High-field magnetoconductance in Anderson insulators

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We report on high-field magnetoconductance measurements made on indium-oxide films as a function of temperature and static disorder. Special emphasis is given to the strong-localization regime where the magnetoconductance reveals a negative contribution associated with a spin-alignment mechanism in addition to the positive contribution associated with orbital, quantum-coherence effects. While the overall features of the theoretically expected effects are observed in our experiments, they depart in certain ways from the detailed predictions. We discuss the merits and shortcomings of current models to describe them, in particular, as they apply to the regime where the localized wave functions become larger than the Bohr radius. The main results of this paper are both quantum interference and spin effects contribute to the magnetoconductance throughout the entire range studied. In the limit of very strong disorder, the quantum interference effects are faithfully described by the Nguyen *et al.* model. The spin effects, on the other hand, show only qualitative agreement with current models which are unable to account for the saturation field being insensitive to changes in disorder. [S0163-1829(96)00344-X]

I. INTRODUCTION

The low-temperature transport properties of dirty metals may be appreciably modified by quantum-coherence and interaction effects. These issues have been intensively investigated for the past 15 years and are, by now, fairly well understood.[1] Quantum-mechanical coherence effects mandate corrections to the Boltzmann theory in this regime. These corrections, usually referred to as weak-localization effects, are believed to be a precursor to the disorder-driven, ''true'' localization in which spatial disorder causes the electronic wave functions to be confined to a microscopic region in space of characteristic size ξ, the localization length. ξ is a function of disorder and diverges at the transition. When ξ becomes the smallest length in the problem, the system is strongly localized and electronic transport is only possible by inelastic hopping. This regime is not as well understood as the diffusive regime despite substantial theoretical and experimental efforts extended over several decades. Nevertheless, in many uniformly disordered electronic systems it is established that transport is governed by variable-range hopping and there is usually a significant magnetoconductance (MC) which can be of either sign depending on the system and measurements conditions. The MC in the insulating regime may originate from suppression of backscattering[2,3] (in analogy with the known mechanism in the diffusive regime). This is apparently the dominant mechanism near the metal-insulator transition where ξ is large.[2,4] In the limit of strong disorder, however, compelling arguments suggest that another MC mechanism, proposed by Nguyen, Spivak, and Shklovskii (NSS),[5] takes over. The NSS mechanism leads to a positive MC even in the presence of spin-orbit scattering. This regime has been studied only in few cases due to the difficulties in measuring highly disordered systems at low temperatures. For either mechanism, the most convincing case for the MC being due to an orbital effect comes from the anisotropy[4,6] of the phenomenon when the system is 2D (or 1D).

Recently, Frydman and Ovadyahu reported on a negative MC component whose origin could be traced to spin mechanism.[7] Such an effect, appropriate for the strong disorder regime, was proposed by Kurobe and Kamimura (KK).[8] The basic assumption of this model is that, for strongly disordered systems, there is a finite density of states near the Fermi level that may be singly occupied, doubly occupied, or unoccupied. The effect is sensitive to the existence of intrasite Coulomb interactions. Applying a magnetic field tends to align the electronic spins which makes the spin conserving transition probabilities between two singly occupied states less favorable (and, at high enough fields forbidden), due to the Pauli exclusion principle. This, in turn, leads to a *negative* and *isotropic* MC which saturates above a characteristic field. Another effect, proposed long ago, namely a field-induced shrinkage of the wave function,[9] is ineffective in our system where the Bohr radius ($\sim$10 Å) is much smaller than the magnetic length associated with the highest attainable field.[4] Also, the feature that clearly distinguishes the observed negative MC from wave-function shrinkage is the tendency to saturate at high fields. This feature seems to be quite common. It has been seen, for example, in MC studies on insulating samples of $Y_{1-x}Pr_xBa_2Cu_3O_7$,[10] CdSe:In,[11] CdSe:Cr,[12] GaAs,[13] Mo_xGe_{1-x},[14] carbon nanotubes,[15] and ReSi (Ref. 16), but its origin was ascribed to spin effects only in few of these cases.

