

Probing frustrated metallomolecular spin Kondo lattice interfaces through anomalous Nernst effect

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Frustrated Kondo spin-lattice systems away from the antiferromagnetic ground state have been found to display strange metal behavior. In two dimensions (2D), where deviations from bulk systems are expected, the emergence of Kondo spin lattices has recently been observed in van der Waals systems. Metallomolecular interfaces of supramolecular lattices have also been suggested as an alternative 2D Kondo spin-lattice system where a single-ion Kondo effect, as well as an inter-spin-site Ruderman-Kittel-Kasuya-Yosida coupling, has been demonstrated in scanning tunneling microscopy studies. Here, going beyond ultrahigh-vacuum STS studies in metallomolecular interfaces, we report a metastable frustrated antiferromagnetic state, with ultrahigh spin-freezing temperatures T_f ranging from 240 to 300 K on the interfaces of highly textured Pt(111) and Pt(111)/Co films with organic molecules grown in an ultrahigh vacuum of approximately 10^{-10} mbar. A signature of the expected strange metal behavior in correlated systems is a large Nernst response. In the vicinity of the spin-freezing transition, we measure a large anomalous Nernst effect in the Pt/Co/molecule heterostructures that we attribute to the frustrated 2D Kondo spin lattice interface, with an anomalous Nernst coefficient of at least $3\mu\text{V/K}$.

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I. INTRODUCTION

Recent scanning tunneling microscopy (STM) studies of Kondo resonance on metallomolecular interfaces have led to observations of Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling between molecular-spin sites, suggesting emergence of a metallomolecular Kondo spin lattice accompanied by long-range magnetic order [1–3]. Kondo spin lattices have led to the emergence of heavy-fermion f -orbital systems that have been subject to prolonged interest [4,5], with antiferromagnetism and superconductivity [6] emerging in these systems in the vicinity of quantum criticality [7]. The relative strength of the RKKY interaction between spin-lattice sites is mediated by the conduction electrons, and the single-ion Kondo interaction at each spin-lattice site dictates the nature of emerging phases, including the quantum phase transitions between them. Frustration has been understood to be another relevant parameter in Kondo spin-lattice systems [8,9], where a strange metal or a spin liquid state is expected in the

so-called localized limit in the vicinity of the antiferromagnetic ground state [10].

Existing evidence in this limit in conventional systems has been obtained on bulk systems such as $\text{Pr}_2\text{Ir}_2\text{O}_7$ [11,12], YbRh_2Si_2 [13,14], CePdAl [15], and CeRhSn [16]. The phase transitions are observed at low temperatures, for example, the freezing temperature T_f for $\text{Pr}_2\text{Ir}_2\text{O}_7$ is 0.3 K [12] and the Néel temperature T_N for CePdAl is ≈ 2.75 K [15]. Looking beyond existing two-dimensional van der Waals systems [17–22], metallomolecular thin film interfaces may offer much higher magnetic transition temperatures [23]. A better fundamental understanding of Kondo spin lattices [24] could also be gained through metal surface choices of different conduction electron bandwidths, but the observations so far have been limited to ultrahigh-vacuum STM experiments [1–3].

Here, we demonstrate magnetic frustration as well as a large anomalous Nernst effect on flat-lying [1,3,25] metal (Cu, Co) and hydrogen (H_2) phthalocyanine (Pc) supramolecular lattice interfaces with textured Pt(111) and Pt(111)/Co (< 2 nm) films. The chamber pressure during the Pc layer sublimation was kept on the order of 10^{-10} mbar to enable a strong π - d orbital hybridization [26] during chemisorption [27] as well as the formation of a molecular lattice [1–3,28]. We deposited a molecular layer thicker than 10 nm on top of Pt and Pt/Co films to ensure a continuous film of flat-lying molecules at the interface. We capped the multilayer with a 15-nm-thick metal film to preserve the high-vacuum hybridized interface under ambient conditions.

