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Magnetic-Field Universality of the Kondo Effect Revealed by Thermocurrent Spectroscopy

Chunwei Hsu,[1] Theo A. Costi,[2,3] David Vogel,[4] Christina Wegeberg,[4] Marcel Mayor,[4,5,6] Herre S. J. van der Zant,[1] and Pascal Gehring[1,7,*]

[1]*Kavli Institute of Nanoscience, Delft University of Technology, Lorentzweg 1, Delft 2628 CJ, The Netherlands*
[2]*Peter Grünberg Institut, Forschungszentrum Jülich, 52425 Jülich, Germany*
[3]*Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany*
[4]*Department of Chemistry, University of Basel, St. Johanns Ring 19, 4056 Basel, Switzerland*
[5]*Institute for Nanotechnology (INT), Karlsruhe Institute of Technology (KIT), P.O. Box 3640, 76021 Karlsruhe, Germany*
[6]*Lehn Institute of Functional Materials (LIFM), School of Chemistry, Sun Yat-Sen University (SYSU), 510275 Guangzhou, China*
[7]*IMCN/NAPS, Université Catholique de Louvain (UCLouvain), 1348 Louvain-la-Neuve, Belgium*

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Probing the universal low-temperature magnetic-field scaling of Kondo-correlated quantum dots via electrical conductance has proved to be experimentally challenging. Here, we show how to probe this in nonlinear thermocurrent spectroscopy applied to a molecular quantum dot in the Kondo regime. Our results demonstrate that the bias-dependent thermocurrent is a sensitive probe of universal Kondo physics, directly measures the splitting of the Kondo resonance in a magnetic field, and opens up possibilities for investigating nanosystems far from thermal and electrical equilibrium.

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Introduction.—The Kondo effect, originally describing the anomalous increase with decreasing temperature in the resistivity of nonmagnetic metals containing a small concentration of magnetic impurities [1–3], is now a ubiquitous phenomenon in physics, forming the starting point for understanding the Mott transition [4], heavy fermions [5], and transport through correlated nanostructures, such as quantum dots [6,7], molecules [8], and adatoms on surfaces [9]. In the so-called "QCD Kondo effect" [10], it also constitutes one of the first known examples of asymptotic freedom [11,12], a property of the strong interaction in particle physics.

A key feature of the Kondo effect is its universality [13–15]. For example, the temperature dependence of the linear conductance $G(T)$ of a spin-1/2 quantum dot is described by a unique universal scaling function $G(T)/G(0) = g(T/T_K)$ of T/T_K, where T is the temperature and T_K is the Kondo scale, and is used as a hallmark for establishing a spin-1/2 Kondo effect in quantum dot systems [16]. The same holds for exotic realizations of the Kondo effect [17–20], with each having its own characteristic set of universal scaling functions. Thus, universality in Kondo systems provides hallmarks for identifying the particular Kondo effect in a given experiment [18,19,21].

In this Letter, we address another aspect of universality of Kondo-correlated quantum dots, namely, the universal magnetic-field scaling in the low-temperature ($T \ll T_K$) Fermi-liquid regime of quantum dots. While our interest is in the thermocurrent, we first specify what we mean by low-temperature magnetic-field scaling in the context of the more familiar differential conductance $G(T, V_{\mathrm{sd}}) = dI/dV_{\mathrm{sd}}$ (derivative of the electrical current with respect to source-drain voltage). Specifically, for the asymmetrically coupled quantum dot device investigated in this Letter [Fig. 1(b)], described within the Anderson impurity model [Fig. 1(c)], $G(T \ll T_K, V_{\mathrm{sd}} \ll T_K)$ is given, for *arbitrary* magnetic fields B, within higher-order Fermi liquid theory [22–24] as

$$\frac{dI}{dV_{\mathrm{sd}}} \propto a_0 - c_T\left(\frac{\pi T}{T_K}\right)^2 - c\frac{V_{\mathrm{sd}}}{T_K} - c_V\left(\frac{V_{\mathrm{sd}}}{T_K}\right)^2, \quad (1)$$

