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Hall effect at a tunable metal-insulator transition

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Using a rotating magnetic field, the Hall effect in three-dimensional amorphous Gd_xSi_{1-x} has been measured in the critical regime of the metal-insulator transition for a constant total magnetic field. The Hall coefficient R_0 is negative, indicating electronlike conductivity, with a magnitude that increases with decreasing conductivity. R_0 diverges at the metal-insulator transition, and displays critical behavior with exponent -1 $[R_0 \sim (H-H_C)^{-1}]$. This dependence is interpreted as a linear decrease in the density of mobile carriers $n \sim R_0^{-1} \sim H-H_C$, indicative of the dominant influence of interaction effects.

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Investigation of the electronic properties at the metal-insulator transition (MIT) is a topic of current interest because the MIT displays characteristics of a $T=0$ K quantum phase transition.[1] To probe the MIT in the critical regime, it is advantageous to be able to control the transition in a continuous way. Most previous studies have examined a series of samples of varying concentration spanning the MIT, introducing uncertainties due to possible inhomogeneity or irreproducibility in the sample preparation process. An elegant solution to these shortcomings arises from using a single sample, which is controlled by an external parameter such as stress,[2] magnetic field[3] or illumination,[4] thereby substantially improving the quality of transport data and allowing measurements of, e.g., the electronic density of states by electron tunneling,[5] or the dielectric constant.[6]

The issue addressed in the present work is the critical behavior of the density of mobile carriers as measured by the Hall effect. While it has already been shown from tunneling experiments that the single-particle density of states vanishes at the MIT,[5] in a correlated electron material such as exists at the MIT, the mobile carrier density is distinct from the single particle electron density of states. According to Altshuler and co-workers,[7] a system that is dominated by electron-electron (e-e) interactions should exhibit critical behavior for the Hall coefficient R_0. Fukuyama has suggested[8] that corrections due to localization should leave R_0 unchanged, a result consistent with calculations (in the absence of electron interactions) by Shapiro and Abrahams.[9] In the two-dimensional (2D) disorder-induced MIT, it has been found theoretically that the Hall coefficient is critical with exponent -1.[10] The material used in this study has a large carrier concentration (it is whether these carriers are mobile or not that we are investigating) at the MIT and therefore should exhibit strong e-e interactions and localization effects. Previous studies of the Hall effect near the MIT for crystalline Ge:Sb and for amorphous Kr-Bi alloys show a divergent Hall coefficient R_0, with a coefficient of -0.7; other work, however, suggests that R_0 is nondivergent.[11–14] We note that in all previous studies, the Hall effect was measured by applying a variable magnetic field, which has the unavoidable consequence of altering the conductivity, even in nominally nonmagnetic systems. This affects the occupation and hence the interactions of the highly correlated states especially near the MIT, possibly leading to the differing results discussed above. In the present work, we use a rotating magnetic field and take advantage of the large negative magnetoresistance of these alloys to measure the Hall effect *at a constant magnetic field*. By this means, we are able to measure R_0 and conductivity simultaneously and continuously through the MIT. It is the goal of this study to determine if R_0 is critical in the presence of both e-e interactions and localization, and to draw conclusions about the dependence of the carrier concentration (as determined by the Hall effect) on the tuning parameter driving the MIT.

The three-dimensional system a-Gd_xSi_{1-x} can be reversibly tuned through the MIT by application of a magnetic field H.[15] In this system, we have measured transport conductivity,[15,16] density of states,[5] magnetization,[17] optical properties,[18] and specific heat[19] in the critical regime. The transport conductivity and the density of states at fixed H on the metallic side of the MIT were shown to vary as $T^{1/2}$ and $E^{1/2}$, respectively, the same dependence as for nonmagnetic systems,[1] with $T=0$ and $E=0$ offsets that depend on H. To study the dependence of the mobile carrier concentration n on the magnetic field H, we measure the Hall coefficient for *fixed* values of H at which the system is well into the metallic state to fixed values of H at which the system is deep into the quantum critical regime. Since the goal is to study the n dependence on H, we cannot use the conventional Hall geometry to determine R_0. Instead, as described in more detail below, we work at constant magnitude of H (hence constant n and magnetization M) and vary the angle between H and the sample plane to obtain R_0. Because of the magnetic nature of the dopant atom Gd ($S=7/2$), we must also consider the contribution of the anomalous Hall effect to this measurement. For reasons to be discussed below in the context of the data obtained in this experiment, we think that the anomalous contribution is significantly smaller than the ordinary Hall effect near the MIT.

