Rapid Communications

Magnetic field dependence of the thermopower of Kondo-correlated quantum dots

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Recent experiments have measured the signatures of the Kondo effect in the zero-field thermopower of strongly correlated quantum dots [Svilans *et al.*, Phys. Rev. Lett. **121**, 206801 (2018); Dutta *et al.*, Nano Lett. **19**, 506 (2019)]. They confirm the predicted Kondo-induced sign change in the thermopower, upon increasing the temperature through a gate-voltage dependent value $T_1 \gtrsim T_K$, where T_K is the Kondo temperature. Here, we use the numerical renormalization group (NRG) method to investigate the effect of a finite magnetic field B on the thermopower of such quantum dots. We show that, for fields B exceeding a gate-voltage dependent value B_0, an additional sign change takes place in the Kondo regime at a temperature $T_0(B \geqslant B_0) > 0$ with $T_0 < T_1$. The field B_0 is comparable to, but larger than, the field B_c at which the zero-temperature spectral function splits in a magnetic field. The validity of the NRG results for B_0 are checked by comparison with asymptotically exact higher-order Fermi-liquid calculations [Oguri and Hewson, Phys. Rev. B **97**, 035435 (2018)]. Our calculations clarify the field-dependent signatures of the Kondo effect in the thermopower of Kondo-correlated quantum dots and explain the recently measured trends in the B-field dependence of the thermoelectric response of such systems [Svilans *et al.*, Phys. Rev. Lett. **121**, 206801 (2018)].

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Introduction. Understanding the thermoelectric transport through gate-tunable Kondo-correlated molecules [1–3], adatoms [4–9], and semiconductor quantum dots [10–14] poses both experimental and theoretical challenges [15–17]. While electrical conductance measurements on nanosystems are standard, applying a quantifiable temperature gradient, and measuring the resulting thermovoltage, across such small systems is experimentally challenging [18–21]. On the theoretical side a description of the Kondo-induced transport at low temperatures [22–24], requires, in general, the use of nonperturbative methods [25–32], and, moreover, the above systems can be routinely driven out of equilibrium [33–35], thus posing additional challenges, such as the description of dissipative processes in the nonequilibrium Kondo effect [36–48]. In this Rapid Communication, motivated by a recent experiment [49], we focus on the magnetic field dependence of the thermopower of Kondo-correlated quantum dots, which, unlike the magnetoconductance [50–52], has received almost no theoretical attention [53–56], although it also exhibits, as we shall show, marked signatures of the Kondo effect.

Specifically, we focus on a gate-tunable quantum dot described by an Anderson impurity model. In zero magnetic field, both its thermopower, $S(T)$, and electrical conductance, $G(T)$, have been thoroughly investigated as a function of temperature and gate voltage, and characteristic signatures of the Kondo effect have been identified in both $G(T)$ [22–24,57] and $S(T)$ [58]. A hallmark of the Kondo effect in $G(T)$ is the lifting of the Coulomb blockade at low temperatures T below the Kondo scale T_K [22–24,57], which has been verified in many experiments [10–14], and reflects the development of the Kondo resonance at the Fermi level upon decreasing temperature. Signatures of the Kondo effect in the zero-field thermopower are more subtle, since the thermopower probes the asymmetry of the Kondo resonance about the Fermi level, and thus, its sign reflects the relative importance, at any given temperature T, of electron- or holelike contributions to the transport integrals in the definition of $S(T)$. It has been found theoretically [58], and verified in recent experiments [49,59], that a hallmark of the Kondo effect in the zero-field thermopower, is a sign change at a characteristic temperature $T_1 \gtrsim T_K$, which is absent in the other regimes [58,59]. Given the above, and since it is well known that a magnetic field has a large effect on the Kondo resonance [60], the question we ask in this Rapid Communication is whether a magnetic field gives rise to additional characteristic signatures of the Kondo effect in the thermopower of quantum dots? We show that this is the case and provide an interpretation of the recent experiment of Svilans *et al.* [49].

