Highly Enhanced Thermopower in Two-Dimensional Electron Systems at Millikelvin Temperatures

Srijit Goswami,[*] Christoph Siegert, Matthias Baenninger, Michael Pepper,[†] Ian Farrer, and David A. Ritchie
Cavendish Laboratory, University of Cambridge, J.J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom

Arindam Ghosh[‡]
Department of Physics, Indian Institute of Science, Bangalore 560 012, India

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We report experimental observation of an unexpectedly large thermopower in mesoscopic two-dimensional (2D) electron systems in GaAs/AlGaAs heterostructures at sub-Kelvin temperatures and zero magnetic field. Unlike conventional nonmagnetic high-mobility 2D systems, the thermopower in our devices increases with decreasing temperature below 0.3 K, reaching values in excess of 100 μV/K, thus exceeding the free electron estimate by more than 2 orders of magnitude. With support from a parallel study of the local density of states, we suggest such a phenomenon to be linked to intrinsic localized states and many-body spin correlations in the system.

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The diffusion thermopower (TP), S_d, in a solid depends on the variation of scattering time or the density-of-states (DOS) in the vicinity of the Fermi energy (E_F), and is expressed by the Mott relation [1]:

$$S_d = \lim_{\Delta T \to 0} \frac{V_{\text{th}}}{\Delta T} = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{d \ln \sigma(E)}{dE} \bigg|_{E=E_F} \tag{1}$$

where V_{th} is the thermovoltage at temperature T, and $\sigma(E)$ is the energy-dependent conductivity. Consequently, TP is highly sensitive to gaps in the DOS or proximity to the band edge in strongly localized systems. This has been exploited in investigating the charge density driven localization transition in a disordered 2DES [2]. Similar TP measurements have been used to explore quantum insulating ground states of a bulk 2D electron gas in high magnetic fields [3]. On the other hand, being a measure of entropy per carrier, the third law of thermodynamics requires TP → 0 as $T \to 0$ in delocalized or metallic systems, though the magnitude of TP depends critically on the energy dependence of the scattering mechanism of conduction electrons. This has direct implications for a large class of highly correlated 3D systems [4–6], such as dilute magnetic alloys, heavy Fermions, or Kondo lattice compounds. Here, the conduction electrons undergo spin-flip scattering at the localized d or f sites, which may lead to very large TP, exceeding several hundred μV/K [7]. In this study, we focus on nonmagnetic delocalized high-mobility 2D electron systems (2DES) at GaAs/AlGaAs interface, which are not expected to contain localized spins (LS), and indeed, earlier measurements of zero magnetic field diffusion TP of 2D electron gases yielded TP $\lesssim 0.4$ μV/K even at T as large as ≈ 3 K [8].

Recently, several experiments on nonequilibrium transport in quasi-one-dimensional quantum point contacts (QPCs) [9,10] and mesoscopic 2DES [11–13] suggest the possibility of LS in these III-V semiconductor-based low-dimensional systems. While the microscopic origin of the LS in such materials remains unclear, measurements in QPCs have revealed an additional contribution to the diffusion TP near the so-called "0.7 state" that is not entirely understood [14]. A more controlled study has been carried out with electrostatically confined single LS in odd-electron quantum dots, where the spin-flip scattering was found to result in large TP ($\simeq 60$ μV/K) at low T [15], which was attributed to spin-entropy transfer. Though GaAs/AlGaAs-based 2DESs form the host in many of these studies, no systematic TP experiments have been reported in unconfined 2DESs at mesoscopic length scales, which could potentially provide crucial insight into the nature and role of intrinsic spins. Here we report the observation of a giant diffusion TP in delocalized (conductivity $\gg e^2/h$) 2DES in high-mobility GaAs/AlGaAs heterostructures at $T < 0.3$ K. We find that at most gate voltages $|S_d|$ *increases* with decreasing T down to the base lattice temperature (T_{latt}) of $\simeq 70$ mK, and thus cannot be explained with free noninteracting electrons in 2D. We complement TP measurements with nonequilibrium transport, and show that these results point strongly towards the existence of LS in 2D mesoscopic systems. Our findings may have a direct impact on the understanding of many experimentally reported, but not fully understood, phenomena in low-dimensional quantum systems, such as the 0.7-state [9] and breakdown of Wiedemann-Franz law in QPCs [16], the zero-bias anomaly [11–13] and anomalous Hall effect in 2DESs at low temperatures [17].

