $T = T_N$ is indicated by a vertical dashed line.

that obtained in an applied field of 30 T (solid red circles) (see Supplemental Material [15] for $S_{ab}(T)$ data at intermediate fields [15]). In zero field, S_{ab} has a small positive value less than 2 μV/K—typical of good metals—while the sign and order of magnitude of S_{ab} at $T = 130$ K are similar to those found in $PdCoO_2$ [26]. In a large magnetic field, as summarized in Fig. 1, the thermopower is almost completely suppressed for $T > 70$ K. Lowering the temperature towards the magnetically ordered phase reduces the magnitude of the field-suppression of S_{ab}.

Such a large suppression of S is not expected in a conventional metal, where the field has a negligible effect on the relative spin-up and spin-down populations of electrons and their entropic current [27]. In strongly correlated electron systems, however, the spin degrees of freedom can give a large contribution to S according to Heikes formula $S = \mu/eT = S_E/e$, where μ is the chemical potential, and S_E is the entropy per charge carrier [28]. Since S_E depends on the spin and configuration degeneracies, the spin entropy term can raise S to order k_B/e and subsequently be suppressed to zero in a magnetic field by a lifting of the spin degeneracy. Such an effect was observed first in Na_xCoO_2 [4] where the field dependence of $S(B)$ for different T was found to be consistent with the variation of the residual spin entropy $S_E(B, T)$ for noninteracting spins in a magnetic field.

The field dependence of $S_{ab}(B)$ in $PdCrO_2$, normalized to its zero-field value, is shown in Fig. 2(a) for constant temperature field sweeps over a range of temperatures 1.2 K $\leq T \leq$ 130 K. As shown in the inset of Fig. 2(b), the form of the field suppression is qualitatively similar to that found in Na_xCoO_2 [4]. Indeed, since both transition-metal oxides form a layered triangular lattice of localized, but frustrated magnetic moments which interact with the conducting d electrons, it might be tempting to assign

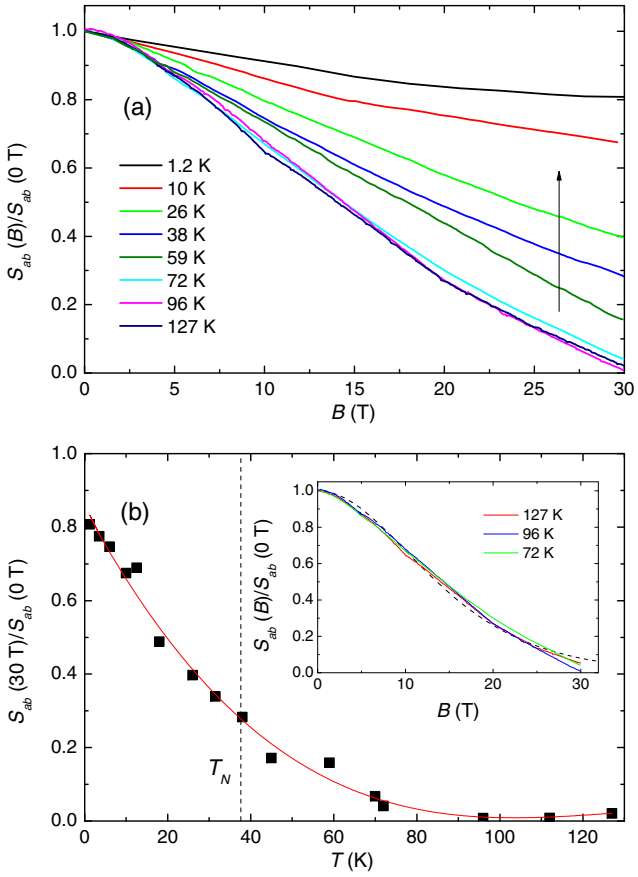

FIG. 2. (a) Magnetic field dependence of S_{ab} as a function of temperature. The suppression of the normalized thermopower by an out-of-plane magnetic field decreases with lowering temperature, as indicated by the arrow. (b) T dependence of the normalized field change of S_{ab} in an applied field of 30 T. Line is a guide to the eye. The inset shows the comparison of a modeled spin entropy in field from Ref. [4] (dashed curve) and the normalized $S_{ab}(B)$ for $T > 30$ K. While the form of the suppression is consistent with the model, its magnitude clearly does not scale with $\mu_0 H/k_B T$.

the same origin to the field suppression of $S(B)$ in both cases. Closer inspection, however, reveals some important differences between the compounds and their thermoelectric response that suggest otherwise.

