Magnetoconductivity near the percolation threshold in granular Sn:Ar mixtures

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We have measured for the first time to our knowledge the magnetoconductivity $\Delta\sigma$ on the metallic side of the metal-insulator transition in a three-dimensional percolating system as a function of the metal volume fraction v. The magnetoconductivity vanishes at the critical metal volume concentration $v_c = 0.26 \pm 0.01$ according to $-\Delta\sigma \propto (v - v_c)^t$ with a critical exponent $t = 1.90 \pm 0.15$ which is slightly higher than the conductivity exponent. The relative magnetoresistivity scales as $\Delta\rho/\rho \propto (v - v_c)^{0.3}$. This behavior is in contradiction to theory which predicted $\Delta\rho/\rho$ to stay constant in the critical region.

Metal-insulator transitions (MIT) in three-dimensional (3D) percolating systems have been mostly investigated by measurements of the transport properties. It is well established that the behavior of the conductivity σ can be described by a power law $\sigma \propto (v - v_c)^t$, where v is the actual metal volume fraction, v_c is the critical metal volume fraction at which the MIT occurs, and t is the critical exponent of the conductivity. Experimental results gave values for the critical exponent t in the range of $t = 1.6$–1.9 (Ref. 1) in agreement with theoretical predictions.[2] Recent Hall-effect data show that the Hall constant R_H diverges at the MIT with $R_H \propto (v - v_c)^{-g}$ where $g = 0.4$–0.5.[3,4] This is in agreement with theoretical models which predicted the critical exponent of the Hall constant to be in the range of $g = 0.3$–1.0.[5–7]

Whereas the MIT in percolating systems seems to be well described in terms of the two magnetotransport parameters σ and R_H, no data are available on the behavior of the magnetoconductivity $\Delta\sigma(B)$ or the relative magnetoresistivity $\Delta\rho(B)/\rho$ approaching the MIT.

Within this article we want to report the first systematic study of the dependence of magnetoconductivity $\Delta\sigma(B)$ and magnetoresistivity $\Delta\rho(B)/\rho$ on conductor volume fraction in a 3D percolating system, namely the granular-metal–rare-gas mixture Sn:Ar. Our results can be compared with a theory recently developed by Bergman for the case of a random mixture of a good and a bad conductor.[8]

Measurements of the longitudinal voltage U_{xx} as a function of the external magnetic field were performed on films with a thickness of 800–1000 Å. The metal was evaporated out of a resistance heated Ta crucible and condensed simultaneously with the rare gas on a sapphire substrate held at a temperature of $\simeq 5$ K. Au electrodes, as contact areas ($\simeq 800$ Å thickness), were evaporated on the substrate before each measurement at room temperature.[3] Cu wires were attached to the Au areas using In as solder. Condensation rates and the thickness of the films were controlled by a quartz-crystal oscillator. Since the Sn:Ar films were unstable against heating above 30 K, all measurements were performed *in situ* in a temperature range of 4.2–10 K. After preparation of the film the sample holder was brought into a field of a superconducting split-coil magnet ($B \leq 5$ T). Since the Sn:Ar films were superconducting ($T_c \simeq 4.5$ K) with the exception of the immediate vicinity of the percolation threshold,[5] the films had to be heated above the transition temperature T_c during the measurement procedure. The longitudinal voltage U_{xx} as a function of the external magnetic field was determined by a dc method. An evaporating mask was used to obtain a certain geometry of the film, i.e., a defined length to width ratio which was about 4:1. This ratio is sufficient to avoid geometrical contributions to the magnetoresistance.[9] All measurements were made in the low-field regime ($\omega_c \tau \ll 1$) with a linear behavior of R_H as a function of the magnetic field and a relative change of the longitudinal conductivity $\Delta\sigma(B)/\sigma \leq 10^{-2}$ at a field of 3 T.