Silicon δ-doped heterostructures with thick (≈ 80 nm) spacer layer of undoped AlGaAs were employed in our experiments. Similar devices were earlier used for nonequilibrium transport and Hall measurements [11,12,17]. The electron mobility in these wafers was in the range of

1–3 × 10^6 $\mathrm{cm}^2/\mathrm{V\,s}$ at the as-grown electron sheet density, $n_s \simeq 1 \times 10^{11}$ cm^2 resulting in a long elastic mean free path ($\gtrsim 10$ μm). A schematic of the device structure for TP measurements is shown in Fig. 1(a) [inset of Fig. 1(c) shows an SEM image of the device]. Central to the design is the 5 μm × 5 μm full gate (FG) which forms the mesoscopic device under study. A voltage (V_{FG}) on this gate tunes n_s in the 2DES directly below, thus allowing for a detailed study of the thermovoltage as a function of E_F of the mesoscopic system. The QPCs on either side of FG serve three purposes: (i) Lateral isolation of the device region from the remaining ungated 2DES, (ii) validation of the measurement technique with known thermoelectric behavior of QPCs, and (iii) electron temperature calibration of the mesoscopic region with respect to T_{latt} using the procedure described in Ref. [18]. An oscillatory heating current (I_h) with frequency $f_h = 7.3$ Hz was used between remote leads 4 and 5. The thermovoltage detection was performed with a lock-in amplifier at $2f_h$ to ensure a purely thermal origin of the signal. Figure 1(b) shows $-V_{\mathrm{th}}$ across QPC$_1$ as a function of the split-gate voltage V_{QPC1} for various I_h (0.2 μA → 0.5 μA) with T_{latt} fixed at the base. The peaks in $|V_{\mathrm{th}}|$ between two consecutive conductance plateaus could be scaled (not shown) for $I_h \leq 0.5$ μA yielding the electron temperature T_m at the center of the mesoscopic region as a function of I_h [see inset of Fig. 1(b)]. The quantitative agreement between measured QPC TP and the energy-derivative of its conductance [through Eq. (1)] supports the independent, noninteracting electron description of the QPC at higher subbands. We also notice that (i) TP in our devices has a purely diffusive origin, and any contribution from phonon drag is negligible, as expected for GaAs-based 2DESs below ~0.3 K [19], (ii) scaling of thermopower for $I_h \leq 0.5$ μA suggests thermal broadening to be negligible at these currents, and (iii) Fig. 1(c) shows the TP of QPC$_2$ below the first subband where a clear deviation from the Mott relation is observed near the 0.7 structure. This deviation has been studied by Appleyard *et al.* [14], though its origin still remains elusive. However, it serves as proof that the observed TP is indeed capable of detecting signatures of many-body spin-correlated states in low-dimensional systems. For subsequent TP measurements of the mesoscopic region, both QPCs were pinched off, and V_{th} was measured after adequate amplification. The temperature difference (ΔT) across the device was estimated as $\Delta T \approx (L/\xi)[T_m(I_h) - T_{\mathrm{latt}}]$, where $L(= 5$ μm) and $\xi(\approx 100$ μm) are the device length and thermal relaxation length in high-mobility GaAs/AlGaAs systems, respectively [18,20]. For electrical transport, we have used a standard ac-dc technique to measure both linear response conductivity ($G|_{V_{\mathrm{ds}}=0}$) and nonequilibrium differential conductivity (dI/dV_{ds}), where V_{ds} is the drain-source bias. The electrical characteristics were recorded at $I_h = 0$, and thermovoltages were measured at $I = 0$.

