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Low-temperature magnetoresistance in insulating a-Gd_xSi_{1-x} alloys

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We report on low-temperature magnetotransport measurements on the amorphous semiconductor a-Gd_xSi_{1-x} with $x \sim 0.13$, on the insulating side of the $T=0$ metal-insulator transition for this material. The samples exhibit a negative magnetoresistance of more than five orders of magnitude at 1 K, which grows exponentially larger at lower temperatures. The temperature dependence of the conductivity displays an activated form indicative of variable range hopping in the presence of a Coulomb gap in all magnetic fields from 0 to 9 T, while the characteristic temperature T_0 of the hopping conductivity decreases from over 300 K in 0 T to 6 K in 8.5 T. This enormous magnetoresistance must arise from an exchange interaction between the conduction electrons and the local Gd moments which are randomly oriented in zero field and become (partially) aligned in 9 T, and its consequent influence on the electron-electron interaction. However, it remains to be determined whether the effect of this magnetic interaction is a relative shift of the Fermi energy relative to the mobility edge and a modification to the Coulomb gap or a change in the bandwidth and its consequent effect on the electronic density of states. [S0163-1829(99)50606-1]

Many materials with intrinsic magnetic moments have been found to possess giant negative magnetoresistance (MR) comparable to or larger than that in artificial magnetic structures,[1,2] due to various types of interactions between the conduction electrons and the local moments. In particular, this class of materials include mixed-valence manganites,[3] some heavy fermion systems,[4] rare-earth chalcogenides,[5,6] and diluted magnetic semiconductors[7] (DMS's). In insulating crystalline DMS's and chalcogenides, the exchange interaction has been shown to cause the formation of bound magnetic polarons (BMP's).[8] Specifically, an electron localized on an impurity atom with a Bohr radius of many lattice constants polarizes the magnetic moments within its orbit and forms a ferromagnetic entity within the paramagnetic or antiferromagnetic background of the material. In some materials the extra binding energy due to the polarization cloud leads to a reduced hopping probability and a large suppression of the conductivity below a temperature comparable to the polaron binding energy. A magnetic field aligns the spins in the background with those in the BMP, reducing the binding energy and resulting in a dramatic increase of the sample conductivity. This mechanism is believed to be responsible for the negative MR, and in some extreme cases the magnetic-field-tuned insulator (I) to metal (M) transition in a variety of crystalline DMS's (Refs. 9 and 10) and rare-earth chalcogenides.[5,6] Microscopic mechanisms behind the negative MR in the heavy fermion systems and the manganites are presumed to be different, but in all cases involve an interaction between conduction electrons and local moments which are aligned by the application of a field. In fact, in the case of doped manganites, there is a growing body of theoretical work[11,12] and experimental evidence[13,14] for electronic phase separation and consequently the formation of magnetic polarons. Inelastic neutron scattering even revealed the existence of coherent ''mobile'' magnetic polarons[13] and a picture of ferromagnetic percolation was suggested for the metal-insulator transition near T_c.

The effect of disorder on the conductivity of materials both in the metallic and the insulating state has been much studied.[15] Amorphous alloys such as a-Nb_xSi_{1-x},

a-Au_xGe_{1-x}, and a-Mo_xGe_{1-x} with $x \sim 0.1-0.15$ have provided systems where the disorder is large. In these alloys changing x allows for a smooth transition from an insulating to a metallic state. It has been shown that the electrical conductivity near the MI transition is dominated by the formation of a Coulomb gap Δ_c in the single-particle electronic density of states (DOS) on the insulating side[16] and a precursor correlation gap on the metallic side.[17] Mott[18] showed that in the disordered insulator at finite temperature T, electrons optimize their hopping probability by seeking accessible states both in space and energy, which results in a variable range hopping conductivity $\sigma(T) \propto \exp(-T_0/T)^{\nu}$ with $\nu = \frac{1}{4}$ for noninteracting electrons in three dimensions and $\nu = \frac{1}{2}$ when a quadratic soft Coulomb gap is present.[19] The effect of adding magnetic interactions to this picture with local magnetic moments is not clear; in particular there is a lack of understanding of the interplay between the magnetic interaction and the electron-electron interaction effects. In the DMS's, there is clearly increased localization and a consequent shift of the MI transition to higher carrier concentrations. In $Gd_{3-x}v_xS_4$ (v = vacancy), where the magnetic field induces an I to M transition, there is evidence for the continued importance of the Coulomb gap or its precursor in both the I and the M states.[6] In at least one of the DMS's it has been suggested that a hard gap Δ_H in the DOS is produced inside the soft Coulomb gap by the magnetic interaction, leading to a simple thermally activated conductivity $\sigma(T) \propto \exp(-\Delta_H/T)$ at low temperature and a crossover to the $\nu = \frac{1}{2}$ behavior at higher temperatures.[10] In this case there is an apparent modification of the DOS from the magnetic interaction and the transport is determined by the relative strength of Δ_H and Δ_c, which depend on the magnetic field as well as the carrier concentration.