There is significant debate as to what constitutes the critical regime. In three dimensions, the Ioffe-Regel[20] limit ($k_F l \approx 1$; where k_F is the Fermi wave vector, l is the mean free path) results in a conductivity, $\sigma = ne^2/(\hbar k_F^2)$ (n is the electron concentration, e is the electron charge, and $\hbar$ is the Planck's constant), below which a classical Fermi-liquid description fails to make sense. This conductivity is material dependant (due to $n/k_F^2 \propto n^{1/3}$) and may be considered a phe-

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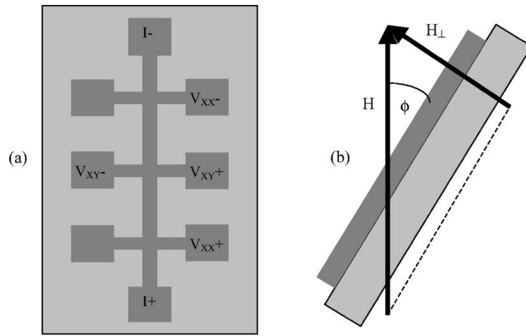

FIG. 1. (a) Schematic top view of sample, (b) Schematic on-edge view of sample rotated at angle ϕ with respect to the applied magnetic field H. $H_\perp = H \sin \phi$ is the magnetic field contributing to the measurement of the Hall coefficient.

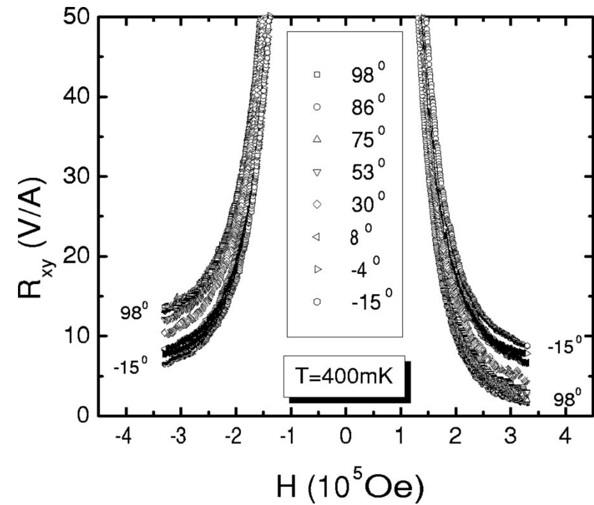

FIG. 2. Transverse resistance R_{xy} vs applied magnetic field H for various angles ϕ.

nomenological upper limit for the conductivity σ of the critical regime. The MIT in Gd_xSi_{1-x} occurs for $x = 0.14$, with an electron concentration that is orders of magnitude larger than that found in crystalline doped semiconductor systems such as Si:P (for Gd_xSi_{1-x}, this electron concentration may be estimated as between 4×10^{20} cm^{-3} from optical absorption measurements[18] and 2×10^{22} cm^{-3} from the Rutherford backscattering spectroscopy measured Gd concentration, assuming three donated electrons per Gd). Consequently, the Fermi temperature and the Ioffe-Regel limit on the conductivity in Gd_xSi_{1-x} is significantly enhanced; taking $n \approx 10^{22}$ cm^{-3} leads to a conductivity at the Ioffe-Regel limit ($\sigma \sim 500\ \Omega^{-1}$ cm^{-1}), about 40 times larger than the equivalent value in Si:P,[2] and allows us to experimentally probe the MIT deeper into the critical regime. Moreover, a high E_F allows for a valid probing of the critical regime at a higher temperature, since the effective temperature T/E_F is substantially reduced compared with the previous studies. In the present study, data have been acquired at $T = 400$ mK (ten times lower than previous data on amorphous systems). Temperatures 10-100 times smaller would be needed to obtain similar effective temperatures for crystalline doped semiconductors.

