Effects of spin-orbit scattering on hopping magnetoconductivity

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The low-field magnetoresistance of Au-doped indium oxide films exhibits a pronounced positive component that persists into the insulating regime. Above a certain degree of disorder, however, only negative magnetoresistance is observed similar to that of the undoped material. For weakly localized samples, it can be shown that the observed magnetoresistance may be accounted for by backscattering in the presence of moderately strong spin-orbit scattering (presumably, off the Au impurities). It is argued that backscattering is still important for samples that are fairly deep in the variable-range-hopping regime, albeit in an indirect way. The apparent insensitivity of the magnetoresistance to spin-orbit scattering in the limit of strong disorder, on the other hand, indicates that neglecting "returning loops" may be justified at that limit.

INTRODUCTION

Quantum-interference effects in the transport properties of disordered conductors have been the subject of extensive investigations. It is now generally accepted that a dominant mechanism for the anomalous magnetoresistance (MR) observed in, e.g., thin metallic samples is an orbital effect associated with suppression of backscattering. The sign of this MR may be negative or positive depending on the strength of spin-orbit scattering.[1]

The occurrence of similar effects in the variable-range-hopping (VRH) regime is less well understood. An orbital, negative MR in the VRH regime has been reported by several researchers[2] and it was recently demonstrated[3] that the coherence length associated with this phenomenon is the hopping length, r, although other possibilities have been considered.[4]

In the present work we give results of MR measurements on thin indium oxide films covering a wide range of disorder. The presence of spin-orbit scattering is shown to be important up to a certain degree of disorder which includes strongly localized samples. Deeper into the insulating regime, however, the orbital MR becomes insensitive to spin-orbit scattering. The implications of these results to theoretical models of hopping magnetoconductivity are discussed.

EXPERIMENTAL TECHNIQUES AND RESULTS

Samples used in this study were 150-Å-thick films of In_2O_{3-x} doped with Au using the following procedure: Pure (99.997%) In_2O_3 was evaporated from an e-gun source onto room-temperature glass slides through suitable stainless-steel masks. This was followed by evaporation of 5 Å mass equivalent of Au from a Knudsen source. The films were then removed from the vacuum system and placed on a hot plate (200 °C) for approximately 1 h to affect crystallization and homogenization. Transmission electron microscopy performed on such films, showed tightly packed In_2O_3 polycrystals with grain sizes of 100–300 Å. The electron diffraction patterns consisted of the previously reported[5] set of rings corresponding to bcc In_2O_{3-x} with no trace of Au precipitation. Spectrophotometry revealed that the Au-doped samples are 10–20 % less transmissive in the visible then the pure In_2O_{3-x} films (a difference easily discernible by the eye). No extra specific absorption modes were detected down to 200 cm^{-1} but some of the In_2O_3 Fröhlich modes[6] in the 600–300-cm^{-1} range were slightly shifted to lower energies which may suggest an intimate contact of the Au atoms with the lattice. The room-temperature Hall effect indicated a carrier concentration of 10^{20} e/cm^3 that is quite close to the value usually found[6] in In_2O_{3-x} samples.

Five different batches of Au-doped In_2O_{3-x} were studied. Within each batch, several samples (in the form of 5×8 mm^2 strips) were measured, having $R_\square$ (samples are identified by their $R_\square$ at $T=4.11$ K) ranging from 1.5 kΩ to 100 MΩ. The different $R_\square$ values were generated by heat treatment as described before[6] in detail for undoped In_2O_{3-x} samples.

Resistance and MR data were taken by a standard four-probe dc technique employing a high-impedance Keithley current source (K220) and electrometer (K617) controlled by a personal computer. The MR data points represent computer-averaged results of two to ten bipolar readings, depending on the noise level. Measurements were made in a ^{4}He immersion cryostat mounted in the air gap of a split-coil electromagnet. The magnetic field was applied perpendicularly to the sample's plane except where otherwise noted. Temperature was measured by means of a calibrated Ge thermometer. In the following, we describe results for one particular batch for which all the diagnostic procedures described above were made simultaneously with the transport measurements.