with field-dependent coefficients a_0, c_T, c, and c_V [25]. The low-temperature magnetic-field scaling that we refer to is reflected in the universal dependence of the curvature coefficients $c_V(B) \propto -\partial^2 G/\partial V_{\mathrm{sd}}^2$ and $c_T(B) \propto -\partial^2 G/\partial T^2$ on B/T_K in the Kondo regime [22–24] [66]. Surprisingly, the exact dependence of c_V and c_T on magnetic field has only recently been calculated via a generalization of Nozières Fermi-liquid theory [67] to nonequilibrium and particle-hole asymmetric situations [22–24]. The results show that c_V and c_T are universal functions of magnetic field which change sign at a universal crossover field $B = B_c$ describing the onset of the splitting of the Kondo resonance in dI/dV_{sd}, in agreement with predictions for B_c for the Kondo model [68]. Nevertheless, establishing this universality and the splitting of the Kondo resonance in dI/dV_{sd} is intrinsically difficult [69–72]. Yet, both serve as

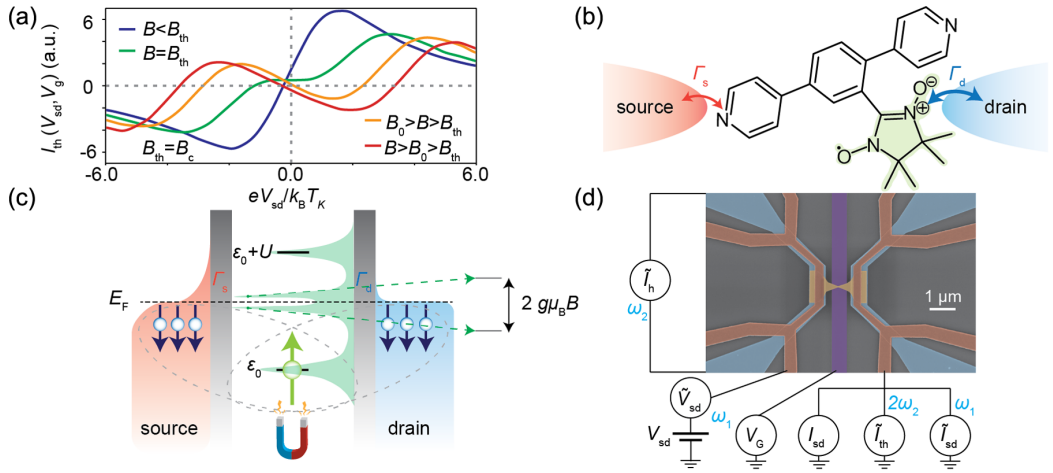

FIG. 1. (a) Sketch of $I_{\rm th}$ vs $V_{\rm sd}$ in the Kondo regime for several B. $(\partial I_{\rm th}/\partial V_{\rm sd})|_{V_{\rm sd}=0}$ is a universal function of B/T_K and changes sign (kink) at a universal field $B = B_{\rm th} = B_c$ [Fig. 4(a)], while $I_{\rm th}(V_{\rm sd} = 0)$ [73] changes sign at $B = B_0 > B_c$ and is nonuniversal [Fig. 4(a), Ref. [74] and Sec. SM.3.5.5 [25]]. (b) Molecular junction of a NNR molecule anchored to source and drain leads. (c) Anderson model of (b) in a magnetic field B. A singly occupied level ε_0 with Coulomb repulsion U and gate voltage $V_g = (\varepsilon_0 + U/2)/\Gamma$ coupled to hot and cold source and drain leads with strength $\Gamma = \Gamma_s + \Gamma_d$ gives rise to a spin-1/2 Kondo effect for $V_g \approx 0$ resulting in a Kondo resonance at the Fermi energy, E_F. The field B splits the up and down levels at ε_0 by $g\mu_B B$ and the Kondo resonance in $dI/dV_{\rm sd}$ by $2g\mu_B B$ [75,76] (for $g\mu_B B \gg k_B T_K$). A thermal bias $\Delta T > 0$ causes a thermocurrent $I_{\rm th}$ to flow between source and drain, measured as described in (d). (d) False-colored scanning electron microscopy image of the thermoelectric device. Bias and thermal voltages are generated by a dc + ac bias voltage source, $V_{\rm sd} + \tilde{V}_{\rm sd}(\omega_1)$, and an ac heater current source, $\tilde{I}_h(\omega_2)$, on the hot left lead. The resulting dc, ac electrical currents and ac thermocurrent, $I_{\rm sd}$, $\tilde{I}_{\rm sd}(\omega_1)$, and $\tilde{I}_{\rm th}(2\omega_2)$, are measured simultaneously on the cold right lead.

