Direct Entropy Measurement in a Mesoscopic Quantum System

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The entropy of an electronic system offers important insights into the nature of its quantum mechanical ground state. This is particularly valuable in cases where the state is difficult to identify by conventional experimental probes, such as conductance. Traditionally, entropy measurements are based on bulk properties, such as heat capacity, that are easily observed in macroscopic samples but are unmeasurably small in systems that consist of only a few particles \cite{1,2}. In this work, we develop a mesoscopic circuit to directly measure the entropy of just a few electrons, and demonstrate its efficacy using the well understood spin statistics of the first, second, and third electron ground states in a GaAs quantum dot \cite{3–8}. The precision of this technique, quantifying the entropy of a single spin-\(\frac{1}{2}\) to within 5\% of the expected value of \(k_B \ln 2\), shows its potential for probing more exotic systems. For example, entangled states or those with non-Abelian statistics could be clearly distinguished by their low-temperature entropy\cite{9–13}.

Our approach is analogous to the milestone of spin-to-charge conversion achieved over a decade ago, in which the infinitesimal magnetic moments of a single spin were detected by transforming them into the presence or absence of an electron charge \cite{14,15}. Following this example, we perform an entropy-to-charge conversion, making use of the Maxwell relation

\[
\frac{\partial \mu}{\partial T} = - \frac{\partial S}{\partial N} \quad (p,N)
\]

that connects changes in entropy, particle number, and temperature (\(S, N, T\), respectively) to changes in the chemical potential, \(\mu\), a quantity that is simple to measure and control.

The Maxwell relation in Eq. 1 forms the basis of two theoretical proposals to measure non-Abelian exchange of Moore-Read quasiparticles in the \(\nu = \frac{5}{2}\) state via their entropy \cite{9,10}. Reference 10 proposes a strategy by which quasiparticle entropy could be deduced from the temperature-dependent shift of charging events on a local disorder potential—a thermodynamic equivalent of the measurements that established the \(e/4\) quasiparticle charge\cite{16}. As a demonstration of the viability and the high accuracy achievable by this technique, we investigate a well-understood system with localized fermions in place of more exotic quasiparticles: a few-electron GaAs quantum dot. The entropies of the first three elec-
electron states in the dot are measured by the temperature-dependent charging scheme laid out in Ref. 10. Applying the language of quantum dots to Eq. 1, the entropy difference between the \( N - 1 \) and \( N \) electron ground states (\( \Delta S_{N-1 \rightarrow N} \) for \( \Delta N = 1 \)) is measured via the shift with temperature in the electrochemical potential, \( \mu_N \), needed to add the \( N \)th electron to the dot.

The measurement relies on the mesoscopic circuit shown in Fig. 1a, using electrostatic gates to realize an electron reservoir in thermal and diffusive equilibrium with a few-electron quantum dot coupled to its right side. The occupation of the dot is tuned with the plunger gate voltage, \( V_p \), and measured using an adjacent quantum point contact as a charge sensor [17–19]. Applying more positive \( V_p \) lowers \( \mu_N \), bringing the \( N \)th electron into the dot when \( \mu_N \) drops below the Fermi level of the reservoir, \( E_F \). The reservoir temperature, \( T \), can be increased above the GaAs substrate temperature by Joule heating from current, \( I_{heat} \), driven through a quantum point contact on the left side. Charge transitions on the dot appear as steps in the charge sensor conductance, \( G_{sens}(V_p) \), thermally broadened by the reservoir temperature (Figs. 1b and c). The gate voltage corresponding to the midpoint of the transition, \( V_{mid} \), marks the electrochemical potential at which the probabilities of finding \( N - 1 \) and \( N \) electrons on the dot are equal.

