

Thermoelectricity of Moiré Heavy Fermions in MoTe₂/WSe₂ Bilayers

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Tunable Kondo lattice and heavy fermion physics have been recently reported in moiré materials, but most of the studies have focused on the electrical and magnetic properties. Quantitative thermoelectric measurements, which can reveal entropic information of the heavy fermions, have yet to be achieved. Here, we report a comprehensive thermoelectric study on the moiré heavy fermion phase realized in hole-doped angle-aligned MoTe₂/WSe₂ bilayers. By electrically gating the material to the Kondo lattice region of the phase diagram, we observe a sign change in the Seebeck coefficient near the Kondo coherence temperature, where the heavy fermion phase with an electronlike Fermi surface evolves into an itinerant Fermi liquid with a holelike Fermi surface. We compare the results with the semiclassical Mott relation and discuss the observed discrepancies. In addition to the thermal dissociation of Kondo singlets in the heavy Fermi liquid, a sign change accompanied by a strong peak in the Seebeck coefficient is also observed near a Zeeman breakdown of the Kondo singlets, signaling an entropy accumulation. Our results provide entropic information on both the formation and breakdown of heavy fermions in moiré semiconductors.

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Recently, tunable Kondo lattice and heavy fermion physics have been reported in both graphene [1–3] and transition metal dichalcogenide (TMD) [4–7] moiré materials. Most of the experimental studies to date have focused on the electrical and magnetic properties. Although photothermoelectric evidence of topological heavy fermions in twisted bilayer graphene has been reported recently [2,3], quantitative thermoelectric measurements of the heavy fermion phase are still missing because of the difficulty in accurate temperature calibration under a driven thermal current. Accurate measurements of the thermoelectric coefficients are rewarding because they can quantify the entropy per particle (the Seebeck coefficient) and/or per vortex (the Nernst coefficient) in the zero-temperature-gradient limit in strongly correlated materials [8–16] (e.g., cuprate high-temperature superconductors [17,18] and heavy fermion materials [19–21]). In heavy fermion

materials, the large Seebeck coefficients due to the heavy electron mass and their temperature and magnetic field dependences can provide invaluable entropic information for the formation of Kondo singlets [22–24]. Performing quantitative thermoelectric measurements on moiré heavy fermion systems is therefore fundamentally important.

In this study, we report measurements of the Seebeck coefficient (S) in the Kondo lattice regime of hole-doped angle-aligned MoTe₂/WSe₂ moiré bilayers. We observe a thermal crossover from an electronlike response ($S < 0$) in the heavy fermion phase to a holelike response ($S > 0$) at temperatures above the Kondo coherence temperature (T^*), where Kondo singlets are dissociated by thermal excitations. The result violates a simple application of the semiclassical Mott relation for S in the heavy fermion phase; possible origins of the violation are discussed. A similar sign change in S and a strong enhancement are also observed near a breakdown of the Kondo singlets by an external Zeeman field at low temperatures. These results are contrasted to the normal behavior of an itinerant Fermi liquid when the Kondo lattice is quenched by gating.

Moiré Kondo lattice in MoTe₂/WSe₂ bilayers—We examine dual-gated Hall bar devices of angle-aligned MoTe₂/WSe₂ moiré bilayers (Figs. 1(a) and 1(b); see fabrication details in Supplemental Material, Sec. 1 [25]), in which Kondo lattice and heavy fermion physics have been reported [4–7]. The dual-gated structure allows