useful experimental hallmarks of the Kondo effect in quantum dots.

Here, we propose a different approach to address magnetic-field scaling in the strong-coupling Kondo regime of quantum dots by employing the recently developed thermocurrent spectroscopy [77]. We experimentally show that the thermocurrent, $I_{\rm th}$, of a molecular quantum dot in the Kondo regime exhibits a clear feature as a function of magnetic field, in the form of a zero-bias ($V_{\rm sd} = 0$) kink appearing for fields B larger than a certain value, which we denote by $B_{\rm th}$. We explain this behavior within higher-order Fermi-liquid theory [22,24] for $V_{\rm sd} \ll T_K$, and an approximate nonequilibrium Green's function approach [78] for $V_{\rm sd} \gtrsim T_K$. Within the former, to leading order in $V_{\rm sd}, T$ and ΔT, where ΔT is the applied thermal bias, we find in the low-temperature strong-coupling regime $\Delta T \ll T \ll T_K$,

$$I_{\rm th}(T, V_{\rm sd}) = \gamma \frac{\pi^2 k_B^2}{3} T \Delta T [s_0(B) + s_1(B) V_{\rm sd}], \quad (2)$$

with constant γ and coefficients $s_0(B)$ and $s_1(B)$ [25]. Remarkably, we show that, (i), $s_1(B)/s_1(0)$ and $c_V(B)/c_V(0)$ are described by essentially the same universal scaling function in the Kondo regime, showing that $(dI_{\rm th}/dV_{\rm sd})|_{V_{\rm sd}=0}$ [$\propto s_1(B)$] probes magnetic-field universality, and, (ii), $B_{\rm th}$ coincides with B_c, thus demonstrating that thermocurrent spectroscopy provides a new route to directly probe the splitting of the Kondo resonance [68] and extract the universal field $B_c = B_{\rm th}$. Our findings are concisely summarized in the sketch in Fig. 1(a). We note, that in contrast to the zero-bias thermocurrent slope, the zero-bias thermocurrent, $I_{\rm th}(T, V_{\rm sd} = 0)$ [$\propto s_0(B)$], measured in Ref. [73] as a function of gate voltage (V_g) and magnetic field and found to change sign at a certain field B_0, is nonuniversal [Fig. 4(a), Ref. [74] and Sec. SM.3.5.5 in the Supplemental Material [25]]. Thus $I_{\rm th}(T, V_{\rm sd} = 0)$ does not provide a hallmark for the splitting of the Kondo resonance and cannot be used to extract B_c, in contrast to the thermocurrent spectroscopy proposed in this Letter.

Experiment and results.—The experiment is carried out on a molecular quantum dot consisting of an organic radical molecule [nitronyl nitroxide radical (NNR)] made up of a backbone and a nitronyl-nitroxide side group where an unpaired electron resides as shown in Fig. 1(b). Such free radical molecules are model systems to study the spin-1/2 Kondo effect [Fig. 1(c)] [79–81]. Furthermore, their asymmetric structure and the additional pyridine anchoring sites allow us to achieve asymmetric and strong coupling between the source and drain leads and the molecule [quantified by couplings Γ_s and Γ_d, Fig 1(b)]. We form a NNR-molecule quantum dot in the thermoelectric device shown in Fig. 1(d) by immersing electromigrated nanogaps in the molecular solution [82]. The thermoelectric device incorporates a local backgate and two microheaters in direct thermal contact with the source and drain leads [see Fig. 1(d) and Sec. SM.1.1 [25]].

