Spontaneous breaking of time reversal symmetry in strongly interacting two-dimensional electron layers in silicon and germanium

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We report experimental evidence of a remarkable spontaneous time reversal symmetry breaking in two-dimensional electron systems formed by atomically confined doping of phosphorus (P) atoms inside bulk crystalline silicon (Si) and germanium (Ge). Weak localization corrections to the conductivity and the universal conductance fluctuations were both found to decrease rapidly with decreasing doping in the Si:P and Ge:P δ-layers, suggesting an effect driven by Coulomb interactions. In-plane magnetotransport measurements indicate the presence of intrinsic local spin fluctuations at low doping, providing a microscopic mechanism for spontaneous lifting of the time reversal symmetry. Our experiments suggest the emergence of a new many-body quantum state when two-dimensional electrons are confined to narrow half-filled impurity bands.

Invariance to time reversal is among the most fundamental and robust symmetries of nonmagnetic quantum systems. Its violation often leads to new and exotic phenomena, particularly in two dimensions (2D), such as the quantized Hall conductance in semiconductor heterostructures [1], the quantum anomalous Hall effect in topological insulators [2] or the predicted chiral superconductivity in graphene [3]. The breaking of time reversal invariance is experimentally achieved either by an external magnetic field or intentional magnetic doping. Here we show that strong Coulomb interactions can also lift the time reversal symmetry in nonmagnetic 2D systems at zero magnetic field.

While bulk P-doped Si and Ge have been extensively studied in the context of electron localization in three dimensions [4–9], confining the dopants to one or few atomic planes (δ-layers) of the host semiconductor has recently led to a new class of 2D electron system [10–18]. Electron transport in these atomically confined 2D layers occurs within a 2D impurity band where the effective Coulomb interaction is parameterized in terms of $U/\gamma$, with $U$ being the Coulomb energy required to add an additional electron to a dopant site, and $\gamma$, the hopping integral between adjacent dopants. Since each dopant P atom contributes one valence electron, the impurity band is intrinsically `half filled’ (schematic in Fig. 1a), which reinforces the interaction effects due to the in-built electron-hole symmetry, and forms an ideal platform to explore the rich phenomenology of the 2D Mott-Hubbard model, ranging from Mott metal-insulator transition (MIT) to novel spin excitations and magnetic ordering [14–17].

In this Letter we show evidence of spontaneously broken time reversal symmetry in 2D Si:P and Ge:P δ-layers as the on-site effective Coulomb interaction is increased by decreasing the doping density of P atoms. Quantum transport and noise experiments indicate a strong suppression of quantum interference effects at low doping densities. We could attribute this to a spontaneous breaking of time reversal symmetry which manifest in an unambiguous suppression of universal conductance fluctuations (UCF) at zero magnetic field.

The preparation of the P δ-layers in Si and Ge have been detailed in earlier publications [10, 11, 18], and parameters relevant to the present work is supplied in the Supplementary Information (SI). The Drude conductivity ($\sigma_D$) of the δ-layers decreases with decreasing doping as $\sigma_D \propto n^{3/2}$ (Fig. 1b), where $n$ is the electron density measured from Hall effect, implying significant scattering from charged dopants [19]. We find $\sigma_D \gg e^2/h$ in all devices, ensuring a nominally weakly localized regime. All electrical transport measurements were carried out in a dilution refrigerator with an electron temperature of 0.15 K using low frequency ac lock-in technique. The electron transport was strictly diffusive with $k_B T \tau_0/h \ll 10^{-2}$, because of short momentum relaxation times $\tau_0 \sim 10 - 100$ fs, and displays negative logarithmic correction to conductivity in the quantum coherent regime (Fig. 1c) [11].

The key advantage of using both Si and Ge as host semiconductors is the factor of three difference in the Bohr radius, $a_B$, which allows us to achieve a wide range of average effective dopant separation ($r_p/a_B^*\gamma$) within the similar range of doping density ($r_p \approx 2/\sqrt{\pi n}$). As shown in the scale bar of Fig. 1b, $r_p/a_B^*$ has an overall range from $\approx 0.6$ to 3. This corresponds to a range of $\gamma \sim 10 - 20$ meV and $\sim 20 - 50$ meV for the Ge:Si and Si:P devices respectively, assuming hydrogenic orbitals [20]. Since $U \sim 200$ meV and $\sim 50$ meV for single P donor in Si and Ge, respectively, the effective on-site Coulomb interaction $U/\gamma$ can be $\gg 1$, particularly in lightly doped Si devices.