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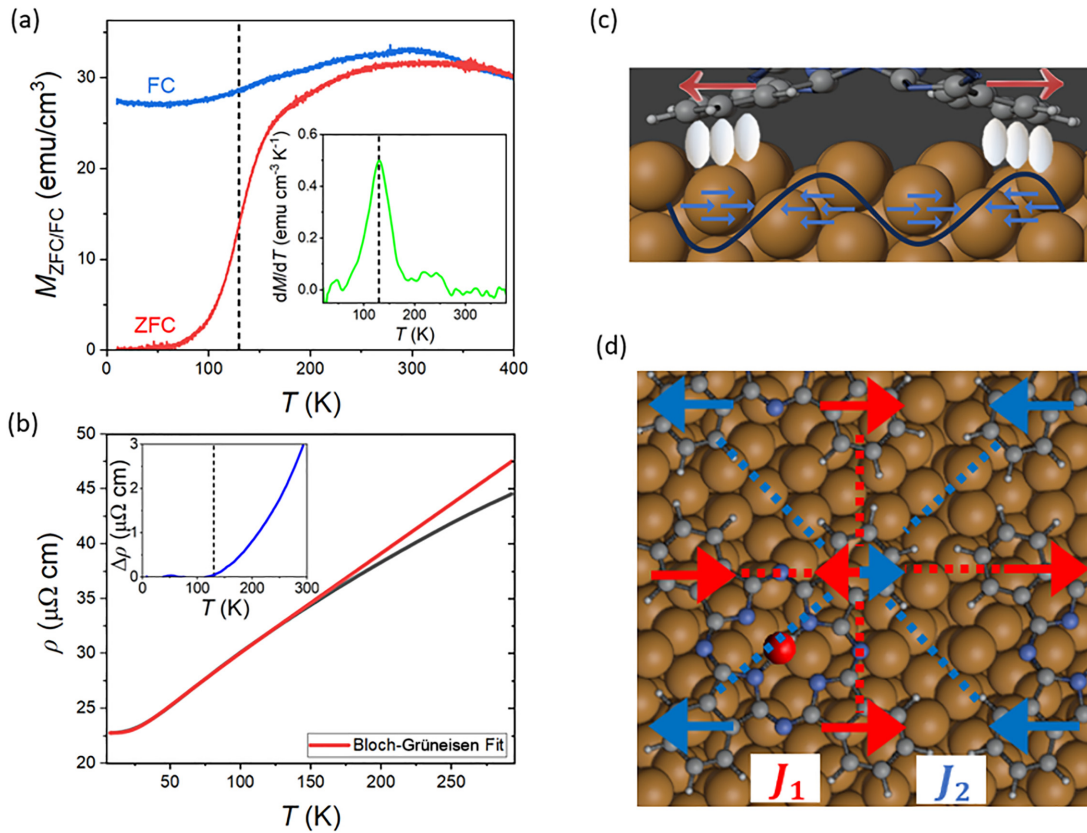


FIG. 1. Frustrated antiferromagnetism onset on (111) textured Pt/molecular interface with a spin freezing point at room temperature. (a) Zero-field-cooled–field-cooled (ZFC-FC) magnetometry curves measured in a 20-mT in-plane field on a Pt(4.2 nm)/CuPc metallomolecular interface, showing a broad spin-freezing transition point peaking at $T_f = 300$ K. The derivative of the ZFC curve as a function of temperature is plotted in the inset and shows a peak at $T = 130$ K. (b) Resistivity versus temperature measured on a strip of capped Pt(4.2 nm)/CuPc interface yielding a $T_D = (163 \pm 1)$ K with the red fit being the Bloch-Grüneisen fit that a Pt film is expected to follow. The deviation from the fit at high temperatures is plotted in the inset (blue curve), with the onset (dashed line) emerging at the dM/dT peak at $T = 130$ K. (c) Side view of hexagon-pentagon carbon units with π orbitals hybridized to Pt(111) surface atoms, leading to spin-polarized charge transfer to hexagon-pentagon units and an RKKY-mediated interaction between them through the metal surface states as suggested by previous scanning tunneling microscopy (STM) studies. (d) Square lattice of spins coupled via RKKY interaction (arrows) formed on hexagon-pentagon units within the molecules, due to spin-polarized charge transfer as suggested by previous STM studies, with the surface states of Pt (atoms depicted in brown) mediating the interaction.