MR data for several $R_\square$ values are shown in Figs. 1, 2,

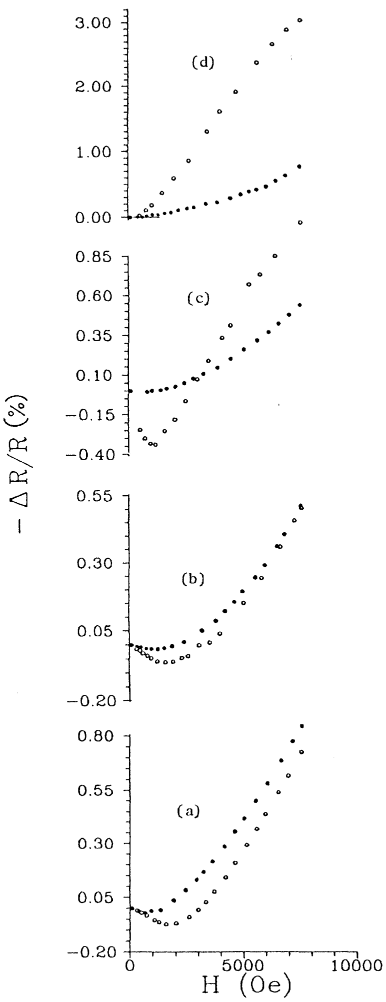

FIG. 1. MR curves for Au-doped samples for various $R_\square$ values (solid circles, $T \simeq 4.1$ K; empty circles, $T \simeq 1.4$ K). (a) A weakly localized sample, $R_\square = 4.5$ kΩ. The other data sets are for samples exhibiting VRH conductivity with the following parameters: (b) $R_\square = 45$ kΩ, $T_0 = 32$ K, $\xi = 200$ Å. (c) $R_\square = 300$ kΩ, $T_0 = 1100$ K, $\xi = 35$ Å. (d) $R_\square = 4.5$ MΩ, $T_0 = 11\,000$ K, $\xi = 12$ Å.

and 4 and some aspects of these are compared with data for undoped films in Fig. 3.

Roughly speaking, the results fall into two qualitatively different groups: Au-doped samples with $R_\square < 1$ MΩ exhibit a pronounced positive component reminiscent of that usually found in metal films in the presence of moderately strong spin-orbit scattering.[7] In fact, up to a certain field, H^*, the MR is *positive*. H^* is well defined experimentally in samples with $R_\square < 500$ kΩ (Fig. 2). This feature is not observed in undoped films,[3] where, independent of $R_\square$, the MR is negative. Au-doped samples with $R_\square > 2$ MΩ, on the other hand, show only *negative* MR within the range of measurements. Au-doped samples in this group could not be told apart from undoped films with comparable $R_\square$ (except for their optical

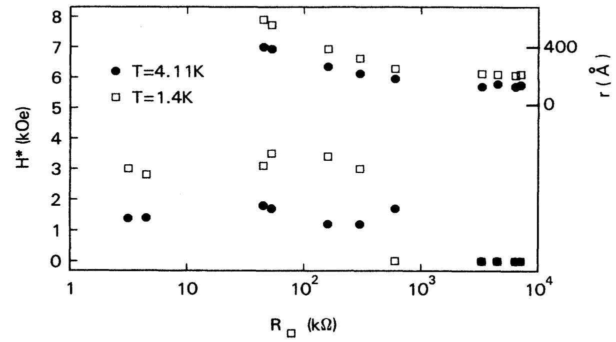

FIG. 2. The dependence of the zero-crossing field, H^* (lower set of data), and the hopping length, r (upper set), on the disorder. Note that most of the variation in r occurs *below* the "critical" $R_\square$. ($H^* = 0$ means that no positive MR could be detected.)

characteristics mentioned above). In particular, such films exhibited only negative MR which also showed field, temperature, and disorder dependences in close similarity with those of undoped films (cf. Figs. 1–3 and Ref. 3).