In a thin film [i.e., two-dimensional (2D) geometry] it is possible to separate, at least approximately, the spin effects from the orbital effects.[7] Spin alignment can be reasonably assumed to be independent on whether the field is perpen-

dicular or parallel to the film plane, and is thus *isotropic* with respect to the field orientation. In contrast, an orbital effect (such as the NSS) depends strongly on the orientation of the field with respect to the film. To be fully effective, NSS requires a flux through an area of width $(\xi r)^{1/2}$ and length r (r is the hopping length). If the thickness of the film is less than $(\xi r)^{1/2}$, such an area is clearly not available in a field parallel to the film, leading to a prominent anisotropy in the MC. This behavior is indeed observed here and in previous experiments.[3,4,6]

It must be stressed that both the KK and the NSS theories ignore *intersite* Coulomb interactions (the KK model considers only *intrasite* interactions). Also, they strictly apply for the limiting case of very strong localization, where ξ is less than the separation between sites. Much of the published studies in this field however, do not satisfy this condition. Covering a wide range of disorder in the present study was motivated by the need to elucidate this often ignored point. We thus should look for a quantitative agreement with KK and NSS in the regime of very large disorder, and hopefully learn experimentally if and how the effects change when the localized functions spread over more than one site. If the effects do correspond to theory in the limit of strong disorder, one can presumably argue that intersite Coulomb interactions in our system are weak. This turns out to be the case for the NSS mechanism but not for KK. The other caveat, namely, problems associated with ξ being larger than the Bohr radius (the intermediate regime of disorder), entails significant and systematic discrepancies between theory and experiment. We shall argue (in Sec. III) that this problem is more general than that involved in the MC. Rather, it reflects on the applicability of the simple percolation picture which underlies most transport theories in the hopping regime, including interpretations of the temperature dependence of the conductivity. In the next section, we give the details of the samples used in this study and the measurements techniques. Section III includes the experimental results, their analysis, and discussion of the theoretical implications.

II. EXPERIMENT

The full details of preparation and characterization of the crystalline In_2O_{3-x} films used in this study was described elsewhere.[17] Two types of samples were used: thin films of thickness $d=50-110$ Å, which are effectively 2D at the temperatures of the measurements, and much thicker films ($d>1200$ Å) which are 3D. (The effective dimensionality depends on the interplay between d and the hopping length, r.) The advantage of using these films is that their spatial disorder, in a given sample, could be varied and fine tuned as has been demonstrated in previous studies.[3,18] This process allowed us, in the 2D samples, to scan a wide regime of disorder, typically covering resistance per square of $R_\square \approx 4$ kΩ to $R_\square \approx 10$ MΩ at $T=4$ K, a range which straddles the transition between weak and strong localization transition (i.e., $R_\square \approx 30$ kΩ) using the *same* film and without breaking the physical continuity of the structure. Our films are thus free of the complications inherent in studies of metallic systems that usually must be made granular to be in the insulating regime. Furthermore, all the relevant system parameters are known from previous studies.[19]

The MC measurements were made in a liquid-helium rig employing a probe that could rotate the sample plane relative to the field direction (fields up to 9 T were employed). This enabled us to compare between parallel and perpendicular MC measurements of the same sample. For the 2D case, this technique helps to separate the isotropic MC associated with spin effects, which are the focus of the present study, from quantum-interference effects.[7] Conductivity and MC studies were usually made by a standard four-probe dc technique employing the Keithley K617 electrometer and the K220 current source. For the most resistive samples, however, we used a two-probe conductance technique biasing the sample with a small voltage source. The latter was always small enough to be in the Ohmic regime. In a few cases this point was further checked by resorting to an ac technique employing an ITHACO 1211 current preamplifier and PAR 124A lock-in amplifier tuned to the measuring frequency (typically, 0.5–8 Hz).

Six different 2D samples were measured during this study with lateral dimensions of 1×1 mm^2 to 6×6 mm^2 and thicknesses ranging from 50 to 110 Å with essentially the same results which attests to the large scale homogeneity of the material. In the following, we focus on the detailed results obtained on the 110 Å sample for which an extensive range of spatial disorder was covered.

III. RESULTS AND DISCUSSION

The overall results of this paper are given in Fig. 1 where the MC of the 2D samples with different degrees of disorder can be compared for both perpendicular and parallel field orientations. In small fields (typically, $H<1$ T), the MC is positive and anisotropic, indicative of an orbital mechanism. These quantum interference effects were extensively investigated previously,[6] mainly at low magnetic fields. The new aspect revealed in this study is the saturation field and its dependence on disorder, as will be discussed below. For now we wish to focus on the high-field regime ($H>4$ T) where the MC acquires a negative component whose magnitude is a sensitive function of the film disorder. For films with $R_\square<20$ kΩ this component is apparently small. In this regime of disorder the well-known weak-localization positive MC dominates in the entire range of fields, and shows typical $\ln(H)$ dependence above $H\approx1$ T. A systematic analysis, which includes also a comparison of the parallel and perpendicular MC data, reveals some deviations from this asymptotic dependence which corresponds to a negative $\Delta G/G$ component of about 2–4% for $R_\square \approx 10$ kΩ at $H=9$ T. This component, which is probably due to the Aronov-Altshuler[20] mechanism, is still smaller than the positive MC. Once $R_\square>30$ kΩ, however, the negative MC component quickly establishes itself and begins to dominate the high-field data.