II. RESULTS AND DISCUSSION

The emergence of Kondo spin lattices has been demonstrated via STM experiments for molecules deposited on Au(111) [1,2] and Ag(111) [3] surfaces, which naturally makes hybridized organic molecular interfaces with Pt(111) a candidate system (see Fig. S1, Supplemental Material [29], for XRD peak). Figure 1(a) shows the post zero-field-cooled (ZFC) and field-cooled (FC) magnetization curves as a function of temperature for a Pt(4.2 nm)/CuPc interface with an in-plane field of 20 mT. Figure S2 [29] shows a similar result for a Pt/H₂Pc interface, ruling out a mechanism related to the presence of metal ions in the molecule. At room temperature, prior to ZFC cooling, the system is weakly magnetic ($M_S = (49 \pm 1)$ emu/cm³ for Pt/CuPc—see Ref. [29], Fig. S3), which we attribute to spin-polarized charge transfer [30] from intrinsically paramagnetic Pt and an enhanced exchange interaction [23,31]. After zero-field cooling, a near-zero-magnetization (or near-zero magnetic susceptibility) state emerges below ≈ 50 K, as would be expected

for an antiferromagnetic ground state. Magnetic susceptibility is peaking at ≈ 300 K as shown in Fig. 1(a) which could be interpreted as a Néel temperature. The rate of change in the magnetic state peaks at 130 K as shown in the inset in Fig. 1(a). During field cooling, the system is already switched to a ferrimagnetic state with a low-temperature magnetization of $M_{FC} \approx 27$ emu/cm³ in a 20 mT applied field. This suggests that the measured ZFC state of zero susceptibility is metastable, as it would be expected for a frustrated antiferromagnet; hence, it would be more appropriate to refer to the peak value of magnetic susceptibility at 300 K as a spin-freezing temperature. The observation of similar results in a Pt(111)/H₂Pc interface (Supplemental Material, Fig. S2 [29]) points to strongly π - d hybridized spin-polarized hexagonal pentagon carbon units as shown in STM experiments [32]. These hybridized units have been found to be good mediators of the substrate metal electron wave functions [33]. This mechanism is depicted in Fig. 1(c), with the lattice arrangement [1–3] of spins transferred to hexagon-pentagon units shown in Fig. 1(d). Using a model of RKKY coupling

in two dimensions [34], and considering the spin-polarization to be localized on the hexagon-pentagon carbon unit [32], we estimate [35] a nearest-neighbor spin-lattice distance of 0.59 nm and a next-nearest-neighbor distance of 0.83 nm on the hexagon-pentagon units (see Supplemental Material [29], Fig. S4). It has been shown that the surface states of Pt(111) cross the Fermi level at two different parts of the Brillouin zone, along $\bar{\Gamma}$ and $\bar{\Sigma}$ [36]. Post hydrogen molecule adsorption, interfacial states were found to be present in the $\bar{\Sigma}$ direction in the Brillouin zone, hence suggesting the relevant k_F value to be one along this direction, which is 10 nm^{-1} [36]. Our structure is not bulk Pt(111) single crystal, but the structure at the surface with the molecular interface is very similar, as the high quality of the (111) texture on Pt film surface is demonstrated through Pendellösung fringes in Ref. [29], Fig. S1. Hence, taking k_F to be 10 nm^{-1} , we calculate the nearest-neighbor exchange parameter $J_1 = -0.052 J_0^2$ (where J_0 is the ferromagnetic π - d orbital exchange interaction constant [37]) and the next-nearest-neighbour exchange parameter $J_2 = -0.028 J_0^2$ (see Ref. [29], Supplemental Note 1). It is known from studies of square lattices that the J_2/J_1 ratio quantifies the degree of frustration in the system. Our value of 0.54 suggests strong frustration effects [38,39], in agreement with the metastability suggested by the magnetometry measurements in Fig. 1(a).

