Current Jets, Disorder, and Linear Magnetoresistance in the Silver Chalcogenides

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The inhomogeneous distribution of excess or deficient silver atoms lies behind the large and linear transverse magnetoresistance displayed by $Ag_{2\pm\delta}Se$ and $Ag_{2\pm\delta}Te$, introducing spatial conductivity fluctuations with length scales independent of the cyclotron radius. We report a negative, nonsaturating longitudinal magnetoresistance up to at least 60 T, which becomes most negative where the bands cross and the effect of conductivity fluctuations is most acute. Thinning samples down to 10 μm suppresses the negative response, revealing the essential length scale in the problem and paving the way for designer magnetoresistive devices.

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The silver chalcogenides provide a striking example of the benefits of imperfection. Perfectly stoichiometric Ag_2Se and Ag_2Te are nonmagnetic, narrow-gap semiconductors whose electron and hole bands cross at liquid nitrogen temperatures. They exhibit negligible magnetoresistance [1], as predicted from conventional theories [2]. By contrast, minute amounts of excess Ag or Se/Te—at levels as small as 1 part in 10 000—lead to a huge and linear magnetoresistance over a broad temperature range [3–8]. The unusual linear dependence on magnetic field down to 100 G indicates a particularly long length scale associated with the underlying physics, while, at high field, a nonsaturating response up to at least 0.5 MG exceeds by a factor of 50 to 100 the expected cutoff where the product of the cyclotron frequency and the scattering rate $\omega\tau = 1$ [9]. This remarkably robust linear magnetoresistive response makes the silver chalcogenides promising candidates for high field sensors. Missing at present, however, is experimental evidence for the pertinent length scales of the inhomogeneities that determine the unusual physics and that are essential to the materials' usefulness.

Abrikosov was the first to stress the importance of the inhomogeneous distribution of the excess or deficient silver ions. In his effective medium theory of quantum linear magnetoresistance [10], disorder and a linear dispersion relation at band crossing [11] combine to produce a linear, rather than a quadratic, magnetic field dependence for the electrical conductivity. As pointed out by Parish and Littlewood [12], fluctuations in the mobility are particularly acute when the gap goes to zero and both positive and negative values can be sampled. Their simulations of large spatial conductivity fluctuations in strongly inhomogeneous semiconductors derive a linear magnetoresistance from the Hall voltage picked up from macroscopically distorted current paths caused by variations in the stoichiometry. The spatial fluctuations in the conductivity are caused by the random distribution of Ag ions, which may take the form of highly conducting nanothreads or lamellae along the grain boundaries of polycrystalline material [13]. The distorted current paths seen in the simulations lead to the emergence of a characteristic length scale that can be associated directly with a magnetic field scale for the onset of linear response.

The exact disposition of the excess or deficient Ag is difficult to resolve via either scattering or imaging techniques. In this Letter, we show that a systematic investigation of the longitudinal magnetoresistance of the silver chalcogenides in magnetic fields up to 60 T can be used to directly identify the physical length scale of interest. Although the transverse magnetoresistance is always positive, a pronounced negative longitudinal magnetoresistance, i.e., the reduction of the sample resistance in a parallel magnetic field, emerges at high field. We observe this effect over a wide temperature and composition range for both n-type and p-type Ag_2Se and Ag_2Te. The appearance of the negative magnetoresistance is due to an effect analogous to current jetting [14,15], but is caused here by conductivity fluctuations. Most significantly, its dependence on sample thickness yields a characteristic length scale set by the spatial inhomogeneity that is as large as 10 μm.

High purity, stoichiometric Ag_2Se was ground and loaded into outgassed, fused silica ampoules inside a helium glovebox, and appropriate weights of Ag or Se were added to reach the desired compositions. The mixture was baked at 50 K above the melting point (1170 K) and left to cool for 24 h in a horizontal position. Polycrystalline samples with typical dimensions $(3.0 \times 1.0 \times 0.4)$ mm^3 were cut perpendicular to the cylindrical axis to avoid possible dopant variations due to temperature gradients inside the furnace. $Ag_{2\pm\delta}Te$ polycrystals were prepared in a similar manner, except that high purity elements of Ag and Te (99.999%, Alfa Aesar) were directly used for melt crystallization. We performed conventional four-probe re-

sistivity measurements $\rho(T, H)$ with electrical contacts placed on the top, bottom, and sides of the samples to measure the ρ_{xx}, ρ_{xy}, and ρ_{xz} components of the resistivity tensor. It is important to note that the van der Pauw method, with contacts placed on the corners of a square plate, cannot be applied to the present situation because it relies on a uniform current distribution to deconvolute the tensor components of ρ. We used a 14/16 T superconducting magnet for low field measurements and the 60 T short-pulsed magnet at Los Alamos National Laboratory for the high field response. The latter provides 16 ms long field pulses from the discharge of a 1 MJ capacitor bank and, when combined with the National High Magnetic Field Laboratory-designed synchronous digital lock-in amplifier with response times much shorter than commercial lock-ins, can provide low-noise data during each 16 ms shot.

