Intermediate phase at the metal-insulator boundary in a magnetically doped two-dimensional electron system

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A magnetotransport study in magnetically doped (Cd,Mn)Te two-dimensional quantum wells reveals an apparent metal-insulator transition as well as an anomalous intermediate phase just on its metallic side. This phase is characterized by colossal magnetoresistancelike phenomena, which may be due to the phase separation of the electron fluid and the associated emergence of ferromagnetic bubbles.

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I. INTRODUCTION

Despite intensive research efforts, the apparent metal-insulator transition (MIT) in two-dimensional electron systems[1] (2DESs) remains one of the most challenging problems of condensed matter physics. Several recent theoretical studies[2–4] suggest the existence of an intermediate phase near the 2D MIT, where the competition between distinct ground states results in their nanoscale phase separation. A large number of possible configurations of these local regions often have comparable energies, resulting in time-dependent phenomena such as slow relaxation, aging, and other signatures of glassy dynamics. Indeed, such manifestations of glassiness and evidence for an intermediate metallic phase have been found recently in a 2DES in Si metal-oxide-semiconductor field-effect transistors (MOSFETs).[5–7] There is also growing evidence that phase separation is responsible for several striking effects in bulk transition metal oxides, such as manganites,[8] cuprates,[9] and similar complex magnetic materials.[10] In that context, magnetically doped 2DESs (M2DESs) in semiconductor heterostructures constitute an ideal system for studying both magnetism and reduced dimensionality effects near the MIT.

M2DESs in (Cd,Mn)Te quantum wells (QWs) are particularly appealing, since the 2D electron density n_s can be changed externally by an electric gate *independent* of the density of magnetic ions, so that interactions and the amount of disorder can be tuned separately. These M2DES have been well characterized in the studies of quantum Hall ferromagnetism[11] and quantum Hall effect.[12] Furthermore, (Cd,Mn)Te has a simple crystal structure and, most importantly, extensive studies have shown[13–16] that the distribution of Mn ions in (Cd,Mn)Te is perfectly random, implying an *absence* of chemical phase separation. Thus, the molecular-beam-epitaxy (MBE) grown (Cd,Mn)Te seems to be chemically and structurally the cleanest system for studying a possible electronic phase separation in a magnetic material.

Here, we report a magnetotransport study of M2DES in (Cd,Mn)Te QWs, which provides experimental evidence of an apparent MIT in M2DES. At sufficiently high temperatures T, the MIT is similar to that observed in low-mobility (high disorder) Si MOSFETs.[5]

However, when T is low enough, a qualitatively distinct transport regime emerges just on the metallic side of the MIT. In this intermediate phase, the resistivity ρ increases dramatically by several orders of magnitude with decreasing T. A magnetic field B applied *parallel* to the 2D plane gives rise to an enormous *negative* magnetoresistance (MR), which drives the system back to metallic conductivity. This is exactly the opposite of the behavior observed in nonmagnetic 2DES near the MIT,[1] but it resembles the phenomena found in manganites and other colossal magnetoresistance (CMR) materials.[8] We propose that they share essentially the same origin: a nanoscale coexistence of competing phases which, in our case, corresponds to the formation of ferromagnetic (FM) metallic bubbles embedded within a carrier-poor, magnetically disordered host. This picture is further supported by the observed strongly nonlinear current-voltage (I-V) characteristics in this regime, where sufficiently high excitation voltages V_{exc} destroy the heterogeneous state and, hence, drive the system back to metallic conductivity.

II. EXPERIMENT

The samples were grown by MBE and contain a 10 nm wide $Cd_{1-x}Mn_xTe$ QW, in which the 2DES is confined by $Cd_{0.8}Mg_{0.2}Te$ barriers.[11,12] A 10 nm thick layer of the front barrier residing 20 nm away from the QW is doped with iodine donors up to $n_I \approx 10^{18}$ cm^{-3}. In 0.5×1 mm^2 Hall bar structures A and B, Mn contents x are 0.015 and 0.005, the peak mobilities 3.0 and 6×10^4 cm^2/V s, and $1.4 \leqslant n_s \leqslant 4.2$ and $4.9 \leqslant n_s \leqslant 6.2\times 10^{11}$ cm^{-2} (controlled by a metal front gate), respectively, as determined from Hall resistance and the periodicity of Shubnikov–de Haas oscillations at $T=4.2$ K. The main experiment was performed in parallel B up to 9 T (the current $I \| B$) and down to either 0.24 K or 0.03 K. $\rho(B,T)$ was measured with a standard low-frequency lock-in technique. For $T \gtrsim 0.3$ K, $\rho(T)$ did not depend on whether the samples were cooled in $B=0$ or $B \neq 0$.