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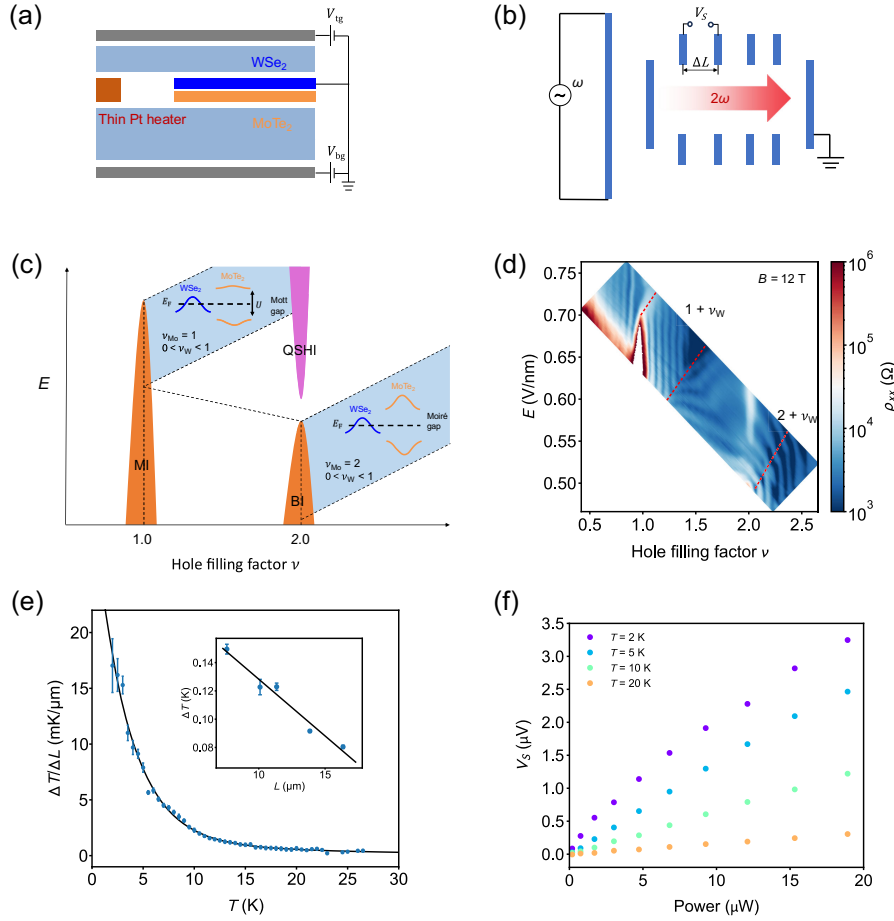


FIG. 1. Thermoelectric measurements on gate-tunable moiré Kondo lattice. (a) Schematic dual-gated angle-aligned MoTe₂/WSe₂ moiré bilayer device. V_{tg} and V_{bg} denote the top and bottom gate voltages, respectively, applied to the few-layer graphite gate electrodes (gray). The blue areas label the h -BN gate dielectrics, which encapsulate the Pt heater. (b) Schematic 2ω method for measurements of the Seebeck coefficient. An ac excitation current at frequency $\omega = 11.137$ Hz is applied to the heater, which generates a temperature gradient and a Seebeck voltage V_S at frequency 2ω (arrow). ΔL is the distance between adjacent probe electrodes. (c) Schematic phase diagram of MoTe₂/WSe₂ in ν and E . MI, BI, and QSHI denote a Mott insulator, a band insulator and a quantum spin Hall insulator. The upper blue shaded region with $\nu = \nu_{Mo} + \nu_W = 1 + \nu_W$ is the Kondo lattice region. U and E_F denote the on site Coulomb repulsion and the Fermi level (dashed line), respectively. Blue and orange bands label the moiré band of the W layer and the lower and upper Hubbard bands of the Mo layer, respectively. The lower blue shaded region with $\nu = 2 + \nu_W$ is used as a control experiment. The blue and orange bands here label the moiré bands of the W and Mo layer, respectively. (d) Longitudinal resistivity ρ_{xx} as a function of ν and E at $T = 2$ K and $B = 12$ T. The phase boundaries of the $1 + \nu_W$ and $2 + \nu_W$ regions are identified from the Landau level features and marked by red dashed lines. (e) Temperature gradient ($\Delta T/\Delta L$) between adjacent probe electrodes as a function of T (see main text and the Supplemental Material for details on calibration). The solid line is a fit to the data by $Ae^{-\alpha T} + Be^{-\beta T}$. Inset: Temperature rise at $T = 2$ K as a function of the distance to the heater. The solid line is a linear fit to the data. (f) Power dependence of V_S measured at $T = 2$ K, 5 K, 10 K, and 20 K. The gate voltages are fixed at $V_{tg} = -3.7$ V and $V_{bg} = 0.5$ V. Thermoelectric measurements were performed in the linear response regime.

independent control of the moiré lattice filling factor (ν) and the perpendicular electric field (E) (Supplemental Material, Sec. 2 [25]). The 7% lattice mismatch between the two TMD layers creates a moiré lattice of period $a_M \approx 5$ nm and a moiré density $n_M \approx 5 \times 10^{12}$ cm⁻² [4–7,33]. The moiré lattice creates flat bands in both the Mo and W layers (we denote MoTe₂ and WSe₂ by Mo and W, respectively, from now on) [34–39]. At low E fields, where the topmost valence band of the Mo layer is higher in

energy than that of the W layer, hole doping the material depletes electronic states only in the Mo layer [35,36]; a triangular lattice Mott insulator emerges at half band filling (i.e., $\nu = 1$) due to the strong on site Coulomb repulsion in the Mo layer [40–43]; the Mo band is split into the lower and upper Hubbard bands. The E field tunes the energy difference between the Mo and W bands through the interlayer Stark effect [34]. At sufficiently high E fields, the W band can be tuned to locate in between the Mo