In a previous study[7] employing deeply insulating samples, the origin of a negative MC similar to the one discussed here has been attributed to the spin-alignment mechanism proposed by KK.[8] The characteristic tendency of the negative MC towards saturation is evident in the data (Figs. 1, 2, and 5). It stands out more clearly at lower temperatures (Fig. 2) as could be expected of a spin-alignment mechanism.

The present experiments demonstrate that these effects persist all the way up to the metal-insulator transition. The

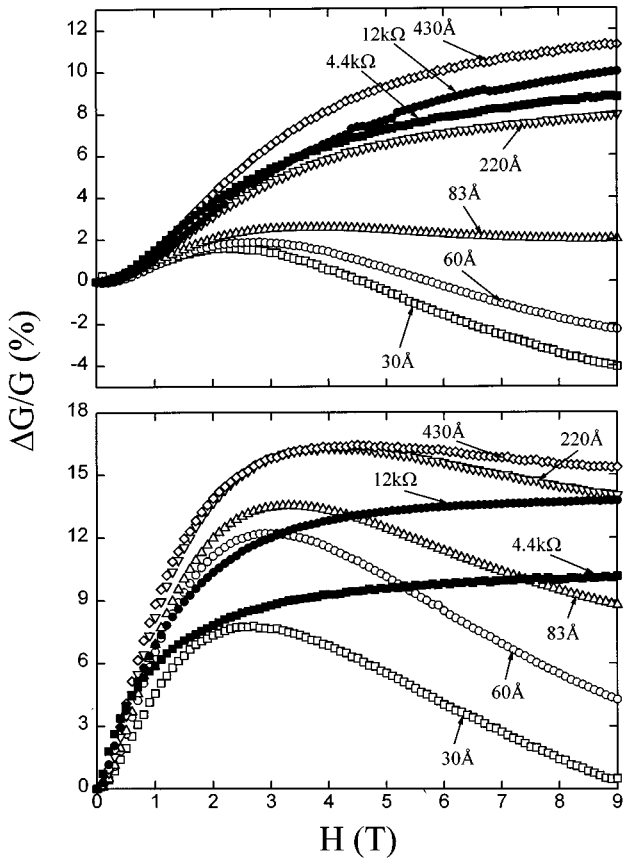

FIG. 1. Perpendicular and parallel MC (lower and upper plots respectively) for 2D In_2O_{3-x} films. $T=2.17$ K. The samples have a thickness, $d=110$ Å, but different degrees of disorder created by the method described in Ref. 18. The samples are labeled by either their localization length, ξ or, for the weakly localized samples (full symbols), by their $R_\square$ at $T=2.17$ K. See Figs. 3 and 4 for the detailed temperature dependence of these samples.

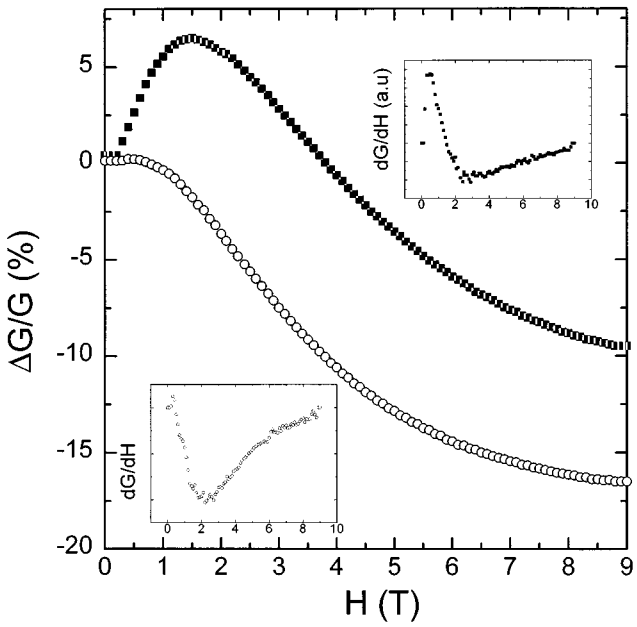

FIG. 2. The MC of a sample with $\xi=30$ Å ($R_\square=0.5$ MΩ at $T=4.1$ K), for a parallel field (empty symbols) and perpendicular field (full symbols), respectively. $T=1.27$ K. Note the pronounced tendency towards saturation for $H>3$ T and the fact that the MC curves for the parallel and perpendicular orientations are of similar shape at this field regime. The latter is further illustrated in the derivative plots of the respective curves showing that the inflection point of $G(H)$ occurs for both orientations at a similar value of H.