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show $\rho(T)$ of structure A at $B=0$ and $B=3$ T for different n_s. The data were obtained with low

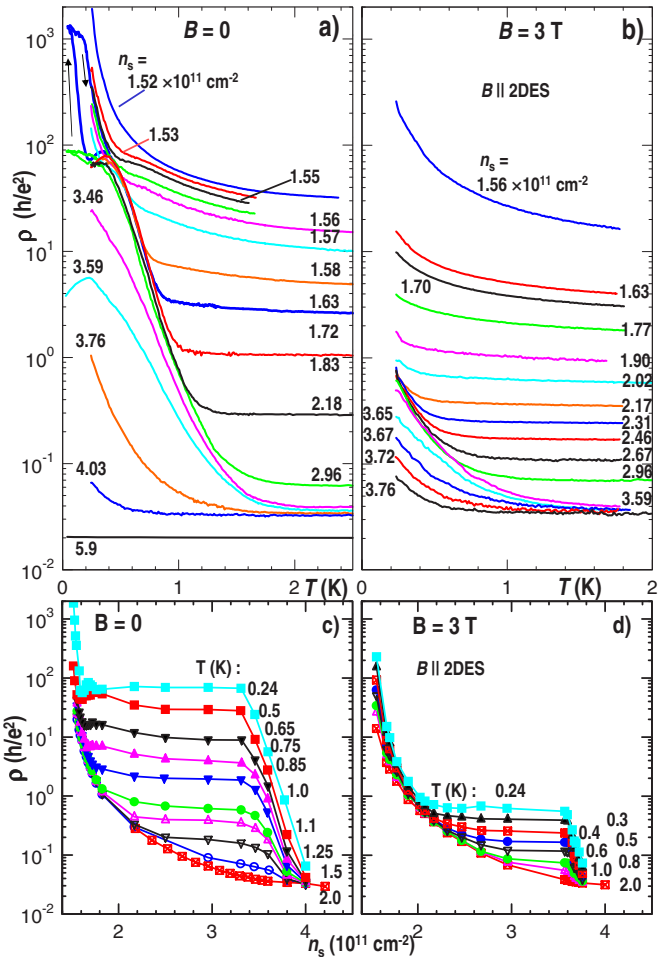

FIG. 1. (Color online) Resistivity as a function of T for different electron densities at $B=0$ (a) and $B=3$ T (b). The same data as a function of n_s at selected T [(c) and (d)]. Except for the lowest trace in (a) all the data are from structure A.

$V_{exc} \sim 10\ \mu$V. At elevated T and high n_s, $\rho(T)$ is weakly metallic, i.e., $d\rho/dT \gtrsim 0$, which is seen better in the scale of Fig. 3(b). At $n_s^* \approx 2.4 \times 10^{11}$ cm^{-2}, $d\rho/dT$ changes sign, which is sometimes attributed to an apparent 2D MIT. In our case, this occurs at a relatively low $\rho \sim 0.2h/e^2$ in comparison with nonmagnetic 2DES.

Generally, the nature and even the existence of the metallic state in 2D are still strongly debated since, according to the scaling theory of localization,[17] for noninteracting systems there can be no metallic state in 2D. However, in recent years, there has been increasing experimental evidence of an apparent metallic state in many 2D systems [e.g., Si-MOSFETs,[18] GaAs (Ref. 19) modulation-doped quantum wells]. Experimentally, there are several criteria used to determine the critical concentration n_c for the metal-insulator transition in 2DES, such as the extrapolation of the hopping activation energy, nonlinear current-voltage characteristics, or the sign of the $d\rho/dT$ dependence. These criteria do not necessarily give the same results. There is now a more or less general consensus that the sign of $d\rho/dT$ is not a reliable way to determine n_c. Therefore, our results for n_c were obtained by using the other criteria mentioned above, and in analogy to the studies of other[20] 2D systems.

