Giant Magnetic Interaction (Condon Domains) in Two Dimensions

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We observe clear evidence for formation of a field-induced two-phase state in Br_2-intercalated graphite. This is the first time such an effect has been seen in a two-dimensional system, and the sharpness of the features is quite extraordinary. We also observe possible manifestations of domain-wall dynamics in the two-phase regime.

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An electron gas in a strong magnetic field B can undergo a series of phase transitions, periodic in $1/B$. These phase transitions are driven by the de Haas–van Alphen (dHvA) effect: Since the field seen by the electrons in $B = H + 4\pi M$, the dHvA magnetization $M = M(B)$ must be found self-consistently and, when plotted versus H, can be multivalued. Minimization of the free energy of the electrons will then lead to first-order transitions, in which the electrons jump from one Landau level to the next. This effect, known as magnetic interaction (MI),[1] has the same periodicity as the dHvA effect. Condon[2] showed that MI is sensitive to the sample demagnetization, so that very different results are found in rod-shaped and disk-shaped samples. In a disk, the first-order discontinuity in M is replaced by a smooth transition occurring in a finite field range. Within this "intermediate state" the electron gas is not spatially homogeneous, but is broken up into domains of two different magnetizations. MI effects, including domain formation, are now well documented in three-dimensional (3D) systems.[3] However, the dHvA magnetization should be greatly enhanced in a two-dimensional (2D) system, essentially because all of the electrons have the same Fermi surface area and hence a common dHvA frequency. This in turn should produce a giant MI effect. In this paper we report the first direct observations of such an effect in a 2D-layered compound, a stage-2 Br_2 graphite intercalation compound (GIC). While the overall features of the observed MI are consistent with the conventional picture, we observe a number of anomalies which suggest that 2D MI possesses a distinct character of its own.

Pure graphite is itself nearly two dimensional, with weakly interacting layers stacked along the c axis, but the two dimensionality is greatly enhanced by intercalating an acceptor compound, such as Br_2. The Br_2 forms uniformly spaced layers along the c axis (the stage number is defined as the number of graphite layers between successive intercalant layers) and, since the intercalant layers are insulating, the conductivity along the c axis is very low. Hence the Fermi surfaces are cylinders, and there are two different areas in a stage-2 compound. In Br_2 these areas correspond to dHvA "frequencies" of about 250 and 950 T (which vary somewhat from sample to sample). Batallan *et al.*[4] report frequencies of 240 and 1050 T, with a large number of "overtones" (harmonics and sum and difference frequencies of the fundamentals). Such overtones are characteristic of MI.

Figure 1 shows dHvA and Shubnikov–de Haas (SdH) oscillations for a stage-2 Br_2 GIC. Data were taken by the field-modulation technique, keeping the ac field small (≤ 30 G) to avoid distorting the oscillations and recording out-of-phase or in-phase pickup in a secondary coil via a lock-in amplifier coupled to an x-y recorder or minicomputer. The samples, grown from highly oriented pyrolitic graphite (HOPG) were about 1 $cm^2 \times 1$–2 mm thick, sealed in Pyrex with an excess of Br_2 and held in place with glass wool. Data were taken with the Pyrex ampoule immersed in liquid He, at $T = 1.6$–4.2 K. We concentrate here on the field range below 7 T, where only the lower frequency oscillations have significant amplitude. To date, we have studied two samples, which both display the giant

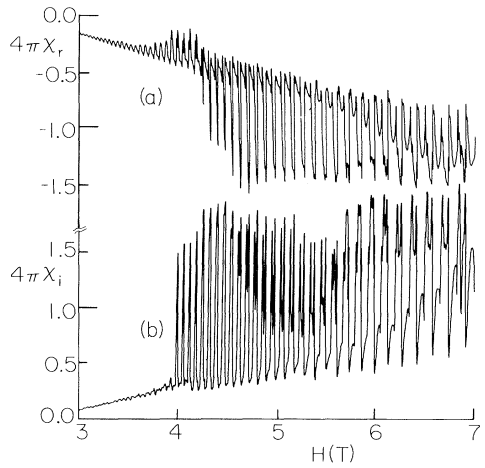

FIG. 1. Magneto-oscillations of stage-2 Br_2 graphite: (a) quadrature and (b) in-phase signals corresponding to real and imaginary parts of susceptibility, respectively, at $f = 933$ Hz, $T = 4.2$ K. (Empty-coil background not subtracted; absolute *sign* of oscillations not determined.)

MI effect, but with some significant differences. Sample 1 shows practically no signs of hysteresis, whereas in sample 2 there is a strong hysteresis—very different structure is observed depending on whether the field is swept up or down. Since GIC often show large sample-to-sample variability (one compound may have several in-plane superlattices), and since hysteresis can be caused by relatively subtle effects of pinning centers, such differences are not surprising. MI is a first-order transition, so that hysteresis effects are certainly possible, although they have not been observed in 3D systems.[3] In this paper we discuss mainly sample 1.