Hubbard bands [4–7] [Fig. 1(c)]. Doping beyond $\nu = 1$ injects holes only in the W layer, which are nearly itinerant because of the significantly more dispersive W band (and thus much weaker correlation) than its Mo counterpart [35]. The system in this regime can be mapped to a moiré Kondo lattice model with a Kondo exchange interaction (J_K) between the local magnetic moments in the Mo layer and the itinerant holes in the W layer [35–39,44,45].

The schematic electrostatics phase diagram of the material is shown in Fig. 1(c). The Kondo lattice region with doping $\nu = \nu_{\text{Mo}} + \nu_{\text{W}} = 1 + \nu_{\text{W}}$ is shaded in blue (ν_{Mo} and ν_{W} denote the filling factor in the Mo and W layer, respectively). The E field span in this region is proportional to the Mott gap ≈ 37 meV in the Mo layer (see Mott gap size estimation in Supplemental Material, Sec. 3 [25]). A heavy Fermi liquid with a large Fermi surface has been reported at temperatures $T \lesssim T^*$; T^* increases with ν_{W} as more itinerant holes are available to Kondo screen the local moments [4]. In addition to thermal dissociation of the Kondo singlets at $T \gtrsim T^*$, an external Zeeman field (B) with Zeeman energy $g\mu_B B \gtrsim k_B T^*$ can also break down the Kondo singlets and destroy the heavy Fermi liquid [46] (g , μ_B , and k_B denote the electron g factor, the Bohr magneton, and the Boltzmann constant, respectively). This Zeeman breakdown has enabled us to visualize the $\nu = 1 + \nu_{\text{W}}$ Kondo lattice region in the resistivity (ρ_{xx}) map under a perpendicular magnetic field $B = 12$ T [Fig. 1(d)]; specifically, we observe vertical stripes in ρ_{xx} arising from the quantized Landau levels of the itinerant holes in the W layer. (See Fig. S1 for additional ρ_{xx} maps at other B fields [25].) Similar stripes in ρ_{xx} are also observed in the $\nu = 2 + \nu_{\text{W}}$ region of the phase diagram. Here the topmost moiré valence band of the Mo layer is fully filled; the lattice of local moments is quenched; the physics is controlled by the itinerant holes remaining in the W layer. We will use this region as a control experiment in the following discussions. The above results are fully consistent with earlier reports [4–7].

Thermoelectric measurements—To quantitatively measure the Seebeck coefficient S of $\text{MoTe}_2/\text{WSe}_2$, we evaporated a platinum heater and encapsulated it between the top and bottom hexagonal boron nitride (h -BN) gate dielectrics [Fig. 1(a)] [13–15]. The high in plane thermal conductivity of h -BN generates a nearly linear temperature gradient across the sample when the heater is turned on [13]; this is confirmed by both finite element analysis (Fig. S2 and Supplemental Material, Sec. 4 [25]) and local temperature measurements [inset of Fig. 1(e), Figs. S3–8 and Supplemental Material, Sec. 5 [25]].

We measured the Seebeck coefficient S by the established 2ω method [13–16,47,48] [Fig. 1(b) and Supplemental Material, Sec. 6 [25]]. We biased an ac current (I_{ac}) at frequency $\omega = 11.137$ Hz in the heater, which induces a 2ω thermoelectric voltage drop (V_S) in the longitudinal direction of the device. To calibrate the local

temperature rise (ΔT) induced by heating, we first measured the T dependence of the two-terminal resistance (heater off) for each transverse electrode pair in the device near the $\nu = 1$ insulating state, where a strong T dependence is observed; we then measured the heating-induced resistance change at 2ω to obtain ΔT . (Note that gate voltages were chosen such that the Seebeck and Nernst responses are negligible compared to the heating-induced resistance change.) For small ω , ΔT is determined by the thermal conductance of the substrate (not the sample) and is thus independent of gating [13–15]. The calibrated ΔT shows a linear dependence on the distance (L) from the heater [inset of Fig. 1(e)]; the resultant temperature gradient ($\Delta T/\Delta L$) decays quickly with T [Fig. 1(e)]. The Seebeck coefficient is $S = [(\Delta V_S/\Delta L)/(\Delta T/\Delta L)]$ (ΔL is the distance between two adjacent probe electrodes; see Fig. 1(b)).