When \( \mu_N \) shifts with temperature, \( V_{mid} \) also shifts; it is the shift in \( V_{mid} \) with temperature that forms the basis of our experiment (Fig. 1c). In practice, charge noise limits the accuracy to which \( V_{mid} \) can be measured. To overcome this, the measurement is done with a lock-in amplifier, oscillating the temperature using an AC \( I_{heat} \) and measuring resultant oscillations in \( G_{sens} \), which we label \( \delta G_{sens} \). As seen in the insets of Figs. 1b and c, the lineshape of \( \delta G_{sens} \) is perfectly antisymmetric when \( \delta S/\delta N = 0 \), but asymmetric when \( \delta S/\delta N \neq 0 \).

The temperature-induced shift in the dot chemical potential with respect to reservoir \( E_F \) can also be understood in terms of detailed balance. At \( V_{mid} \), where probabilities for \( N \) and \( N - 1 \) electrons on the dot are equal, the tunnel rates \( \Gamma_{in} = \Gamma_{N-1 \rightarrow N} \) and \( \Gamma_{out} = \Gamma_{N \rightarrow N-1} \) must also be equal. These rates depend on the number of available states in the tunneling process, and therefore on the degeneracies, \( d_{N-1} \) and \( d_N \), of the \( N - 1 \) and \( N \) ground states [20, 21]. The condition \( \Gamma_{in} = \Gamma_{out} \) leads to a simple relationship between degeneracy and the thermally broadened Fermi function, \( f(\mu_N - E_F, T) \): \( d_{N-1}/d_N = f/(1 - f) \). Using the Boltzmann entropy, \( S_N = k_B \ln d_N \), this relationship becomes \( \Delta S_{N-1 \rightarrow N} = (\mu_N - E_F)/T \), clearly demonstrating the connection between entropy, temperature, and the shift in \( \mu_N \) at \( V_{mid} \). Previous experiments have explored the relationship between tunnel rates and degeneracy using time-resolved transport spectroscopy and by coupling quantum dots to atomic force cantilever oscillations [8, 22–24]. The approach presented here is a thermodynamic analogue, and extends entropy measurements to a wider set of applications where tunneling processes may not be observable in real-time.

The dot was tuned such that the source was weakly tunnel-coupled to the reservoir with the drain closed. The conductance of the charge sensor was tuned to \( G_{sens} \sim e^2/h \), where it was most sensitive to charge on the dot. The addition of the first electron to the dot was marked by a decrease in \( G_{sens} \) that is consistent with a thermally-broadened two-level transition (Fig. 2a):

\[
G_{sens}(V_p, \Theta) = G_0 \tanh \left( \frac{V_p - V_{mid}(\Theta)}{2\Theta} + \gamma_1 V_p + G_2 \right)
\]

where \( G_0 \) quantifies the sensor sensitivity, \( \Theta = \frac{k_B T}{\alpha e} \) is

FIG. 2. Entropy measurement for a single spin-\( \frac{1}{2} \)

(a) Charge sensor data for \( N = 0 \rightarrow 1 \) at two temperatures set by DC current through the QPC heater. (b) Transition width, \( \Theta \), was linear in \( T_{QPC} \) above 100 mK, and \( I_{heat} = 0 \). Lever arm \( \alpha \) is calculated by fitting a straight line to this region. (c) Lock-in measurement of \( \delta G_{sens} \) with \( \delta T = 32 \) mK, determined from the calibration in panel (d). Fits to \( \delta G_{sens} \) (Eq. 3) are shown with \( \Delta S/k_B \) as a free parameter (solid) and fixed at \( \Delta S/k_B = 0 \) (dashed). (d) \( \Theta \) grows with DC current through the QPC heater. A fit to \( T^2 = a T_{QPC}^2 + b I_{heat}^2 R_{QPC} \) is used to convert between \( I_{heat} \) and \( \delta T \), where \( T_{QPC} \) is the mixing chamber temperature [25]. (e) Entropy measurements were independent of the magnitude of \( I_{heat} \) oscillations over a large range. The top axis indicates the corresponding magnitude of \( \delta T \), while the right axis shows the entropy signal converted to a gate voltage shift per unit temperature. Error bars show 95% confidence intervals calculated with the bootstrap method.
the thermal broadening expressed in units of gate voltage, \( \alpha \equiv \frac{1}{k_B} \frac{\partial V}{\partial T} \) is the lever arm, \( \gamma_1 \) reflects the cross capacitance between the charge sensor and plunger gate, and \( G_2 \) is an offset. Figure 2a shows two such transition curves with thermal broadening set by \( I_{\text{heat}} \). For \( I_{\text{heat}} = 0 \), \( \Theta \) followed \( T_{\text{MC}} \) down to approximately 100 mK (Fig. 2b), validating the approximation of thermal broadening used throughout this experiment.