The anomalies we report are only observed when the sample is subjected to high-field "annealing"—the sample must either be cooled in a field or, if cooled to 4.2 K in zero field, must be held at $h \geq 10$ T for periods ~ 1 h for the very sharp steps to appear. We believe that this is related to the overcoming of pinning by defects—in 3D, the MI oscillations show a superlinear increase of amplitude with ac field, which has also been attributed to depinning effects.[3,5] At low frequencies, we also observe similar pinning effects—the oscillations appear nearly sinusoidal at low modulation amplitude, but sharpen up as the amplitude is increased. In the range of ~ 300–1000 Hz, the steps are sharp and nearly independent of modulation amplitude. The sharp onset of the large oscillations is very sensitive to the anneal, and can be varied from about 2–10 T.

If the sample is not annealed, normal, nearly sinusoidal dHvA oscillations are observed at low frequencies. Above ~ 1 kHz, the phase of the oscillations (relative to the ac field) begins to shift, and SdH oscillations occur in quadrature with the dHvA signal. This frequency dependence is consistent with estimates of sample skin depth. It is interesting to note that the SdH and dHvA oscillations are also 90° out of phase with respect to one another as a function of dc field. Such behavior is expected for a 2D system. In the annealed samples there is a large phase change on entering of the domain state. This shows up as a nearly discontinuous jump in the in-phase signal (Fig. 1). The phase change occurs because the domain-wall motion cannot perfectly follow the changes in applied field.[5] The domain walls acquire an inertia due to impurity pinning, and their motion is further damped by eddy currents. The net result is that the magnetization becomes a complex number. As expected for large MI, the susceptibility $\chi > \frac{1}{4}\pi$.

The steps of the magnetization on entering the domain state are extremely abrupt. Figure 2 shows that the width of the step is entirely determined by the modulation envelope. The central section of Fig. 2 shows the oscilloscope trace of the pickup voltage in the secondary coil. The notch-shaped distortion of the

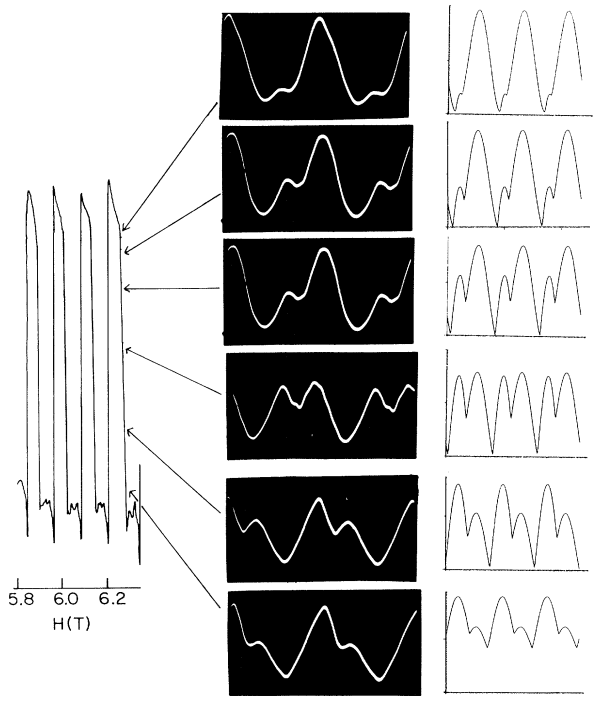

FIG. 2. Structure of a single step. Left-hand side, expanded view of in-phase oscillations in range 5–6 T. Center, oscilloscope traces of pickup-coil voltage (for sinusoidal input voltage) as step is traversed. Right-hand side, model of oscilloscope scan. Signal is composed of two parts: response of coil to sample magnetization which has a slope discontinuity (V shape) plus featureless background signal 90° out of phase.

sine wave is due to discontinuity in slope as the modulation field sweeps through the V of the sawtooth M. The right-hand portion of Fig. 2 shows that the form of the distortion is exactly as expected from this simple picture. (There is a small, in-phase component in the pickup voltage, due to imperfect cancellation of the direct coupling between drive and pickup coils.)

Figure 3(a) shows that, as the modulation amplitude is reduced, the steps sharpen up and additional fine structure is revealed. This effect is not always observed, and its origin is unknown at present, but is reminiscent of the magnetic excitons observed in Bi.[6] Figure 3(c) shows the measured width of the abrupt rise between two steps at 5.6 T. Most of the width is due to the modulation field, and the residual width is less than 5 G (!), which is comparable to the field homogeneity. The sharpness of the structure in field is quite unexpected—any inhomogeneity in the carrier density would wash it out—and suggests that the phase transition imposes a macroscopic coherency in the sample.