First, in Na_xCoO_2 ($x = 2/3$), one third of the Co ions are in a Co^{4+} configuration, giving rise to a band of mobile but AFM-coupled charges (with spin $s = 1/2$) moving through a magnetically inert background of $s = 0$ moments localized on the Co^{3+} sites. Wang *et al.* showed that the spin entropy term associated with these mobile spin excitations accounts for almost all of S at 2 K and a dominant fraction at 300 K [4]. In $PdCrO_2$, on the other hand, the sea of conduction electrons is comprised uniquely from the $4d/5s$ states of the Pd ions, as shown convincingly in recent QO studies [12], while the Cr^{3+} states are (Mott) insulating and therefore cannot, by themselves, respond to a thermal gradient. Thus, while there is significant spin

entropy in the system, it does not contribute to the thermopower of $PdCrO_2$, and correspondingly, there is no enhancement in S_{ab}.

Second, the temperature evolution of the suppression is markedly different in the two compounds. In Na_xCoO_2, the relative suppression of S in field grows with decreasing T as the contribution of the spin entropy term becomes ever more dominant. In $PdCrO_2$, by contrast, the field suppression becomes more pronounced with *increasing* temperature. Hence, the H/T scaling observed in Na_xCoO_2 [4] fails in $PdCrO_2$ [see inset to Fig. 2(b)]. As illustrated in the main panel of Fig. 2(b), where the ratio $S(B = 30\ \mathrm{T})/S(B = 0\ \mathrm{T})$ is plotted, a total suppression of S_{ab} is only observed for $T \geq 2T_N$. Below this temperature scale, $S(30\ \mathrm{T})$ remains finite and grows in magnitude with decreasing temperature.

Large magnetothermopower is often observed when the zero-field thermopower itself has an enhanced value, e.g., as found in semiconductors, and is commonly attributed to the effects of a magnetic field on the respective mobilities of the electron and hole carriers [29]. A sizeable MTP is also found in systems exhibiting a large magnetoresistance [30]. Neither of these scenarios apply to $PdCrO_2$. As mentioned above, the zero-field thermoelectric response in $PdCrO_2$ is that of a single-band metal with a high carrier density. Moreover, at these elevated temperatures where the suppression of the thermopower is most complete, the in-plane magnetoresistance of $PdCrO_2$ is very small, of order 5% or less in a field of 30 T [15]. Thus, it would appear that the large MTP in $PdCrO_2$ stems from a different origin. Before discussing this in more detail, however, we first complete the summary of our experimental findings, some of which have an important bearing on this discussion.

Figure 3 shows the variation of S_{ab}/T in $PdCrO_2$ between 1 K and 130 K, both in 0 T and 30 T, on a semilogarithmic scale. Over the entire temperature range, S_{ab}/T in zero field increases with decreasing temperature. Above T_N, S_{ab}/T appears to exhibit a logarithmic enhancement which we attribute to the scattering of electrons on short-range magnetic correlations in the PM phase that are also presumed to be responsible for changes in the zero-field [8] or low-field [31] transport properties. The established FS topology of $PdCrO_2$ makes the electron states highly sensitive to scattering processes, either from magnons or spin fluctuations, associated with the AFM wave vector Q (illustrated in the right inset of Fig. 3) [32]. In an isotropic antiferromagnet, the scattering probability W_q with momentum-transfer q, while vanishing for $q \to 0$, is formally divergent for $q \to Q$, $W_q \propto 1/\omega_q \propto |q - Q|^{-1}$ [33]. Above a threshold temperature $T^* \sim 20$–50 K (see Ref. [15] and below for a fuller description), "singular" scattering off such FS hot spots can lead to a contribution to $S \propto \ln T^*/T$ [15], qualitatively consistent with what is found here in zero field above T_N (Fig. 3).