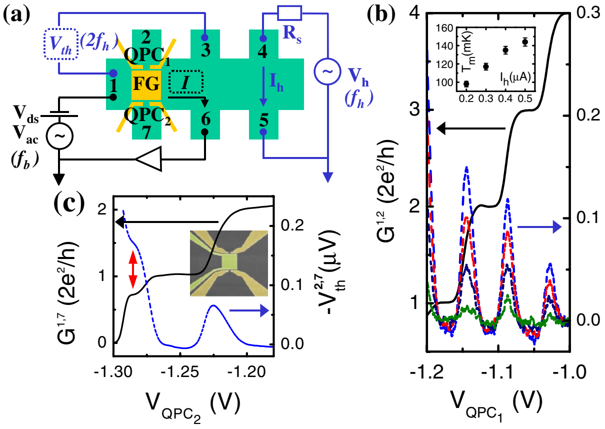

FIG. 1 (color online). (a) Schematic of the experimental setup used to perform thermovoltage (blue lines) and conductance (black lines) measurements. (b) Characteristic peaks in V_{th} as a function of the QPC gate voltage (V_{QPC_1}) for heating currents (I_h) ranging from 0.2 μA (bottom trace) 0.5 μA (topmost trace). (Inset) Electron temperature at the center of the 2D mesoscopic region obtained from the QPC TP analysis (see text). (c) A similar trace for QPC$_2$, where a distinct 0.7 structure is observed. Arrows mark its position in conductance and the corresponding deviation of thermovoltage from the semiclassical expectation. (Inset) scanning electron microscopy image of the device with a central 5 μm × 5 μm gate used to form the mesoscopic system.

Figure 2(a) shows the measured thermovoltage of the mesoscopic device (at various I_h) as a function of V_{FG} at $T_{\mathrm{latt}} \simeq 70$ mK. We concentrate on the delocalized regime of the 2DES, where $G \sim (2 - 20) \times e^2/h$ [inset of Fig. 2(a)], which corresponds to $V_{\mathrm{FG}} \gtrsim -0.52$ V, or equivalently, $n_s \gtrsim 1.8 \times 10^{10}$ cm^{-2}. A reproducible fluctuating behavior in V_{th} rides on an overall background that increases with decreasing n_s. Note that the decreasing magnitude of V_{th} with increasing V_{FG} ensures that the contribution to V_{th} from the ungated part of the 2DES is negligible, and the measured V_{th} arises predominantly from the TP of the mesoscopic region. The V_{th} vs V_{FG} traces could be collapsed onto a single trace by normalizing each V_{th} with the corresponding ΔT obtained from QPC calibration. Strikingly, Fig. 2(b) indicates the TP of our mesoscopic device to exceed 100 μV/K at low n_s, which is unexpectedly large for a delocalized system at sub-Kelvin temperatures and zero magnetic field. While the qualitative agreement with the energy derivative of linear conductivity [dotted line in Fig. 2(b)] provides further support for the diffusive origin of TP, the scaling also confirms that increasing I_h does not lead to thermal broadening or substantial lattice heating.

Increase in TP with decreasing n_s can be envisaged for free degenerate electrons, as well as for systems close to a localization transition [2,21,22]. The latter is plausible at $G \ll e^2/h$, when the 2DES becomes inhomogeneous deep into the band tail, and transport is dominated by classical percolation [22–24]. In our case however, $G \gg e^2/h$, and direct Hall measurements also indicate the charge

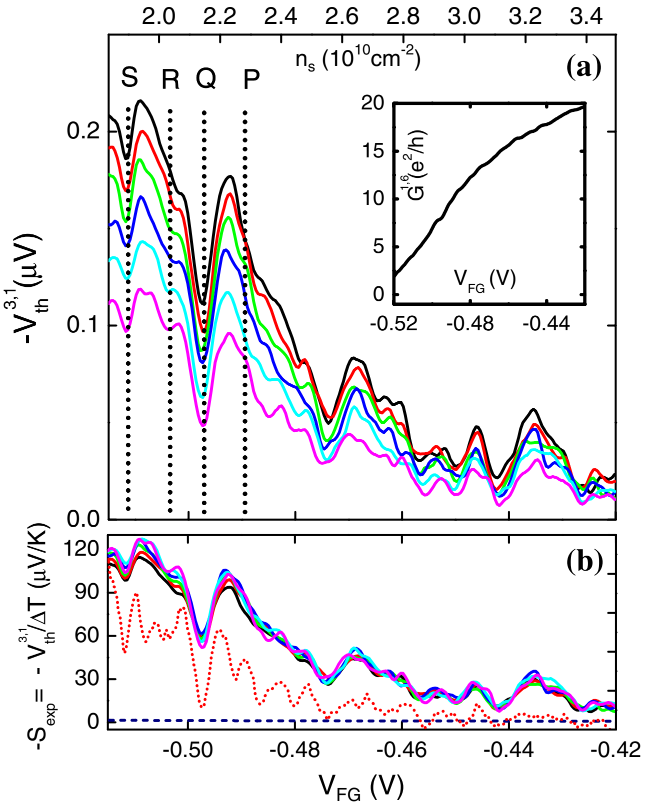

FIG. 2 (color online). (a) Variation of $-V_{th}$ as a function of V_{FG} for I_h ranging from 0.15 μA (bottom trace) to 0.25 μA (top trace). The top axis shows the corresponding electron sheet density (n_s) of the mesoscopic system. (Inset) equilibrium conductance (G) in this V_{FG} range. (b) Curves in (a) scaled by ΔT to obtain thermopower (S_{exp}). (Dotted line) calculated $d\ln G/dV_{FG}$ (arb. units) using G from the inset in (a), confirming the diffusive origin of TP. (Dashed line) Expected TP in the free electron picture.