Model and transport calculations. We describe the thermoelectric transport through a strongly correlated quantum dot within a single level Anderson impurity model, $H = H_{\rm dot} + H_{\rm leads} + H_{\rm tunneling}$. Here, $H_{\rm dot} = \sum_\sigma \varepsilon_0 n_{0\sigma} - g\mu_B B S_z + U n_{0\uparrow} n_{0\downarrow}$ describes the quantum dot with energy level ε_0 and local Coulomb repulsion U in a magnetic field B, with $S_z = \frac{1}{2}(n_{0\uparrow} - n_{0\downarrow})$. $H_{\rm leads} = \sum_{k\alpha=L,R\sigma} \epsilon_{k\alpha} c^\dagger_{k\alpha\sigma} c_{k\alpha\sigma}$ describes conduction electron leads ($\alpha = L, R$), with kinetic energies $\epsilon_{k\alpha}$, and $H_{\rm tunneling} = \sum_{k\alpha\sigma} t_\alpha (c^\dagger_{k\alpha\sigma} d_\sigma + d^\dagger_\sigma c_{k\alpha\sigma})$ describes the tunneling of electrons from the leads to the dot with amplitudes $t_{\alpha=L,R}$. In the above, $n_{0\sigma} = d^\dagger_\sigma d_\sigma$ is the number operator for electrons on the dot, $d^\dagger_\sigma$ (d_σ) and $c^\dagger_{k\alpha\sigma}$ ($c_{k\alpha\sigma}$) are electron creation (annihilation) operators, and we assume a constant density of states, $\rho_\alpha(\omega) = \sum_k \delta(\omega - \varepsilon_{k\alpha}) = 1/(2D) \equiv N_F$ for both leads, with $D = 1$ the half-bandwidth. The strength of correlations is characterized by U/Γ, where $\Gamma = 2\pi N_F(t_L^2 + t_R^2)$ is the tunneling rate, taken throughout as $\Gamma = 0.002D$. We

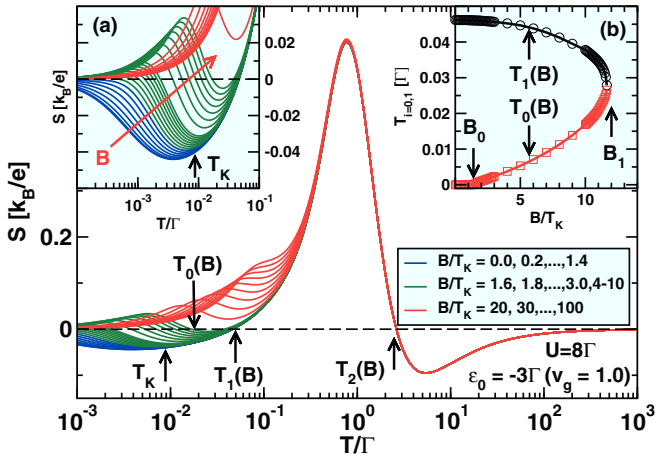

FIG. 1. Thermopower S (in units of $k_B/e = 86.17\ \mu$V/K) vs temperature T/Γ of a Kondo-correlated quantum dot for increasing values of the magnetic field B/T_K [$U = 8\Gamma$, $\varepsilon_0 = -3\Gamma$ ($v_g = 1.0$), $T_K/\Gamma = 4.32 \times 10^{-3}$]. For $B < B_0 \approx 1.45T_K$ (blue solid lines), two sign changes are found at $T_1(B)$ and $T_2(B)$, whereas for $B_0 < B < B_1$ (green solid lines) an additional sign change occurs at a temperature $T_0(B)$, and for $B > B_1$ (red solid lines), only the sign change at $T_2(B)$ is present. Inset (a): Evolution of the Kondo-induced thermopower peak with increasing B (red arrow). Inset (b): $T_0(B)$ and $T_1(B)$ vs B/T_K. B_0 and B_1 are also indicated. NRG parameters: discretization parameter $\Lambda = 4$, z averaging [67,68] with $N_z = 4$, retaining $N_{\text{states}} = 900$ states.