We investigated the Pt/molecular interface further using electrical transport measurements [Fig. 1(b)]. Single-ion Kondo effects, typically observed as an increase in the low-temperature resistivity [40], were not present. This absence is expected in the limit where RKKY interactions dominate over single-ion Kondo effects [10,41]. The resistivity vs temperature of both a Pt/CuPc interface and the Pt reference samples can be described by the Bloch-Grüneisen law (see Ref. [29], Fig. S5 and Supplemental Note 2), with a Debye temperature of $T_D = (163 \pm 1) \text{ K}$. Interestingly, there is a deviation from this behavior starting at 130 K for the Pt/CuPc interface [Fig. 1(a), inset, dashed line]. This onset seems correlated with a change in the magnetic susceptibility of the film [dashed line in Fig. 1(a)]. Chemisorbed CuPc molecules have been previously shown to be nondynamic at low temperatures [42], so we ascribe the deviation from Bloch-Grüneisen law and the finite magnetization onset at 130 K to a vibrational coupling between Pt and the hybridized molecules as the metal phonon modes are fully occupied approaching the Debye temperature. Therefore, the lower-than-expected resistance in the Pt/molecule interface can be attributed to enhanced parallel conduction across the molecule hybridized to metal which becomes comparable to that of across the Pt metal channel and is thermally activated. An Arrhenius fit to the resistivity deviation from the Bloch-Grüneisen fit yields a thermal activation energy of $(76 \pm 1) \text{ meV}$ (inset in Fig. 1(b), and Ref. [29], Fig. S6). This number is in a good agreement with the 70–120 meV energy gap previously measured in organic charge transfer interfaces [43].

An important question is whether a frustrated antiferromagnetic state could also emerge at a Pt(111)/Co(1.7 nm)–organic molecule interface. These systems show a perpendicular magnetic anisotropy at room temperature [44], and x-ray magnetic dichroism sum-rule analysis, post field cooling, has indicated a scaling relation that we attribute to the frustrated

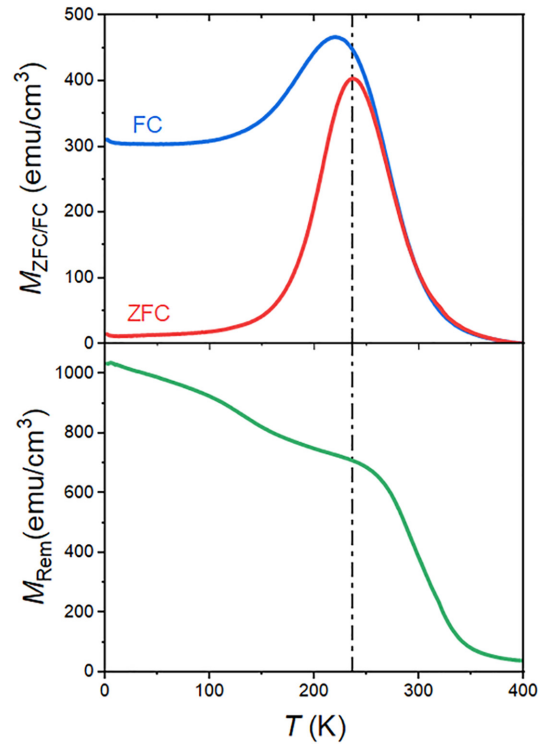


FIG. 2. In-plane frustrated antiferromagnetism onset in perpendicular magnetic anisotropy Pt/Co bilayers with a copper phthalocyanine interface. A Pt/Co(1.7 nm)/CuPc interface has an out-of-plane easy axis at high temperatures with negligible in-plane remanence after saturation. Zero-field-cooled–field-cooled in-plane magnetometry curves (upper graph, 20 mT) and in-plane remanence measured at zero field after field cooling in a 2 T field (lower graph) yield a freezing temperature of $T_f \approx 237 \text{ K}$ that coincides with the in-plane magnetization onset and higher remanent state attributed to the formation of the Kondo spin lattice.

ferromagnetic Kondo spin lattice [44]. The ZFC-FC magnetometry curves measured for a Pt/Co(1.7 nm)/CuPc interface in an in-plane magnetic field of 20 mT are shown in the upper panel of Fig. 2. A transition with $T_f = 237 \text{ K}$ is apparent, with very low magnetization measured below T_f in the ZFC curve, suggesting a compensated frustrated state, as in the case of Pt/molecule interfaces. Surface states on Co films have been studied extensively [45,46], and these states mediate the RKKY interaction between hexagon-pentagon units. Similarly to Pt/CuPc, a deviation from the Bloch-Grüneisen law is measured for Pt/Co/molecular interfaces (see Ref. [29], Fig. S7), suggesting that vibrational modes in the metal suppress the molecular lattice spin interactions above T_f . The ZFC spin-freezing peak is much sharper for Pt/Co/molecule than in Pt/molecule interfaces, which we attribute to the emergence of a perpendicular spin orientation on the Pt/Co interface above T_f [44]. The bottom panel in Fig. 2 shows the in-plane magnetic remanence measured on the same sample at zero field after a 2 T field cooling, where we can observe that the system switched to an in-plane ferromagnetic state for $T \leq T_f$ (see Ref. [29], Fig. S8, for magnetic hysteresis above and below T_f).