distribution to be uniform [17]. Moreover, the TP never reverses its sign, ruling out sequential or cotunneling effects in unintentional quantum dots within the mesoscopic region [24,25]. In the free electron scenario with scattering from the dopant potential, Eq. (1) estimates the TP to be $\approx -\pi^2 k_B^2 T(p+1)/3|e|E_F$ $(p \approx 1.5)$ [26], which is nearly 2 orders of magnitude lower than what is observed experimentally [dashed line in Fig. 2(b)].

The temperature dependence of V_{th} provides further evidence of the nontrivial origin of enhanced TP in our devices. This is shown in Fig. 3(a) for a selected range of V_{FG} centered around a local minimum ($V_{FG} \approx -0.497$ V). Increasing T_m from 98 mK to 228 mK (at constant $I_h = 0.2$ μA), washes out strong fluctuations in V_{th}, and results in a decrease in its overall magnitude for $T_m \gtrsim 150$ mK. Two distinct behaviors were observed [see Fig. 3(b)]: At V_{FG}, labeled A and C, away from the local minimum, $|V_{th}|$ increases monotonically with decreasing T_m, but at the minimum (B) $|V_{th}|$ saturates at an intermediate temperature, and even decreases when T_m is reduced further. Clearly, this is not expected either in the free electron scenario where $|V_{th}| \propto T_m$ [black dotted line in Fig. 3(b)], or close to localization where TP is expected to increase with decreasing T_m at all V_{FG} [2,21,22].

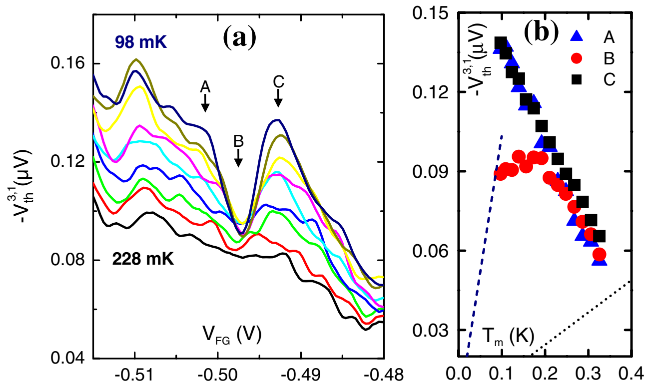

FIG. 3 (color online). (a) $-V_{th}$ versus V_{FG} for 98 mK $\leq T_m \leq$ 228 mK. (b) Distinct T dependences for positions marked A, B, and C in (a). Calculated T dependence expected for a free electron system (dotted) and Kondo lattice system for $T < T_K$ (dashed).

The feasibility of spontaneous spin effects in high-mobility nonmagnetic 2DESs have been discussed widely [11,13,27], where both the background disorder and many-body exchange interaction play crucial roles. While the signature of exchange-driven Stoner ferromagnetism on the TP of a 2DES at low T is not clear, a disorder-induced two-component fluctuation in the conduction band may lead to LS embedded inside the delocalized Fermi sea [13]. This may form a scenario similar to Kondo lattice compounds or dilute magnetic alloys. V_{FG} tunes the Fermi wave vector, and consequently the relative scales of Kondo coupling and RKKY magnetic exchange, which have characteristic signatures in the DOS near E_F. In nonequilibrium electrical transport, such low energy structures in the DOS are often manifested as a zero-bias anomaly (ZBA) in dI/V_{ds} around $V_{ds} = 0$. Figure 4 shows two different forms of ZBAs in our device as V_{FG} is changed, alternating between a single-peak at $V_{ds} = 0$ (representing the Kondo resonance at individual LS) and a double-peaked ZBA that has a shallow minimum close to E_F (indicating finite interspin interaction and local magnetic ordering [13]). We note that both temperature and V_{FG} dependence of V_{th} are directly connected to the structure of ZBAs. The strong minima observed in $|V_{th}|$ at -0.497 V and -0.510 V (indicated by S and Q in Figs. 2 and 4) correspond to strong single peaked ZBAs, i.e., when interspin exchange is small and Kondo-coupling dominates. Analogous to heavy Fermions or dilute magnetic alloys, the T dependence of $|V_{th}|$ in this state is nonmonotonic [6,28] (see trace B in Fig. 3), and shows a downturn just below $T_K \approx 0.2$ K (T_K was determined independently from both equilibrium and nonequilibrium transport. For details see [12]). When spin-spin exchange is strong, illustrated by traces at P and R with split ZBA, $|V_{th}|$ increases monotonically down to base temperature, implying that the