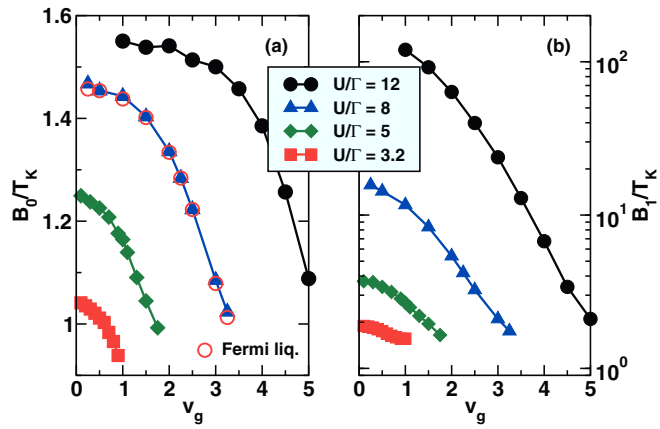

FIG. 2. NRG results (filled symbols) for (a) B_0/T_K vs v_g and (b) B_1/T_K vs v_g (in the Kondo regime), for a range of U/Γ. Open circles in (a): Fermi-liquid result for B_0/T_K at $U/\Gamma = 8$. NRG parameters as in Fig. 1.

solve H using the numerical renormalization group (NRG) technique [25–28,61], and exemplify results primarily for $U/\Gamma = 8$, or for $U/\Gamma = 3.2$, relevant to the experiment [49]. With the dimensionless gate voltage $v_g \equiv (\varepsilon_0 + U/2)/\Gamma$, the particle-hole symmetric (or midvalley) point $\varepsilon_0 = -U/2$, where $n_0 = \sum_\sigma n_{0\sigma} = 1$, occurs at $v_g = 0$. The Kondo scale, T_K, is obtained from the $T = B = 0$ spin susceptibility χ_0 via $\chi_0 = g\mu_B/4k_BT_K$, and is comparable to $k_BT_{K1}/\Gamma = \sqrt{U/4\Gamma}\exp(-\pi|\varepsilon_0||\varepsilon_0 + U|/\Gamma U)$ from perturbative scaling [62,63]. The thermopower $S(T) = -I_1/|e|TI_0$ [58,64,65] is calculated by evaluating the transport integrals $I_{m=0,1} = \gamma\int_{-\infty}^{+\infty} d\omega(-\partial f/\partial\omega)\omega^m A(\omega, T)$, with $\gamma = \pi\Gamma/2h$, directly from the discrete Lehmann representation of $A(\omega, T) = \sum_\sigma A_\sigma(\omega, T)$ [66], where $A_\sigma(\omega, T)$ is the spin-resolved local level spectral function. In the following, we focus on $v_g > 0$. Results for $v_g < 0$ follow by particle-hole symmetry: $S_{-v_g}(T) = -S_{+v_g}(T)$.

Thermopower in a magnetic field. Figure 1 shows the effect of a magnetic field on the temperature dependence of the thermopower for typical parameters in the Kondo regime [69]. It is useful to first briefly recapitulate the behavior of $S(T)$ at $B = 0$ [58] (also shown in Fig. 1): for $v_g > 0$ the thermopower, $S(T)$, exhibits a (negative) Kondo-induced thermopower peak at $T \approx T_K$ and two sign changes at the gate-voltage dependent temperatures $T_1 \gtrsim T_K$ and $T_2 \gtrsim \Gamma$, which are characteristic of the Kondo regime, and are absent in the other regimes, where $S(T)$ is of one sign [58]. Unlike T_K, neither T_1 nor T_2 are low-energy scales, since they are not exponentially small in U/Γ [58,59]. They are nevertheless closely connected to Kondo physics [58,59]. For example, the sign change at T_1 results from a rearrangement of spectral weight in the asymmetrically located Kondo resonance with increasing temperature [59].

