Electron Localization in a Magnetic Semiconductor: $Gd_{3-x}v_xS_4$

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The metal-insulator transition in the magnetic semiconductor $Gd_{3-x}v_xS_4$, where v stands for vacancies, has been studied by tuning through the transition with the application of a magnetic field at low temperatures. For two samples with $x = 0.321$ and 0.325 the transition is continuous. $\sigma(T\rightarrow 0)$ is linear in $H-H_c$ which implies that $\sigma(T\rightarrow 0)\propto (E-E_c)$, where H_c is a critical field and E_c is the mobility edge. This is consistent with new scaling theories of both localization and interaction effects.

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Current understanding of transport near the metal-insulator transition in three-dimensional materials, including heavily doped crystalline semiconductors and disordered amorphous alloys, derives from several recent theoretical developments. The first is the idea of minimum metallic conductivity

$$\sigma_{\min}(T=0)\simeq C(e^2/ha_E) \tag{1}$$

derived by Mott,[1,2] where $C\simeq 0.025-0.05$ is a constant depending on coordination, the estimated reduction in density of electron states near the mobility edge, E_c, and degree of compensation. a_E is the distance between electrons $\simeq n_c^{-1/3}$, where n_c is the net carrier concentration at which the transition occurs [i.e., $E_F(0)=E_c$ where $E_F(0)$ is the Fermi energy] and is related to the Bohr radius a_B of the impurity wave function by $n_c^{1/3}a_B \sim 0.25$.[3]

Whereas the concept of $\sigma_{\min}$ predicts a discontinuous jump in low-temperature conductivity as the carrier concentration is increased through n_c, Abrahams *et al.*[4] and later Imry[5] concluded, using scaling arguments, that

$$\sigma(T=0)\propto[E_F(0)-E_c]^{\nu}\propto[\ln(g_0/g_c)]^{\nu}, \tag{2}$$

where $g_0 \gtrsim g_c$ is a generalized conductance near the transition and $\nu\sim 1$. Equation (2) may also be written[6] as

$$\sigma(T=0)=\sigma_{\min}(n/n_c-1)^{\nu} \tag{3}$$

to make explicit the dependence on carrier concentration, n. No discontinuity is therefore expected according to the scaling theory.

Recent low-temperature (millikelvin) experiments show no discontinuity and no unique experimental value for ν.[6-9] Since both the scaling result and $\sigma_{\min}$ are based on a theory of noninteracting electrons, interactions have been invoked either for the occurrence of a continuous transition or for a departure from $\nu=1$. For example, in uncompensated Si:P, Rosenbaum *et al.*[6] and Paalanen *et al.*[7] observe $\nu=0.48\pm0.07$ and explain the deviation from $\nu=1$ by invoking electron-electron interactions. The presence of interaction effects was demonstrated by tunneling experiments in granular Al[8] and in amorphous NbSi[9] in agreement with theoretical predictions of a gap by Mc Millan.[10] However, ν was found to be close to 1 in these experiments. More recently, Grest and Lee,[11] also considering the combined effects of interaction and localization, have predicted $\nu\sim 0.6$ in zero magnetic field H, and $\nu\simeq 1$ for $H\neq 0$. The effects of magnetic order (with or without H) on transport in disordered magnetic metals have been discussed qualitatively by Imry.[12] In particular, he notes that magnetic order may affect g_0-g_c.

The purpose of this Letter is to describe transport measurements in the magnetic semiconductor $Gd_{3-x}v_xS_4$ [13-15] (v = vacancy) where it is possible to tune through the insulator-metal transi-

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tion by applying a magnetic field. $Gd_{3-x}v_xS_4$ is the analog of a classical compensated semiconductor. As was first recognized by Cutler and Mott,[16] the random distribution of vacancies at the rare-earth (Gd) sites leads to fluctuating repulsive potentials and tailing of the conduction band in which the electronic states are localized. As emphasized by Mott and Kaveh,[17] interaction effects will be minimized in strongly compensated semiconductors. For these conditions a discontinuity in $\sigma(T\to 0)$ should be observable for conduction bands formed from nondegenerate ground states.