We recently showed[20] that an amorphous alloy with a local magnetic moment, a-Gd_xSi_{1-x} with x from 0.11 to 0.15, exhibits giant negative MR while its nonmagnetic analog a-Y_xSi_{1-x} only possesses a small positive MR similar to what was previously seen in other nonmagnetic alloys.[15] This material bears some resemblance to the DMS materials, but the greater disorder of the amorphous structure leads to several crucial differences. In particular, the carrier concentration of interest, near the MI transition, is approximately four orders of magnitude larger ($10^{22}\,cm^{-3}$ versus $10^{18}\,cm^{-3}$). The higher concentration and greater disorder means that the electron-electron correlation effects are crucial, as is seen in the nonmagnetic alloys, and as will be shown in the magnetic systems as well. It also means that the Fermi energy and temperature are large, so that Coulomb correlation effects are seen at relatively high temperatures. In amorphous materials, instead of an electron localized on a single impurity donor or defect with a Bohr radius of many lattice constants, the MI transition occurs due to a collective action of many scattering centers and could take place even if a single impurity has no bound states; hence the localization length depends on a variety of factors such as the dielectric constant and the proximity to the MI transition. The high carrier concentration also implies that the ratio of the number of magnetic moments to carriers is close to unity instead of the 10^3-10^4 for the DMS's. Therefore, the BMP is a difficult concept to take literally in these amorphous materials since it is hard to imagine a picture of a single localized electron

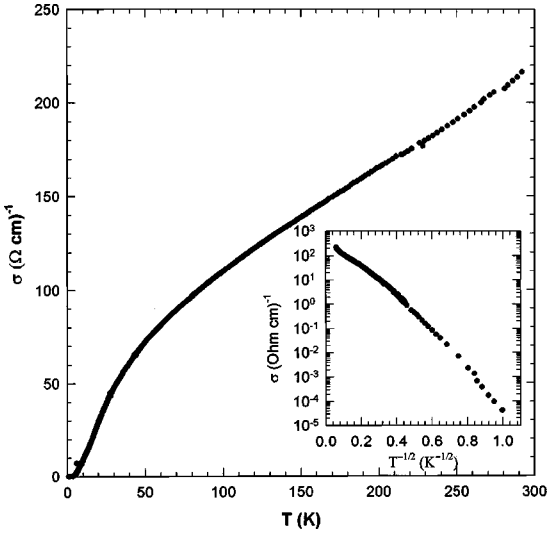

FIG. 1. σ as a function of T for an a-Gd_xSi_{1-x} sample with $x \sim 0.13$. Inset: The low-temperature σ plotted logarithmically versus $T^{-1/2}$.

polarizing the nearby moments. If the BMP are to remain a valid concept, they are certainly overlapping since the localization length is large near the MI transition and many conduction electrons (and moments) must exist within it. Hence one may be able to view the effect as the formation of a polaron band localized by disorder. The materials discussed in this paper are insulators, but within a localization length the overlapping electron wave functions could be considered in terms of a metallic state with an effective mass which depends on the polarization of the local magnetic moments. It is thus possible that the concepts developed for heavy fermions of a field- and temperature-dependent effective mass are a better way of understanding the present materials. The magnetotransport in these materials also bears some resemblance to that of the manganites in their paramagnetic state, above the magnetic ordering temperature. Therefore, the IM transition in a-Gd_xSi_{1-x} as a function of increasing x may have some relevance to the physics of the IM transition in the manganites near T_c.