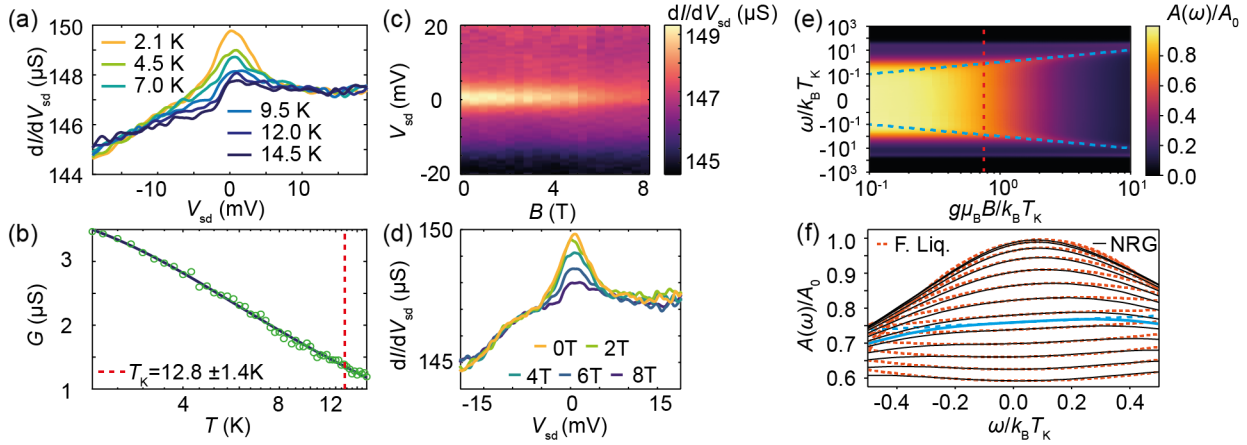

FIG. 2. (a) dI/dV_{sd}, of the molecular quantum dot vs V_{sd} at different temperatures. (b) Peak conductance of Kondo resonance vs temperature and fit to a spin-1/2 Kondo model yielding $T_K \approx 12.8$ K, see Secs. SM.1.3 and SM.2.3. (c)–(d) dI/dV_{sd} vs V_{sd} at different magnetic fields. (e) NRG Spectral function $A(\omega)/A_0$ with $A_0 = 1/2\pi\Gamma$ of the Anderson model vs energy (ω) and magnetic field for $V_g = (\varepsilon_0 + U/2)/\Gamma = -1$ and $U/\Gamma = 8$. Vertical dashed line: splitting field $B_c = 0.75 k_B T_K/g\mu_B$. Blue dashed lines: positions $[\omega(B) = \pm g\mu_B B]$ of the split Kondo peaks in $A(\omega)$ for $B \gg B_c$ [75,76]. (f) $A(\omega)/A_0$ from line cuts in (e) at $g\mu_B B/k_B T_K = 0, 0.1, \ldots, 0.7, 0.75, 0.8, \ldots, 1.2$ (solid lines), compared to $A(\omega)/A_0$ from Fermi-liquid theory (dashed lines). Blue lines: $B = B_c$. A g factor of $g = 2$ is used, as measured by electron paramagnetic resonance (Sec. SM.4.2 [25]).

Evidence for a Kondo effect is shown by the strong suppression of the zero-bias peak in the measured dI/dV_{sd}, both as a function of increasing T [Figs. 2(a)–2(b)] and B [Figs. 2(c)–2(d)]. The T dependence of the zero-bias peak height [Fig. 2(b)] is well described by the numerical renormalization group (NRG) conductance of a spin-1/2 Kondo model and yields $T_K = 12.8$ K (Sec. SM.1.3 [25]). Based on the structure of the molecule, an asymmetric coupling is expected. Assuming, $\Gamma_d \gg \Gamma_s$ (see Sec. SM.3.5.3 in Ref. [25] for $\Gamma_d \ll \Gamma_s$), we find $\Gamma_s/\Gamma_d \approx 0.017$. An underscreened Kondo effect [18,19], requiring a larger molecular spin ($S > 1/2$), is excluded, since such an effect results in a split Kondo resonance in dI/dV_{sd} starting already at zero field, which is not observed in Figs. 2(c)–2(d). Thus, a single-level Anderson model describing a $S = 1/2$ Kondo effect [Fig. 1(c)] is justified by the data. In the remainder of this Letter, the base temperature is kept at $T \approx 2$ K $\ll T_K$ while the thermocurrent is measured for a small thermal bias $\Delta T \approx 0.6$ K $\ll T \ll T_K$ so that we probe the strongly- coupled Kondo regime (see Secs. SM.2.4-5 and Secs. SM.3.6.4-5 of the Supplemental Material [25] for thermal bias and temperature effects).