We contrast in the two panels of Fig. 1 the normalized magnetoresistance $\Delta\rho(H)/\rho(0)$ of n-type $Ag_{2+\delta}Se(\delta = 0.0001)$ under transverse and longitudinal magnetic fields $0 \leq H \leq 55$ T. The longitudinal and transverse magnetoresistance differ in both magnitude and functional dependence. The longitudinal response is an order of magnitude smaller than its transverse counterpart and crosses through zero to become negative at high field. Both quantities refuse to saturate at the highest fields and assume an essentially linear dependence on field (except at the lowest temperatures where quantum oscillations appear to dominate). Similar negative longitudinal magnetoresistance was observed on all $Ag_{2+\delta}Se$ samples investigated (δ varying from -0.002 to 0.001). Although the negative longitudinal magnetoresistance is consistently larger in p-type material, there is no systematic dependence on δ.

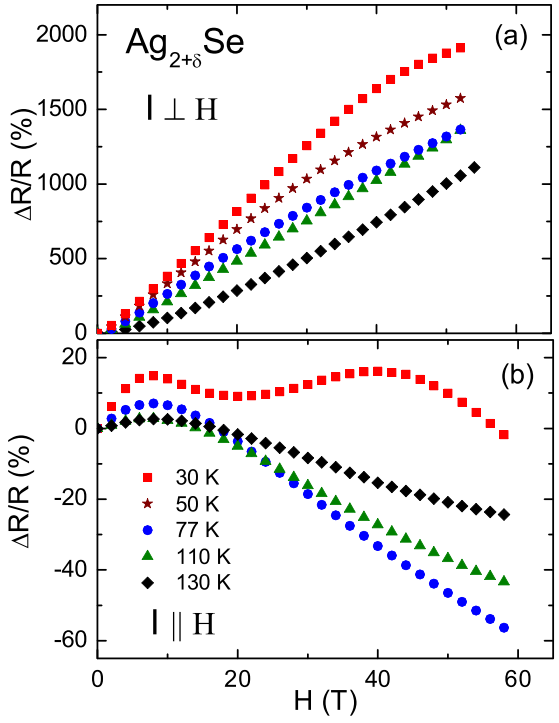

FIG. 1 (color online). Normalized magnetoresistance of n-type $Ag_{2+\delta}Se$ under (a) transverse and (b) longitudinal magnetic field at a series of temperatures. The transverse response is large, positive, and essentially linear with magnetic field, while a crossover to linear but negative magnetoresistance occurs in the longitudinal configuration.

The pronounced negative component in the longitudinal magnetoresistance is consistent with the picture of inhomogeneous conductance. In such a model, the combination of inhomogeneities and high magnetic fields leads to current paths that behave in a counterintuitive manner, at times flowing perpendicular to both the applied voltage and the magnetic field [12]. This provides a linear contribution to the transverse magnetoresistance due to Hall resistances associated with the perpendicular flows.

In the present longitudinal geometry, where the magnetic field is parallel to the applied current, the perpendicular current will flow across the thickness of the sample, bending the equipotential lines. The voltage drop across the length of the sample will be reduced and a negative magnetoresistance can emerge. This is analogous to the effect known as current jetting [14,15], differing only in the fact that here it is a manifestation of inhomogeneous conductivity rather than simple geometric effects. The maximum in the negative magnetoresistance at $T = 77$ K corresponds to the crossover from intrinsic to extrinsic semiconducting behavior. At this point the distortions in the current should be strongest as the fluctuations in the conductivity can veer from positive to negative.