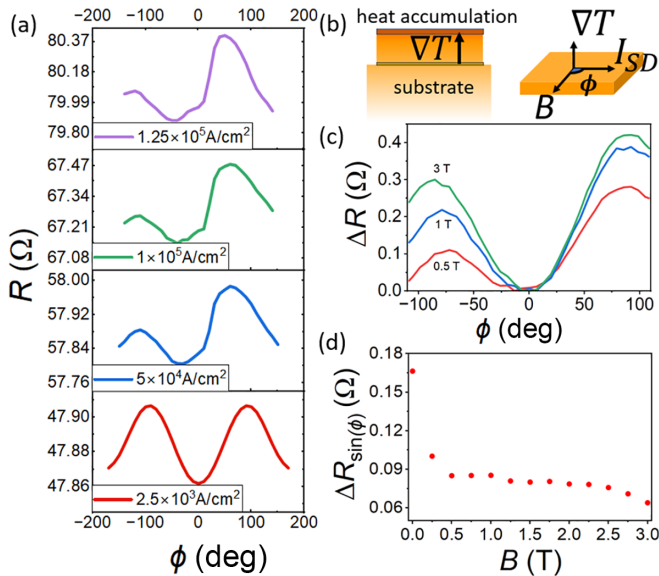


FIG. 3. Joule-heating-induced Nernst effect superimposed to anisotropic magnetoresistance on a Pt/Co/molecular interface. (a) DC measurements (averaged opposite polarities of current) of the planar anisotropic magnetoresistance (AMR) at various current densities as a function of planar angle (ϕ) between current and magnetic field, where $B = 3$ T, on a Pt/Co(1.7 nm)/CoPc metallomolecular structure at $T = 150$ K. (b) Schematically illustrated (i) Joule-heating-induced perpendicular to sample plane temperature gradient generated due to heat accumulation and (ii) source-drain current, temperature gradient, and magnetic field. (c) AMR curve measured with a current density of 6.5×10^5 A/cm² at varying magnetic fields on a Pt/Co(1.4 nm)/H₂Pc metallomolecular interface at $T = 250$ K. (d) Magnetic field dependence of fitting extracted amplitude of a Nernst-effect-induced $\sin(\phi)$ term on a H₂Pc metallomolecular interface at $T = 250$ K.

Near quantum phase transitions, away from the frustrated antiferromagnetic ground state, non-Fermi-liquid behavior is expected [8,10], and it has been found in the form of the emergence of a strange metal state [47]. A characteristic signature of a strange metal state that is widely observed in cuprates as well as Kondo spin-lattice systems is a linear temperature-dependent resistivity [41]. However, given the interfacial nature of our metallomolecular Kondo spin-lattice system, we have significant parallel conduction from Pt/Co layers and the molecular layers (above 130 K), leading to the system obeying Bloch-Grüneisen law and deviating from it above 130 K. An alternative powerful probe of emergence of a strange metal state is a manifestation of a sizable (anomalous) Nernst effect [21]. Thus, to probe the phase transition further, we studied the Joule-heating-induced Nernst effect, here superimposed on anisotropic magnetoresistance (AMR) [48–50] measurements for Pt/Co/molecular systems as shown in Fig. 3(a). The emergence of an unusual sinusoidal term [51] in the AMR curves at high current densities is due to a Nernst effect where the thermal gradient perpendicular to the sample plane is induced by Joule heating (Figs. 3(a) and 3(b), and Ref. [29], Supplemental Note 3) [48–51]. To clarify the nature of this asymmetry in the magnetoresistance with the magnetic field polarity, its magnetic field dependence was

studied [see Fig. 3(c)]. The asymmetric term was found to dominate approaching zero magnetic field in the absence of AMR, hence confirming the anomalous nature of the Nernst effect [Fig. 3(d)].