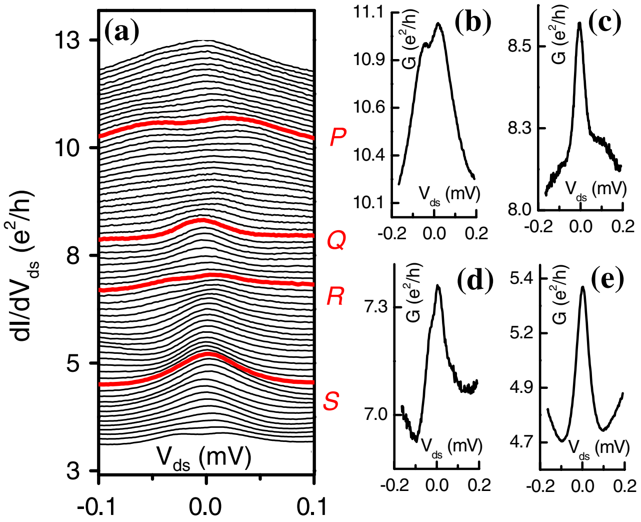

FIG. 4 (color online). (a) Modulation of the ZBA with V_{FG} varying from -0.482 V (top) to -0.516 V (bottom). Bold plots correspond to V_{FG} values marked P-S in Fig. 2(a). P,R(Q,S) show the strongest double (single) peaked ZBAs. (b)–(d) Individual ZBA corresponding to P-S, respectively, in (a).

giant TP observed at low temperatures may arise from scattering of the conduction electrons by unscreened magnetic moments in the 2DES.

Many quantitative approaches towards thermal transport coefficients in Kondo lattice systems exist in the periodic Anderson model [28], although primarily in the context of heavy atom intermetallic compounds [4,7]. Nevertheless, in the absence of magnetic interactions, a universal behavior of TP (for $T \ll T_K$) in these systems can be expressed as $S_d = -\alpha(k_B/|e|)(T/T_K)$ [28], where $\alpha \sim \mathcal{O}[1]$. An estimate with $\alpha = 2$, and typical experimental $T_K = 0.25$ K is shown as the dashed line in Fig. 3(b). Despite uncertainties in the numerics, the asymptotic behavior of trace B is encouraging, though measurements need to be extended to lower electron temperatures for more quantitative conclusions. Two important points to note: (i) Unlike many Kondo lattice systems, or even semiconductor quantum dots [15], we do not find any change in the sign of V_{th} within the experimental temperature range, remaining negative throughout. This indicates that the average energy of the quasiparticles is negative, as would be expected for a quasiballistic electronic system in the Kondo regime [15]. (ii) We find that the modulations in V_{th} can be traced to V_{FG} as large as -0.42 V, where the large zero-bias conductance $G|_{V_{\mathrm{ds}}=0} \approx 20 \times e^2/h$) makes the ZBA essentially undetectable. This establishes the greater sensitivity of TP over electrical transport in detecting anomalies in the DOS near E_F.

In conclusion, we have measured unexpectedly large values of diffusion thermopower (in excess of 100 μV/K) in delocalized 2D mesoscopic electron systems. Below 0.3 K, the thermopower was found to increase with decreasing temperature indicating the failure of non-interacting electron model in this regime. We suggest that the observed enhancement in thermopower may be related to the formation of localized spins in the system, and draw analogies between nonmagnetic high-mobility electron devices and Kondo lattice compounds.

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*sg483@cam.ac.uk.

†Present address: Department of Electronic and Electrical Engineering, University College, London.

‡arindam@physics.iisc.ernet.in.

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