The data in Fig. 2c, and corresponding fits, illustrate a measurement of \( \Delta S_{0\rightarrow 1} \) across the 0 \( \rightarrow \) 1 electron transition. The lock-in measurement of \( \delta G_{\text{sens}} \), due to temperature oscillations \( \delta T \), yields the characteristic peak-dip structure seen in Fig. 2c.

The expected lineshape of such a curve is \( \delta G_{\text{sens}} = \frac{\partial V}{\partial T} \delta T \), with \( G_{\text{sens}} \) defined by Eq. 2. This lineshape depends explicitly on \( \Delta S \); recognizing (via Eq. 1) that
\[
\delta G_{\text{sens}}(V_p, \Theta) \propto -\delta T \left[ \frac{V_p - V_{\text{mid}}(\Theta)}{2\Theta} - \frac{\Delta S}{2k_B} \right] \times \cosh^{-2} \left( \frac{V_p - V_{\text{mid}}(\Theta)}{2\Theta} \right) + \text{const.}
\]

As expected from Figs. 1b and c, \( \delta G_{\text{sens}}(V_p) \) is antisymmetric around \( V_{\text{mid}} \) for \( \Delta S = 0 \), and asymmetric for \( \Delta S \neq 0 \). A fit of the data in Fig. 2c to Eq. 3 yields \( \Delta S_{0\rightarrow 1} = (1.02 \pm 0.03)k_B \ln 2 \), closely matching the expected \( \Delta S_{0\rightarrow 1} = S_1 - S_0 = k_B \ln 2 \) for transitions between an empty dot with zero entropy \( (S_0 = 0) \) and the two-fold degenerate one-electron state \( (d_1 = 2) \) with entropy \( S_1 = k_B \ln 2 \).

It is important to note that \( \Delta S \) is extracted from fits to Eq. 3 based solely on the asymmetry of the lineshape, with no calibration of measurement parameters (such as \( \delta T \) or the lever arm \( \alpha \)) required. We can, however, estimate \( \alpha \) and \( \delta T \) by determining \( \Theta \) from fits to Eq. 2 for varying substrate temperature (Fig. 2b) and \( I_{\text{heat}} \) (Fig. 2d). Measurements of \( \Delta S \) remained constant over a broad range of \( B \) (Fig. 2e), as expected for temperatures low enough not to excite orbital degrees of freedom on the dot.

Confirmation that the measured \( \Delta S \) derives from spin degeneracy is seen through its evolution with in-plane magnetic field, \( B_\parallel \). Figure 3a compares \( \Delta S(B_\parallel) \) for the 0 \( \rightarrow \) 1 and 2 \( \rightarrow \) 3 transitions, both of which correspond to transitions from total spin zero to total spin one-half. The entropies of the one- and three-electron states go to zero as Zeeman splitting lifts the spin degeneracy, following the Gibbs entropy for a two-level system:

\[
S = k_B \sum_{i=\pm} p_i(B_\parallel, T) \ln p_i(B_\parallel, T)
\]