The "history" of measurements in this batch was as follows: The as-prepared film had $R_\square = 4.5$ kΩ and was measured first. Heat treatment was then used to generate the samples with $R_\square$ of 300, 4500, 650, 7100, 54, 3300, 3.5, and 6400 kΩ, respectively, measured in this order. Clearly, the salient features of the transition observed in Figs. 2 and 3 are independent of history. That, along with the diagnostic tests alluded to above, suggests that this transition is "disorder driven" rather than an artifact

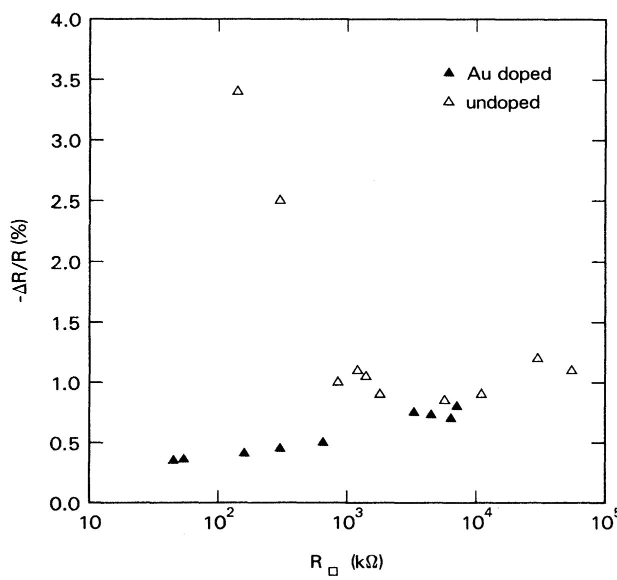

FIG. 3. MR for $H = 6.5$ kOe and $T \simeq 4$ K as a function of disorder for doped and undoped In_2O_{3-x} samples. Note the similarity above $R_\square > 1$ MΩ (and compare with Fig. 2).

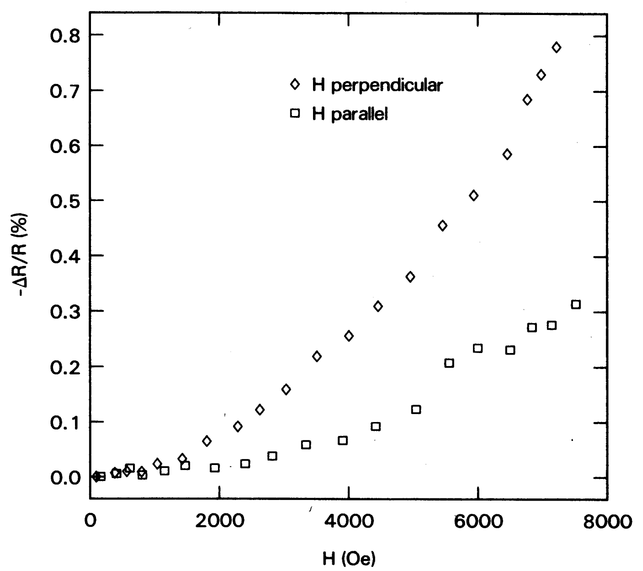

FIG. 4. MR (at $T=4.11$ K) for a doped sample with $R_\square=6.4$ MΩ illustrating the orbital origin of the effect (cf. Ref. 3).

due to, e.g., Au expulsion or segregation to grain boundaries which could hardly be expected to be reversible. To better understand the role played by the Au impurities, several additional batches with increasing amounts of Au were made and measured. For batches with Au doping exceeding 15 at. %, the above-mentioned reproducibility did not hold. Heat-treatment cycles resulted in Au precipitation and the optical transmission of such films increased upon rinsing in acetone. This apparently suggests a Au miscibility limit in In_2O_{3-x} of the order of 10%. Since In_2O_{3-x} has a 10% oxygen deficiency[5] it seems plausible to assume that most of the Au atoms in our studied films reside on oxygen voids. It was also found that the Au inclusion allows less latitude in the range of $R_\square$ realizable with heat treatment: The lowest $R_\square$ achievable within a given batch increased with the Au concentration. The choice of the particular ($\simeq 2\%$) Au concentration was largely dictated by the desire to be able to include weakly localized samples in the batch to serve as a useful reference as will become clear below.

