Anomalous thermoelectric effects and quantum oscillations in the kagome metal CsV₃Sb₅

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The kagome metal compounds AV₃Sb₅ (A = K, Rb, and Cs) feature a wealth of phenomena including non-trivial band topology, charge density wave (CDW), and superconductivity. One intriguing property is the time-reversal symmetry breaking in the CDW state without local moments, which leads to anomalous transport responses. Here, we report the investigation of magneto-thermoelectric effects on high-quality CsV₃Sb₅ single crystals. A large anomalous Nernst effect is observed at temperatures below 30 K. Multiple Fermi surfaces with small effective masses are revealed by quantum oscillations in Nernst and Seebeck signals under high magnetic field. Furthermore, we find an unknown frequency, and attribute it to the magnetic breakdown across two smaller Fermi surfaces. A gap around 20 meV can be resolved from the breakdown threshold field, which we propose to be introduced by the CDW. These results shed new light on the CDW-related phenomena, particularly in AV₃Sb₅ compounds.

Condensed matter systems with kagome lattices have attracted significant interest owing to their rich physics. With the special two-dimensional corner-sharing triangular network, the electronic structure of the kagome lattice holds Dirac cones, flat bands, and enhanced correlation [1, 2]. Further inclusion of other collective orders in metallic kagome Dirac cones, flat bands, and enhanced correlation [3–10]. Recently, a new kagome metal family materials can give rise to more exotic quantum states and the inclusion of other collective orders in metallic kagome Dirac cones, flat bands, and enhanced correlation [1, 2]. Furthermore, the electronic structure of the kagome lattice holds the special two-dimensional corner-sharing triangular net-...
genic temperature. These features can provide the preferable condition for large ANE and magnetic breakdown, which are discussed in the following parts.

Figures 2(a) and (b) show the magnetic field dependence of the Nernst signal $S_{xy}/T$ for samples #1 and #2 at selected temperatures, respectively. In the semiclassical one-band theory, the Nernst thermopower $S_{xy}$ evolves with magnetic field as $S_{xy} = S_0 \mu B/[1 + (\mu B)^2]$, where $\mu$ is the carrier mobility. It has a peak at $B = 1/\mu$ and tends to zero under higher field.[31] The Nernst signal of both samples #1 and #2 show a weak peak below 1 T, and the peak field value increases with temperature. The low peak field underlines the high mobility of the charge carriers. At low temperatures, the Nernst signals display intense quantum oscillations, and tend to a nonzero constant in high-field region, which is an obvious anomalous component. As the temperature increases, the anomalous component gradually vanishes. At higher temperatures, a linear behavior with a negative slope becomes noticeable, which is caused by the multiband effect.[32,33]

To further reveal the origin of the ANE, we extract the anomalous Nernst component by the linear extrapolation of $S_{xy}(B)$ to zero field, as shown in Fig. 2(c). Although the anomalous component for sample #1 is larger than that of sample #2, they have a similar temperature dependence in the overlapping range. The ANE component has almost no temperature dependence at low temperatures, and rapidly decreases at higher temperatures until about 30 K. The Nernst curves can also be described by an empirical expression [28]:

$$S_{xy}(B) = S_{N_{xy}} \frac{\mu B}{1 + (\mu B)^2} + S_{A_{xy}} \tanh \left( \frac{B}{B_0} \right),$$

where $S_{N_{xy}}$ and $S_{A_{xy}}$ are the ordinary and anomalous Nernst signal amplitudes, respectively. $B_0$ is the saturation field of the anomalous component. This expression fits well with the Nernst signals for low temperatures, and some examples of the fits are shown in Fig. 2(d). They result in a very high mobility of $\mu \sim 10^5 \text{cm}^2\text{V}^{-1}\text{s}^{-1}$ for sample #1 and an order of magnitude lower for sample #2. The mobility difference can also be perceived by comparing the Nernst signals of the two samples at same temperature, where the quantum oscillations of sample #1 are obviously greater than those of sample #2. These features suggest the enhancement effect of the mobility to the ANE [28].

Figures 3(a) and (b) show the Seebeck coefficient $S_{xx}$ for both samples at selected temperatures. Also, the Seebeck signals of the two samples have similar magnetic field dependence and close values. The temperature dependence of the zero-field $S_{xx}$ for both samples are shown in Fig. 3(c). It is negative for $T > 6$ K, indicating the dominant electron carriers, but becomes positive for lower temperatures, proving the existence of two types of carriers. Moreover, for temperatures below $\approx 20$ K the Seebeck signal has a mustache shaped profile around zero field, which cannot be explained by the conventional one-band model [34]. Considering the multiband nature of the system, we use the modified expression:

$$S_{xx}(B) = S_1 \frac{1}{1 + (\mu_1 B)^2} + S_2 \frac{1}{1 + (\mu_2 B)^2} + S_\infty \frac{(\mu B)^2}{1 + (\mu B)^2},$$

(2)

where $S_1$ ($S_2$) and $\mu_1$ ($\mu_2$) are the zero-field Seebeck coefficients and mobility of the first (second) carrier, respectively, and $S_\infty$ is the limiting value when $B \to \infty$. The expression can well fit the Seebeck signals, as shown in Fig. 3(d).