$Gd_{3-x}v_xS_4$ forms a complete series of solid solutions in which the end members Gd_2S_3 ($x=\frac{1}{3}$) and Gd_3S_4 ($x=0$) are an antiferromagnetic insulator and a ferromagnetic metal, respectively. The Th_3P_4 type structure[18] is preserved for all x, despite the fact that the number of vacancies can be varied by $\sim 2.3\times 10^{21}$ cm^{-3}. Resistivity measurements[14,15] on samples with electron concentrations $n\sim 8.7\times 10^{19}$ to 2.5×10^{20} cm^{-3} indicate activated transport down to 4.2 K. For $H \geq 32$ kOe, however, the sample with largest n becomes metallic. This is exactly the opposite of what happens in, e.g., In-Sb,[19] where a positive magnetoresistance results from the shrinkage of impurity state orbits in high applied fields. In fact, all samples described in Refs. 12 and 13 show large negative magnetoresistances. This unusual behavior for carrier concentration $n>n_c$ $\sim 8\times 10^{18}$ cm^{-3}, estimated from paramagnetic $Ce_{3-x}v_xS_4$,[16] indicates the importance of magnetic interactions in determining the binding energies of the carriers. Localization occurs because of the combined action of the random potential fluctuations and the magnetic interaction of the conduction electron with its Gd^{3+} neighbors. This latter effect may be described as a ferromagnetic polaron in an antiferromagnetic lattice. An applied field polarizes the matrix ferromagnetically, thereby unbinding the polaron and producing the observed negative magnetoresistance. Since this effect is isotropic, the direction of the applied field (apart from small demagnetizing field corrections) is inconsequential and the present experiments were performed with $H \perp I$, the current.

Penney *et al.*[15] have examined both transport and magnetic data to characterize the polaron. Their conclusions for the insulator side of the transition include the following: (a) The magnetic polaron is large, extending over many lattice and vacancy sites; (b) the polaron hopping energy in the absence of an applied magnetic field, H, is of order 100 K, decreasing with increasing n; (c) the polaron hopping energy decreases linearly with increasing H. In simple terms, the field-dependent transport is due to the variation of the quantity $|E_c(H) - E_F(0)|$ with increasing H.

Just as it is possible to tune through the metal-insulator transition in Si:P with stress,[7] a similar effect can be accomplished with field in the present experiment. We shall demonstrate that the transition is continuous at temperatures as low as 54.8 mK and that the conductance σ varies linearly with applied magnetic field H above a critical field H_c. We also show that this linearity in H implies a linear dependence on energy near the mobility edge E_c. This result is in agreement with recent scaling theories.[4,11]

In the following, we describe the results of magnetoresistance measurements on two samples, described in Table I, in which the Fermi energy, E_F, is very close to, but below, E_c in the absence of an applied field. Small single crystals were shaped to approximate parallelepipeds and four 2.5×10^{-3}-cm Cu leads were attached to the samples with indium for four-probe resistivity measurements. This procedure provided Ohmic contacts with an average experimental volume of $(1.7\pm 0.3)\times 10^{-4}$ cm^3, the large estimated error resulting from the size of the indium contacts defining the position of the voltage probes. The temperature dependence between 0.3 and 4 K of sample No. 1 is shown in Fig. 1 for selected fields close to the transition. The minimum metallic conductivity from Mott's formula, Eq. (1), with $a_E \lesssim (8\times 10^{19})^{-1/3}$ and $C\sim 0.005$ gives $\sigma_{min} \gtrsim 50$ $(\Omega\ cm)^{-1}$ and is indicated in the picture. Clearly the resistivity remains finite for $\rho(T=300$ mK) considerably above σ_{min}^{-1}. Only the curves for the lowest three fields do not appear to extrapolate to some finite values as $T\to 0$ ($1/T\to\infty$). Since our modest temperature interval (one decade) does not permit an accurate enough determination of the temperature dependence to extrapolate to $T=0$, we plot in Fig. 2 the magnetic field dependence of $\sigma(T=300$ mK) as rep-

TABLE I. Carrier concentration n (from Ref. 14) and vacancy content x of the two $Gd_{3-x}v_xS_4$ samples.

