Entropy Measurement of a Strongly Coupled Quantum Dot

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The spin 1/2 entropy of electrons trapped in a quantum dot has previously been measured with great accuracy, but the protocol used for that measurement is valid only within a restrictive set of conditions. Here, we demonstrate a novel entropy measurement protocol that is universal for arbitrary mesoscopic circuits and apply this new approach to measure the entropy of a quantum dot hybridized with a reservoir. The experimental results match closely to numerical renormalization group (NRG) calculations for small and intermediate coupling. For the largest couplings investigated in this work, NRG predicts a suppression of spin entropy at the charge transition due to the formation of a Kondo singlet, but that suppression is not observed in the experiment.

Entropy is a powerful tool for identifying exotic quantum states that may be difficult to distinguish by more standard metrics, like conductance. For example, bulk entropic signatures in twisted bilayer graphene indicate that carriers in some phases with metallic conductivity retain their local moments, as would normally be associated with a Mott insulator [1–3]. Entropy has also been proposed as a tell-tale characteristic of isolated non-abelian quasiparticles, whether Majorana modes in a superconductor [4, 5] or excitations of a fractional quantum Hall state [6–8], distinguishing them from abelian analogs.

Quantifying the entropy of single quasiparticles is challenging due to the small signal size, of order k_B, but first steps in this direction have been made in recent years [9, 10]. Ref. 9 employed Maxwell relations to measure the $k_B \ln 2$ spin entropy of a single electron confined to a quantum dot (QD) in GaAs via the temperature-induced shift of a Coulomb blockade charge transition. That approach relied on the assumption of weak coupling between the QD and the reservoirs, in order to fit based on the specific charging lineshape known for that regime. In that weak-coupling regime, spin states are pristine enough to serve as spin qubits [11–17] but the underlying physics is very simple.

The weak-coupling approach of Ref. 9 is not applicable to a broad class of mesoscopic devices[18], which limits its value in probing the complex Hamiltonians that may be implemented in such systems. For example, a single-impurity Kondo effect may be realized when the localized spin is strongly coupled to a reservoir [19, 20]. Recently, more complicated structures including multiple dots have been engineered to host multi-channel Kondo states [21, 22], or a three-particle simulation of the Hubbard model [23]. Entropy measurements made on any of these systems would offer a significant advance in their understanding.

Here, we develop a universal protocol for mesoscopic entropy measurement that forgoes the simplifying assumptions of Ref. 9, then apply it to investigate the entropy of the first electron as it enters a quantum dot when strongly hybridized with a reservoir. The protocol is based on a Maxwell relation appropriate for mesoscopic systems, where the free energy includes both local and global terms. Expressed in integral form, the relation

$$\Delta S_{\epsilon_1 \to \epsilon_2} = -\int_{\epsilon_1}^{\epsilon_2} \frac{dN(\epsilon)}{dT} d\epsilon, \quad (1)$$

provides access to the entropy change, ΔS, of the QD-lead system as a function of the gate-tuned QD energy ϵ, based on measurements of the change in average QD occupation, N, with temperature, T [5, 18, 24]. Eq. 1 is related to the more conventional Maxwell relation, $\partial s/\partial \mu = \partial n/\partial T$, that applies to macroscopic systems with particle density n and entropy density s, here replacing the reservoir chemical potential μ with the dot energy ϵ [24].