$R(T)$ data for the films in the vicinity of this transition are given in Fig. 3. Comparing the data in this figure with the MC in Fig. 1 one notes that the appearance of a positive MC component coincides, within the resolution of the experiment, with the onset of significant deviations from a $\ln(T)$ dependence of the films resistance. We recall that for weak-localization $R(T)$ in 2D is given by $R(T)=R_0+R_\square f(X)\ln(T)$, where $f(X)$ stands for the combined localization and interaction corrections to the resistance in this regime (X stands for the various material and disorder-dependent parameters inherent in these theories).[1] Deviation from this logarithmic dependence is a characteristic hallmark of the transition to strong localization.[21] The $R(T)$ of samples with $R_\square>30$ kΩ can be interpreted with the Mott variable range hopping law, which in 2D is

$$R(T)=R_0\exp\left(\frac{T_0}{T}\right)^{1/3}, \quad \text{with} \quad K_BT_0=\frac{3}{N(0)\xi^2 d}, \quad (1)$$

where $N(0)$ is the density of states [$N(0)$ in In_2O_{3-x} is (Ref. 19) 2×10^{32} erg^{-1} cm^{-3}]. The various ξ's assigned to the samples in Fig. 1 were estimated[22] on the basis of their $R(T)$ data (Fig. 4) through Eq. (1). One question often raised in connection with such a procedure is whether a fit of the data to the exponent $\frac{1}{3}$ is compelling. In our case it is not, *per se*, but the value of ξ, extracted from fits to any other power, e.g., $\frac{1}{4}$, yields hopping lengths that are larger than the film thickness and are thus inconsistent with a 3D hopping law. The connection between the appearance of a negative MC component and the transport mechanism is also observed in the data for the 3D samples illustrated in Fig. 5. Again, for weak disorder, the MC behaves in accordance with the weak-localization theory, scaling like $H^{1/2}$ at high fields while for the bulk resistivity, ρ just bigger than a critical value (the critical resistivity, ρ_C of 3D In_2O_{3-x} is (Ref. 19) 2.2×10^{-2} Ω cm), a clear negative MC component takes over at higher fields. Note that in this 3D case the *resistance* of the film where the negative MC appears is only 4 kΩ, almost an order of magnitude *smaller* than in the 2D series. This corresponds to the system being *insulating* (for which the condition in 3D is $\rho>\rho_C$, while in 2D it is $R_\square\approx30$ kΩ).[21]

We attempted to analyze the MC data as a function of disorder for several 2D films measured in the temperature range 1–4 K and field range 0–9 T, in a similar vein as was done in Ref. 7. The procedure used in this analysis (described in the Appendix), yields two characteristic fields, H_{NSS} and H_S. These are the fields where the quantum-interference and the spin-alignment mechanisms are expected to saturate, respectively. H_{NSS} obeys[5,7]