The quantum oscillations can provide more information on the electronic structure. Figure 3(a) shows the oscillatory parts $\Delta S_{xx}$ and $\Delta S_{xy}$ at 2.4 K. Although composed by multiple frequencies, the oscillations have one primary frequency for both $\Delta S_{xx}$ and $\Delta S_{xy}$ as marked by the dashed lines. As shown in Fig. 3(b), the fast Fourier transformation (FFT) of

![FIG. 1. (Color online) (a) Temperature dependence of the resistivity of a typical sample with the RRR as high as 325. The kink at 94 K corresponds to the CDW transition. The inset shows the lower-temperature resistivity with a sharp superconducting transition at 3.5 K. (b) Resistivity at 2 K divided by the normal-state resistivity $\rho(5 \text{ K})$ as a function of the magnetic field. The high RRR and magnetoresistance, sharp superconducting transition, and intense quantum oscillations suggest the high quality of the samples.](image-url)
FIG. 3. (Color online) (a, b) Magnetic field dependence of the Seebeck signals for sample #1 and #2, respectively. (c) Temperature dependence of the Seebeck coefficient at the zero field for both samples. (d) Seebeck signal of sample #1 with the fitting lines of the two-band expression (2).

\( \Delta S_{xx} \) reveals four main frequencies, which are \( F_\alpha = 18 \) T, \( F_\beta = 28 \) T, \( F_\gamma = 72 \) T, and \( F_\delta = 91 \) T, consistent with those in the SdH oscillations [21] [34]. From the Onsager relation \( F = (h/2\pi e) S_T \) [35], these low frequencies correspond to four small FSSs. Moreover, there is an additional significant frequency at approximately 46 T in the FFT of Nernst oscillations, and this frequency is less notable in the Seebeck and SdH oscillations [Fig. 4(c)]. This additional frequency is not a harmonic one of other frequencies but roughly equals to the sum of \( F_\alpha \) and \( F_\beta \). Besides, it only occurs in the field region > 3 T, as shown in Fig. 4(c). We attribute it to the magnetic breakdown across the orbits \( \alpha \) and \( \beta \). This quantum tunneling effect suggests their adjacent positions in the Brillouin zone [33].

The Lifshitz-Kosevitch (LK) theory describes the evolution of the quantum oscillations with the temperature and magnetic field [35], where the cyclotron effective mass and Dingle temperature are involved. For the oscillations in the thermoelectric coefficients, we fit the temperature dependence of the amplitudes of the oscillations using the following expression [36]:

\[
\frac{A}{T} \propto \frac{\lambda}{\sinh(\lambda)},
\]

where \( A \) is the amplitude of \( \Delta S_{xx} \) or \( \Delta S_{xy} \), \( \lambda = 2\pi^2 k_B m^* T/ehB, \) and \( m^* \) is the cyclotron effective mass. For \( B, \) we use the average of the field range of oscillations, \( 1/B = 1/B_1 + 1/B_2 \). Figure 4(d) shows the selected fitting results. Both \( \Delta S_{xx} \) and \( \Delta S_{xy} \) give consistent effective masses: \( m_{\alpha}^* = 0.039 m_0, m_{\beta}^* = 0.043 m_0, m_{\gamma}^* = 0.058 m_0, \) and \( m_{\delta}^* = 0.054 m_0 \). These light effective masses are close to the ones obtained from SdH oscillations [21].

Figure 4(e) shows the index plot of the primary oscillation shown in Fig. 4(a), and the \( (\mu_0 H)^{-1} \) values correspond to the maxima of the \( \Delta S_{xx} \) or \( \Delta S_{xy} \). The slope of the index plot gives a frequency of 72 T, which is consistent with the value of \( F_\gamma \) obtained by the FFT. Because \( S_{xx} \) and \( S_{xy} \) are the diagonal and off-diagonal terms of the tensor \( S \), respectively, the maxima in \( \Delta S_{xy} \) typically have a 1/4 phase shift relative to \( \Delta S_{xx} \) [31]. However, there is no phase shift between \( \Delta S_{xx} \) and \( \Delta S_{xy} \) in CsV₃Sb₅, at least at the frequency \( F_\gamma \). The reason remains unclear and requires further investigation.