A closer look at the field dependence of dI/dV_{sd} in Figs. 2(c)–2(d), indicates that the expected splitting of the Kondo peak at $B_c \approx 7.15$ T [83] is not observed. This is in part due to a large non-Kondo (field and temperature independent) contribution in Figs. 2(a) and 2(d), which may mask the appearance of a splitting at zero bias. In addition, the largest field used, $B = 8$ T, was only marginally larger than B_c. For a device where higher fields relative to B_c could be applied, such a splitting is observed (Sec. SM.2.1 [25]). Despite these complications in extracting B_c from dI/dV_{sd} for the device studied, there is also a general problem in doing so, which can be appreciated by attempting this from exact theoretical results. This is illustrated in Figs. 2(e)–2(f) which show the spectral function $A(\omega = eV_{\mathrm{sd}}) \sim dI/dV_{\mathrm{sd}}$ within the NRG method and within Fermi-liquid theory. While the precise value of B_c is impossible to determine visually in Fig. 2(e) (vertical dashed line), it can be deduced from the line cuts in Fig. 2(f) as the field where the curvature of $A(\omega)$ vanishes. However, such accuracy in second derivatives of $A(\omega = eV_{\mathrm{sd}}) \sim dI/dV_{\mathrm{sd}}$ is difficult to attain from experimental data with finite error bars.

Thermocurrent spectroscopy resolves the above difficulty. Figures 3(a)–3(b) show the measured thermocurrent

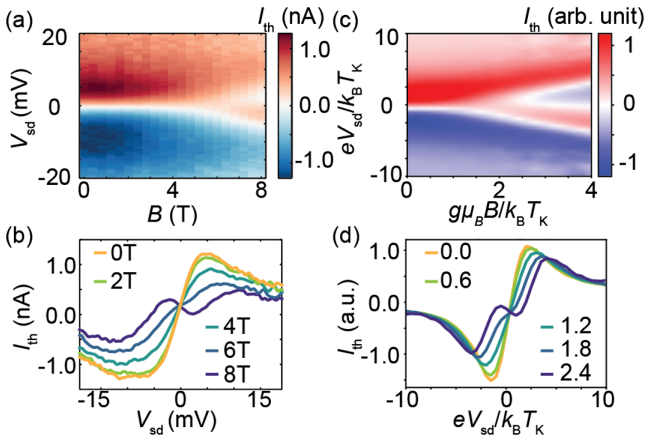

FIG. 3. (a)–(b) Measured I_{th}, vs V_{sd} at different magnetic fields. (c)–(d) Calculated I_{th} vs V_{sd} at different magnetic fields $g\mu_B B/k_B T_K$ for the Anderson model in Fig. 1(c) with $V_g = -2.5$, $U/\Gamma = 8$, $\Delta T/T_K = 0.2$, and $T/T_K = 0.5$.