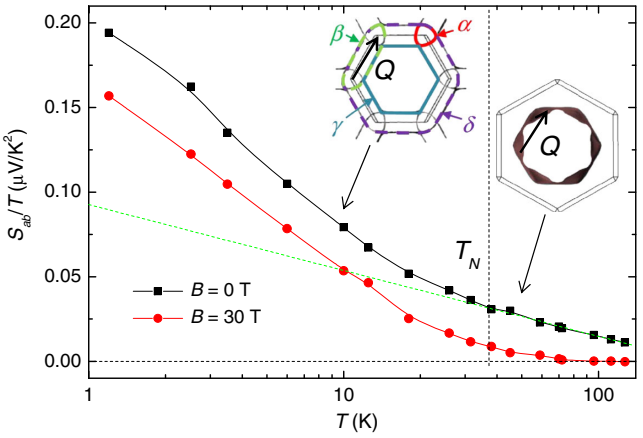

FIG. 3. S_{ab}/T as a function of temperature, plotted on a semilogarithmic scale, in zero field and at $B = 30$ T. The dashed line is a logarithmic fit to S_{ab}/T above T_N that is associated with the scattering of itinerant electrons on spin fluctuations with a wave vector Q [34], as indicated in the Fermi surface model shown in the right inset [32]. Below T_N, the enhancement is attributed to a dominant contribution from the α pocket in the reconstructed zone, labeled in the left inset [11].

At lower temperatures, where the Cr^{3+} spins order, S_{ab}/T increases more rapidly as the conduction electrons finally undergo FS reconstruction [11,12] (note the upward deviation from the dashed line in Fig. 3 below T_N). The existence of small pockets is confirmed by the observation of QO in the high-field thermopower below T_N, shown in Fig. 4. Both the QO frequency $F = 710$ T and the obtained mass $m^* = 0.31(3)\ m_e$ [15] agree with those previously attributed to the small α pocket in the reconstructed zone [11,12]. Assuming that the α pocket is the dominant contribution to S_{ab} in the low-T limit (given that it has the smallest $T_F = 3000$ K), we use a simple Drude model to estimate $S_{ab}/T = 140$ nV/K, comparable to the measured

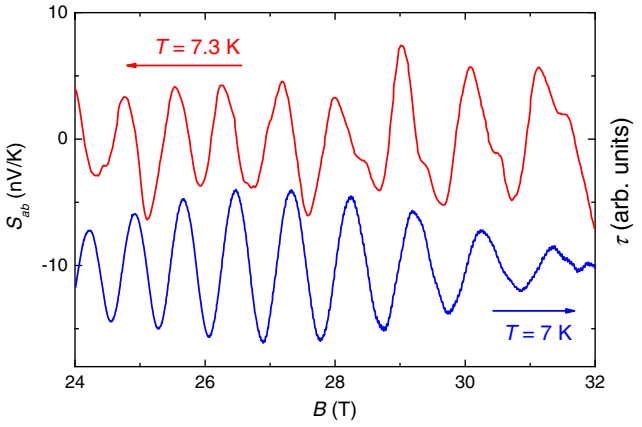

FIG. 4. Slow quantum oscillations in S_{ab} (obtained by subtracting a polynomial background) and in magnetic torque (obtained previously [11]) originating from a small electron pocket in the reconstructed Fermi surface.

value at 1.2 K of 200 nV/K. Thus, we can attribute the additional enhancement of S_{ab}/T below T_N to FS reconstruction. In contrast to the thermoelectric response of other systems (such as the parent pnictide $BaFe_2As_2$ [35]) that undergo a magnetic zone folding, the change in S/T here is very gradual.