To clarify the role of the phase transition, we measured the ANE at three different temperatures (all post field cooling at 3 T): before, during, and after the phase transition, as shown in Fig. 4. This was carried out with a current density of 3.7×10^5 A/cm², which keeps the temperature increase of the sample below 20 K in Pt/Co(1.4 nm)/H₂Pc interfaces (see Ref. [29], Fig. S9, for temperature interpolation). The largest change in normalized resistance is observed at a cryostat temperature of 250 K, in the vicinity of the easy axis switching for these samples (see the inset remanence curve in Fig. 4), and suggests that the anomalous Nernst effect (ANE) is linked to the phase transition, as it would be expected for a strange metal state emerging in the vicinity of a Fermi surface transition at the metallomolecular Kondo spin-lattice interface [21,52,53].

The evolution of the anomalous Nernst voltage at different current densities measured at a cryostat temperature of 250 K is plotted in Fig. 5(a). Plotting the obtained electric fields as a function of the square of current yields a linear relationship, as shown in Fig 5(b), which further evidences the thermoelectric nature of the effect. To estimate the anomalous Nernst coefficient S_{ANE} , we performed finite element modeling (FEM) of the Joule heating (see Methods, and Ref. [29], Fig. S10). Using the FEM-modeled temperature drop across the organic layer [Fig. 5(c)] to obtain varying values of ∇T_z at different current densities, we obtained the maximum thermal gradient and therefore the lowest S_{ANE} estimates for different current densities as shown in Fig. 5(d). The anomalous Nernst coefficient S_{ANE} is calculated using the expression $E_{\text{ANE}} = S_{\text{ANE}} \nabla T_z$. We find S_{ANE} to be 3.0 $\mu\text{V/K}$ at the peak E_{ANE} value of 7 V/m, calculated using a FEM-estimated ∇T_z value of 2.3×10^6 K/m. This is considerably higher than, e.g., the values for chiral kagome antiferromagnets Mn₃Ge [54] and Mn₃Sn [55] and is comparable to systems such as the canted antiferromagnet YbMnBi₂ [56] and the iron-based binary ferromagnet Fe₃Ga [57]. Previously, an approximately 0 value of the anomalous Nernst effect ($E_{\text{ANE}} \approx 0$ V/m) had been measured on in-plane magnetized Pt/Co films in experiments utilizing a perpendicular heat gradient [48]. Contrary to our experiments, an ANE attributed to magnetization was measured utilizing a planar heat gradient and 10 repeats of a perpendicularly magnetized Pt/Co layer [58]. Here, we report a value of S_{ANE} that is approximately a factor 3 larger at a remanent field of 3 mT. Furthermore, the common orientation of the magnetization and thermal gradient [44], at a current density of 6.5×10^5 A/cm², where the maximum Nernst coefficient is measured, imply that this effect is linked to large entropy of charge carriers around the quasiparticle breakdown point or, namely, a strange metal state emergence during the phase transition at the metallomolecular interface rather than the presence of a finite magnetization.

III. CONCLUSIONS

In conclusion, we have demonstrated the emergence of frustrated spin order in metallomolecular interfaces grown on