We performed several tests to ensure the reliability of the Seebeck measurement. First, the thermoelectric measurements were performed in the linear response regime. This is illustrated by the linear dependence of V_S on the heater power in Fig. 1(f). (See Fig. S10 for additional power dependence measurements [25].) The temperature difference between adjacent electrodes in the longitudinal direction is also very small compared to the sample temperature T [Fig. 1(e)]. Second, we performed additional grounding tests and frequency dependence measurements to rule out measurement artifacts (Fig. S9 [25]); we also ensured a 90° phase shift in V_S relative to I_{ac} in all measurements. Third, we confirmed the reliability of our Seebeck measurements in the quantum Hall regime under high B fields. The measured Seebeck coefficient is in good agreement with the expected quantized value $S = \{[(k_B/e) \ln 2]/[n + 1/2]\} \approx [(59.7)/(n + 1/2)] \mu\text{V/K}$ at the n^{th} quantum Hall state [49–51] (Fig. S11 and Supplemental Material, Sec. 7 [25]; e is the electron charge). All these demonstrate the reliability of our measurements. See the Supplemental Material for details on device fabrication, measurements, and simulations [25].

Thermoelectric response of moiré heavy fermions—Figures 2(a) and 2(b) show the dependence of S and ρ_{yx} , respectively, on ν and E at $T = 2$ K (ρ_{yx} is the Hall resistivity measured at $B = 2$ T). The regions of interest, $\nu = 1 + \nu_{\text{W}}$ and $\nu = 2 + \nu_{\text{W}}$, are labeled according to the high B -field map of ρ_{xx} in Fig. 1(d). We observe the same sign for S and ρ_{yx} in both the $\nu = 1 + \nu_{\text{W}}$ and $\nu = 2 + \nu_{\text{W}}$ regions; specifically, $S, \rho_{yx} > 0$, corresponding to holelike transport, and $S, \rho_{yx} < 0$, corresponding to electronlike transport, are observed at $\nu = 2 + \nu_{\text{W}}$ and $\nu = 1 + \nu_{\text{W}}$, respectively.

We compare the measured Seebeck coefficient S to a simple application of the semiclassical Mott relation for metals $S_{\text{Mott}} = -(\pi^2 k_B^2 T/3e)[d \ln \sigma_{xx}/d\epsilon]_{\epsilon=\epsilon_F} \approx -(\pi^2 k_B^2 T/3e)[d \ln \sigma_{xx}/d\nu]\{N(\epsilon_F)/n_M\}$ [52,53]. Here, $\sigma_{xx} = (1/\rho_{xx})$ is the sample conductivity, ϵ_F is the electron Fermi energy, and $N(\epsilon_F) = (m^*/\pi\hbar)$ is the density of state