versus bias voltage and magnetic field, while Figs. 3(c)–3(d) show analogous theory results within an approximate nonequilibrium Green's function approach (Sec. SM.3.6 [25]). First, notice that the large non-Kondo contribution to the differential conductance [Figs. 2(a) and 2(d)], is absent in the thermocurrent Fig. 3(b), with the latter being largely symmetric in magnitude around zero bias, in agreement with theory [Fig. 3(d)]. This is because the thermocurrent effectively measures a difference of electrical currents (in the presence or absence of thermal bias), and thus filters out the non-Kondo contributions. By the same token the thermocurrent therefore probes universal aspects of Kondo physics more precisely than the differential conductance. Second, we now see a clear feature, in the form of a zero-bias kink with a negative slope of the thermocurrent, appearing in $I_{\rm th}$ at a field $B = B_{\rm th}$. This is qualitatively captured, together with the behavior at $V_{\rm sd} \gtrsim T_K$, by the approximate approach [Figs. 3(c) and 3(d)]. However, the precise field at which this feature occurs and its connection to B_c requires a more exact theory, which is provided by the higher-order Fermi-liquid theory [Eq. (2) and Sec. SM.3.5 [25]]. Preempting the result of this theory, we note that analyzing the experimental data in Figs. 3(a) and 3(b) for the slope of the thermocurrent $dI_{\rm th}/dV_{\rm sd}(V_{\rm sd} = 0)$ at zero-bias voltage as a function of magnetic field, we find that this slope vanishes (i.e., the kink appears) at $B_{\rm th} \approx 6.6$ T. This value is within 10% of the expected $B_{\rm c} \approx 7.15$ T and already suggests that $B_{\rm th} = B_c$, and, hence, that the splitting of the Kondo resonance can be directly measured in the bias voltage dependence of $I_{\rm th}(V_{\rm sd})$.

Equation (2), with $s_i(B)$, $i = 0, 1$ evaluated exactly for all B within the NRG (Sec. SM.3.5 [25]), allows us to address the experimentally observed sign change of $[\partial I_{\rm th}(V)/\partial V_{\rm sd}]_{V=0} \propto s_1(B)$ upon increasing B above $B_{\rm th}$ (the "kink") and to extract $B_{\rm th}$. Figure 4(a) shows the normalized zero-bias thermocurrent slope $\propto s_1(B)/s_1(0)$, the normalized zero-bias thermocurrent $\propto s_0(B)/s_0(0)$, and the normalized curvature coefficient $\propto c_V(B)/c_V(0)$ as a function of B and for a range of V_g in the Kondo regime. First, notice that both $c_V(B)/c_V(0)$ and $s_1(B)/s_1(0)$ [in contrast to $s_0(B)/s_0(0)$] are universal scaling functions of $g\mu_B B/k_B T_K$ with only a weak dependence on V_g [inset Fig. 4(a)], and while distinct, they lie within about 1% of each other [inset Fig. 4(a) and Fig. S13 [25]]. Thus, measuring the field dependence of $s_1(B)$ via thermocurrent spectroscopy, requiring only a first derivative with respect to bias voltage, equivalently probes the magnetic-field universality from an electrical conductance measurement, which, however, requires a second derivative with respect to bias voltage and is consequently less accurate. Furthermore, since both $s_1(B)$ and $c_V(B)$ change sign at the same magnetic field, i.e., $B_{\rm th} = B_c \approx 0.75 k_B T_K/\mu_B$, thermocurrent measurements of Kondo correlated quantum dots at finite bias voltage provide a new way to determine the splitting of the Kondo resonance via a sign change in the slope of the thermocurrent with respect to bias voltage.