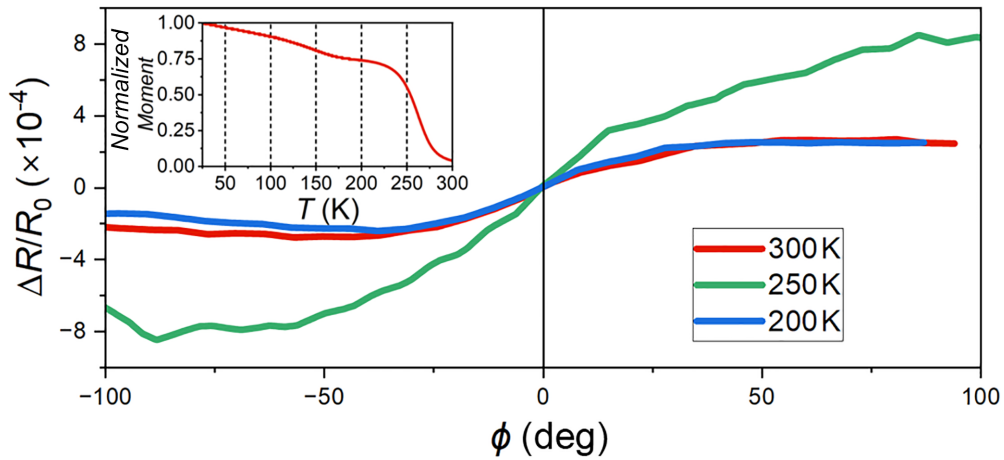


FIG. 4. Maximized anomalous Nernst effect in the vicinity of the spin-freezing transition. Normalized change in resistance as a function of angle (ϕ) between remanent planar magnetic field and current due to anomalous Nernst effect measured at three different temperatures around the easy axis switching region post field cooling at 3 T (see inset for the in-plane remanence curve for the corresponding Pt/Co(1.4 nm)/H₂Pc film) with a current density of 3.7×10^5 A/cm².

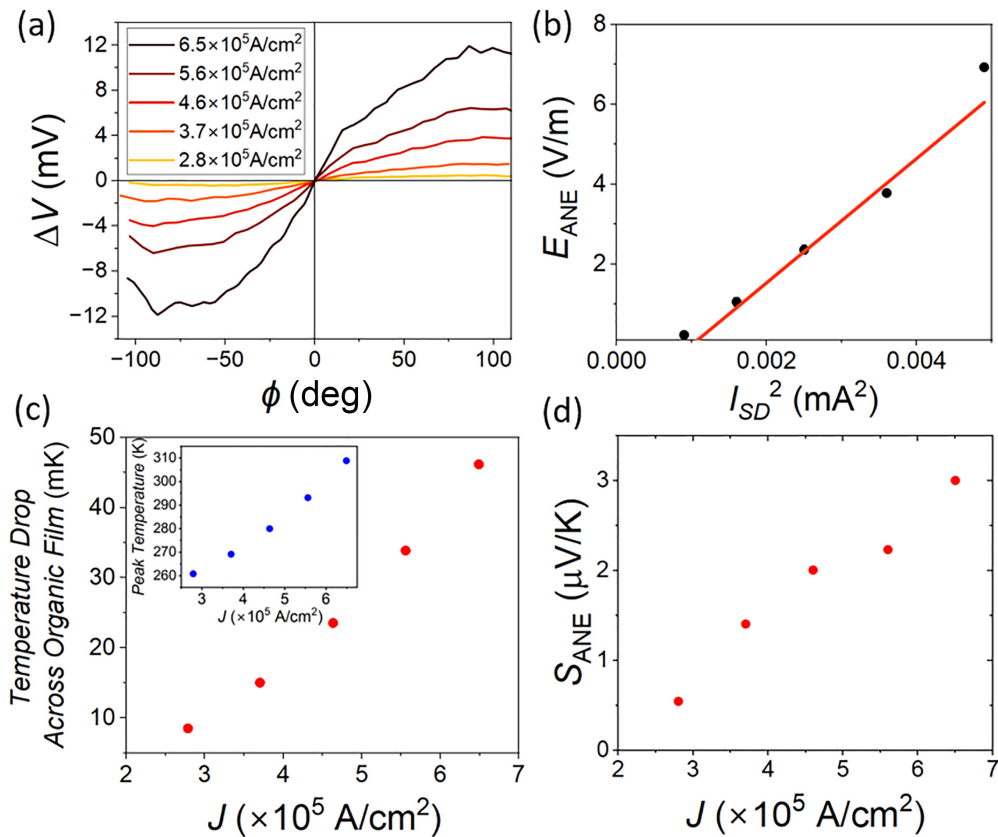


FIG. 5. Extracted Nernst coefficients for maximized Joule-heating-induced anomalous Nernst effect measured at $T = 250$ K. (a) Measured change in DC longitudinal voltage at $T = 250$ K as a function of planar angle (ϕ) between remanent magnetic field and current across a device at different Joule heating current densities. (b) Extracted anomalous Nernst effect field plotted as a function of the square of the source-drain current, with measured (E_{ANE}) values approaching 7 V/m, yielding a linear fit to the square of the source-drain current. (c) Finite-element-modeled temperature drop across the organic film as a function of current density, with peak temperature accumulating at the top of the film plotted in the inset. (d) Estimated anomalous Nernst coefficient (S_{ANE}) at $T = 250$ K as a function of current density, with maximum (S_{ANE}) being 3 μ V/K.