$$H_{NSS}=\frac{\Phi_0}{\xi^2}\left(\frac{T}{T_0}\right)^{1/2}, \quad (2)$$

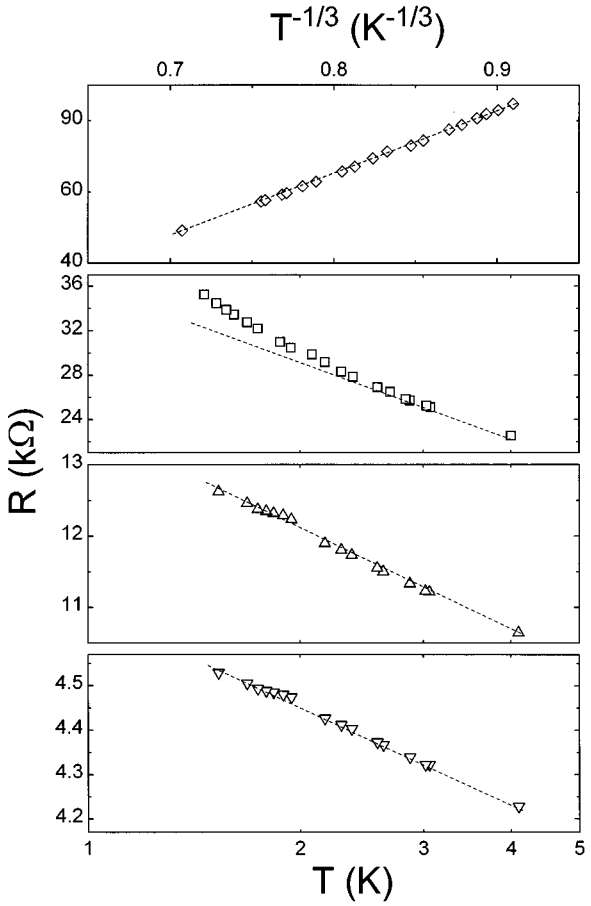

FIG. 3. Resistance versus temperature of four of the samples that are nearest to the weak-localization to strong-localization transition. Note the correlation between the appearance of a negative MC (cf. Fig. 1) and the crossover to hopping behavior. The two bottom samples are in the weakly localized regime. The third (the sample with ξ=430 Å in Fig. 1), already shows deviations from the ln(T) behavior and, at the same time, exhibits a significant negative MC component (see bottom plot in Fig. 1). The top curve (for a slightly more disordered sample in this series), is plotted differently to illustrate the 2D variable-range-hopping law that characterized samples with $R_{\square}>30$ kΩ.

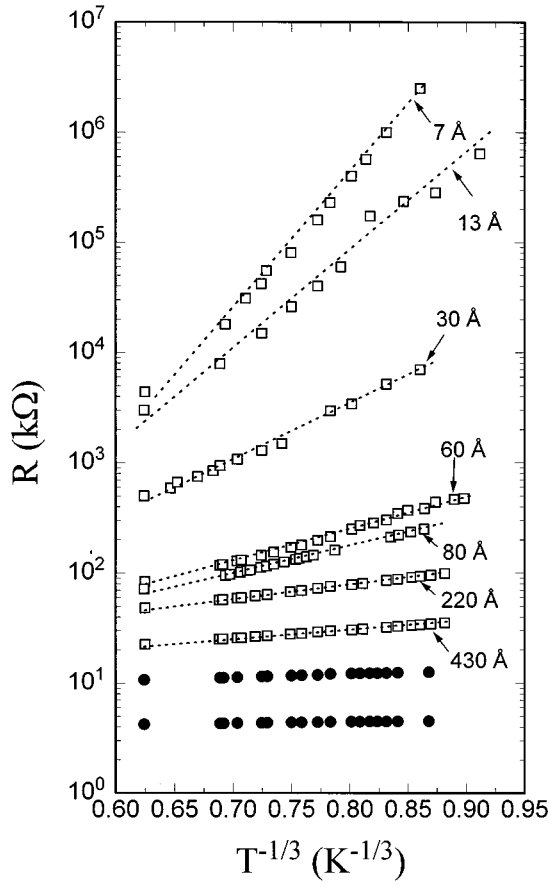

FIG. 4. Resistance versus temperature for the In_2O_{3-x} films used in this study. The plots for all samples with $R_{\square}>30$ kΩ are consistent with the 2D nature of the hopping process as given by Eq. (1) and are labeled by the values of ξ extracted from it. For comparison, the two weakly localized samples are also depicted (full symbols).

where Φ_0 is the flux quantum.

Assuming the applicability of the KK theory, H_S is given by the following expression:[7,8]

$$g\mu_B H_S = ak_B T\left(\frac{T_0}{T}\right)^{1/3}, \tag{3}$$

where μ_B is the Bohr magneton, a is a constant of the order of unity, and T_0 is defined in Eq. (1). These expressions imply a specific dependence on both temperature and static disorder (the latter enters through T_0, and thus ξ). The temperature dependence due to these mechanisms has been studied before[7] and, on the whole, reasonably good agreement was found between theory and experiment. As we shall show now, this is not the case for the dependence on static disorder. The values of the two saturation fields as a function of film disorder, as obtained from our analysis, are plotted in Fig. 6.