In Fig. 1d, we show the transverse magnetic field ($B_\perp$) dependence of the quantum correction to conductivity, $\sigma_{QF}(B_\perp) = \sigma(B_\perp) - \sigma(0) - \sigma_d$, where $\sigma_d = -(\sigma_0^2/n^2e^2)B_\perp$, is the classical correction to the Drude conductivity. Due to diffusive nature of our devices the quantum correction from the electron-electron interac-
The Fermi energy, $E_F$, lies near the center of the impurity band whose width is determined by the hopping integral, $\gamma$. (b) The Drude conductivity $\sigma_D$, as a function of $n$ for SiP and GeP devices. The range of the effective dopant separation, $r_p/a_B$, and the device nomenclature are shown in the shaded panel on the right, where HD, MD and LD correspond to high density, medium density and low density respectively. The corresponding densities are 2.5, 1.1 and $0.5 \times 10^{14}$ cm$^{-2}$ respectively for Si and 1.35, 0.46 and $0.32 \times 10^{14}$ cm$^{-2}$ respectively for Ge. (c) The temperature dependence of conductivity, $\sigma$ (scaled by the Drude conductivity, $\sigma_D$) for heavily and lightly doped $\delta$-layers in Si and Ge. (d) The quantum correction to conductivity, $\sigma_{QI}$ (obtained from measured magnetoconductivity after eliminating the classical contribution) as a function of perpendicular magnetic field, $B_{\perp}$, at 0.28 K for Si_HD, Si_MD and Si_LD. The phase breaking field, $B_\phi$, is shown by vertical lines. The solid black lines are fits using Eq. 1 in the main text.

The quantum correction cannot be due to finite experimental range ($\approx 0 - 14$ T) of $B_{\perp}$, which exceeds both $B_\phi$ and $B_0 (= \hbar/4eD\tau_0)$, the upper cutoff field due to quantum relaxation) by factors of 1000 and 2 respectively even for the least doped devices at 0.28 K (Table I in SI). Spin-orbit interaction is also known to be small for P-doped (bulk) Si and Ge, and independent of the density of the dopants. Any long range magnetic order is also unlikely because the Hall resistance was found to vary linearly with $B_{\perp}$ at all $T$ (see SI, section S7) in all our devices. [14].

The suppression of quantum correction to conductivity has been observed in low density electron gases in Si MOSFETs near the apparent MIT [23] although its microscopic origin remains unclear with both temperature dependant screening of disorder and interaction driven spin fluctuations suggested as competing mechanisms.
However, the formation of local magnetic moments in the presence of strong Coulomb interactions, is known to occur in three dimensional P-doped Si close to the MIT \[7–9\]. These moments serve to remove the time reversal symmetry, suppressing the coherent back-scattering of electrons. In 2D, the possibility of localized spin excitations at the Mott transition has been suggested theoretically \[17–25\], but without any experimental evidence so far.

To probe whether the observed suppression of localization correction indeed manifests a breaking of the time reversal symmetry, we have measured the UCF as a function of \(T\) and \(B_\|\) from slow time-dependent fluctuations in the conductance \(G\) of the \(\delta\)-layers which represents the ensemble fluctuations via the ergodic hypothesis \[22,26,29\]. The time dependent conductance fluctuations (inset of Fig. 2a) are analyzed to obtain the power spectral density, \(S_G\), which on integration over the experimental bandwidth gives the normalized variance, \(N_G = \int S_G G^2 df = \langle \delta G^2 \rangle / \langle G \rangle^2\) as shown in Fig. 2a (see Ref. [30] and SI, section S3 for details). Fig. 2b shows \(N_G\) as a function of \(T\) for Si_HD. For \(T \gtrsim 15\) K, \(N_G\) increases with decreasing \(T\), which is a hallmark of UCF. In this regime, one expects \(N_G \propto L_\phi^4 n_T \propto 1/T\), where \(L_\phi(\propto T^{-0.5})\) and \(n_T(\propto T)\) are the phase coherence length and density of active two level fluctuators \[27\] (Fig. 2b).

The absolute magnitude of \(N_G\) in all devices correspond to the change in conductance by \(\sim O(\hbar^2/l)\) due to a single fluctuator within a phase coherent box (see SI, section S5), establishing the observed noise to be indeed from mesoscopic fluctuations.

As a function of \(B_\|\), the magnitude of UCF is expected to decrease by an exact factor of two at two field scales, first at \(B_\| \sim B_\phi\) when the time reversal symmetry, and hence the Cooperon (self-intersecting diffusion trajectories) contribution, is removed \[26,31,32\] and second at \(B_\| \sim B_Z = g_B T/g\mu_B\) due to removal of spin degeneracy \[26,32,33\], where \(g\) and \(\mu_B\) are the \(g\)-factor and \(\mu_B\) respectively. The inset of Fig. 2d shows schematically the two reductions in UCF magnitude as a function of \(B_\|\).

Fig. 2d shows that the UCF magnitude in heavily doped Ge_HD (violet symbols) consists of both factors of two reduction at \(B_\| \approx B_\phi\) and \(B_\| \approx B_Z\), corresponding to the removal of time reversal symmetry and spin degeneracy, respectively, whereas the lightly doped devices, such as Si_MD, shows almost no variation in the UCF magnitude on the scale of \(B_\phi\) but decreases by a factor of two at \(B_\| \approx B_Z\). To confirm this scenario, we have also recorded the variation of \(N_G\) in Si_MD as a function of parallel magnetic field, \(B_\|\), which couples only to spin degree of freedom (Fig. 2c). The factor of two reduction at \(B_\| \sim B_Z\) (shown by vertical arrows in Fig. 2c) for \(T = 0.5\) K and \(4.2\) K establishes that the \(1/f\) noise in our devices indeed arises from the UCF mechanism.