We first confirm that the data match well to single-

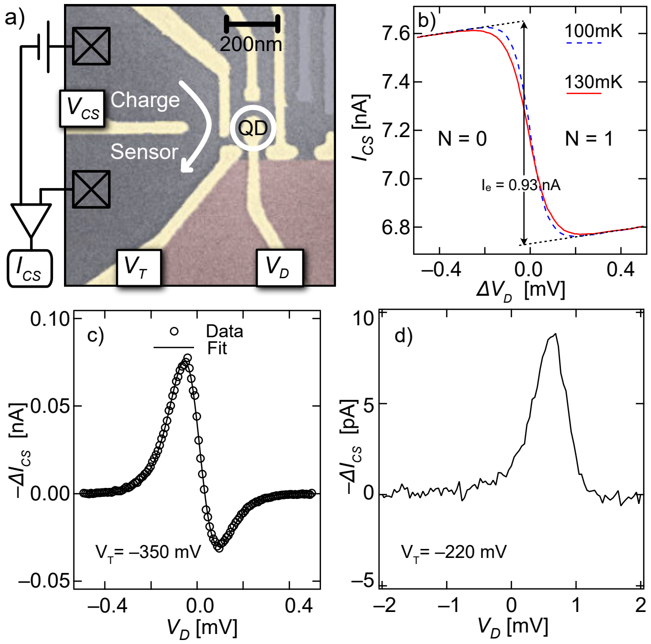

FIG. 1. a) Scanning electron micrograph of the device. Electrostatic gates (gold) define the circuit. Squares represent ohmic contacts to the 2DEG. The thermal electron reservoir (red) was alternated between base and elevated temperatures. b) Current through the charge sensor, I_{CS}, for the $0 \rightarrow 1$ charge transition in a weakly coupled regime, separated into the unheated (100mK) and heated (130mK) parts of the interlaced measurement [25], showing the single electron step height I_e. c,d) Change in I_{CS} from 100 to 130 mK, for weak (c) and strong (d) coupling between QD and reservoir. c) includes fit to weakly-coupled theory

particle approximations when the coupling, Γ, between dot and reservoir is weak ($\Gamma \ll k_B T$), then show that the onset of entropy as the electron enters the dot is strongly modified when $\Gamma \gtrsim k_B T$. The measurement of this modified entropy signature is the primary result of this work, offering clear entropic evidence of the effect of strong reservoir coupling on the quantum state.

Measurements were performed on a mesoscopic circuit (Fig. 1a) in a GaAs 2D electron gas [24, 25], including the QD, a charge sensing quantum point contact, and an electron reservoir that can be rapidly Joule-heated above the chip temperature T to an elevated $T + \Delta T$. Coupling between the QD and the thermal reservoir is via a single tunnel barrier, with Γ controlled by V_T. The QD energy ϵ was tuned using gate voltage V_D. Throughout this paper we report V_D with respect to to the midpoint of the $N = 0 \rightarrow 1$ charge transition, $\Delta V_D \equiv V_D - V_D(N = 1/2)$. N in the QD was monitored via the current, I_{CS}, through the charge sensor (Fig. 1b)[26], which was biased with a DC voltage typically 100 μV. Changes in occupation, N, were scaled from I_{CS} using I_e, the net drop in I_{CS} across a $1e$ charge transition[24]. Fig. 1b illustrates weakly coupled $N = 0 \rightarrow 1$ transitions at $T = 100$ mK and $T + \Delta T = 130$ mK. Throughout this paper both T and $T + \Delta T$ were calibrated by fitting to thermally broadened charge transitions; except where noted, $T = 100$ mK with $\Delta T \sim 30$ mK. Measurements at T and $T + \Delta T$ were interlaced by alternated Joule heating of the reservoir at 25Hz to reduce the impact of charge instability, then averaged over several sweeps across the charge transition, see Ref. 24.

Figure 1c shows the change in detector current from 100 to 130 mK, $\Delta I_{CS}(V_D) \equiv I_{CS}(T + \Delta T, V_D) - I_{CS}(T, V_D)$, scanning across the $0 \rightarrow 1$ transition in the weakly coupled regime. Note that $-\Delta I_{CS}$ is plotted instead of ΔI_{CS} in order to connect visually with ΔN, which increases when I_{CS} decreases. As in Ref. 9, the lineshape of $\Delta I_{CS}(V_D)$ in Fig. 1c may be fit to a non-interacting theory for thermally-broadened charge transitions to extract the change in entropy across the transition, ΔS_{fit}, not requiring calibration factors or other parameters (see Ref. 9 for details). For the data in Fig. 1c, this yields $\Delta S_{\text{fit}} = (1.02 \pm 0.01) k_B \ln 2$, where the uncertainty reflects standard error among 5 consecutive measurements at slightly different V_T.