where \( p_{\pm}(B_\parallel, T) = (1 + e^{\frac{-\Delta S_{\pm 0}(B_\parallel)}{k_B T}})^{-1} \) are the probabilities for the unpaired electron to be in the spin up or spin down states at a given field and temperature. Fits to Eq. 4, with the ratio \( g/T \) and an added scaling \( \Delta S(B = 0) \) as free parameters, give \( \Delta S_{0\rightarrow 1}(B = 0) = (0.94 \pm 0.03)k_B \ln 2 \) and \( \Delta S_{2\rightarrow 3}(B = 0) = (0.98 \pm 0.02)k_B \ln 2 \) (Fig. 3b), and reflect the collapse to zero at high field as spin degeneracy is broken. This collapse can also be seen qualitatively, in the crossover from asymmetric to antisymmetric lineshapes of \( \delta G_{\text{sens}}(V_p) \) (Figs. 3b and c). Estimating an average \( T \) for each data set using the calibration in Fig. 2d yields \( |g| = 0.48 \pm 0.02 \) and \( |g| = 0.44 \pm 0.01 \) for the 0 \( \rightarrow \) 1 and 2 \( \rightarrow \) 3 transitions, respectively. Errors in the g-factor measurement are likely due to the difficulty of estimating temperature oscillations. Still, the g-factors are consistent with reported values [26–28] and the value measured separately in Fig. 3e using bias spectroscopy.

The 1 \( \rightarrow \) 2 transition can be understood as the inverse of the 0 \( \rightarrow \) 1 transition for \( B_\parallel < 5 \) T, comparing Figs. 3a and 4a. For relatively low fields, the two-electron ground state remains a spin singlet with zero
FIG. 4. Entropic signature of a singlet-triplet crossing (a) Change in entropy, extracted from \( \delta G_{\text{sens}} \) fits at varying in-plane field. Dashed line shows fit to Eq. 4, allowing for an offset from \( \Delta S = 0 \) away from the degenerate points to compensate for non-linearities in the charge sensor. Values stated for \( \Delta S \) are with respect to the vertical offset apparent in the data. (b), (c), and (d) show characteristic \( \delta G_{\text{sens}} \) traces from which the data in (a) were extracted. These data points are show as large markers in (a). (e) Bias spectroscopy data for the \( N = 1 \rightarrow 2 \) transition. Transitions to the two-electron triplet state correspond to the lines appearing at \( V_{SD} = \pm 320 \mu \text{eV} \). Dashed line at \( V_{SD} = 1250 \mu \text{eV} \) shows where data in (f) are taken. (f) Fixed bias data in in-plane field. Triplet level is split into \( |T_{\uparrow}\rangle \) and \( |T_{\downarrow}\rangle \) levels with a third \( |T_{\text{middle}}\rangle \) level not visible here. At 8.4 T \( |T_{\uparrow}\rangle \) becomes degenerate with \( |S\rangle \), \( |g| = 0.40 \pm 0.04 \) is determined using \( |T_{\uparrow}\rangle \) and \( |T_{\downarrow}\rangle \) fits (dashed).

The field-dependent entropy measurement for the \( 1 \rightarrow 2 \) transition can again be fit using Eq. 4, with probabilities as before for the one-electron states and

\[
p_{|S\rangle}(B_0, T) = (1 + e^{-\frac{\Delta S_k B_0}{k_B T}})^{-1}, \quad p_{|T_{\uparrow}\rangle}(B_\parallel, T) = (1 + e^{\frac{\Delta S_k B_\parallel - \Delta ST}{k_B T}})^{-1}
\]

for the two-electron states, where \( \Delta ST \) is the singlet-triplet splitting at zero field. From the fit, we find \( \Delta S_{1\rightarrow2} \approx 0.07 \) eV.