Measurements were made using Hall bars as shown in Fig. 1(a). These were fabricated using a lift-off technique. A film of ~200 nm of Cu was deposited on a SiN-covered Si surface, lithographically patterned, and etched into an inverse Hall bar pattern. Onto the resulting inverted Cu pattern ~100 nm of a-Gd_xSi_{1-x} was deposited, with $x \sim 0.14$, using electron-beam coevaporation of Gd and Si in an ultrahigh vacuum deposition system. The remaining Cu was then etched in a $FeCl_3$ solution, leaving the desired a-Gd_xSi_{1-x} Hall bar. The Hall bars have a current carrying strip and three pairs of voltage probes at three different locations along the strip, allowing for simultaneous measurements of the longitudinal ($R_{xx} = V_{xx}/I$) and the transverse ($R_{xy} = V_{xy}/I$) resistance. All resistances are measured using a low-frequency ($f < 30$ Hz) ac method.

Samples were measured in a ^{3}He cryostat at $T = 400$ mK in applied magnetic fields H to 33 T at the National High Magnetic Field Lab in Tallahassee, Florida. In order to measure R_0 in a system, where there is a magnetoresistance and n may depend on H, we developed a technique to measure R_0 in constant H by adjusting the *angle* ϕ between H and the sample plane *in situ* using a rotating stage, as shown in Fig. 1(b). We have experimentally verified that the change in longitudinal resistance R_{xx} of three-dimensional a-Gd_xSi_{1-x} is independent of the direction of H.[21] The transverse voltage due to the Hall effect, however, depends on the perpendicular magnetic field $H_\perp = H \sin \phi$ [see Fig. 1(b)]. Thus, by measuring R_{xy} in fixed H at different angles ϕ, the Hall coefficient can be determined. In all figures, the field shown is the applied field, not corrected for demagnetization that is negligible compared to the fields shown in this paper, reaching a theoretical maximum of less than 5.6 kOe.

Figure 2 shows the measured R_{xy} vs H for various angles ϕ. Because of the extremely large magnetoresistance, a small misalignment of voltage leads will cause the measured R_{xy} to contain a component of R_{xx}. R_{xx} is, however, symmetric in H, while R_{xy} is antisymmetric. In order to extract the Hall-effect contribution, we determine the antisymmetric (odd) component of the data in Fig. 2, $R_{xy,odd}(H) = 1/2[R_{xy}(H) - R_{xy}(-H)]$. We experimentally confirmed that the even component was indeed proportional to the previously measured $R_{xx}(H)$ and gave a geometric factor for the voltage lead misalignment, which was well within the size of the leads.

Figure 3 shows the odd component of the transverse resistance, $R_{xy,odd}$, versus H for various angles ϕ. For $H < H_C \sim 100$ kOe, the sample is insulating and the measurement breaks down. As expected, at angles close to $\phi = 0°$, the odd contribution vanishes since the Hall effect disappears for H parallel to the film. For larger ϕ, however, we find a significant odd contribution. For positive H, the voltage due to the Hall effect in the data close to $\phi = 90°$ is negative, which indicates that R_0 is negative (electronlike). We note that the largest odd contribution occurs for $\phi = 86°$ and the magnitude of the contribution declines from that value at $\phi = 98°$ as may be expected, as the maximum should occur at $\phi = 90°$. Data at constant H but different angles (indicated

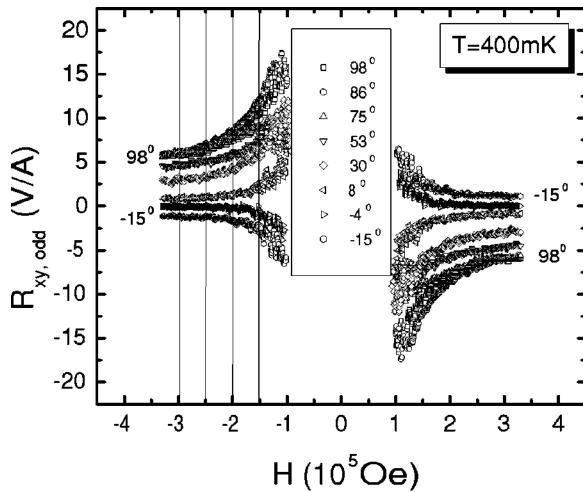

FIG. 3. Odd component of the transverse resistance, $R_{xy,odd}$, versus H for various angles ϕ.

by the vertical lines) yield $R_{xy,odd}$ vs $H_\perp = H \sin\phi$.