No.	x	n (cm^{-3})
1	0.321	$(2.5\pm 0.2)\times 10^{20}$
2	0.325	$(1.6\pm 0.5)\times 10^{20}$

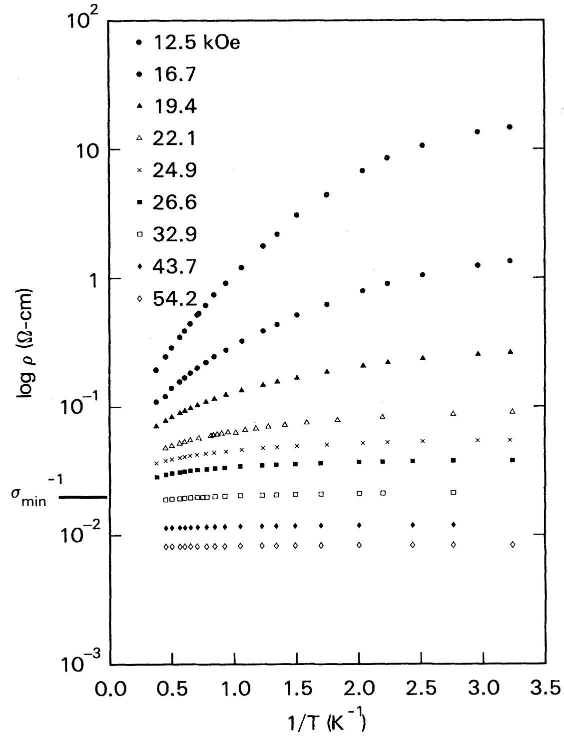

FIG. 1. Temperature dependence of magnetoresistivity of $Gd_{3-x}v_s S_4$ sample No. 1. An estimate for σ_{min}^{-1} is indicated and discussed in the text.

resentative of $\sigma(T=0)$. This is certainly a valid approximation for fields higher than ~25 kOe (see Fig. 2) and leads to the remarkable result that $\sigma(T=300$ mK) is linear in H for 25 kOe $< H < 55$ kOe, varying smoothly through the estimated values for σ_{min} with a dependence given by

$$\sigma(T=300\ \text{mK}) = A(H-H_c) = [3.4\pm0.7\ (\Omega\ \text{cm kOe})^{-1}] \times [H-19.0\pm0.1\ \text{kOe}].$$

The error in A again reflects the inaccuracy in estimating the distance between voltage probes. The results shown in Fig. 3 demonstrate that the effect is also observed in a more lightly doped sample No. 2.

Here, we have plotted measurements at three different temperatures, $T=55$, 94, and 550 mK. Clearly the curve is not altered between 55 and 94 mK. This demonstrates that temperature-dependent effects are negligible in defining the continuous linear dependence

$$\sigma(T=55\ \text{mK}) = [4.50\pm0.1\ (\Omega\ \text{cm kOe})^{-1}] \times (H-39.5\pm0.5\ \text{kOe})$$

between 40.7 and 62.4 K and for $0.1\sigma_{min} \leq \sigma \leq 2\sigma_{min}$.

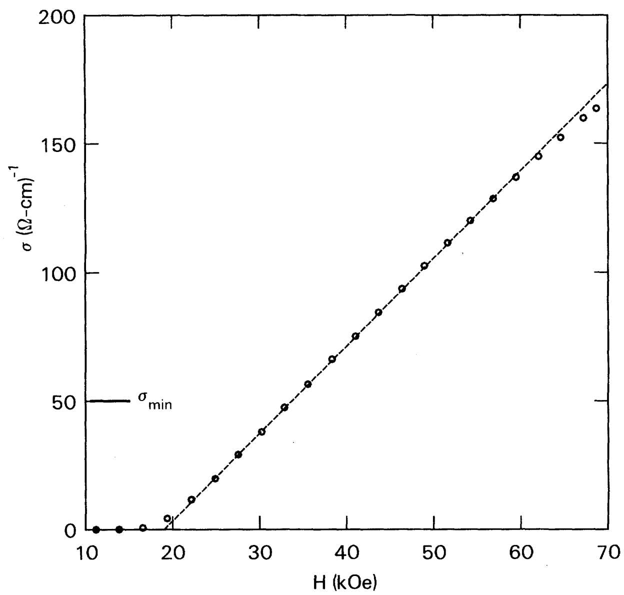

FIG. 2. Magnetic field dependence of conductivity of $Gd_{3-x}v_x S_4$ sample No. 1 at $T = 300$ mK. The fitted line obeys the equation