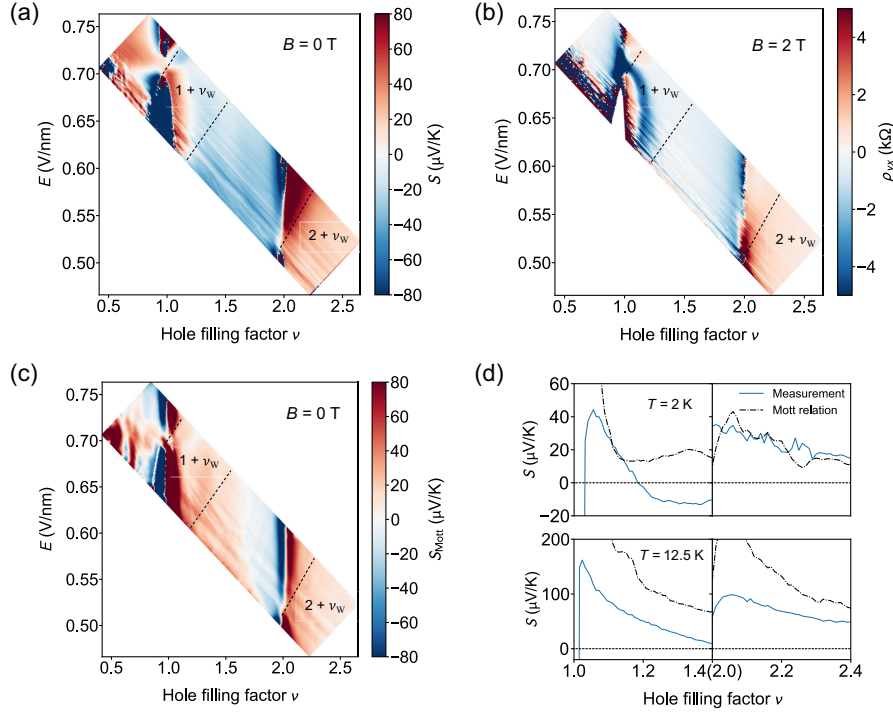


FIG. 2. Seebeck coefficient in a moiré Kondo lattice. (a) Seebeck coefficient S as a function of ν and E at $T = 2$ K and $B = 0$ T. (b) Hall resistivity ρ_{yx} (measured at $B = 2$ T) as a function of ν and E at $T = 2$ K. (c) The Seebeck coefficient S_{Mott} obtained using a simple application of the Mott relation as a function of ν and E (see main text). The dashed lines in (a)–(c) mark the phase boundaries of the $1 + \nu_W$ and $2 + \nu_W$ regions. (d) Comparison of the measured Seebeck coefficient S (solid lines) and the calculated S_{Mott} (solid-dotted lines) at $\nu = 1 + \nu_W$ (left panel, line cut at $E = 0.68$ V/nm) and $\nu = 2 + \nu_W$ (right panel, line cut at $E = 0.52$ V/nm) for $T = 2$ K (top) and 12.5 K (bottom).

at ϵ_F for the W layer with an effective hole mass $m^* \approx 0.5m_0$ obtained from T -dependent Shubnikov-de Haas measurements (m_0 is the free electron mass) [4]. Figure 2(c) shows the calculated map for S_{Mott} ; representative linecuts along the center of the $\nu = 1 + \nu_W$ and $\nu = 2 + \nu_W$ regions are shown in Fig. 2(d). Whereas a good agreement between S and S_{Mott} is observed at $\nu = 2 + \nu_W$, the two show opposite signs in most of the regions at $\nu = 1 + \nu_W$. In other words, while S_{Mott} predicts holelike transport in the W layer for both $\nu = 1 + \nu_W$ and $\nu = 2 + \nu_W$, the measured Seebeck coefficient shows electronlike transport in the $\nu = 1 + \nu_W$ region.

To better understand the origin of the disagreement between S and S_{Mott} , we perform T -dependence studies at both $\nu = 1 + \nu_W$ and $\nu = 2 + \nu_W$. We first examine the T dependence of S at different fillings for $\nu = 2 + \nu_W$ in Figs. 3(a) and 3(b) ($E = 0.5$ V/nm). We also compare the results to the metallic T dependence of ρ_{xx} at the same fillings in Fig. 3(c). The Seebeck coefficient S shows a T -linear dependence at low T and a decreasing slope with increasing ν_W . This T dependence is well described by the Mott relation for itinerant holes in the W layer, which predicts $S = (\pi^2/2)[k_B/e]\{T/T_F\} \approx 425(T/T_F)$ $\mu\text{V/K}$ for impurity-dominated scattering at low T [54]. Here, $T_F \equiv (\epsilon_F/k_B) = (\hbar^2\pi n_M\nu_W/m^*k_B)$ is the Fermi temperature of

the itinerant holes. Fitting the data to the Mott relation gives an effective mass (m^*/m_0) [inset of Fig. 3(b)] in good agreement with that obtained from T -dependent Shubnikov-de Haas measurements [4]. The deviation from the T -linear dependence at $T \gtrsim 10$ K is possibly caused by phonon drag [55,56] and/or flat band effects (Fig. S13 and Supplemental Material, Sec. 8 [25]).

Next, we study the T dependence at $\nu = 1 + \nu_W$ in Figs. 3(d)–3(f) ($E = 0.68$ V/nm). The Seebeck coefficient S scales linearly with T at low T ; the absolute slope increases with increasing ν_W ; it also changes sign at a characteristic temperature T^* that increases with ν_W . This contrasts with that at $\nu = 2 + \nu_W$, where no sign change is observed. We mark the location of T^* in the T dependence of ρ_{xx} in Fig. 3(f). Interestingly, it corresponds to the previously reported Kondo coherence temperature, where a peak or bump in ρ_{xx} is observed and below which coherent Fermi liquid transport emerges [4–7]. The results strongly suggest the emergence of heavy fermions at $T \lesssim T^*$ as the origin of the observed behavior of the Seebeck coefficient S and its disagreement with S_{Mott} at $\nu = 1 + \nu_W$.