For $B > 0$, the above picture is modified as follows: initially, for low fields $B \lesssim T_K$, the thermopower $S(T)$ has a similar temperature dependence as for $B = 0$, with two sign changes at $T_1(B)$ and $T_2(B)$, where $T_1(B)$ and $T_2(B)$ are the finite-B analogs of the two temperatures T_1 and T_2 where $S(T)$ changes sign at $B = 0$. The main effect of B on $S(T)$ in this low-field limit is to shift the Kondo-induced peak in $S(T)$ at $T \approx T_K$ to higher temperatures and to reduce it in amplitude with increasing B, while leaving its sign unchanged [see Fig. 1(a)], a trend also seen experimentally in heavy fermion systems [70]. Once B exceeds a gate-voltage dependent value, B_0, the thermopower exhibits an additional sign change at a temperature $T_0(B) < T_1(B) < T_2(B)$. While $T_0(B)$ and $T_1(B)$ have a significant B dependence [Fig. 1(b)], $T_2(B)$ (for the present parameters) is essentially B independent (Fig. 1). Further increasing B towards the gate-voltage dependent value B_1 results in a merging of $T_0(B)$ and $T_1(B)$ to a common value at $B = B_1$ [Fig. 1(b)]. For the parameters in Fig. 2, for example, we have $T_0(B_1) = T_1(B_1) \approx 0.027\Gamma$ and $B_1 \approx 12T_K \gg B_0 \approx 1.45T_K$. For still larger B values $B > B_1$ (and for v_g still in the Kondo regime), only the sign change at T_2 remains. Thus, in the Kondo regime, a sign change in $S(T)$ at $T = T_0(B)$ for $B_0 < B < B_1$ is an additional characteristic feature of the Kondo effect in $S(T)$.

Of particular interest for experiments are the magnitudes of B_0 and B_1 for quantum dots with different values of U/Γ. These are shown in Figs. 2(a) and 2(b), respectively, for gate voltages in the Kondo regime. While B_0 is of order T_K for all values of U/Γ, B_1 is typically much larger, being of order T_1 for $U/\Gamma \gg 1$, and only approaching values of order T_K for smaller values of U/Γ [71]: e.g., for the moderately correlated quantum dot of the experiment [49], with $U/\Gamma = 3.2$, we have that $1.5T_K \lesssim B_1 \lesssim 2.0T_K$ and $0.94T_K \lesssim B_0 \lesssim 1.04T_K$ for gate voltages in the Kondo regime. The value of B_0 can also be extracted from Fermi-liquid theory, since the opposite signs of the thermopower for $B < B_0$ and $B > B_0$ persist to asymptotically low temperatures $T \ll T_K$ [Fig. 1(a)]. At such low

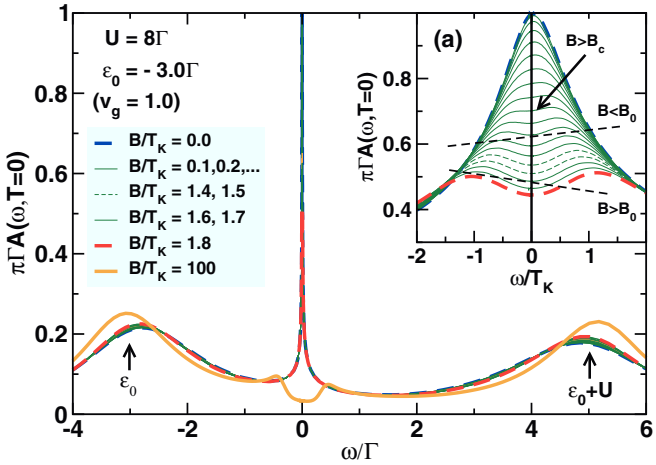

FIG. 3. Spectral function $\pi\Gamma A(\omega, T = 0)$ vs ω/Γ for increasing B/T_K in the Kondo regime. Inset (a): Evolution of the low-energy Kondo resonance with increasing B, showing that it splits at a field $B_c \approx 0.75T_K$ below the field $B_0 \approx 1.45T_K$ at which the slope of $\pi\Gamma A(\omega, T = 0)$ at the Fermi level [and hence $S(T \to 0)$] changes sign. Dashed lines indicate the slope of $\pi\Gamma A(\omega, T = 0)$ at the Fermi level for $B < B_0$ and $B > B_0$. All parameters as in Fig. 1.