highly textured Pt(111) layers with and without an intrinsically magnetic sub 2-nm Co layer. The interfaces, grown in ultrahigh vacuum (10^{-10} mbar), show two-dimensional Kondo spin-lattice effects previously observed in scanning tunneling microscopy experiments. Single-ion Kondo effect [40] signatures are absent from transport measurements, as expected in the localized limit of dominating RKKY interaction between spin sites. In comparison with previous studies, the unconventional systems reported here show very high spin-freezing temperatures ranging from 240 to 300 K. A giant Joule-heating-induced ANE is observed near the spin-freezing transition, which we attribute to a strange metal state onset in the vicinity of the Fermi surface transition [28]. The enhanced thermoelectric properties lead to a S_{ANE} approaching $3 \mu\text{V/K}$ around room temperature with measured current densities of the order of 10^5 A/cm^2 . This large coefficient, comparable with colossal ANE systems [56,57] and combined with the possibility of tuning the transitions around room temperature, bring Kondo spin lattices closer to applications.

IV. METHODS

Thin film growth and characterization. Thin film structures were grown on 0.65-mm-thick *c*-plane sapphire films. The textured Pt(111) films ($\approx 4 \text{ nm}$) (see Ref. [29], Fig. S1, for the x-ray diffraction peak) were grown at 500°C with *e*-beam evaporation at a growth rate of $\approx 0.1 \text{ \AA/s}$. For samples also possessing the Co layer, the substrate was then cooled down to room temperature and the Co layer was grown also with the *e*-beam evaporation technique at a rate of $\approx 0.1 \text{ \AA/s}$. Within the same chamber, organic molecule layers were sublimed onto the Co or Pt surface at a pressure of $\approx 5 \times 10^{-10}$ mbar with a rate of $\approx 0.3 \text{ \AA/s}$ also at room temperature and until a thickness of 20 nm was reached as monitored by a quartz crystal monitor. The cap layer was magnetron sputtered on top of the thin film structure, with the material used as the cap being Cu for Pt/Co/molecule interfaces and Nb for Pt/molecule interfaces with a thickness of $\approx 15 \text{ nm}$.

Magnetometry. Magnetization measurements were carried out using a SQUID-magnetometer (MPMS Q.D.) which offers a resolution better than 10^{-8} emu accompanied by temperature control.

Electronic transport. Four-probe resistance measurements were carried out in continuous-flow cryostats on devices of narrow cleaved strips of films, with a Keithley 6221 DC and AC current source with a Keithley nanovoltmeter being used for Joule-heating-induced anomalous Nernst effect measurements.

Finite element modeling. Finite element modeling was performed using the Joule-heating multiphysics interface on the commercially available COMSOL multiphysics package. Electrically insulating boundary conditions were assumed for outer edges of the sample, except at the electrically contacted edges of the heterostructure, where a fixed-current terminal and ground were supplied. The relative electrical resistivity of the oxidized copper layer, in comparison to the other metallic layers, was increased from nominal copper resistivity until the maximum temperature gradient across the organic film was achieved. Then, the resistivity of all current-carrying layers was increased to match the average resistivity of the experimental sample. For computational efficiency, the thermal conductivity of the substrate was increased so that the equivalent thermal resistance of the sapphire bulk could be obtained with only 500-nm modeled thickness. An additional thermally resistive layer was added, below the sapphire substrate, which accounted for the any other thermal resistances present between the base of the substrate and a fixed temperature point. The base edge of this layer was held fixed at 250 K, and its thermal resistance was modified until the peak temperature of the modeled system matched that of the experimental sample (as informed by resistance versus temperature measurements). All other outer layers had thermally insulating boundary conditions.

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DATA AVAILABILITY

The data that support the findings of this article are openly available at the Research Data Repository of the University of Leeds [59].

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