The data in Fig. 6 expose serious discrepancies between the dependence on disorder expected by the theoretical models and the experimental results. Considering first the saturation field for quantum interference, $H_{\rm NSS}$, one notes that it does increase systematically and monotonically with disorder. However, for most of the range, the dependence is considerably weaker than the $1/\xi$ law inferred from combining Eqs. (1) and (2). This finding reenforces the conclusion[4] that the NSS mechanism, in its simple form, works only in the limit $\xi\rightarrow\xi_0$ (ξ_0 is the Bohr radius). For the disorder range $\xi\leqslant$20 Å the data for $H_{\rm NSS}$ are indeed consistent with the theoretically expected ξ dependence (cf. Fig. 6). That this is the disorder regime where the NSS does work is in very good agreement with the study of the effect of spin-orbit scattering on the MC by Shapir and Ovadyahu.[23]

The KK model, on the other hand, fails to describe the ξ dependence of the data in terms of H_S even in the strong disorder regime. The saturation field in this case, H_S, seems to be essentially disorder *independent*, and thus at variance with the $\xi^{-2/3}$ law expected by Eqs. (1) and (3). We have considered the possibility that this discrepancy is an artifact of the specific analysis used here. However, another estimate of H_S, independent of that described in the Appendix is readily obtainable from the *raw data* and it shows the same

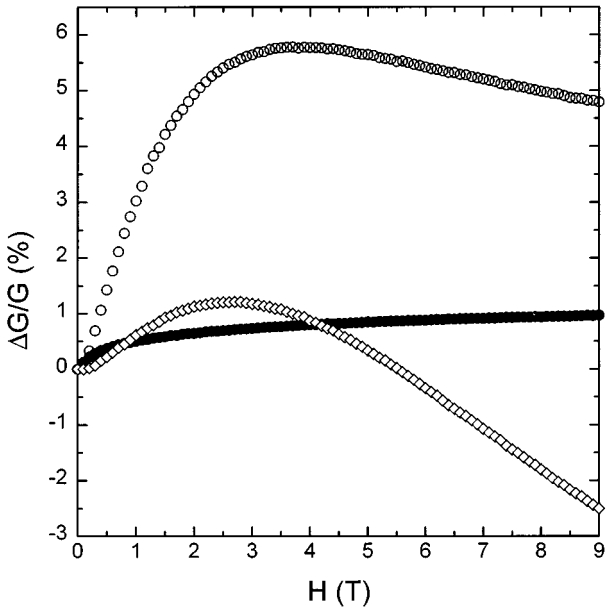

FIG. 5. The MC of 3D In_2O_{3-x} samples for A weakly localized sample (full circles) with room-temperature resistivity, $\rho=1.3\times10^{-3}$ Ω cm ($T=1.5$ K), just-insulating sample (empty circles) with $\rho=3\times10^{-2}$ Ω cm ($T=1.5$ K), and strongly localized sample (empty diamonds) with $\rho=2.2\times10^{-1}$ Ω cm ($T=2.17$ K).

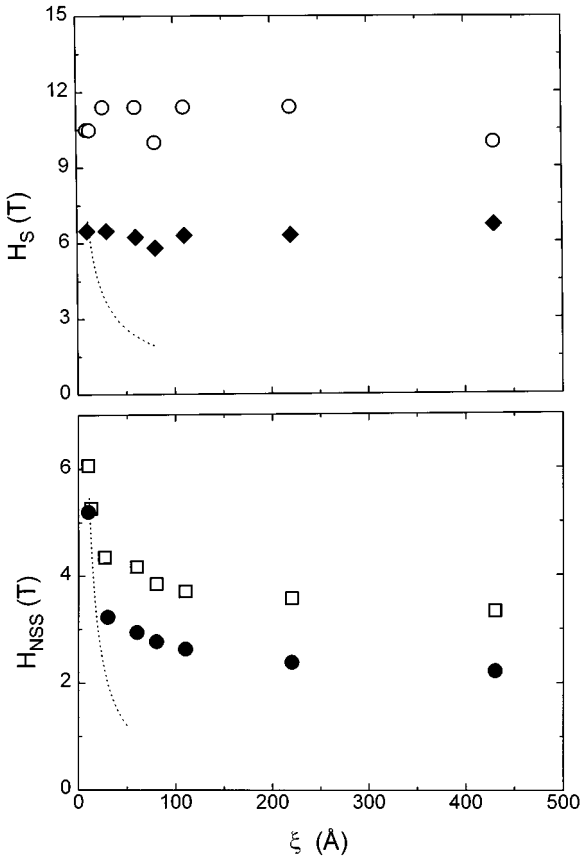

FIG. 6. The dependence of the saturation fields H_S and $H_{\rm NSS}$ on the localization length ξ. Data are taken from analysis of the MC at $T=4.11$ K (empty symbols) and at $T=2.17$ K (full symbols). The dotted curves are the theoretical dependencies calculated according to Eqs. (2) and (3) [with T_0 given by Eq. (1)] for the NSS and KK models, respectively (for $T=2.17$ K).