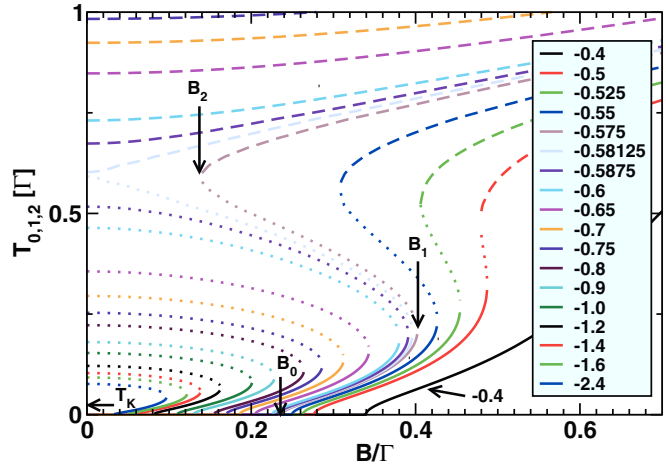

FIG. 4. $T_{i=0,1,2}(B)$ vs B/Γ for $U/\Gamma = 5$ and different ε_0/Γ, listed in the legend, and ranging from $\varepsilon_0/\Gamma = -2.4$ ($v_g = 0.1$) in the Kondo regime, to $\varepsilon_0/\Gamma = -0.4$ ($v_g = 2.1$) in the mixed valence regime. Solid lines: $T_0(B)$; dotted lines: $T_1(B)$; dashed lines: $T_2(B)$. Values of B_0, B_1, and B_2 are indicated for $\varepsilon_0/\Gamma = -0.575$ in the "weak Kondo regime." Horizontal arrow: midvalley T_K.

temperatures, a Sommerfeld expansion for the thermopower gives

$$S(T) \approx -\frac{k_B}{|e|}\frac{\pi^2}{3}k_B T \frac{1}{A(0,0)}\frac{\partial}{\partial\omega}A(\omega, T = 0)|_{\omega=0}, \qquad (1)$$

which also makes clear the physical significance of B_0 as the field where the slope of the spectral function at the Fermi level changes sign. Making use of the Fermi-liquid expressions for $A(\omega, T)$ to leading order in ω and T from Ref. [48], we evaluate the above expression and find that the sign of the low-temperature thermopower is determined by the factor $s(B) = \sum_\sigma \sin(2\delta_\sigma)\chi_{\sigma\sigma}(T = 0)/\sum_\sigma \sin^2(\delta_\sigma)$, where $\delta_\sigma = \pi n_{0\sigma}(0)$ is the spin $\sigma = \uparrow, \downarrow$ conduction electron phase shift, and $\chi_{\sigma\sigma}(T)$ is a static susceptibility defined by $\chi_{\sigma\sigma}(T) = \int_0^{1/T} d\tau \langle n_{0\sigma}(\tau)n_{0\sigma}(0)\rangle$. The latter quantities can be evaluated essentially exactly within the NRG for arbitrary B, and thereby allow B_0 to be extracted via $s(B = B_0) = 0$. A comparison between the NRG and the Fermi-liquid results for B_0 vs v_g is shown in Fig. 2 for the case of $U/\Gamma = 8$. The excellent agreement provides a check on the validity of the NRG results for S.

Experiments on quantum dots often measure the field $B_c \approx 0.75T_K$ at which the Kondo resonance splits in a magnetic field [local minimum in $A(\omega, T = 0)$ with positive second derivative at $\omega = 0$]. In contrast, B_0 represents the field at which $S(T \to 0)$ changes sign [resulting from a change in sign of the slope of $A(\omega, T = 0)$ at $\omega = 0$; see Eq. (1)]. How do B_c and B_0 compare? We find that B_0, while being of order T_K, is generally larger than the field $B_c \approx 0.75T_K$ at which the Kondo resonance splits in a magnetic field [see Fig. 2(a)]. To illustrate this, we show in Fig. 3 the $T = 0$ NRG spectral function for increasing B values [72–75]. The detailed behavior of the low-energy Kondo resonance in a magnetic field in Fig. 3(a) shows that the value of B_0 exceeds $B_c \approx 0.75T_K$ [76]. For $U/\Gamma \gg 1$, we see from Fig. 2(a) that B_0 can be up to twice as large as B_c, whereas for $U/\Gamma = 3.2$, relevant to the experiment of Ref. [49], B_0 can be up to 30%–40% larger than B_c.