To determine the central result, the Hall coefficient R_0, $R_{xy,odd}$ determined in a manner described in the preceding paragraph, is plotted versus $H_\perp$ for various H in Fig. 4. The Hall coefficient R_0 is related to the slope $\Delta R_{xy,odd}/\Delta H_\perp$ according to $R_0 = t\Delta R_{xy,odd}/\Delta H_\perp$, where $t=100$ nm is the sample thickness. As H is reduced and the material approaches the insulating regime, we observe a concomitant increase in R_0.

In a magnetic metallic system, the Hall resistivity is expressed as

$$R_{xy,odd}\,t = R_0 H_\perp + R_S M_\perp, \tag{1}$$

where $M_\perp$ is the perpendicular component of magnetization. The first term describes the ordinary Hall effect with $R_0 = (ne)^{-1}$ (e is the electron charge). The second term is the anomalous Hall effect, a result of interactions with magnetic

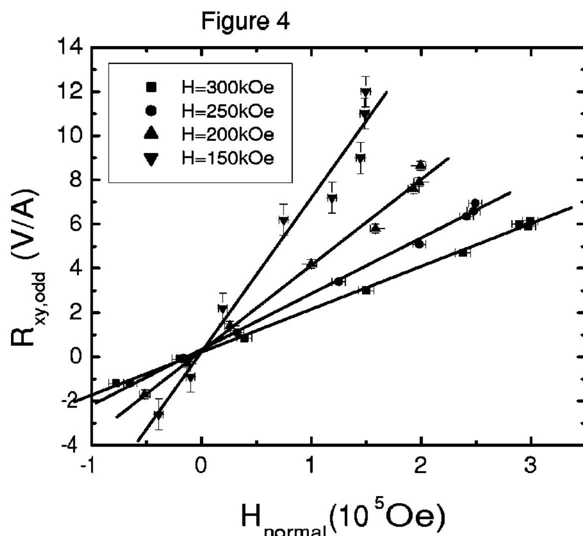

FIG. 4. Odd component of the transverse resistance, $R_{xy,odd}$, versus normal magnetic field $H_\perp$ for various applied magnetic fields H.

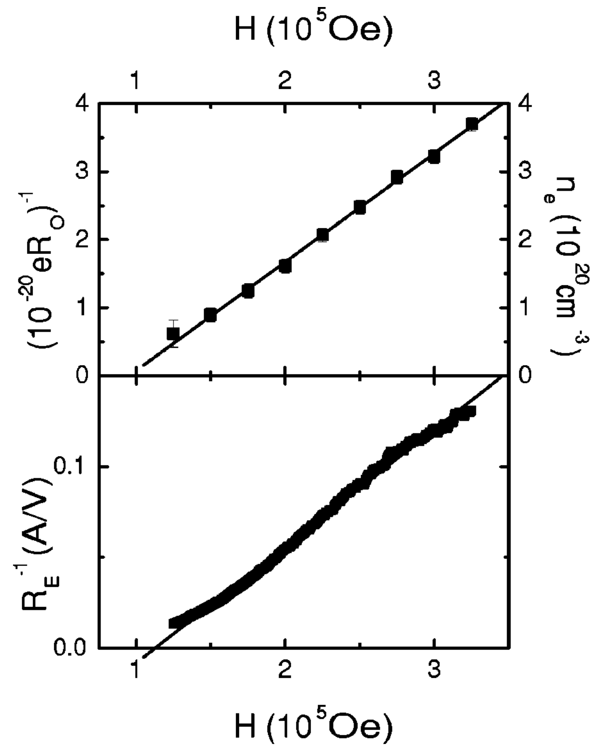

FIG. 5. Top: Inverse Hall coefficient (in units of e) and electron concentration $n_e = (te\Delta R_{xy,odd}/\Delta H_\perp)^{-1}$ vs H. Bottom: Inverse of $R_{xy,even}$ vs H. $R_{xy,even}(H) = 1/2[R_{xy}(H) + R_{xy}(-H)]$ is due to the (tiny) misalignment of transverse voltage probes and provides a simultaneous measurement that is proportional to the longitudinal magnetoresistance $R_{xx}(H)$. Both sets of data are linear and go to zero ($R_{xy,odd}$ and $R_{xy,even}$ divergent) for $H = 100$ kOe.

moments, with the anomalous Hall coefficient R_S.[22] Since there is no magnetic anisotropy in these samples, $M_\perp = M \sin\phi = (M/H) \times H \sin\phi = (M/H) \times H_\perp$, so Eq. (1) can be rewritten as

$$R_{xy,odd}\,t = (1/ne + R_S M/H) H_\perp. \tag{2}$$

At a fixed value of total field, H, n, R_S, M, and hence M/H are constant. Thus, both terms give a linear dependence on $H_\perp$ *for data taken in constant* H, as seen in Fig. 4. The dependence of R_0 ($\propto 1/n$) and R_S on field H is the issue to be determined.