FIG. 4. (Color online) (a) Oscillation parts of both \( S_{xx} \) and \( S_{xy} \) at 2.4 K. A primary frequency of \( F_\gamma = 72 \) T can be identified and the peaks are marked by dashes lines. (b) FFT spectrum of the Seebeck and SdH oscillations with four peaks are labeled. The highest peak \( \gamma \) corresponds to the primary frequency shown in (a). (c) FFT spectrum of the Nernst oscillations obtained from different magnetic ranges. A fifth peak with frequency close to \( F_\gamma + F_\beta \) appears but is absent below 3 T. (d) Amplitudes of the four main peaks as a function of the temperature, with the solid lines representing the LK fitting. The effective masses obtained from \( \Delta S_{xx} \) and \( \Delta S_{xy} \) have very close values. (e) Index plot obtained from the primary oscillation of \( \Delta S_{xx} \) shown in (a). (f) FS mapping measured by the ARPES at 33 K with the Brillouin zone and high-symmetry points superposed. (g) Possible sketch of FSSs \( \alpha \) and \( \beta \), with a breakdown path marked by the dashed lines.
Understanding the band structure in the CDW state of the AV$_3$Sb$_5$ compounds is a key topic. Although density-functional-theory calculations can well explain the band structure observed in angle-resolved photoemission spectroscopy (ARPES) experiments \[14\], the four oscillation frequencies in the transport measurements remain poorly understood \[34\]\[37\]. It suggests the delicate effect of the CDW modulation to the band structure. The electronic structure of undistorted CsV$_3$Sb$_5$ has three types of bands in the vicinity of the Fermi level: a parabolic electronic band near the $\overline{\Gamma}$ point, multiple Dirac bands around the $\overline{K}$ points, and saddle points or van Hove singularities at the $\overline{M}$ points \[14\]\[15\], as shown in Fig. \[4\]. The CDW transition in the AV$_3$Sb$_5$ system is commonly believed to be driven by the Peierls instability related to FS nesting \[15\]. The direct consequence of this instability is the gap opening on the FSs, which has been demonstrated by several experimental techniques \[17\]\[19\]\[38\]\[43\]. Moreover, the CDW gap has a strong momentum dependence along the FSs of the Dirac bands \[41\]\[42\]. The small $\epsilon_g$ CDW gap has a strong momentum dependence along the FSs and may originate from a single band split by the CDW gap. Figure \[4g\] shows a possible sketch of the FSs $\alpha$ and $\beta$, together with a breakdown path. The gap ($\epsilon_g$) between them can be related to the threshold field of magnetic breakdown ($B^* \sim 3$ T) by $\hbar \omega_c \geq \epsilon_g^2 / E_F$, where $\omega_c = eB^* / m^*$ \[35\]. With $E_F \sim 50$ - 80 meV given by the oscillations, the gap $\epsilon_g$ can be estimated less than $\sim$ 20 meV. This value is consistent with the CDW gap obtained from the spectroscopy experiments.

In the CDW state of AV$_3$Sb$_5$ compounds, an unconventional chiral charge order was discovered \[16\]\[44\], and an accompanied chiral flux phase was proposed to account for the TRSB \[45\]\[46\]. For CsV$_3$Sb$_5$, the TRSB has been confirmed by a $\mu$SR experiment \[24\], along with the AHE and our ANE \[21\]\[44\]. Intriguingly, the TRSB signal in the $\mu$SR experiment, the AHE, and the ANE appear below different temperatures. A local field within the kagome plane is detected by the $\mu$SR below 70 K ($< T_{CDW}$), and an additional perpendicular component appears below 30 K. However, the AHE starts at $T_{CDW}$, whereas in our data the ANE becomes obvious below 30 K. The different temperature regimes where AHE and ANE are noticed may be due to their different sensitivities in this system. The ANE appears at the close temperature as the out-of-plane field in the $\mu$SR data, suggesting a possible relationship between the ANE and a hidden chiral flux phase.

In summary, we conducted a systematic investigation on the magneto-thermoelectric effect of the kagome metal CsV$_3$Sb$_5$. The Nernst signal shows a large anomalous component below 30 K, which suggests the time-reversal symmetry breaking may be accompanied by a hidden chiral flux phase appearing at the same temperature. The quantum oscillations in the thermoelectric coefficients reveal multiple Fermi surfaces with small effective masses. Two small Fermi surfaces that are split from a single Dirac band by a charge density wave gap are indicated by the magnetic breakdown effect. Our results are significant for further study on the chiral flux phase, fine electronic structure, and novel superconductivity in the charge density wave state of kagome metal.

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