Even in the crystalline magnetic semiconductors, the microscopic mechanism behind the degradation of conductivity has not been determined beyond ambiguity in all of the systems. Specifically, in some systems it is still not clear whether the BMP lead to a narrowing of the bandwidth, in addition to the shift of the Fermi energy E_F and the mobility edge E_c, which is well accepted.[8] A further interesting aspect of the a-Gd_xSi_{1-x} alloys reported here when compared to the DMS is that the direct Gd-Gd exchange interaction is negligible due to the f-electron character, unlike the large antiferromagnetic exchange interaction between the Mn in the DMS's.

In order to investigate the interplay between the magnetic interaction and the electron-electron correlation effects, it is important to study the transport at low temperatures where both effects are well developed. In this paper we report a detailed study of the low-temperature (down to 155 mK) magnetotransport behavior of the a-Gd_xSi_{1-x} with $x \sim 0.13$ on the insulating side of the $T=0$ MI transition for this material in zero field. We note that a-Y_xSi_{1-x} and a-Nb_xSi_{1-x} at $x \sim 0.13$ are on the metallic side of this transition at all

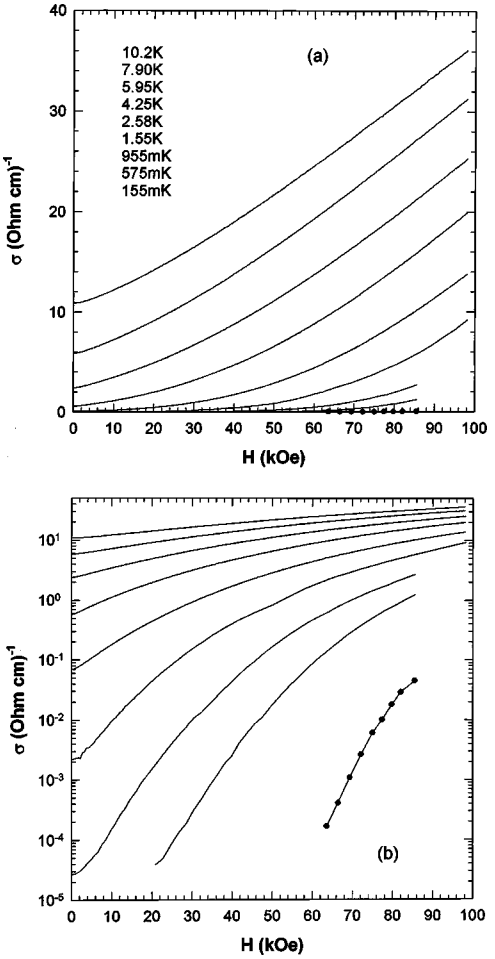

FIG. 2. (a) σ versus applied magnetic field at different temperatures; (b) the same data plotted on logarithmic scale.

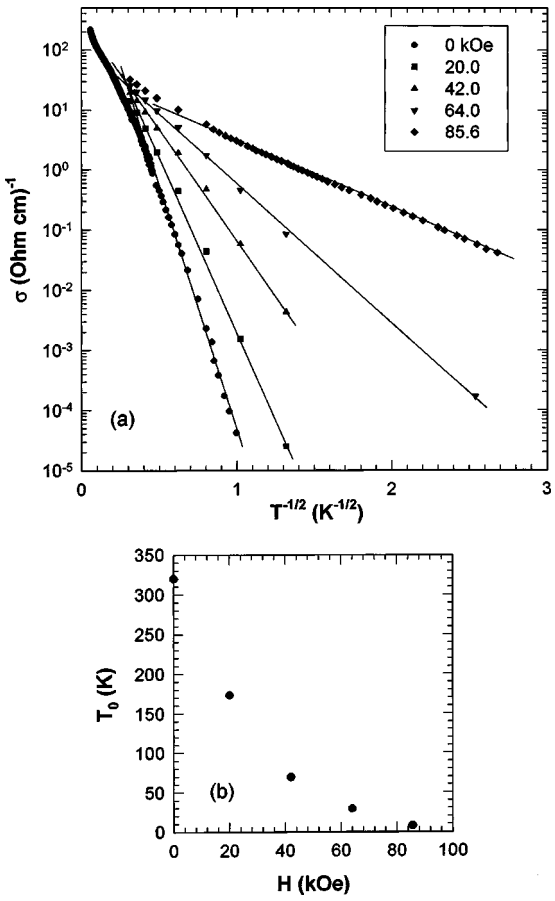

FIG. 3. (a) $\log_{10}\sigma$ as a function of $T^{-1/2}$ for the same sample in different fields; (b) T_0 extracted from above plotted against H.

fields and that at high temperatures (above ~70 K) the conductivity of the a-Gd_xSi_{1-x} sample is similar to that of the nonmagnetic samples. In other words, the insulating behavior of the a-Gd_xSi_{1-x} sample does not develop until the temperature drops below ~70 K, which is suggestive of a magnetic binding energy as seen in the DMS's. In a magnetic field of 9 T, the a-$Gd_{0.13}Si_{0.87}$ samples exhibit a dramatic enhancement of conductivity, as high as five orders of magnitude at 1 K and exponentially larger at lower temperatures.