The inset of Fig. 2 shows a schematic visualization of distorted current paths in the presence of a longitudinal magnetic field. In addition to the negative magnetoresis-

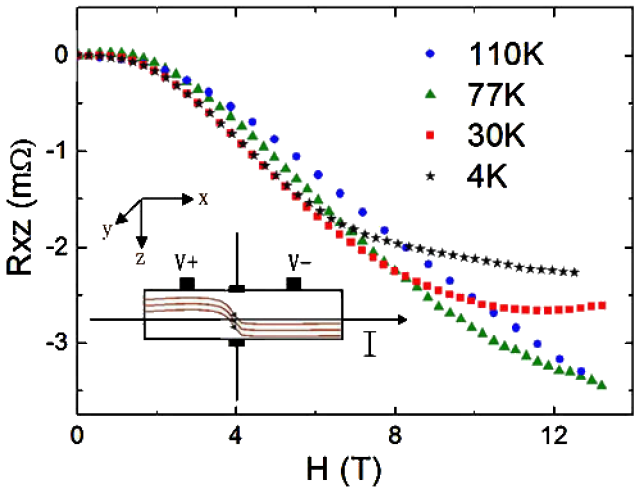

FIG. 2 (color online). A voltage drop across the thickness of p-type $Ag_{2-\delta}Se$ develops under a longitudinal magnetic field, revealing distorted current flows across the sample as the source of the negative magnetoresistance (illustrated in the inset).

tance, we expect a voltage drop across the sample thickness for the depicted current pattern. In other words, in contrast to what is observed in ordinary polycrystalline semiconductors, a nonzero tensor component $\rho_{xz}(H)$, known as the transverse-longitudinal coupling, should become apparent due to the inhomogeneous nature of our system. We plot in the main panel of Fig. 2 the voltage drop across a p-type sample as a function of magnetic field for $0 \leq H \leq 14$ T. A conventional six-probe configuration was used to allow for later correction $[\rho_{xz}(H) - \rho_{xz}(0)\frac{\Delta\rho_{zz}(H)}{\Delta\rho_{zz}(0)}]$ of any possible lead misalignment. The observed voltage drop provides clear evidence of the predicted current distortion in our system. The sign and functional form varies from sample to sample depending on the direction of the bend in the current and the location of the voltage leads. Again, the maximum voltage drop found at $T = 77$ K indicates that the most crooked current paths occur near band crossing.

The formation of spatial inhomogeneities in the silver chalcogenides is closely related to the clustering of Ag ions at lattice defects or grain boundaries when quenched from the high temperature α phase to the semiconducting β phase with lower Ag solubility [13]. The ultimate unit size of these clustering networks sets a lower limit for the effective length scale of the current distortion, in the sense that the effect of spatial randomness would be completely suppressed if the size of the system becomes comparable to that length scale in one dimension. As a result, the negative longitudinal magnetoresistance, a direct manifestation of inhomogeneity along the sample thickness, should diminish in thinner crystals. We investigate the thickness dependence of the negative longitudinal magnetoresistance by mechanically shaving the same p-type sample used for the $\rho_{xz}(H)$ measurement of Fig. 2. We plot in Fig. 3 the evolution of the longitudinal magnetoresistance at various sample thicknesses: 400, 50, and 15 μm. The negative component decreases as the sample grows thinner and eventually disappears at 15 μm, indicating a characteristic length scale of order 10 μm. At the same time, no change was found in the transverse magnetoresistance at all thicknesses investigated, indicating that the strain-induced magnetoresistance associated with grinding is negligible, and in agreement with the claim that the linear transverse effect results from the current distortion within a 2D plane [12].

Experimental visualization of Ag clustering was provided recently by von Kreutzbruck *et al.*, who report micron scale silver lamellae in $Ag_{2+\delta}Se$ with extremely high silver excess, $\delta \approx 1.3$ [16]. Such high doping levels give rise to a relatively large diameter, interconnected silver clusters, and an onset of percolation phenomena, well away from the regime covered in this study, but the silver threads observed with high resolution TEM are consistent with the tens of microns length scales derived from the longitudinal magnetoresistance.

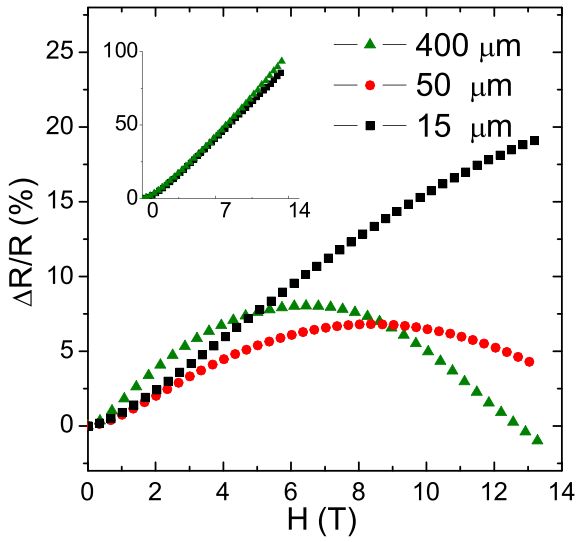

FIG. 3 (color online). Normalized longitudinal magnetoresistance vs sample thickness for $Ag_{2-\delta}Se$ at $T = 30$ K. The negative component of the magnetoresistance decreases with thinning, vanishing by 15 μm. This sets the inhomogeneity length scale in the problem. Inset: The transverse magnetoresistance is independent of sample thickness, shown here for 15 and 400 μm.