The limitation of this approach is illustrated by the very different lineshape in Fig. 1d, reflecting the $0 \rightarrow 1$ transition when the QD is strongly coupled to the reservoir. Fitting the data in Fig. 1d to thermally-broadened theory would yield a meaningless (and incorrect) $\Delta S_{\text{fit}} > 10 k_B \ln 2$ for the entry of the spin-1/2 electron. For a quantitative extraction of entropy beyond the weakly-coupled regime of Fig. 1c, we instead follow the integral approach in Eq. 1 that makes no assumptions on the nature of the quantum state. Evaluating Eq. 1 provides a measurement of $\Delta S(\epsilon)$ that is continuous across the charge transition, rather than just comparing $N = 0$ to $N = 1$ values.

Before moving to the quantitative evaluation of entropy, we note that the different lineshapes of $\Delta I_{CS}(V_D)$ in Figs. 1c and d—the peak-dip structure in Fig. 1c contrasting with the simple peak in Fig. 1d—can be understood as representing two temperature regimes for the Anderson impurity model. Fig. 1c represents the high temperature limit, where dN/dT is approximately a measure of the energy derivative of the density of states in the QD, and thus exhibits positive and negative lobes. At sufficiently low temperatures, the exact solution [27], and the resulting Fermi liquid theory [28] predict a positive dN/dT for all values of the chemical potential, from the empty level to the Kondo regime through the mixed-valence regime, with a peak expected at a chemical potential corresponding to $T_K(\epsilon) \sim T$, where the entropy is expected to crossover from $S = 0$ to $S = k_B \log 2$. Fig. 1d, corresponding to a measurement where $T \ll \Gamma$, demonstrate such all-positive dN/dT.

We next describe the evaluation of Eq. 1 from experimental data. $dN(\epsilon)/dT$ is approximated by the ratio $\Delta N(V_D)/\Delta T = -\Delta I_{CS}(V_D)/(I_e \Delta T)$. ΔT is expressed

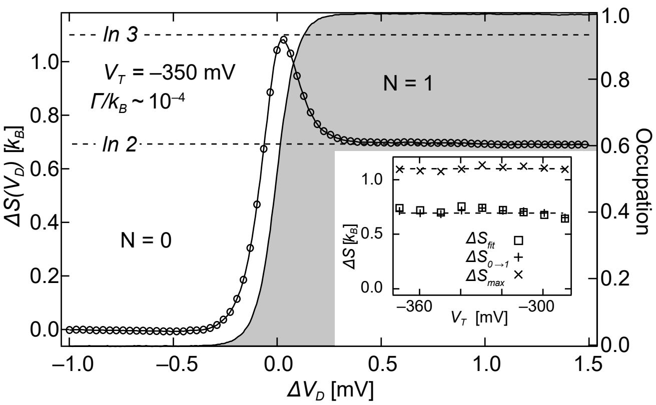

FIG. 2. Change of S in the QD across the $N = 0 \to 1$ transition, obtained by integrating $\Delta I_{CS}(V_D)$ (Fig. 1c) following Eq. 1. Dot occupation across the transition is shown in grey. Data obtained in the weakly coupled limit, $V_T = -350$ mV corresponding to $\Gamma/k_BT \sim 1 \times 10^{-4}$. $\Delta S_{0\to1} = (0.99 \pm 0.02)k_B \ln 2$ is the net change ΔS across the complete transition. Inset: comparison of $\Delta S_{\rm fit}$, $\Delta S_{0\to1}$, and ΔS_{max} (see text) for V_T covering approximately $10^{-5} < \Gamma/k_BT < 10^{-1}$.

in units of gate voltage using the corresponding lever arm[24] so that the integral may be evaluated over V_D, giving $\Delta S(V_D)$. We begin by confirming the integral approach in the weakly-coupled ($\Gamma \ll k_BT$) regime, where the physics is simple.