At the transition into the domain state, a very complicated structure can often be observed on the oscillo-

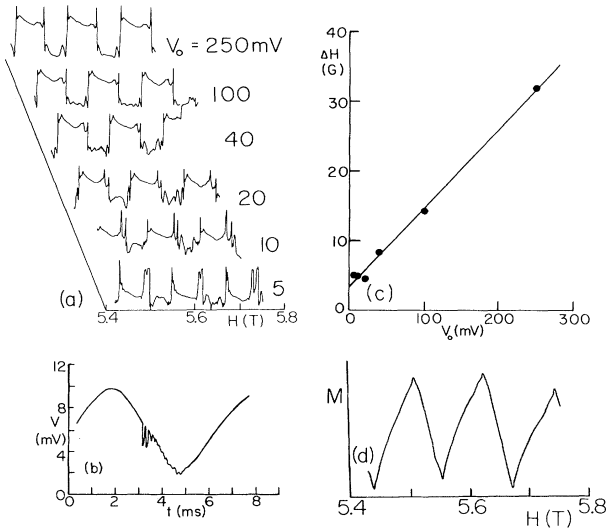

FIG. 3. Fine structure of steps. (a) Sharpening of steps and appearance of fine structure as modulation amplitude (drive coil input $= V_0 \sin\omega t$) is reduced. (b) Oscilloscope trace of pickup signal near a high-field step. (c) Modulation amplitude dependence of width of a single step. (d) Integral of first trace of (a), showing $M(H)$.

scope trace, as shown in Fig. 3(b). The exact shape depends on field, frequency, and modulation amplitude, but it very often appears to be a "ringing." We believe that we may be exciting a normal mode of the domain wall vibration.

The dHvA effect in 2D has previously been studied in Si inversion layers[7] and GaAs quantum well structures[8] where the carrier density is too low to show significant MI. Acceptor GIC's have higher carrier densities and often show dHvA overtones characteristic of MI, but never as strongly as in the Br_2-GIC. These samples display almost ideal MI, with no evidence of impurity or inhomogeneity broadening. Using the GIC band parameters, we estimate that strong MI effects should be observed for fields below 7 T ($4\pi\, dM/dH = 1$ at about 4.5 T).[9] The pulselike structure in dM/dH [Figs. 1 and 3(a)] corresponds to the V-shape $M(H)$ curves [Fig. 3(d)] expected for strong MI.[2,3] The domain state bears a strong resemblance to the intermediate state of a type-I superconductor: The spikes in dM/dH on entering and leaving the domain state [Fig. 3(a)] are observed in the intermediate state as discontinuities in M, due to the interface surface energy.[10] Other similarities include hysteresis, nonlinear I-V curves, and the use of ac fields to overcome pinning.

Vagner, Maniv, and Ehrenfreund[11] recently pointed out that MI should be strikingly different in a 2D system. In particular, they stress that the *domains pin the Fermi level between Landau levels* —a condition necessary for the observation of the quantum Hall effect[12]. Since, within each domain, the Landau levels are either empty or completely filled, the longitudinal resistivity vanishes. Hence the net sample resistance in the domain phase comes only from domain boundary scattering, which should be weak. Our present experiments afford no direct evidence for such an effect. In particular, the fact that the magnetization becomes complex in the domain phase makes it extremely difficult to separate out changes in the magnetoresistance.

However, there are a number of novel and unexplained features of our data which require a more detailed theoretical understanding of this unusual phase in two dimensions. In particular, why is the transition so sharp? Why does it become weaker at very low frequencies? What is the origin of the fine structure observed in Fig. 3(a)? What is the domain-wall resonance responsible for the ringing seen in Fig. 3(b)?

Finally, we briefly address the problem as to why giant MI occurs in Br_2-GIC, and has not been so far observed in other GIC. We believe that this is chiefly due to the size of the Fermi surfaces involved. Strong MI occurs when the magnetization M becomes comparable to the separation ΔB between oscillation maxima. If too few holes are involved (small Fermi surface), M can never be as large as ΔB. On the other hand, when the Fermi surfaces are too large, the oscillations are broadened by collisions, again leading to small values of M. The optimum Fermi surface size appears to be in the range of $\sim$ 100–300 T. While some mixing frequencies are present in AsF_5-GIC (indicative of weak MI), the principal frequencies fall well outside of this range. In HNO_3 and H_2SO_4 GIC, frequencies of $\sim$ 100 T are observed and indeed show strong harmonic content. However, these frequencies are associated with superlattice effects, and are destroyed by magnetic breakdown[13] before domain formation can be observed. A secondary factor may be that Br_2 GIC have a structural phase transition which, although it occurs near 200 K, is very sluggish and is incomplete at liquid He temperatures. It may be that this ordered phase is in some way susceptible to field annealing.

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