In clever experiments, Solin and co-workers have crafted composite materials where current deflections around highly conducting impurities combine a geometrical magnetoresistance with the intrinsic physical magnetoresistance of $Hg_{1-x}Cd_xTe$ to greatly enhance the response [17]. Such an approach points to the general promise of disordered media, but differs from the silver chalcogenides in underlying mechanism (dependence on the intrinsic physical magnetoresistance of the semiconductor $Hg_{1-x}Cd_xTe$), functional form (quadratic with field), saturation at relatively low fields ($\sim$1 T), and macroscopic homogeneity of the current flow.

To this point, we have concentrated on silver selenide as the representative material for large, linear, and nonsaturating magnetoresistance. We complement our study of $Ag_{2\pm\delta}Se$ with a systematic investigation of the longitudinal magnetoresistance in $Ag_{2\pm\delta}Te$. As shown in Fig. 4, even larger negative responses with smaller crossover fields were found for n- and p-type $Ag_{2\pm\delta}Te$ ($\delta = 0.0005$). The measurement of the $\rho_{xz}(H)$ components also shows behavior similar to that depicted in Fig. 2.

The negative longitudinal magnetoresistance universally observed in doped silver chalcogenides appears to be well described by spatial fluctuations in the conductivity. The associated length scale, which dominates the transport mechanism and sets the effective dimensionality, was determined to be approximately 10 μm by investigating the dependence of the magnetoresistive response on sample thickness. The surprisingly large length scale opens the gate to artificial fabrication of conducting networks with

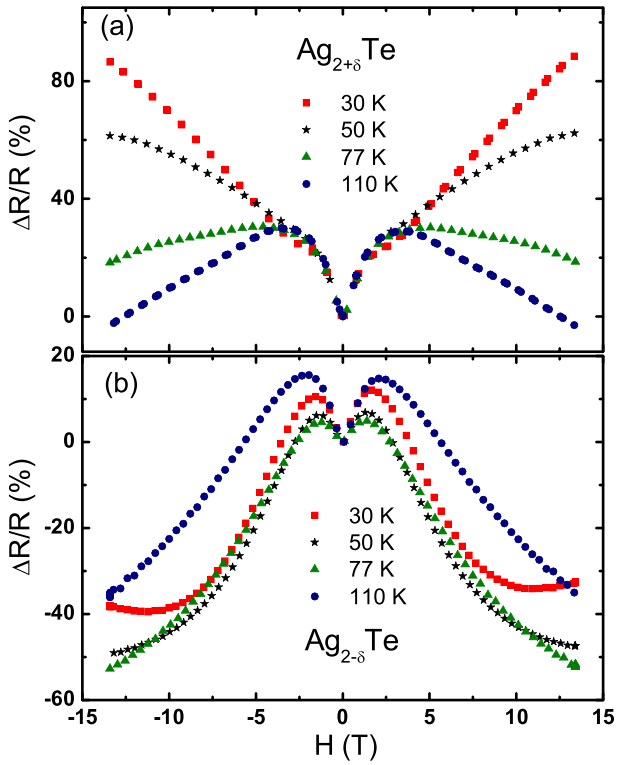

FIG. 4 (color online). Normalized longitudinal magnetoresistance as a function of magnetic field for (a) electron- and (b) hole-doped silver telluride at a series of temperatures. Pronounced negative magnetoresistance emerges in both limits.

micron scale unit size for enhanced magnetoresistive effects. The narrow-gap semiconductor, InSb, may be a promising candidate for such devices. A galvanomagnetic study of *n*-type indium antimonide published almost 50 years ago [18] shows negative longitudinal magnetoresistance of the type we observe in the silver chalcogenides (but absent in modern epitaxial InSb wafers). Harnessing the current jetting effects due to inhomogeneities could linearize and extend the range of the material's transverse magnetoresistive response.

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