Figure 2 shows the entropy change across the $N = 0 \to 1$ charge transition for such a weakly-coupled transition, calculated from the data in Fig. 1c using Eq. 1. The resulting $\Delta S(\epsilon)$ indicates that the change in dot entropy is non-monotonic as the first electron is added, reaching a $k_B \ln 3$ peak before settling to $k_B \ln 2$. The $k_B \ln 3$ peak just above $\Delta V_D = 0$ reflects a combination of charge and spin degeneracy in the middle of the charge transition, with three microstates $\{|N=0\rangle, |N=1,\uparrow\rangle, |N=1,\downarrow\rangle\}$ all equally probable. Charge degeneracy is gone after the transition, but spin degeneracy remains, leaving two microstates $\{|N=1,\uparrow\rangle, |N=1,\downarrow\rangle\}$. The net change in entropy from beginning to end, $\Delta S_{0\to1} = (0.99 \pm 0.02)k_B \ln 2$, is nearly identical to the $\Delta S_{\rm fit} = (1.02 \pm 0.01)k_B \ln 2$ from Fig. 1c, despite different sources of error for the two approaches.

The inset to Fig. 2 compares the fit and integral approaches for weakly-coupled charge transitions covering four orders of magnitude in Γ, tuned by V_T (see Fig. 3b inset for calibration of Γ). The consistency between $\Delta S_{0\to1}$ and $\Delta S_{\rm fit}$ over the full range of weakly-coupled V_T, in addition to the fact that ΔS_{max} remains k_B ln 3 throughout this regime, confirms the accuracy of the integral approach. Small deviations from $\Delta S_{0\to1} = \Delta S_{\rm fit} = k_B \ln 2$, such as that seen around $V_T = -330$ mV, are repeatable but sensitive to fine-tuning of all the dot gates; we believe they are due to extrinsic degrees of freedom capacitively coupled to the dot occupation, such as charge instability in shallow dopant levels in the GaAs heterostructure.

After confirming the accuracy of Eq. 1 in the weakly coupled regime, we turn to the regime $\Gamma \gtrsim k_BT$ ($V_T > -280$ mV), where the influence of hybridization is expected to emerge. Fig. 3 shows the crossover from $\Gamma \ll k_BT$ to $\Gamma \gg k_BT$, illustrating several qualitative features. The $k_B \ln 3$ peak in $\Delta S(\mu)$ decreases with Γ, until no excess entropy is visible at the charge degeneracy point for $\Gamma/k_BT \gtrsim 5$ (Fig. 3a). This suppression of the entropy associated with charge degeneracy originates from the broadening by Γ of the $N = 1$ level due to hybridization with the continuous density of states in the reservoir [5]. At the same time, the total entropy change $\Delta S_{0\to1}$ remains $\sim k_B \ln 2$ over the entire range of Γ explored in this experiment, reflecting the entropy of the spin-1/2 electron trapped in the QD.

To make quantitative comparison between theory and experiment, we employ NRG simulations [29, 30] that yield N as a function of T and ϵ_0, where $-\epsilon_0$ is the depth of the dot level below the reservoir chemical potential μ. From $N(T, \epsilon_0)$, dN/dT and thereby ΔS are extracted via Eq. 1. To make a direct comparison with the experiment, $\Delta\epsilon_0 \equiv \epsilon_0 - \epsilon_0(N = 1/2)$ is defined like ΔV_D, centred with respect to the charge transition. NRG parameters are calibrated to match those in the measurements by aligning the occupation $N(\Delta\epsilon_0)$ with the measured $N(\Delta V_D)$[24], from which the appropriate Γ/T calculation may be selected and the precise connection between $\Delta\epsilon_0$ with ΔV_D is ensured. As seen in Fig. 3b, the data/theory agreement in terms of dot occupation is within the experimental resolution, giving confidence that measured and calculated ΔS may be compared directly.

Figure 3c illustrates NRG predictions for $\Delta S(\epsilon_0)$ over the range of Γ accessible in our measurements. Matching the data, the peak in entropy due to charge degeneracy is suppressed for $\Gamma > k_BT$, while the net entropy change across the transition remains $k_B \ln 2$. At the same time, a qualitative difference between data and NRG is the shift to the right seen in NRG curves for higher Γ (Fig. 3c), but not observed in the measurements (Fig. 3a). This relative shift of NRG with respect to data is not explained by an offset of $\Delta\epsilon_0$ with respect to ΔV_D, as the two are aligned by the occupation data (Fig. 3b).