In a Kondo lattice, the coherent scattering of itinerant electrons with a lattice of local magnetic moments creates Kondo singlets and electrons with a renormalized heavy mass at $T \lesssim T^*$ [23,57]. In this heavy fermion phase, the

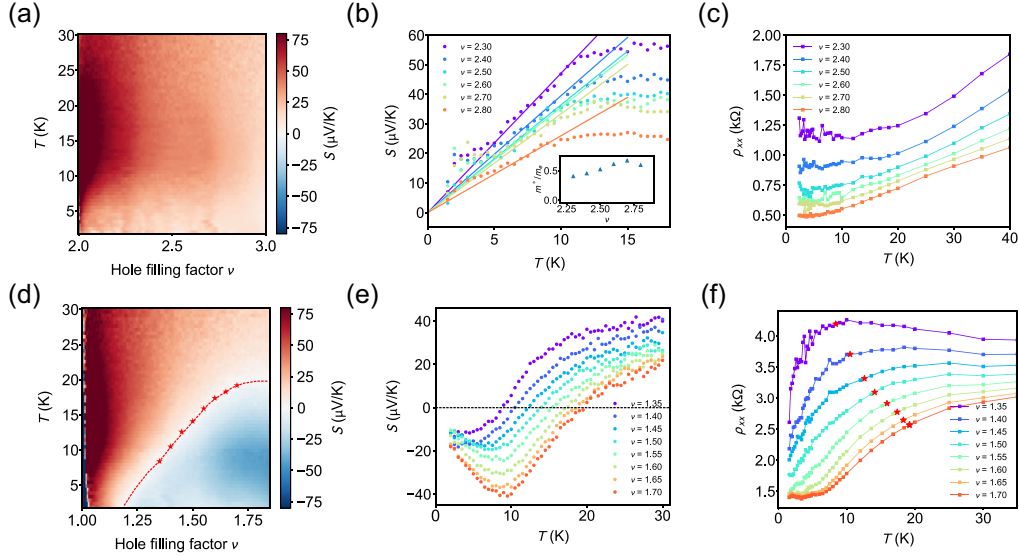


FIG. 3. Temperature-dependent thermoelectric response for heavy fermions. (a),(d) Seebeck coefficient S as a function of ν and T at $E = 0.50$ V/nm [(a), the $2 + \nu_W$ region] and at $E = 0.68$ V/nm [(d), the $1 + \nu_W$ region]. (b),(e) Selected line cuts at constant fillings from a (b) and from d (e). Solid lines in b are linear fits to the data in the low-temperature regime. Inset in (b): Extracted effective mass from the slope of the linear fits as a function of ν . The dashed line in (e) marks the sign change in S . (c),(f) Temperature dependence of ρ_{xx} at the same fillings and E field as b (c) and as e (f). The stars in (d) mark the locations for the sign change in S , which correlate with the Kondo coherence temperature T^* marked by the same stars in (f).

local moments are absorbed by the itinerant electrons, producing a heavy Fermi liquid with a large Fermi surface below the crossover temperature scale T^* . In $\text{MoTe}_2/\text{WSe}_2$, the total hole density of the heavy Fermi liquid is therefore $(1 + \nu_W)n_M$ (rather than $\nu_W n_M$), which is equivalent to an electron density $(1 - \nu_W)n_M$ [4]. This explains the electronlike Seebeck coefficient ($S < 0$) and Hall response ($\rho_{yx} < 0$) observed at $T \lesssim T^*$. [The extracted Hall density is also in good agreement with $(1 - \nu_W)n_M$, as demonstrated by earlier studies [4].] With increasing temperature beyond the crossover temperature scale T^* , the Kondo singlets are gradually dissociated by thermal excitations; the local moments are released [10], resulting in a change in size of the Fermi surface from $(1 + \nu_W)n_M$ to $\nu_W n_M$; thus a sign change in S at $T \gtrsim T^*$, where a better agreement between S and S_{Mott} is observed [lower panel in Fig. 2(d) and Fig. S12 [25]]. Similar T dependence is also observed in many Ce-based heavy fermion materials, including CeGe_2 [58], Ce_2N_7 [59], CeCu_2Si_2 [21,60], and CeAl_2 [61], and has been taken as thermoelectric evidence of heavy fermions.