$$\sigma = [3.4\ (\Omega\ \text{cm kOe})^{-1}]\ [H-19.0\pm0.1\ \text{kOe}].$$

Considerable deviations from linearity are, however, observable near and below 50 kOe in the 550-mK curve. This thermal smearing also may account for part of the rounding in Fig. 2 near H_c although our experience with other samples of the more highly doped material indicate that inhomogeneities also contribute (the rounding below 40.7 kOe in the 55-mK data of Fig. 3 may be due to this effect). Deviations from linearity

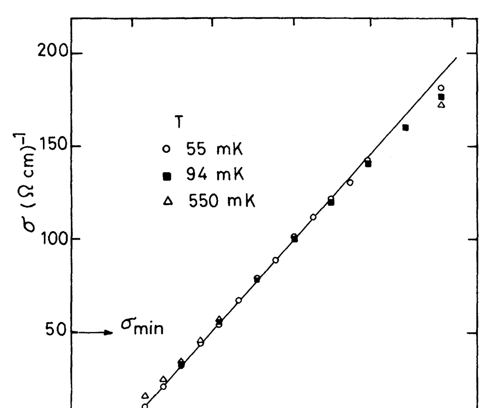

FIG. 3. Magnetic field dependence of conductivity of $Gd_{3-x}v_x S_4$ sample No. 2 at $T = 55$, 94, and 550 mK. The fitted line obeys the equation

$$\sigma = [4.5\ (\Omega\ \text{cm kOe})^{-1}]\ [H-39.5\pm0.5\ \text{kOe}].$$

in H above ~60 kOe in both Figs. 2 and 3 are due to saturation effects where magnetization is no longer proportional to H.[20]

The relationship $\sigma(T\to 0)=A(H-H_c)$ is strongly reminiscent of Eq. (2) with $\nu=1$. A direct comparison requires, however, a knowledge of the functional dependence of E_c upon H. It is worthwhile to reiterate at this point that in the present experiment n is fixed. In our view H reduces the magnitude of the potential fluctuations and, therefore, $|E_c-E_F(0)|$. The functional dependence of $|E_c-E_F(0)|$ on H in the metallic range is not known from direct experiment. We note, however, the following: For sample No. 2, $E_F(0)$ ~ 2000 K; the hopping energy ΔE for this sample is given in Ref. 15 to be 70 K and we see from the present data, Fig. 3, that this energy corresponds to $H_c \simeq 40$ kOe. Thus, the linear behavior which extends over roughly 25 kOe in sample No. 2 represents only a fractional energy change of about 2% of $E_F(0)$. Furthermore, it is known from Ref. 15 that, on the insulating side, $|E_c-E_F(0)|$ is linear in H for fields in excess of approximately 6 kOe. We consequently resort to the scaling results[4] according to which $|E_c-E_F(0)|$ varies smoothly through the transition and argue that also for $E_F(0)-E_c>0$, from Eq. (2)

$$\sigma(T=0)\propto[E_F(0)-E_c]^\nu \equiv A(H-H_c)^\nu. \qquad (4)$$

Therefore, $\nu=1$ observed in the present magnetoresistance measurements may be compared directly to the exponents derived from the scaling theories.

In conclusion, it has been demonstrated that the metal-insulator transition in the magnetic semiconductor $Gd_{3-x}v_xS_4$ is continuous and obeys a critical law, Eq. (4), with $\nu=1$. These results would appear to disagree with the predictions of Mott and Kaveh[17] for compensated semiconductors. However, conduction-band degeneracy which can mask a discontinuous transition[17] cannot be ruled out. Finally, although the physical arguments resulting in Eq. (4) are based strictly on concepts of localization, we cannot, at present, distinguish between localization[4,5] and interaction effects in the presence of a magnetic field.[11]

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