Instead, the shift of NRG curves to the right (to larger chemical potential) with increasing Γ is explained by the virtual exchange interactions underlying the Kondo effect, which form a quasi-bound singlet state between the localized spin and a cloud of delocalized spins in the reservoir at temperatures below T_K. This state has no magnetic moment [31] and, in the case of a single-electron QD, zero entropy. Thus, due to the Kondo effect, we expect the entropy to remain zero for all dot energies that obey $T < T_K(\epsilon_0)$. Since $T_K \propto e^{-\pi(\epsilon_0-\mu)/\Gamma}$ in the (experimentally relevant) large-U limit, where U represents

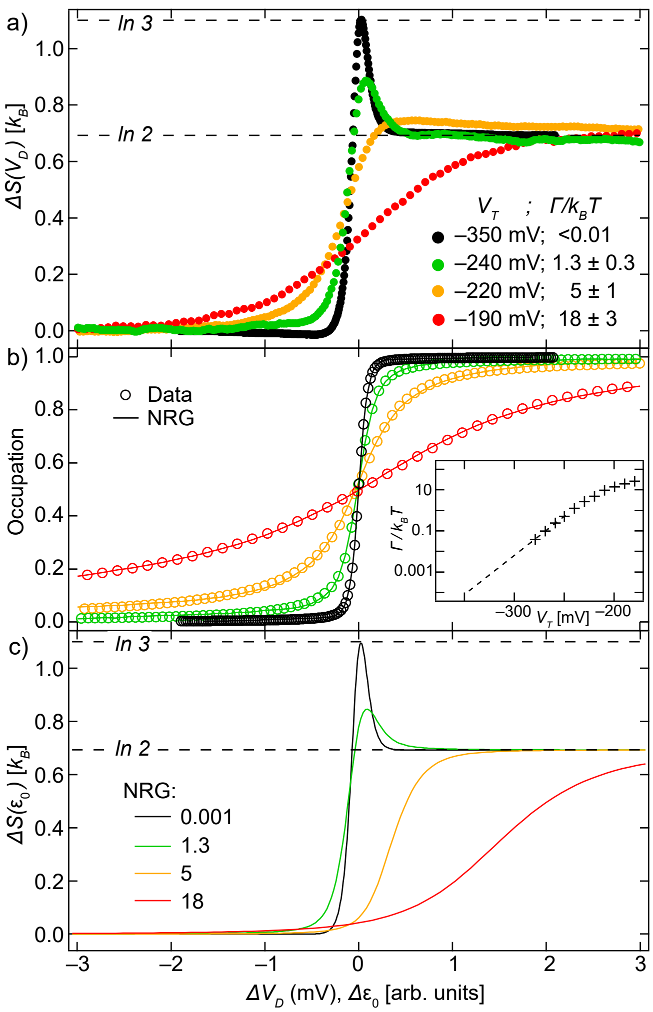

FIG. 3. Evolution of $S(\epsilon)$ from the weak (black) to strong (red) coupling regimes, comparing data (panel a) to NRG calculations (panel c). Measurements of occupation across the charge transition are fit to NRG (panel b), leaving no free fit parameters for the $S(\epsilon_0)$ calculation. Panel b inset: Coupling strength of the QD to the reservoir, Γ/k_BT, extracted from fits, across the full range of V_T. Values $\Gamma/k_BT \ll 1$ cannot be measured directly and are extrapolated (dashed line).

the QD charging energy, we expect the onset of $k_B \ln 2$ entropy to shift to larger values of ϵ as Γ increases, as seen in the NRG results.

It remains a puzzle why the strong suppression of entropy right at the charge transition, seen in NRG calculations for $\Gamma/k_BT \geq 5$, is not observed in the data. It is possible that the charge measurement itself can lead to dephasing of the Kondo singlet[32–34]. In order to test for charge-sensor dephasing in our measurement, the experiment was repeated at charge sensor biases from 300 μV down to 50 μV, but no dependence on the bias was seen in the data [24]. In the future, experiments that allow simultaneous transport and entropy characterization of the Kondo state may help to resolve this puzzle.

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