lack of sensitivity to ξ as that derived by the above analysis: This independent estimate of H_S is based on locating the inflection point[24] of $G(H)$ versus H as illustrated in Fig. 2. An approximate percolation treatment, using the KK model, shows that this inflection point should essentially scale with H_S. The fact that the two different schemes yield the same conclusion for the behavior of H_S presumably suggests that the problem is not due to the specific analysis used.

For the sake of further discussion, the values for H_S of all the samples in this series are exhibited in Fig. 7. Note that these values scale as $\mu_B H\propto k_B T$ which, incidentally, lends further support to our contention that this MC involves spin alignment.

The failure of both models to account for the ξ dependence of the MC over most of the range of ξ studied should not be too surprising. In the first place, the localization radius is an average quantity, and does not apply to individual localized wave functions. It is well known that percolative transport in disordered systems does not select *average* values of Miller-Abrahams (MA) (Ref. 25) resistances but rather certain optimal values. While all appearances indicate that, nevertheless, a percolation theory based on the implicit assumption that all localized wave functions are characterized by ξ seems to work as far as dc conductivity is concerned, it is not clear that such a theory will also work for more subtle effects such as the magnetoconductance.[3] As mentioned above, NSS was shown[23] to be inadequate when $\xi\gg\xi_0$ even when the magnetic length, $l_H(c\hbar/eH)^{1/2}$ is larger than ξ. This is due to the neglect of backscattering suppression on the scale of ξ which should be quite prominent when $l_H<\xi$ and $\xi\gg\xi_0$.[3,4,23] Regarding KK it should be borne in mind that, as derived, it applies strictly in the limit of very strong localization, where the localized wave function coincides with the microscopic length (i.e., ''atomic'' function ξ_0). Once localization extends over more than a single microscopic function, the Pauli principle no longer prohibits the occupation of a localized state by two or more parallel spins. KK's total prohibition of singly occupied to singly occupied (s→s) transitions at high fields is thus no longer valid. In a similar vein, the occupation of a localized state near the Fermi level is not restricted to 0, 1, and 2 when ξ extends over more than a single microscopic function. The different possible types of transitions near the Fermi level are then not properly enumerated by KK. Their solution of the percolation problem thus becomes inadequate as the number of possible types of transitions grows rapidly as ξ increases beyond ξ_0.

These reservations are certainly relevant for the $\xi\gg\xi_0$ regime. Figure 7 suggests, however, that the KK model fails not only for $\xi\gg\xi_0$, but also when $\xi\rightarrow\xi_0$, where the NSS *does* work. So, either the KK is more vulnerable to deviations from the strict $\xi\sim\xi_0$ condition than the NSS or, the spin-alignment mechanism that we observe is due to a different mechanism. In the following we consider, as an example, a simple scenario that combines these alternatives.

As remarked above, as soon as ξ extends even over only *two* sites, the s→s channel is merely impeded, not totally eliminated by strong fields. Notice first, that this sudden departure expected form the KK model is much more dramatic

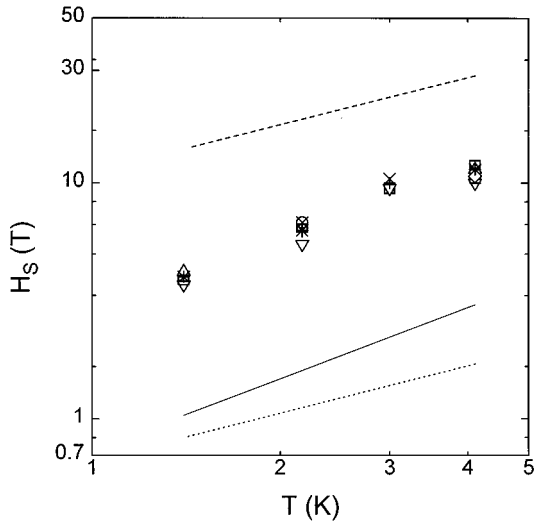

FIG. 7. The dependence of the spin-alignment saturation field, H_S on temperature. Data are included (and almost overlap) for samples with ξ=7, 13, 27, 30, 60, 80, 110, 220, and 430 Å, at four different temperatures. The full line depicts the law $\mu_B H = k_B T$. The dotted and dashed lines are plotted according to the KK model [Eq. (3)] and encompass the range over which the data ought to have spread for the range of ξ=7 to 430 Å.