The critical density n_c for the MIT obtained from the extrapolation of the hopping activation energy, as well as from the saturation of the I-V characteristics,[20] is substantially lower $n_c = 1.7 \times 10^{11}$ cm$^{-2} < n_s^*$. It corresponds to $\rho_c \sim 2h/e^2$, which is similar to that found in nonmagnetic 2D systems. The n_c depends on magnetic field. At $B=3$ T, for example, we find $n_c(B=3\ \text{T}) = 2.1 \times 10^{11}$ cm^{-2}, indicating that, for high enough T, B shifts the system toward an insulating phase just as in standard nonmagnetic materials.

However, below some $T^*(n_s)$, a dramatic upturn of $\rho(T)$ by almost 3 orders of magnitude is observed for moderate n_s at $B=0$ [Fig. 1(a)]. Here, $\rho(T)$ increases down to ≈ 0.3 K, goes through a maximum, and either continues to grow or decreases as T is lowered. Around 0.3 K, a strong resistance noise is observed (not shown), while for $T \lesssim 0.3$ K, ρ becomes hysteretic with respect to both T and B. Moreover, for $0.3\ \text{K} < T < T^*$, $\rho(T)$ collapse onto almost the same curve for a wide range of densities $1.6 \lesssim n_s \lesssim 3.3 \times 10^{11}$ cm^{-2}. This striking behavior is further highlighted in Fig. 1(c), which shows clearly that ρ does *not* depend on n_s at a fixed T. Importantly, these densities $n_s \gtrsim n_c = 1.7 \times 10^{11}$ cm^{-2}, i.e., they belong to the metallic side of the MIT. The same dramatic upturn of ρ and a lack of $\rho(n_s)$ dependence are also clearly seen in B, but at a slightly lower T^* and in a narrower range of densities: e.g., at $2.1 \lesssim n_s \lesssim 3.0 \times 10^{11}$ cm^{-2} for $B=3$ T [Figs. 1(b) and 1(d)] where, again, these $n_s \gtrsim n_c(B=3\ \text{T})$.

Figure 2 depicts the MR of our devices. At high T, and at low T for either high or low n_s, a relatively weak positive MR is visible. In the weakly localized regime, where $k_F \ell > 1$ (k_F, Fermi wave vector; ℓ, mean free path), this positive MR is ubiquitous in diluted magnetic semiconductors (DMSs), such as (Cd,Mn)Te, in the paramagnetic phase.[21–24] It originates from the giant spin splitting Δ_s of the electron states, which considerably affects quantum corrections to the conductivity brought about by the effect of disorder modified electron-electron interactions.[25] The same mechanism is believed to be responsible for positive MR in Si MOSFETs (Ref. 26) and other nonmagnetic 2DES, in which Δ_s is appropriately large. Since in DMS Δ_s is proportional to the magnetization M of the Mn spins, this positive MR scales with B and T like the Brillouin function $B_{5/2}[B/(T+T_0)]$.[13,15] Here, $T_0(x) > 0$ describes a reduction of M by intrinsic antiferromagnetic (AF) couplings between the Mn ions, and its value is well known from extensive magneto-optical and magnetic studies in (Cd,Mn)Te layers.[13] In particular, $T_0(x=0.005) = 0.17$ K and $T_0(x=0.015) = 0.5$ K. Figure 2(c) shows the results of the MR calculations carried out according to this model[21,22,25] and without any fitting parameters, together with the experimental results for structure B in which $k_F \ell \ll 1$, so that the theory should apply quantitatively. Indeed, there is a good agreement between measured and calculated positive MRs, at low B.