The samples were made by electron-beam coevaporation of Si and Gd in a UHV chamber as described in Ref. 20. The substrates, Si covered with 4000 Å of amorphous Si nitride, were held at room temperature during growth. They were cut into 1 mm×3 mm stripes and leads were attached by cold-pressed indium for four-probe resistance measurement. The samples are extremely stable in air and with respect to thermal cycling, which enabled us to perform multiple measurements from room temperature down to 150 mK. For the resistance measurements care was taken to ensure that they were made in the Ohmic regime. Typically a current of 10 nA was used at room temperature and reduced to a few tenths of a nA at lower temperatures. At the lowest temperatures the I-V curves became nonlinear at even smaller current, in which case I-V curves were recorded at individual points of different field strengths and the resistances were obtained by determining the slope of the I-V at zero bias. We have measured two samples on the insulating side showing similar results.

Figure 1 shows the temperature dependence of the conductivity σ in zero field. $\sigma(T)$ is typical of what has been previously observed for a-Gd_xSi_{1-x} with a nearly linear T dependence at high T and a steeper decrease in conductivity below ~70 K. This sample is clearly on the I side of the MI transition as indicated by the vanishing σ at $T=0$ and an exponential T dependence at low T as illustrated in the inset. A function of $\exp(-T_0/T)^{\nu}$ with $\nu=\frac{1}{2}$ gives the best fit to the data below 5 K, over 6 decades of conductivity.[21] This temperature dependence is characteristic of a conduction mechanism of variable range hopping in the presence of a soft Coulomb gap (Δ_c). The nonmagnetic analogs such as a-Nb_xSi_{1-x} (Ref. 17) and a-Y_xSi_{1-x} (Ref. 20) with x ~0.13 have conductivity remarkably similar to that found for a-Gd_xSi_{1-x} at high temperatures but exhibit metallic behavior ($\sigma=\sigma_0+A\sqrt{T}$) at low temperatures. For even lower values of x (e.g., $x=0.11$), the nonmagnetic analogs become insulating with a similar low temperature $\nu=\frac{1}{2}$ T dependence. The difference is presumably due to the interaction of the electrons with the local magnetic moments.

Figure 2 displays the magnetoconductance of this sample at different temperatures. The sample was zero-field-cooled down to 155 mK and then the resistance was measured as a function of magnetic field up to 8.5 T. At $T=155$ mK, the magnetoconductance did not show any hysteresis in high fields but the conductivity became unmeasurably low (limited by the input impedance, 10^9 Ω, of the preamplifiers we

used) at low fields. In all of the measurable range, σ is symmetric in fields of opposite direction. σ was also measured with the field parallel and perpendicular to the film plane and the current direction, with negligible difference. The curves shown here represent one branch of the field sweeps with H applied perpendicular to both the film plane and the current. The magnitude of the magnetoconductance of the a-Gd_xSi_{1-x} is extremely large. The vast variation of the sample conductivity in a magnetic field is best illustrated on the logarithmic scale as shown in Fig. 2(b). At $T=955\,\mathrm{mK}$, σ is increased by more than five orders of magnitude by a field of 8.5 T. At low temperatures and especially at low fields, $\sigma(H)$ shows a substantial nonlinearity. The degree of nonlinearity decreases as the temperature and the magnetic field increase. Attempts to fit $\sigma(H)$ to a simple power-law behavior (H or H^2) did not yield satisfactory results. We also plotted σ as a function of induced magnetization M for different samples, arguing that M is the appropriate independent variable as it measures the degree of local moment alignment, but this did not yield a collapsed set of data even for a single sample at different temperatures.