Finally, since the essential signatures of the Kondo effect in the thermopower are the sign changes at the temperatures $T_0(B)$, $T_1(B)$, and $T_2(B)$, we present in Fig. 4, for $U/\Gamma = 5$, their detailed evolution with magnetic field and local level position (i.e., gate voltage) in the Kondo regime and show how this evolution is modified upon entry to the mixed valence regime at $\varepsilon_0/\Gamma \approx -0.5$. As outlined in the context of Fig 1(a), deep in the Kondo regime, $T_0(B)$ [$T_1(B)$] increase (decrease) with increasing B up until $B = B_1$ when $T_0(B)$ and $T_1(B)$ merge, while for $B > B_1$ only the sign change at $T_2(B)$ remains. Outside the Kondo regime, $\varepsilon_0/\Gamma \gtrsim -0.5$, the temperatures $T_0(B)$, $T_1(B)$, and $T_2(B)$ coalesce into a single temperature $T_0(B)$. For a narrow range of level positions, $-0.6 \lesssim \varepsilon_0/\Gamma \lesssim -0.5$ ($1.9 \lesssim v_g \lesssim 2.1$), between the Kondo and mixed valence regimes, which we term the "weak Kondo regime," a more complex evolution of $T_1(B)$ and $T_2(B)$ with B is observed: $T_1(B)$ and $T_2(B)$ bifurcate from a common (gate-voltage dependent) value at $B = B_2$ with $0 \leqslant B_2 \leqslant B_1$. The coalescence of $T_0(B)$, $T_1(B)$, and $T_2(B)$ to the single temperature $T_0(B)$, then occurs upon entry to the mixed valence regime $\varepsilon_0/\Gamma \approx -0.5$ when $B_2 = B_1$.

Comparison with experiment. As detailed elsewhere [71], results such as those in Fig. 4 allow the full gate-voltage dependence of the thermoelectric response of Kondo-correlated InAS quantum dots at different magnetic fields [49] to be explained. Here, we consider a simpler quantity, the slope of the linear response thermocurrent $I_{th}/\Delta T \equiv G(T)S(T)$ at $v_g = 0$, i.e., $\sigma(T) = d[G(T)S(T)]/dv_g|_{v_g=0}$, where $G(T)$ is the electrical conductance [77], and compare this with corresponding results from Ref. [49]. Evidently, since $G(T)$ is always positive and symmetric in v_g, $\sigma(T)$ exhibits the same Kondo-induced sign changes at T_0, T_1, and T_2 as $S(T)$

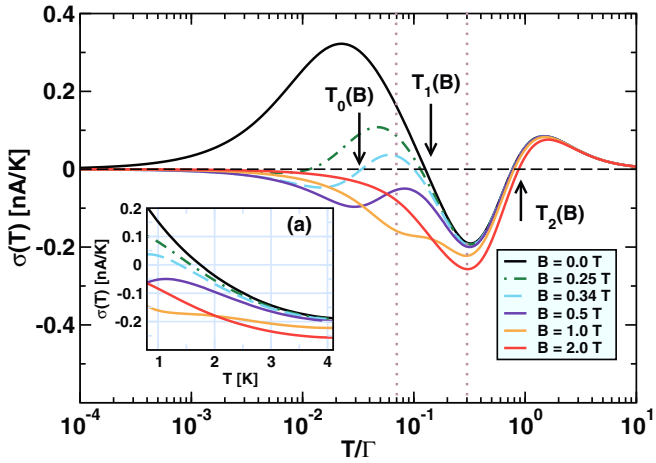

FIG. 5. Main panel: $\sigma(T) = d[G(T)S(T)]/dv_g|_{v_g=0}$ vs T/Γ for the four field values $B = 0T,\ 0.5T,\ 1.0T$, and $2.0T$ of the experiment [49] and two additional field values $B = 0.25T$ and $0.34T$ between $0T$ and $0.5T$. The vertical dotted lines denote the lowest and highest temperatures of the experiment. Inset (a): same as main panel, with temperature in units of K in the range (1–4 K) of the experiment and using a linear scale. For $B = 0T < B_0(v_g = 0) \approx 0.23T$ sign changes at $T_1(B)$ and $T_2(B)$ are seen; for $B = 0.5T,\ 1.0T$, and $2.0T$ we have that $B > B_1(v_g = 0) \approx 0.4T$ and only the sign change at $T_2(B) \approx 10$ K is observable.

at small finite v_g and thereby it suffices to determine the Kondo-induced signatures in the thermopower.