However, exclusively in the range of T and n_s where the lowering of T results in a strong increase of $\rho(T)$, the MR is dominated by a strong negative component. The negative MR is not observed on the insulating side of the MIT, where localization is driven by nonmagnetic disorder and visible already at high T. A sizable negative MR shows up only if

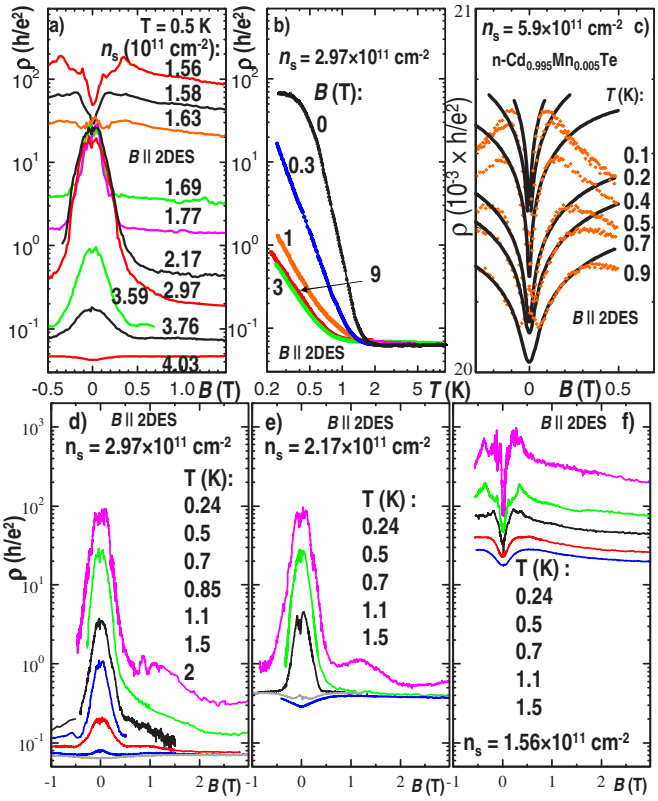

FIG. 2. (Color online) (a) Resistivity $\rho(B)$ for different n_s at $T=0.5$ K. (b) $\rho(T)$ at different B. (c) $\rho(B)$ at different T measured (symbols) and calculated (lines) for structure B. [(d), (e), and (f)] $\rho(B)$ measured at different T for $n_s=2.96$, 2.17, and 1.56 $\times 10^{11}$ cm^{-2}, respectively.

magnetic effects lead to T-dependent localization, so that B can drive the system back to the metallic phase. Remarkably, this negative MR does not scale with the paramagnetic $B_{5/2}$, which suggests an involvement of different magnetic phases.

A dramatic upturn of $\rho(T)$ and the related negative MR have been observed in bulk DMS (Refs. 21, 24, and 27–29) below a characteristic temperature T^* [e.g., $T^* \approx 1.5-2.0$ K in bulk n-$Cd_{1-x}Mn_xTe$, but with a much higher $x=0.047, 0.06$]. These effects were attributed[21,29] to the formation of bound magnetic polarons (BMPs)—clouds of Mn ions mutually polarized by an electron bound to *a single* donor, despite the fact that they are observed from near the MIT to deep into the metallic phase, where all the donors are ionized. This is even more puzzling in a modulation-doped n-(Cd,Mn)Te QW, where Mn are electrically neutral and the ionized donors are far from the conducting channel, so that the BMP effects are expected to be considerably less important than in three-dimensional (3D).

On the other hand, the tendency toward electronic phase separation should be more pronounced in 2D than in 3D.[30] We propose, therefore, that similar to the case of manganites,[8] the intermediate phase in M2DES contains bubbles of different electronic phases, which account for the CMR-like behavior. Recent spectroscopic observation of excitonic transitions which persist deep into the metallic phase in similar n-(Cd,Mn)Te QW (Ref. 31) strongly supports our model.

In general, the question arises whether the observed phenomena do not result from an existence of chemical clustering. However, almost three decades of intensive transport, structural, magnetic, and spectroscopic studies have proven that distribution of Mn ions in II-Mn-VI materials is perfectly random.[15,32]

In particular, Mn concentrations obtained from magneto-optical studies[13] of Zeeman splitting of band electrons which, via s-d exchange is proportional to the magnetization of the Mn moments, agree well with concentration of isolated Mn calculated with assumption of their random distribution on cation sites. Moreover, these studies were performed on the samples grown by the same MBE facility as these used in the present paper.