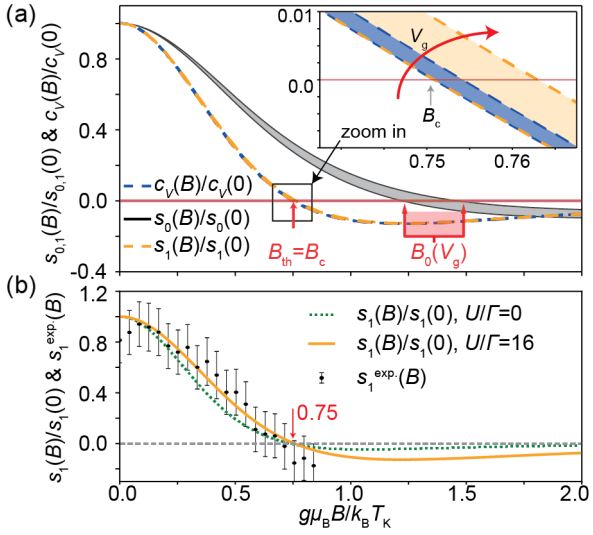

FIG. 4. (a) Normalized zero-bias thermocurrent slope $s_1(B)/s_1(0)$ and curvature coefficient $c_V(B)/c_V(0)$ vs $g\mu_B B/k_B T_K$, for gate voltages $1 \le V_g = (\varepsilon_0 + U/2)/\Gamma \le 5$ in the Kondo regime exhibiting scaling collapse [Anderson model in Fig. 1(c) with $U/\Gamma = 16$]. Also shown is the nonuniversal normalized zero-bias thermocurrent $s_0(B)/s_0(0)$ with a sign change at a strongly V_g-dependent B_0. Inset: $c_V(B)/c_V(0)$ and $s_1(B)/s_1(0)$ in the region around $B = B_{\rm th} = B_c \approx 0.75 k_B T_K/g\mu_B$, and their V_g dependence. (b) Least-squares fit of the experimental zero-bias thermocurrent slope $s_1^{\rm exp.}(B)$ to the universal curve for $s_1(B)/s_1(0)$. Error bars denote 1σ confidence intervals (see Sec. SM.1.4 [25]). A fit to the noninteracting case with $U = 0$ and $\varepsilon_0 = -0.1\Gamma$ (green dotted line) yielded an rms deviation that was 194% larger than for the Kondo scaling curve. The estimated experimental $B_{\rm th} \approx 0.69 k_B T_K/g\mu_B$ is close to theory ($0.75 k_B T_K/g\mu_B$).

In Fig. 4(b) we show a direct comparison between theory and experiment for the slope of the zero-bias thermocurrent as a function of magnetic field. The experimental data follows well the universal curve for $s_1(B)/s_1(0)$, and the aforementioned value extracted from this data for $B_{\rm th} \approx 6.6$ T ($g\mu_B B_{\rm th}/k_B T_K = 0.69$), is consistent with the expected splitting field of $B_{\rm th} \approx 7.15$ T ($g\mu_B B_{\rm th}/k_B T_K \approx 0.75$). The largest available field, 8 T, did not allow accessing the minimum of the $s_1(B)$ vs B curve or the slow increase of $s_1(B)$ to zero at $B \gg T_K$. The agreement between theory and experiment at the largest fields measured $B > B_{\rm th}$ is reduced, but still within the error bounds of the experimental data. The extracted $B_c = B_{\rm th}$ from the thermocurrent validates the theory prediction with higher accuracy than has so far been reported (see Sec. SM.3.5.2 [25]). The large energy level separation in a molecular quantum dot grants the observed good agreement between the theory and experiment, even under a simple single-level assumption in the transport window [Fig. 1(c)].

Conclusion.—In summary, we have studied the effect of a magnetic field on a Kondo-correlated molecular quantum dot via nonlinear thermocurrent spectroscopy.

We demonstrated theoretically and confirmed experimentally, that the nonequilibrium thermocurrent, via its zero-bias slope $s_1(B)$, exhibits universal Fermi-liquid magnetic-field scaling, and that the vanishing of $s_1(B)$ at $B = B_{\mathrm{th}}$ with $B_{\mathrm{th}} = B_c$, directly probes the splitting of the Kondo resonance. Since the thermocurrent is largely robust against parasitic conductive phenomena, it provides a more clear cut signature of this hallmark than is available from conductance measurements only. The ability to tune thermal and voltage bias, as well as temperature and magnetic field, opens up possibilities for using thermocurrent spectroscopy to yield insights into Kondo physics of nanoscale systems and may prompt theoretical investigations to address the largely unexplored area of nanosystems far from thermal and electrical equilibrium.

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*pascal.gehring@uclouvain.be

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[25] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.128.147701 for derivation of Eqs. (1) and (2); expressions for $a_0(B)$, $c_T(B)$, $c(B)$, $c_V(B)$, and $s_0(B)$, $s_1(B)$; evaluations; further experimental details; fitting procedures; synthesis and characterization; results at finite temperature or thermal bias; and including Refs. [26–65].
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