DISCUSSION

Two questions naturally arise: What is the origin of the difference between the MR of the Au-doped samples versus that of the undoped ones and why is this difference disorder dependent? Since the origin of the orbital MR in samples with $R_\square > 30$ kΩ is, as yet, an unresolved issue, it is clear from the outset that no definite answer can be given. Nevertheless, it may be possible to gain some insight into the physics involved by *attempting* to fit the MR data to the well-established theories of weak localization. The MR that is pertinent (but expected to strictly hold only for the diffusive regime) is given by[8]

$$\Delta R/R = 3\delta_1 - \delta_2 \qquad (1a)$$

with

$$\delta_i = (e^2 R_\square / 2\pi h)[\ln(H/H_i) + \psi(\tfrac{1}{2} + H_i/H)] , \qquad (1b)$$

where ψ is the digamma function, $H_1 = H_{in} + (4/3)H_{so}$, $H_2 = H_{in} = \phi_0/4L_{in}^2$, and $H_{so} = \phi_0/4L_{so}^2$. L_{in} and L_{so} are the inelastic and spin-orbit diffusion lengths, respectively. For strongly localized samples, L_{in} in Eq. (1) is not a physically meaningful concept and it is natural to replace it by some function of the hopping length, r. In_2O_{3-x} films (both Au doped and undoped) with $R_\square > 30$ kΩ exhibit $R(T)$ that follows the VRH behavior: $R(T) = R_0 \exp(T_0/T)^{1/3}$. A typical $R(T)$ for Au-doped sample is shown in Fig. 5 (cf. Ref. 3 for similar data for undoped In_2O_{3-x} films). We used the measured T_0 to calculate r through[9] $r = \xi(T_0/T)^{1/3}$, $k_B T_0 = 3/N(0)\xi^2 d$ where ξ is the localization length, d is the film thickness, and $N(0) \simeq 10^{32}$ erg^{-1} cm^{-3} is the bulk density of states of In_2O_{3-x}. The dependence of r on $R_\square$ is depicted in Fig. 2. To make contact with the data, the fitting attempts focused on reproducing the value of the "zero-crossing" field, H^*, which proved to be an *extremely* sensitive function of H_{in}/H_{so}. It is straightforward to fit the MR data of the weakly localized samples such as those shown in Fig. 1(a). For example, $H_{in} \simeq 1$ kOe and $H_{so} \simeq 0.65$ kOe reproduce H^* at $\simeq 1.2$ kOe, and with the *measured* $R_\square \simeq 4.5$ kΩ, Eq. (1) gives $\Delta R/R = -0.68\%$ (at $h = 6.5$ kOe) in good agreement with the experimental value of -0.64%. Note that $H_{in} \simeq 1$ kOe implies $L_{in} \simeq 1000$ Å which is consistent with previously published values for this material[10] at similar $R_\square$ and temperature. Setting $H_{so} = 0$ gives (at $H = 6.5$ kOe) $\Delta R/R = -2.85\%$. That looks promising in terms of suggesting a possible physical reason for the difference between the Au-doped and undoped films. Difficulties were encountered, however, in extending the analysis to the samples with $R_\square > 30$ kΩ in which we are primarily interested. (Some of these difficulties can be readily appreciated from the evolution of the MR curves in Fig. 1 without any computation.) Several fitting procedures were tried (including some that involve terms due to mag-

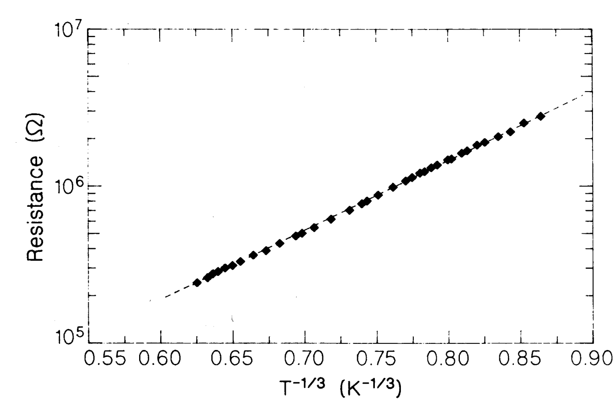

FIG. 5. Resistance as a function of temperature for a typical, Au-doped sample in the VRH regime [same sample as in Fig. 2(c)].