Another proof of textbooklike random Mn distribution comes from the studies of the so-called magnetization steps in DMSs. They were studied intensively in different bulk DMSs (Ref. 14) and confirmed in 2D (Cd,Mn)Te heterostructures.[16] In particular, the probability that a Mn ion is isolated is $P_1=(1-x)$,[12] while $P_2=12x(1-x)$ (Ref. 18) describes the probability that Mn belongs to a nearest neighbor (NN) pair. Since antiferromagnetic exchange between NN Mn dominates, pairs do not contribute to the total magnetization unless the field is strong enough to override internal AF coupling. For (Cd,Mn)Te and NN pairs, it occurs at $B \approx 10$ T, and magnetization increase is indeed observed at this field in (Cd,Mn)Te. Importantly, experimentally observed magnetization gain agrees very well with that predicted within random-distribution model.

Additionally, in $Cd_{1-x}Mn_xTe$, Mn ions are electrically neutral (their $3d^5$ bands are deep below the valence band) and do not serve as dopants, in contrast to III-Mn-V DMS as well as transition metal oxides. Importantly, this makes it possible to change electron and Mn concentrations independently. Thus, MBE grown $Cd_{1-x}Mn_xTe$ seems to be chemically and structurally the cleanest material to study magnetic and electric clustering. This situation is dramatically different in III-V or II-IV DMSs where precipitates of other phases often control magnetic properties.

Having thus established that chemical clustering is not present in our samples, we turn to identify the nature of the electronic phases in question. We note that, according to the Zener theory of carrier-mediated FM, a ferromagnetic transition is expected to occur at low T in bulk zinc-blende DMS,[33] such as (Cd,Mn)Te. Recent Monte Carlo simulations[34–36] indicate a formation of isolated ferromagnetic bubbles and the CMR-like behavior at $T^* \gg T_C$ (T_C, Curie temperature) even in the absence of attractive impurity potentials, provided that AF couplings are strong enough. At $B=0$, the FM bubbles are oriented randomly, which diminishes percolation and thus enhances resistance. Since the magnetic field aligns the bubbles, a strong negative MR follows. These effects are expected to exist in both 3D and 2D cases but should be stronger in 2D than in 3D, in agreement with our results.

We notice also that in the case of 3D manganites, the correlated disorder of the lattice is crucial to destroy homogeneous FM state and to produce substantial CMR effect from clustering and charge ordering.[37] The same theoretical studies show that the electronic phase separation in 2D materials does not need such a trigger.

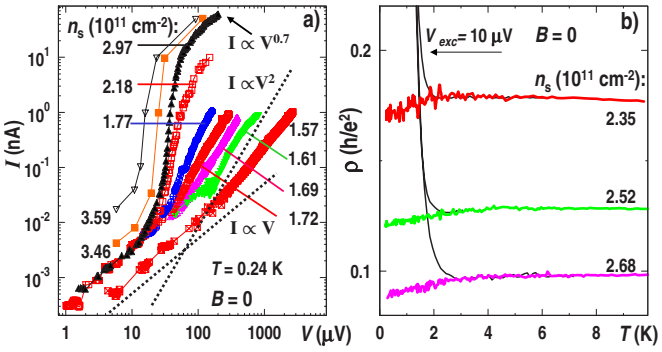

FIG. 3. (Color online) (a) *I-V* characteristics at different n_s. (b) $\rho(T)$ for different n_s at high (thick) and low (thin lines) excitation voltages V_{exc}= 500 and 10 μV, respectively.