Figure 3(a) displays $\sigma(T)$ in different magnetic fields, plotted as $\log_{10}(\sigma)$ versus $T^{-1/2}$. At low temperatures ($T<4\,\mathrm{K}$) the linearity of these curves shows that $\nu=\frac{1}{2}$ gives the best description of the data under all magnetic fields. In Fig. 3(b) we plot the characteristic temperature T_0 as a function of the magnetic field. T_0 decreases from 320 K in 0 T to less than 10 K in 8.5 T. Clearly the enormous field enhancement of the conductivity here does not result in a magnetic-field-driven I to M transition in this sample, although it seems likely that higher fields would result in such a transition. This conclusion is based on the metallic behavior of the a-Y_xSi_{1-x} and a-Nb_xSi_{1-x} alloys of comparable composition; if the Gd moments were perfectly ferromagnetically aligned by a large field, we suspect that the a-Gd_xSi_{1-x} would be metallic. This dependence of T_0 on H means that the MR grows exponentially with decreasing temperature due to the different exponential behavior of the $H=0$ and $H=8.5\,\mathrm{T}$ data. An extrapolation of the $H=0$ data to 155 mK and the measured value in $H=8.5\,\mathrm{T}$ gives a magnetoresistance ($\Delta R/R$) of 10^{17}.

The persistence of the $\nu=\frac{1}{2}$ behavior for all magnetic fields indicates that in a-Gd_xSi_{1-x} the electron-electron interaction dominates the transport, with no sign of a magnetic hard gap. The magnetic interaction between the conduction electrons and the local magnetic moments affect the transport behavior through its influence on the electron-electron interaction. We suggest that this interplay could occur in two ways: The first scenario is a shift of the location of the mobility edge E_c relative to the Fermi energy E_F. In crystalline DMS and chalcogenides where the BMP are well defined, the formation of the BMP reduces the energy of the occupied states which effectively drops E_F below E_c. The reduced kinetic energy can also be understood as an increase in the carrier effective mass and a degradation of the carrier mobility. In amorphous materials the formation of the BMP is not as well defined because of the reasons discussed before. However, regardless of the microscopic picture, the magnetic interaction introduces an additional random fluctuation scattering potential (in zero field) which could shift E_c relative to E_F, and thereby reduce the localization length ξ and decrease the dielectric constant κ. A magnetic field aligns the moments and reduces the disorder potential, increasing ξ and κ. The tremendous effect the magnetic interaction has on the transport properties of a-Gd_xSi_{1-x} in this scenario comes from its influence on the electron-electron interaction, which modifies the activation temperature $T_0=2.8e^2/k_B\kappa\xi$ and the Coulomb gap $\Delta_c=e^3(N_0/\kappa^3)^{1/2}$, where N_0 is the DOS without interaction, through an alteration of ξ and κ. This effect, in essence, includes a combination of a renormalization of the carrier effective mass and the consequent effect on the carrier mobility, as well as a modification of the effect of the Coulomb interaction on the DOS. The magnetic field may alternatively lead to a change in the bandwidth itself, which should result in a significant change in N_0 and the effective mass.[11,12,22] With the present data we cannot make a definitive statement whether the behavior of a-Gd_xSi_{1-x} is due to a modification of Δ_c from poor screening or any significant change in N_0 itself. An independent determination of the DOS such as by an electron tunneling measurement is needed and currently underway.

In summary, we have carried out low-temperature magnetotransport measurements on insulating a-Gd_xSi_{1-x} with $x\sim0.13$. The samples exhibit an enormous enhancement of conductivity in a magnetic field. The magnetotransport properties of the a-Gd_xSi_{1-x} appear to be determined by an intricate balance and interplay between the magnetic exchange interaction and the electron-electron interaction. The behavior of the field and temperature dependence of the conductivity showed marked differences from those for its crystalline counterparts. The Coulomb correlation is found to dominate the transport at low temperatures so that σ retains an $\exp\{-(T_0/T)^{1/2}\}$ form in the full range of magnetic fields we have investigated, although we anticipate that higher fields would result in a metallic form for σ. T_0 is strongly field dependent, changing by nearly two orders of magnitude between 0 and 8.5 T. It has yet to be determined whether the dramatic effect the magnetic impurities and the magnetic field have on the sample conductance results from a modification of the electron-electron interaction and its effect on the electronic DOS via a relative shift of the positions of E_F and E_c, or a change in the bandwidth and the consequent variation in the DOS and effective mass. It is possible that some combination of these effects determines the behavior of the a-Gd_xSi_{1-x} system.

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