We conclude with a few notes to encourage the application of this entropy measurement protocol to other mesoscopic systems. The crucial ingredients in achieving the high accuracy reported here were i) the ability to oscillate temperature rapidly enough to avoid 1/f noise, ii) the ability to measure charging transitions without perturbing the localized states, and iii) the fact that the charging transitions were thermally broadened. Criterion iii) enabled the entropy determination purely by asymmetry, without the need to know \( \delta T \) or other measurement parameters accurately, yielding an uncertainty less than 5%. With this level of precision, it should be possible, for example, to distinguish the \( \frac{1}{2} k_B \ln 2 \) entropy of a non-Abelian Majorana bound state from the \( k_B \ln 2 \) entropy of an Andreev bound state at an accidental degeneracy[11, 12]. Similarly, the \( S = \frac{1}{2} k_B \ln 2 \) two-channel Kondo state could be clearly distinguished from fully screened \( (S = 0) \) or unscreened \( (S = k_B \ln 2) \) spin states[13].

Methods The device was built on a AlGaAs/GaAs heterostructure, hosting a 2D electron gas with density and mobility at 300 mK of \( 2.42 \times 10^{11} \text{cm}^{-2} \) and \( 2.56 \times 10^{6} \text{cm}^{2}/(\text{V} \text{s}) \) respectively, determined in a separate measurement. Mesas and NiAuGe ohmic contacts to the 2DEG were defined by standard photolithography techniques, followed by atomic layer deposition of 10 nm HfO\(_2\) to improve the gating stability in the device. Fine gate structures, shown in Fig. 1a, were defined by electron beam lithography and deposition of 3/18 nm Ti/Au.

The measurement was carried out in a dilution refrigerator with a two-axis magnet. The 2DEG was aligned parallel to the main axis with the second axis used to compensate for sample misalignment. In practice, out-of-plane fields up to 100 mT showed no effect on our data. A retuning of the quantum dot gates was necessary to capture the bias spectroscopy data in Figs. 3d,e and 4e,f. The rightmost gate (Fig. 1a) on the quantum dot was used to tune between the one and two lead configurations, for the entropy and bias spectroscopy measurements respectively. This tuning had a significant effect on the shape of the potential well, accounting for variations in parameters such as \( g \) and \( \Delta ST \) between the two measurement configurations. Charge sensor conductance was measured using a DC voltage bias of 200–350 \( \mu \text{V} \); we
find that Joule heating through the sensor does not affect our reservoir temperatures up to $V_{\text{sens}} \sim 500 \text{ mV}$. The DC current ($I_{\text{sens}}$) was measured using an analog-digital converter while AC current ($\delta I_{\text{sens}}$) was measured using a lock-in amplifier. The DC conductance reported here is $G_{\text{sens}} = I_{\text{sens}}/V_{\text{sens}}$ while the oscillations are defined as $\delta G_{\text{sens}} = (\delta I_{\text{sens}})/V_{\text{sens}}$.

The temperature of the reservoir was raised above the substrate temperature using $I_{\text{heat}}$ at AC or DC, with the QPC heater set by gate voltages to 20 kΩ. Applying AC current at $f_{\text{heat}} = 48.7$ Hz yields an oscillating Joule power, $P_{\text{heat}} = I^2_{\text{heat}}R_{\text{QPC}}$. To leading order this gives oscillations in temperature, and therefore $\delta G_{\text{sens}}$, at $2f_{\text{heat}}$. These are captured by the lock-in amplifier at the second harmonic of $I_{\text{heat}}$. Except where noted, measurements of $\Delta S$ were made at $\delta T \sim 50 \text{ mK}$, although the error bars in Fig. 2 demonstrate that the measurements would have been just as accurate with $\delta T$ set to 30 mK. The fixed pressure condition of Eq. 1 is met by working well below the Fermi temperature of the 2DEG, $T_F \sim 100 \text{ K}$, where degeneracy pressure dominates [30].

Data Availability Data generated for, and analyzed in, this study are available at https://github.com/nikhartman/spin_entropy. The repository also contains all code necessary to complete the analysis and create each of the figures in this manuscript.

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Author Contributions NH and CO fabricated the mesoscopic device. GaAs heterostructures and their characterization were provided by SF, GG, and MM. SL and MS worked on early versions of the experiment and provided helpful discussion. NH performed measurements and analyzed data. Manuscript written by NH and JF with additional feedback from all authors.

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