than is expected for NSS. For the latter, only some detail in the scattering is involved at this point because the field for which $l_H < \xi$ will be attained is inaccessibly large (to fulfill the condition $l_H < \xi$ for $\xi \leqslant 30$ Å one needs $H \geqslant 100$ T!). The KK type MC for $\xi > \xi_0$ is difficult to handle theoretically, but to illustrate how similar physics can affect H_S we present a simple percolation model with negative magnetoconductance which saturates at a field $H_S \propto k_B T$, *independent* of disorder. The model is based on antiferromagnetic interaction between spins on neighboring localized states. The simplest version of the model includes only singly occupied and unoccupied sites. Consider an electron hopping from site a to an unoccupied site b which is in the vicinity of another occupied site c. In the presence of a magnetic field, the electrons in a and in c are aligned by the field. The (spin conserving) hop from a to b is impeded by the antiferromagnetic interaction between b and c, which favors an antiparallel spin in b. A jump from a to b requires an activation by the smaller of the energies corresponding to the antiferromagnetic interaction (i.e., singlet-triplet splitting) between b and c, and $g\mu_B H$, the energy needed to reverse one of the spins in a or c. In the absence of a field, the spins in a and c are just as likely to be antiparallel as parallel, resulting in a 50% chance that the hop can be easily accomplished. Notice that the situation differs from that of KK in that the field here only reduces the MA conductance between a and b but does not eliminate it. In this situation, when $g\mu_B H$ is the important energy, *every* MA conductance will be altered to saturation at $g\mu_B H > k_B T$, so no further change in the percolation path will occur at such a field. It is possible that such a model is actually responsible for the observed negative MC.

In summary, we have presented and analyzed high-field MC data with an emphasis on the behavior as a function of static disorder. In the limit of strong disorder (and *only* in this limit), we confirm that the NSS mechanism is associated with a saturation field controlled by the hopping length as suggested by theory.[5] Another MC mechanism, by all indications due to a spin-alignment mechanism, has the opposite sign and it becomes prominent once the system just crosses from the weakly localized regime into the insulating phase. The latter feature may be taken as indicative of the importance of inelastic processes (in contrast with the orbital effects which is quantum coherent by its nature). The detailed processes underlying the spin mechanism are not yet fully elucidated as current theories (KK) fail to faithfully describe the dependence of the saturation field on disorder. The problem of many-body spin effects and quantum interference have been recently considered by Zhao *et al.*[26] but without an explicit treatment of the $\xi > \xi_0$ regime. We have offered a simple variant of the KK model that may lead to an effect with the observed insensitivity to disorder as experimentally observed. Further studies, both experimental and theoretical are needed to test this possibility.

ACKNOWLEDGMENTS

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APPENDIX

The fitting procedure involved several steps. First, one subtracts from the raw MC data a trial function, $\Delta G(H)$, for the spin-alignment component. The *same* $\Delta(H)$ is used for both the parallel and perpendicular field data. This assumes that $\Delta G(H)$ is isotropic, and that the spin alignment and orbital mechanism are additive. The assumption of additivity is nontrivial as spin-flip events may destroy phase coherence and thus wipe out the quantum interference effects.[27] The trial function has to be such that the two sets of ''new'' data points (one for each field orientation), resulting from subtracting it from the raw data, can be made to overlap by multiplying the H scale for the parallel MC by a *single*[28] number, β^{-1}. (β is the ''anisotropy parameter'' and should be a monotonously increasing function of r/d.[6]) This step of the analysis yields β^{-1} and the saturation field, H_S. In the second step, the ''fictitious'' MC generated by subtracting $\Delta G(H)$ from the raw data is fitted to the numerical simulation of the NSS model[29] from which the value of $H_{\rm NSS}$ is determined. As illustrated in Ref. 7, physically reasonable parameters can be obtained in this procedure. To test the sensitivity of this procedure to the particular choice of $\Delta G(H)$, several trial functions were tested. All of these $\Delta G(H)$ had the asymptotic behavior of $\Delta G \propto H^2$ for small H and saturation for large fields (e.g., $\Delta G \propto H^2/(H_S^2 + H^2)$, $\Delta G \propto \arctan^2[H/H_S]$ as well as the function used by Meir[30]). The particular choice of the function did not change the trend of H_S versus disorder reported above.

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