Although the above picture qualitatively explains the observed T dependence of S at $\nu = 1 + \nu_W$, extracting quantitative information remains challenging. For instance, if we insist on using $S_{\text{Mott}} \approx (\pi^2/2)[k_B/e]\{T/T_F\}$ to describe the T -linear dependence in the heavy fermion phase, we would end up with a decreasing T_F with increasing ν_W , which would contradict the observed dependence of $T^* \sim T_F$ on ν_W [Fig. 3(d)]. (Note that T^* is expected to increase with ν_W as more itinerant holes are

available to Kondo screen the local moments [38,39,45].) The breakdown of S_{Mott} here may not be surprising as the step from $S_{\text{Mott}} = -(\pi^2 k_B^2 T/3e)[d\ln\sigma_{xx}/d\epsilon]|_{\epsilon=\epsilon_F}$ to $S_{\text{Mott}} \approx -(\pi^2 k_B^2 T/3e)[d\ln\sigma_{xx}/d\nu]\{N(\epsilon_F)/n_M\}$ relies on the assumptions of 1) a rigid band, $\sigma_{xx}(\mu, \epsilon) = \sigma_{xx}(\mu + \epsilon, 0)$ [2] and 2) a density-independent scattering rate (μ is the electron chemical potential). These assumptions are known to fail in many strongly correlated materials [62], although the original Mott relation $S_{\text{Mott}} = -(\pi^2 k_B^2 T/3e)[d\ln\sigma_{xx}/d\epsilon]|_{\epsilon=\epsilon_F}$ remains robust as long as a quasiparticle description of transport is valid. Because we cannot directly evaluate $(d\ln\sigma_{xx}/d\epsilon)|_{\epsilon=\epsilon_F}$ using experimental data, numerically solving the Anderson/Kondo lattice model is required to quantitatively describe the T dependence of S in the heavy fermion phase.

Thermoelectric response near Zeeman breakdown of Kondo singlets—In addition to the breakdown of Kondo singlets by thermal excitations, the Kondo singlets can also be dissociated by an external B field or a Zeeman energy $g\mu_B B \gtrsim k_B T^*$ [46]. Figures 4(a) and 4(b) show the dependence of ρ_{yx} and S , respectively, on ν and B in the $\nu = 1 + \nu_W$ region ($T = 2$ K and $E = 0.68$ V/nm). Both ρ_{yx} and S show a sign change with increasing B field; the sign change boundary is slightly different for the two quantities (solid and dashed lines), but the characteristic B field increases with increasing ν_W in both cases. A Landau fan also emerges immediately beyond the characteristic B field (B_c) for ρ_{yx} (solid line). Interestingly, B_c closely traces the peak position of S in Fig. 4(b). The appearance of a