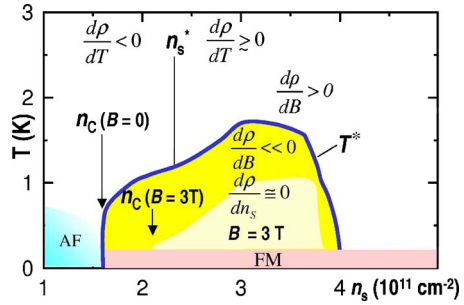

FIG. 4. (Color online) Different transport regimes in the n_s-T plane determined from the measurements in structure A. AF and FM states are sketched tentatively. FM ordering is expected in the metallic phase at low enough T based on the studies of 2D *p*-(Cd,Mn)Te QW (Refs. 36 and 42) and 3D *p*-(Zn,Mn)Te (Refs. 33 and 43), although other states might be possible. For $n_s<n_c$, we expect AF correlations to dominate.

The lack of $\rho(n_s)$ dependence in the intermediate phase [Fig. 1(c)], where magnetic effects take control over charge transport, is also consistent with the Zener theory of ferromagnetism,[33,36] which predicts that FM ordering temperature depends merely on the carrier density of states (DOS) and magnetic susceptibility of the Mn ions. However, DOS does not depend on n_s in 2D, resulting in this striking behavior. At the lowest n_s, where carriers become localized, the local FM order is destroyed, and the intrinsic AF interactions between the Mn ions dominate.[36]

In general, a heterogeneous state may be expected to exhibit glassy behavior and nonlinearities.[10] Indeed, signs of glassiness, such as ρ noise and hysteretic behavior [Fig. 1(a)] are present in our system. We have established that the *I-V* characteristics are strongly nonlinear [Fig. 3(a)]. In particular, while the Ohmic regime is observed for all values of n_s for V_{exc} below ≈ 10 μV, a strong nonlinear behavior is clearly seen for higher V_{exc}. For low n_s, the nonlinear characteristics obeys $I \propto V^2$, but in the intermediate, heterogeneous state, a much steeper dependence is first observed, which is then followed by $I \propto V^{0.7}$. This striking behavior resembles depinning of colloids,[38] Wigner glass to liquid transitions modeled for disordered 2DES,[39] as well as Wigner crystal depinning in quantum Hall systems.[40] In particular, the exponents 2 and 0.7 were found[38] in the *I-V* characteristics of colloidal dynamics and correspond to plastic and elastic depinnings, respectively. It is also possible that the observed nonlinearity stems from the current-induced rotation of FM domains, though the current density used here is 3 orders of magnitude lower than that employed for domain rotation in (Ga,Mn)As.[41] A sufficiently high V_{exc} destroys the heterogeneous state and drives the system back to metallicity, as expected: for $V_{exc} > 100$ μV $\gtrsim kT^*/e$, the metallic behavior ($d\rho/dT>0$) is clearly seen down to the lowest T for $n_s \gtrsim 2.2\times 10^{11}$ cm^{-2} [Fig. 3(b)].

Figure 4 shows the range of T and n_s where the intermediate phase is observed in our M2DES. As it is expected, in general,[3] a heterogenous state exists in the metallic phase just above the MIT: as the density of carriers n_s and their Fermi energy E_F are reduced, a tendency toward magnetic ordering is revealed just prior to the MIT. Moreover, since FM correlations are mediated by mobile carriers, the local magnetic ordering disappears as the number of mobile carriers is further reduced to zero as $n_s \rightarrow n_c^+$, in agreement with our results. In contrast, BMPs are most stable within the insulating phase and, even if they persist into the metallic phase, one would expect a decrease of T^* when n_s increases. This is exactly the opposite of what is observed in the experiment. It is interesting that the phase diagram (Fig. 4) reveals a striking qualitative similarity to that proposed for CMR materials,[8] cuprates,[44] and, in general, for model systems that form heterogeneous phases.[45]

IV. CONCLUSIONS

In summary, we have observed the emergence of an anomalous intermediate phase exhibiting colossal magnetoresistancelike phenomena in a M2DES just on the metallic side of the MIT. We attribute our findings to the competition between AF exchange characterizing the insulating phase and the FM correlations induced by itinerant electrons, resulting in the formation of FM metallic bubbles embedded within a carrier-poor, magnetically disordered matrix. Similarities to other systems, including nonmagnetic 2DES,[5–7] suggest a common origin of complex behavior near the MIT in a variety of materials.

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