Figure 5 shows the measured inverse Hall coefficient in units of $(e\Delta R_{xy,odd}t/\Delta H_\perp)^{-1}$ versus H for 100 kOe $< H <$ 330 kOe, as well as the inverse of $R_{xy,even}(H) = 1/2[R_{xy}(H) + R_{xy}(-H)]$, which as previously discussed is proportional to the longitudinal magnetoresistance $R_{xx}(H)$. Both sets of data are linear and go to zero ($R_{xy,odd}$ and $R_{xy,even}$ divergent) for $H = 100$ kOe, the MIT previously found from conductivity measurements for this sample. The linear dependence of the longitudinal conductivity on field was shown and discussed in Ref. 15. By assuming that $R_{xy,odd}$ is dominated by the ordinary Hall effect near the MIT, we have a linear dependence of the concentration of

mobile carriers, n, on the applied magnetic field H, the driving parameter of the transition, with values as shown in Fig. 5.

We now argue that these results are dominated by the ordinary Hall effect and not by the anomalous Hall effect. For metallic systems, where n is large, the anomalous Hall coefficient dominates, as experimentally observed by Gambino and McGuire for a-Gd_xGe_{1-x} alloys.[23] They found $R_S \approx 5\times10^{-10}$ Ω cm/G (corresponding to $n \approx 2.8\times10^{22}$ cm^{-3}) and $R_0 \approx -2.2\times10^{-12}$ Ω cm/G for $x=0.3$, still well on the metallic side of the MIT, i.e., the ordinary Hall coefficient is $\approx 6\%$ of the anomalous Hall coefficient. For concentrated conventional magnetic metals, the anomalous Hall effect is even more dominant, typically a factor of $10-100$ larger than the ordinary Hall effect ($R_SM/R_0H \sim 10-100$). In the present experiment, however, two factors drastically reduce this ratio: both n and M are greatly decreased from the concentrated state. In the limit of $n \to 0$, $R_0 \to \infty$. The dependence of R_S on n is not clear; assuming the anomalous Hall effect is dominated by skew scattering, as is likely,[24] $R_S \propto n^{2/3}$ in a free-electron picture, hence R_S is not divergent. Magnetization M vs H has been measured on these alloys to 250 kOe, using the magneto-optic Kerr effect.[25] Using this data, and an assumption of $R_S \propto n^{2/3}$, we have the ordinary Hall effect dominating the anomalous Hall effect by several orders of magnitude for the range of data shown. Furthermore, it would be difficult to conceive of a model in which $(R_SM/H)^{-1}$ was linear in H near the MIT (i.e., $1/R_SM$ constant in H), requiring a peculiar cancellation of the separate strong dependences of M and R_S on H. We therefore conclude that the natural and self-consistent solution is that at the MIT the ordinary Hall effect R_0 dominates, leading to the conclusion that the number of mobile carriers (n) depends linearly on the driving parameter in the quantum critical regime, $(H-H_C)$.

In conclusion, while the precise conditions for an experimental realization of a system in the critical regime of a quantum critical transition are under debate,[2] we have investigated a tunable system with conductivities up to two orders of magnitude below the Ioffe-Regel limit.[20] In this system, we have measured transport,[15,16] tunneling,[5] magnetization,[17] optical conductivity,[18] and specific heat.[19] Adding to this body of information, we have shown here that the Hall coefficient R_0 is a critical quantity, with critical exponent -1 $[R_0 \sim (H-H_C)^{-1}]$. This is in agreement with theoretical expectation for a system dominated by e-e interaction.[7] Interpreting R_0 as dominated by the ordinary Hall effect, we find that the number of mobile carriers, $n \sim R_0^{-1}$, depends linearly on the driving parameter of the transition, $(H-H_C)$. In the data presented here we do not see any evidence for the anomalous Hall effect,[22] an observation we can understand by considering the low mobile carrier concentration and the reduced density of magnetic scatterers of Gd_xSi_{1-x} at the MIT.

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