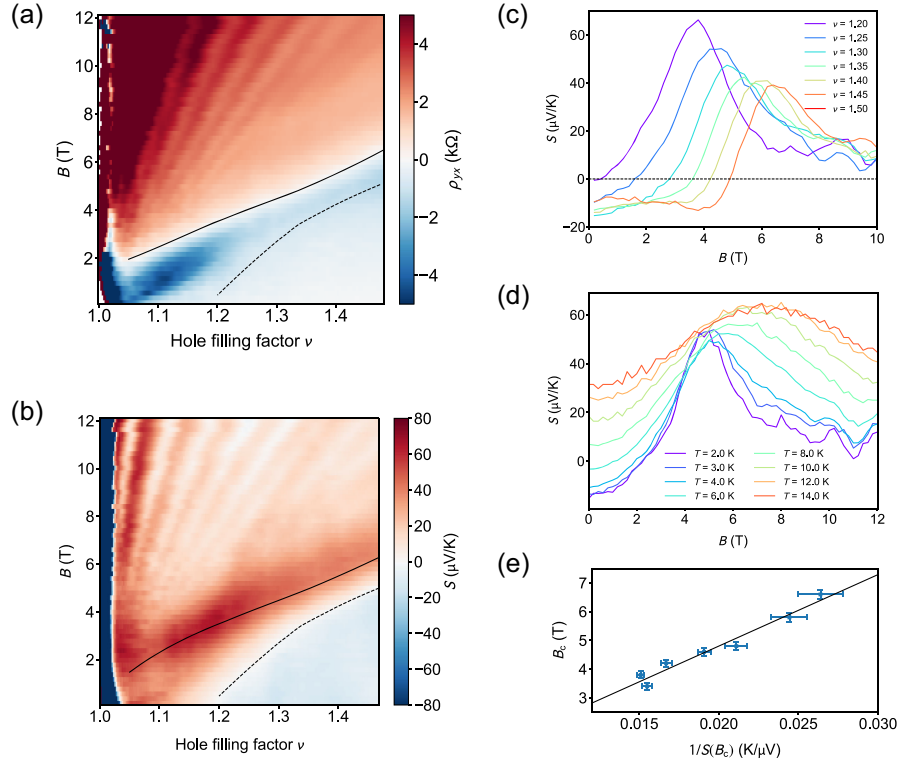


FIG. 4. Thermoelectric response near Zeeman breakdown of Kondo singlets. (a),(b) ρ_{yx} (a) and S (b) as a function of ν and B at $E = 0.68$ V/nm and $T = 2$ K. The Zeeman breakdown occurs at the characteristic B field, B_c , for ρ_{yx} (solid lines). The sign change in S (dashed lines) happens at a slightly lower B field than B_c , likely due to particle-hole asymmetry. (c) Selected line cuts from (b) at constant fillings showing the evolution of the Seebeck peak with filling. (d) The Seebeck coefficient S as a function of B at varying temperatures ($\nu = 1.3$ and $E = 0.68$ V/nm). (e) Inverse relationship between B_c and the peak value $S(B_c)$ at $T = 2$ K. The solid line is a linear fit to the data. The error bar is estimated from the noise level of the Seebeck measurement and the uncertainties in determining the peak position and height of the Seebeck coefficient.

Seebeck peak at $B = B_c$ is further illustrated by the selected linecuts at constant fillings in Fig. 4(c). The peak broadens with increasing temperature [Fig. 4(d)]. We also found an inverse relationship between B_c and the peak value $S(B_c)$ at $T = 2$ K, as demonstrated in Fig. 4(e).

As demonstrated by earlier studies [4,5], the Hall density for the heavy Fermi liquid ($\rho_{yx} < 0$) is equal to $-(1 - \nu_W)n_M$ (the $-$ sign is for electron density); it jumps to a small hole density $\nu_W n_M$ for the itinerant Fermi liquid ($\rho_{yx} > 0$) at $B > B_c \sim (k_B T^*/g\mu_B)$, where the holes in the W layer are decoupled from the local moments in the Mo layer. Because of the much smaller effective mass in the itinerant Fermi liquid, a Landau fan is immediately observed at $B > B_c$. Moreover, the Hall density jump (with B field) gets sharper with decreasing T , suggesting that the Zeeman breakdown at $B \approx B_c$ is a quantum phase transition rather than a smooth crossover [4].

This conclusion is further supported by the observed Seebeck peak and its thermal broadening near B_c . In a simple Fermi liquid with a single Fermi surface and energy-independent scattering mean free path, S has been shown to provide a powerful measure of the entropy per fermion [8,9,12,63]. Although this is no longer strictly correct when these approximations are relaxed, Behnia *et al.*

have demonstrated a phenomenological scaling law $(S/C_e)N_A e \sim \pm 1$ for S and C_e (the electronic specific heat capacity) in many heavy fermion materials (N_A is the Avogadro number) [12]. The observed Seebeck peak at $B = B_c$ therefore suggests an enhancement in $C_e \propto N(\epsilon_F)$ or in the manybody electronic density of states $N(\epsilon_F)$ at the Zeeman breakdown, and therefore supports the scenario of a quantum phase transition [24]. This entropic interpretation $S \sim \pm(C_e/N_A e)$ also provides a natural explanation for the inverse relationship between $B_c \sim (k_B T^*/g\mu_B) \propto [1/N(\epsilon_F)]$ and the peak value $S(B_c) \propto C_e \propto N(\epsilon_F)$ observed in Fig. 4(e). Note that an enhancement in C_e together with an anomaly in the magnetic susceptibility have been reported in the magnetic Kondo breakdowns of several Ce-based heavy fermion materials [64–67]; the results support that the breakdown is a metamagnetic transition rather than a smooth crossover.

Conclusion—In conclusion, we have demonstrated thermoelectric evidence of gate-tunable heavy fermions in angle-aligned WSe₂/MoTe₂ moiré bilayers. The sign and magnitude of the Seebeck coefficient contain important information on the size of the Fermi surface and the entropy per particle. We have observed the breakdown of the Kondo

singlets induced by both thermal excitations and an external Zeeman field. The results have also been reproduced in another device (Fig. S14 [25]). Future thermoelectric measurements down to the mK temperature range should be pursued to further reveal the nature of the Zeeman Kondo breakdown, e.g., the possible existence of a quantum critical point [24] and of a continuous metamagnetic phase transition [46].

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Y. Z. and W. Z. fabricated the devices. Y. Z. and W. Z. performed the electrical and thermoelectrical transport measurement and analyzed the data with the help of Z. H. K. W., and T. T. grew the bulk *h*-BN crystals. Y. Z., W. Z., J. S., and K. F. M. designed the scientific objectives and oversaw the project. All authors discussed the results and commented on the manuscript.

Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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