Since the reduction in UCF at \(B_\| \approx B_\phi\) is associated only to removal of the fundamental time reversal symmetry of the underlying Hamiltonian \[31\], its absence in the lightly doped \(\delta\)-layers is unique, and has not been previously observed in interacting 2D systems in semiconductors \[34,39\]. To elaborate, we have compiled the \(B_\|\)-dependence of \(N_G\) normalized by \(N_G\phi\), where \(N_G\phi\) is the value of \(N_G\) at \(B_\| \gg B_\phi\) but \(< B_Z\), for all devices in Fig. 3. \(N_G\phi\) was chosen at \(B_\| \sim 20B_\phi\) which was \(< B_Z\) for all the devices at all temperatures. The peak in \(N_G\) around \(B_\| = 0\) is progressively suppressed with decreasing doping density, and eventually for \(r_P/a_\|^2 > 1.5\), the Cooperon contribution to UCF noise at low \(B_\|\) becomes immeasurably small, implying a spontaneous breaking of time reversal symmetry even at \(B_\| = 0\) (Inset of Fig. 3).

To explore the origin of lifting of the time reversal symmetry in the \(\delta\)-layers, we subjected the devices to in-plane magnetic field, \(B_\|\), that resulted in a nonmonotonic magnetoconductivity in the lightly doped \(\delta\)-layers. The logarithmic increase in the magnetoconductivity at large \(B_\|\), as shown in Fig. 4a, was observed in all devices irrespective of doping level, and known to represent suppression of weak localization due to the finite width of the \(\delta\)-layers \[36\]. However, the negative magnetoconductivity around \(B_\| = 0\) often indicates the presence of local moments, because localization strengthens as phase coherence increases with the freezing of spin-flip scattering \[36,37\]. In such a case, the activated spin-flip processes across the Zeeman gap, leads to magnetoconductivity decreasing linearly with \(B_\|\) as \(\Delta \sigma(B_\|) = -\eta B_\|/T\), where \(\eta \sim (e^2 g_{imp} \mu_B^2) / (h k_B)\), and \(g_{imp}\) is the \(g\)-factor of the magnetic impurity \[36\]. As shown in Fig. 4c, we indeed find the \(\Delta \sigma(B_\|, T) \propto B_\|/T\) in Si_LD. The negative magnetoconductivity in \(B_\|\) is entirely absent in the heavily doped devices (Fig. 4b). This establishes that the spin fluctuations are entirely due to strong Coulomb interactions, and hence observable only in the lightly doped devices.
The compelling analogy with the bulk P-doped Si and theoretical exploration. Whether atomically confined Si:P and Ge:P crystals. The universal conductance fluctuations and in-plane magnetoconductivity suggest that local spin fluctuations in the presence of strong Coulomb interaction play an important role in the lifting the time reversal symmetry. Whether this indeed leads to a true interaction-induced metallic ground state in two dimensions needs further experimental and theoretical exploration.

Finally, to estimate the fraction of P-dopants that host a local moment, we compare the estimated \( \tau_s^{-1} \) in lightly doped Si\(_{\text{LD}}\) \((n = 5 \times 10^{13} \text{ cm}^{-2})\) with (1) the total momentum relaxation rate \( \tau_0^{-1} \approx 10^{14} \text{ s}^{-1} \) from the experimental Drude conductivity, although this involves scattering from neutral defects as well, and (2) calculated momentum relaxation rate \((\approx 2 \times 10^{13} \text{ s}^{-1})\) expected purely from the P-dopants (charged impurities) (see calculation details in Ref \[19\] and SI, section S6). This gives a bound between 2%–10% of the P-dopants to host local moments which is consistent with the fraction expected for half-filled impurity bands in bulk Si:P \[2\]. Importantly, while the weak localization correction is reduced only partially (30% in Si\(_{\text{LD}}\)), the UCF noise due to the Cooperons is completely suppressed for the weakly doped devices. It is possible that because the UCF noise involves interference between two Feynman propagators, it is more likely to be affected by the localized spins than the WL correction which is determined by a single self intersecting propagator. Note that we have not discussed spatial inhomogeneity or clustering in the distribution of dopants which can lead to coexistence of localized and delocalized phases \[14\], impact of multiple valleys \[33, 40\], or the inter-site Coulomb interaction \[34, 35\] which are unlikely to affect the time reversal symmetry.

In summary, magnetoconductivity and noise measurements reveal an unexpected spontaneous breaking of time reversal symmetry in 2D electron systems hosted in atomically confined Si:P and Ge:P crystals. The universal conductance fluctuations and in-plane magnetoconductivity suggest that local spin fluctuations in the presence of strong Coulomb interaction play an important role in the lifting the time reversal symmetry. Whether this indeed leads to a true interaction-induced metallic ground state in two dimensions needs further experimental and theoretical exploration.
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