Let us now turn to discuss the origin of the unusual field dependence of S_{ab} in $PdCrO_2$. The observed FS reconstruction indicates strong coupling between the itinerant electrons and the local moments on the Cr^{3+} sublattice. According to specific heat and susceptibility data [8], there is a broad region in temperature above T_N, extending up to 150 K (i.e., $4T_N$), in which short-range correlations among the frustrated spins persist. This is also confirmed by the observation of diffuse magnetic scattering in Refs. [7,8,32], discussed in more detail in the Supplemental Material [15]. Such an extended range of critical behavior is a feature of quasi-2D triangular [36–38] and kagome [39] lattice magnets.

Short-range magnetic order in the PM phase, with a correlation length $\xi \gg a$, the lattice parameter, can in principle persist up to a temperature scale of the AFM exchange energy, i.e., $T \sim J \gg T_N$. As was shown in Ref. [40], the character of electron-magnon interactions does not change markedly at T_N assuming that $\xi \gg k_F^{-1}$ which for metals is the same as $T < J$. Below a threshold temperature $T^* \sim (\Delta/E_F)J \sim 20$–$50$ K, where Δ is the AFM gap and E_F the Fermi energy, the thermopower will be dominated by the usual diffusive term that is weakly dependent on magnetic field. Above T^*, however, the strong electron-magnon interaction with $q \to Q$ will become relevant [33], giving rise to a magnon-drag contribution S_g to the total thermopower, which can become significant provided that the magnon-electron scattering is comparable to the scattering of magnons by defects, which seems a reasonable assumption in a clean, stoichiometric $PdCrO_2$. The magnon drag term will be strongly dependent on the spin-wave spectrum which is also very sensitive to magnetic field [41]. According to the theory described in Ref. [42], AFM magnons become unstable with respect to two-magnon decay processes at high enough magnetic fields. This effect leads to a strong suppression of magnon drag and its contribution to the thermoelectric power which explains qualitatively the dramatic growth of the MTP at higher temperatures. This picture should be contrasted with the typical magnon-drag scenario proposed in AFM and FM metals where a strong MTP is only seen *below* the magnetic transition and is absent above it [43–46]. The persistence of such a term in $PdCrO_2$ above T_N is then a direct consequence of the highly frustrated nature of the magnetic order arising from the combined effects of geometrical frustration and low dimensionality.

Another interesting possibility for the large field suppression of S at elevated temperatures is a reduction of the scattering amplitudes related to the interaction between the itinerant electrons and the emergent spin chirality. At temperatures above T_N, the fluctuating Cr spins are easily aligned in a field, forming new spin textures which give rise, owing to their triangular arrangement, to a finite scalar spin chirality. Indeed, analysis of electron spin resonance experiments in $PdCrO_2$ have shown evidence for spin relaxation processes involving Z_2 vortices associated with chiral fluctuations of the 120° spin structure extending up to 300 K [47]. Moreover, the unconventional AHE observed in $PdCrO_2$ above T_N has been attributed to a strong coupling of the itinerant electrons to the emergent field-induced spin chirality [11]. The evolution of the field-induced suppression of the thermopower in $PdCrO_2$ suggests that both phenomena may be linked to the same physics, since below T_N, where the 120° spin structure becomes increasingly more resilient to an applied field, both the MTP and AHE are correspondingly reduced. The form of the in-plane resistivity in $PdCrO_2$ above T_N has also been attributed to magnetic scattering off short-range magnetic correlations among the frustrated spins [8]. It is therefore tempting to attribute the suppression of $S_{ab}(B)$ to a similar effect. For the effect to grow with increasing temperature, however, either the coupling of the conduction electrons to the underlying spin textures would have to become stronger, or the induced chirality more pronounced as T increases.

In summary, we have uncovered a marked suppression of the in-plane thermopower of the metallic frustrated antiferromagnet $PdCrO_2$ in high magnetic fields up to 32 T. Certain features of the thermoelectric response suggest that this suppression is in the metallic, rather than in the spin entropic contribution to S_{ab}. The temperature evolution of the suppression, in particular, implies a dominant magnon-drag contribution that persists far beyond T_N due to the thermally-robust interaction between the conduction electrons and the short-range magnetic correlations.

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