We focus on device QD1a of Ref. [49], which has $U = 3.5$ meV and $\Gamma = 1.1$ meV ($U/\Gamma = 3.2$), resulting in a mid-valley $T_K^{\rm expt} \equiv T_{K1} \approx 1.0$ K. From the value of Γ, and the measured g factor $g \approx 9$ for InAs quantum dots [14,49], we carry out calculations for $\sigma(T)$ vs T/Γ at the experimental field values $B = 0.0T,\ 0.5T,\ 1.0T$, and $2.0T$.

Figure 5 shows $\sigma(T)$ at the four experimental field values and at two additional ones at $B = 0.25T$ and $0.34T$ (to be discussed below) and over the whole temperature range. Figure 5(a) restricts to the measured temperature window 1–4 K and can be directly compared with Fig. 4(f) of Ref. [49]. The possible sign changes in $\sigma(T)$, like in $G(T)S(T)$, can be understood depending on whether (i) $B < B_0$, (ii) $B_0 < B < B_1$, or (iii) $B > B_1$. The sign change at $T_2(B) \approx 10$ K lies outside the measurement window, and will not be considered further [71]. Starting with $B = 0T \lesssim B_0(v_g = 0) \approx 0.23T$ [Fig. 2(a)], we observe the expected sign change in $\sigma(T)$ upon increasing T through $T_1(0)$, as seen also in experiment. For the cases $B = 1.0T$ and $2.0T$, we have $B > B_1(v_g = 0) \approx 0.4T$, and in accordance with theory, no sign change is observed and none is found in experiment (within the measurement window). For the case $B = 0.5T > B_1(v_g = 0) \approx 0.4T$ [Fig. 2(b) and Ref. [71]], a qualitatively similar behavior of $\sigma(T)$ vs T is observed in both theory and experiment (linear at higher T with a leveling off at the lowest T), but in contrast to theory, which does not predict a sign change of $\sigma(T)$ at this field value, the measurement finds a sign change, interpreted as $T_1(B) \approx 2.5$ K $> T_1(0) \approx 1.8$ K. The latter increase of $T_1(B)$ with B, however, is inconsistent with theory, which predicts the opposite trend [see Fig. 1(b) and Fig. 4], and, inconsistent with measurements on all other devices in Ref. [49], which agree with our predicted trend whenever a sign change at $T_1(B)$ is present. To summarize, our comparison with experiment shows that the Kondo-induced sign change at $T_0(B)$ in $\sigma(T)$ has not been measured (in contrast to the one at T_1 for $B = 0$). In order to observe the sign change at $T_0(B)$ for device QD1a, one needs to use smaller fields in the range $B_0 \approx 0.23T \leqslant B \leqslant B_1 \approx 0.4T$. Two fields, $B = 0.25T$ and $0.34T$, satisfying this condition, and exhibiting the predicted sign change at $T_0(B) \approx 0.16$ and 0.45 K, respectively, are shown in Fig. 5.

Conclusions. In summary, we investigated the magnetic field dependence of the thermopower of a Kondo-correlated quantum dot by applying the NRG technique to the Anderson impurity model in a magnetic field. In the Kondo regime, we found, in addition to the known sign changes of $S(T)$ at T_1 and T_2, an additional sign change at a temperature $T_0(B) < T_1(B) < T_2(B)$ for magnetic fields B exceeding a gate-voltage dependent value B_0. The field B_0, of order T_K in the Kondo regime, is comparable, but quantitatively different, to the field B_c for the splitting of the Kondo resonance. Our results are of relevance in the light of recent advances in characterizing the thermoelectric properties of nanodevices [20,21,35,49,59]. They explain, for example, the essential observations (Fig. 5 and Ref. [71]) in the field dependence of the thermoelectric response of Kondo-correlated quantum dots [49], and could serve as a guideline for interpreting future experiments on field-dependent thermoelectric transport through such systems.

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