netic scattering). Since one can only hope to get qualitative answers from this procedure, we list only the main results. The starting point was replacing $L_{\rm in}^2$ in Eq. (1) by either r^2 or[11] $r^{3/2}\xi^{1/2}$. $H_{\rm so}$ was then adjusted to account for the observed H^*. This brought up an interesting feature common to all these fitting attempts: As long as H^* could be identified (i.e., for films with $R_\square$ in the range 4.5 to 600 kΩ), $H_{\rm so}$ grew, with disorder, at least as fast as $H_{\rm in}$. Within the context of the backscattering picture, this observation would seem to imply that the disorder reduces both the phase-coherent and the spin-orbit scattering areas involved in the MR which, by itself, is quite plausible. But, it also means that there ought to be an appreciable multiplicative difference (a factor of 3–4 at $H=6.5$ kOe in our numerical calculations) between the MR of Au-doped versus undoped films. This difference is not expected to diminish with $R_\square$ as long as $H_{\rm so}/H_{\rm in}$ does not. The data (Fig. 3), however, suggest a continuous evolution and asymptotic insensitivity of the MR to the presence of spin-orbit scattering. It is noteworthy that the "crossover" region seems quite sharp and it occurs at a regime where the phase-coherent length r is only weakly disorder dependent (Fig. 2). The failure of our fitting attempts makes it clear that the MR for samples that are deep in the VRH regime is not describable by a simple variant of Eq. (1). It is plausible to assume, though, that the weakly localized behavior (i.e., the backscattering mechanism) *does* persist into the VRH regime but is either, rapidly (but continuously) modified or else replaced by another mechanism. This assumption is based on the similarity of the MR curves in Fig. 1(b) [and even 1(c)] to that of 1(a) as well as on the intuitive notion that the physics involved cannot change discontinuously.

We are not aware of a scenario for a modified backscattering that would account for the insensitivity to spin-orbit scattering. The other possibility, i.e., the existence of an additional mechanism that gradually takes over, calls for some specific assumptions: To account for our data, this mechanism has to have the following features.

(1) It is an orbital effect (to account for the anisotropy cf. Fig. 4) but is, apparently, insignificant in the weakly localized regime.

(2) At sufficiently strong disorder, it should give rise to a negative MR.

(3) It should be insensitive to spin-orbit scattering.

A quantum-interference mechanism that has these features has been recently considered by a number of researchers.[11] The relevant interference is conceived to be between different Feynman trajectories involved in determining the spatial overlap of pairs of sites that are r apart. This kind of (oriented-path) interference is similar in nature to that known to lead to conductance fluctuations and therefore it is unimportant for metals in the thermodynamic limit. The MR for *macroscopic* samples is believed to arise from a nonlinear averaging which is peculiar to the VRH process. Both features (1) and (2) are explicit consequences of these theoretical models but the latter are not yet refined enough to enable a quantitative comparison with experimental data. The qualitative prediction of an orbital, negative MR in the VRH regime is insufficient to distinguish these models from backscattering. It is with this respect that the question of spin-orbit sensitivity may provide a lucid test of the theory.

Shklovskii[12] maintains that the oriented-path interference is essentially insensitive to spin-orbit scattering due to the small momentum transfer involved (as opposed to backscattering). This should hold true when $(r/\xi)^{1/2} \gg 1$, since the relevant area for the oriented-path interference is an ellipse of length r and width $(r\xi)^{1/2}$. The "crossover" we are observing seems to occur for $r/\xi > 10$, which looks reasonable. In the limit of strong disorder, then, our results are consistent with the theory on all three accounts.

However, that still leaves the suspicion that, *below* a certain degree of disorder, on which the present experiments place only a lower bound, neglecting backscattering is not justified even though r/ξ is appreciably larger than unity. As we argue below, that is not necessarily surprising. Backscattering is certainly important on a scale smaller[13] than ξ. As long as $\xi > a_0$ (a_0 is the Bohr radius), there must be some delocalizing (or "antidelocalizing") effect due to the field. Such an effect should be easier to observe near the metal-insulator transition where ξ is fairly large (and, hence, the magnetic field needed to obtain an appreciable effect is fairly small[13]), as was recently demonstrated by Roy *et al.*[14]

It has been argued[3] that one expects the direct effect of backscattering to become increasingly less important as r/ξ grows larger. The "delocalizing" effect of the field is associated with a reduction in the quantum-mechanical probability of the electron to "return." In the strongly localized regime this probability is already close to unity due to short-range ($\simeq\xi$) scattering and quantum interference on the scale of r (such that $r \gg \xi$) is only a small added correction. The oriented-path interference, on the scale of r, is likewise an exponentially small entity. However, its contribution modulates the probability of *forward scatter* which, in the VRH regime, is, by itself, an exponentially small function of r/ξ. This qualitative argument[3] merely means that as r/ξ becomes larger than unity, the forward-scattering interference must become the dominant one as far as *observability* of orbital MR effects is concerned. (It is tacitly assumed, as suggested by the theoretical models,[11] that a nonlinear ensemble averaging is inherently involved.) But, returning loops, specifically ignored by these models, may still be important even in the $r \gg \xi$ case. This should be considered as an indirect effect due to backscattering and it may be operative as long as $\xi > a_0$. We refer to the effect of backscattering (important on scales $<\xi$) on the probability to forward scatter (on scales of $\simeq r$). The latter is the outcome of compounding many (elastic) tunneling events along each Feynman trajectory. The number of such steps is, typically, larger than r/ξ. Clearly, the probability for the local (scale of $<\xi$) events will be affected by backscattering; when a magnetic field is applied, it will be enhanced or suppressed depending on the local relative strength of spin-orbit scattering. Due to the smallness of the relevant area ($\simeq\xi^2$) these local effects are individually small. Nevertheless, they may be amplified to an observable magnitude through the compounded probability associated with the forward scattering since the underlying time-reversal symmetry dictates a common trend in the local steps. More formally, the effective tunneling matrix elements, V, which enter in the Nguyen *et al.*[11] model (where even small returning loops are ignored), will be renormalized if returning loops *are* present and this renormalization may be sensitive to the details of the local effect of the field. If this is indeed true, then the current versions of the oriented-path models[11] are strictly valid only in the $\xi/a_0 \simeq 1$ limit. We note that the formal justification for neglecting "returning loops" in the oriented-path models rests on a numerical simulation[12] for a system where $\xi \simeq a_0$ and $r/\xi > 20$ which, to our knowledge, may exclude from comparison most of the experimental results currently in print. It is emphasized that, in the strongly localized regime, it is r/ξ that appears to be the physically relevant parameter for quantum-interference phenomena: The probability amplitudes for the quantum interference, of either kind, as well as their relative significance, depend on r/ξ. That does not mean that r/ξ is the only relevant parameter for the problem at hand even for the noninteracting system. In fact, the arguments raised above suggest that ξ/a_0 should also be considered. It is rather that $R_\square$ is certainly not expected to be the "universal" single parameter for the strongly localized system. Inasmuch as the contribution of backscattering to $\Delta R/R$ vis-à-vis that of the oriented-path interference is concerned, r/ξ appears to be a natural measure of disorder. This point should be borne in mind when comparing results obtained on systems with highly disparate carrier densities. To illustrate, the GaAs specimens reported by Laiko *et al.*[2] had $R_\square \simeq 10^7$ Ω for r/ξ of 5–6. This should be compared with an In_2O_{3-x} film with $R_\square$ that is 4 orders of magnitude smaller for the *same* r/ξ. Similarly, the sample of PbTe studied by Poyarkov *et al.*[15] had r/ξ of only[12] 3–4 and it is dubious whether it can be treated as being in the limit of very strong disorder despite the huge ($>10^{11}$ Ω) value of the resistance measured.

In summary, we have presented experimental results pertaining to the influence of spin-orbit scattering on the MR in the VRH regime. The insensitivity of the MR to spin orbit at sufficiently strong disorder seems to suggest the relevance of the oriented-path mechanism in this limit. At the same time, attention is called to the possible role of backscattering for the intermediate degree of disorder. Our experiments may be interpreted as indicating that returning loops may significantly influence the magnetotransport in the VRH regime. We are not able to determine whether a "critical disorder" is involved, but only place a lower bound on its value. It is important to note that our study is limited to the case of only moderately strong spin-orbit scattering. One cannot rule out the possibility that the critical disorder (whether measured by r/ξ or ξ/a_0) is a function of the spin-orbit scattering strength. Clearly, further comparison with theory must await a more realistic treatment of the problem.

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