The behavior of the field-induced change of the conductivity $\Delta\sigma(B = 2\text{ T})$ as a function of the distance from the critical metal volume fraction $v_c = 0.26$ is shown as a double logarithmic plot in Fig. 1. The straight line in Fig. 1 is a result of a least-squares fit to the data with $-\Delta\sigma \propto (v - v_c)^{1.9}$ with an uncertainty in the exponent of ± 0.15. This value of the critical exponent of $\Delta\sigma(B)$ is slightly higher than that of the conductivity exponent t with $t = 1.6 \pm 0.1$ in Sn:Ar.[4,10] A more sensitive representation of the experimental data is shown in Fig. 2. The relative magnetoresistivity as a function of the distance from v_c decreases with decreasing metal volume fraction according $\Delta\rho/\rho \propto (v - v_c)^{0.3}$. This experimental results may be compared with a theory recently developed by Bergman[8] in which scaling arguments are used to predict the behavior of the magnetoconductivity $\Delta\sigma(B)$ and the relative magnetoresistivity $\Delta\rho/\rho$ at the percolation threshold in a random mixture of a good conductor with a conductivity σ_M and a bad conducting material with a conductivity σ_I. In the region above the threshold $v > v_c$ the effective magnetoconductivity $\Delta\sigma_e$ in such a system is given by the following equation:

$$\Delta\sigma_e = (\Delta\sigma_{M\perp} + \Delta\sigma_{M\parallel})(v - v_c)^t + (\Delta\sigma_{I\perp} + \Delta\sigma_{I\parallel})(v - v_c)^{-s} + \frac{(\lambda_M - \lambda_I)^2}{\sigma_M}, \quad (1)$$

where λ_M and λ_I are the Hall conductivities of the two components. The indices $\perp$ and $\parallel$ denote the direction of current flow (transverse $\perp$ and longitudinal $\parallel$) with respect to the magnetic field. In addition to the conduc-

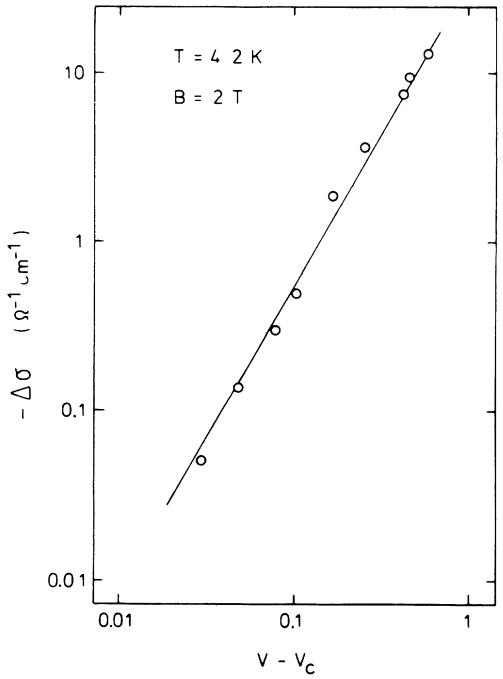

FIG. 1. Magnetoconductivity $\Delta\sigma$ as a function of the distance from the critical metal volume fraction $v_c = 0.26$ at a magnetic field of 2 T and a temperature of 4.2 K. The solid line is a result of a least-squares fit to the data.

tivity exponent t and the critical exponent s, which is correlated to the correlation length exponent $\nu(3D) = s = 0.84$,[11] a new critical exponent $t_{M\perp}$ is introduced by this theory which describes the critical behavior of the second-order Hall contribution to the low-field magnetoconductivity. The value of $t_{M\perp}$ is predicted to lie in the range of $1.3 \leq t_{M\perp} \leq 2.0$.

In the case of the metal rare-gas mixture Sn:Ar the second term of Eq. (1) vanishes since condensed Ar is an ideal insulator with $\sigma_I = \Delta\sigma_I = \lambda_I = 0$. The prefactor λ_M^2/σ_M in the third term is of the order of 10^{-3} $(\Omega\,\mathrm{cm})^{-1}$ whereas the value of the magnetoconductivity of pure Sn is $\Delta\sigma_M \approx 100$ $(\Omega\,\mathrm{cm})^{-1}$. Therefore, the third term in Eq. (1) is negligible with respect to the first term. Thus, the effective magnetoconductivity $\Delta\sigma_e$ should exhibit the same critical behavior as the normal conductivity σ. The value of the critical exponent $t = 1.6 \pm 0.1$ determined in the conductivity experiment is slightly below that value of $t = 1.90 \pm 0.15$ resulting from the measurement of the magnetoconductivity but it is nevertheless in the range of $1.6 \leq t \leq 2.0$ as predicted by percolation theory.[2] Due to the special properties of Sn:Ar, i.e., the very low value of the ratio λ_M^2/σ_M in comparison to $\Delta\sigma_M$, it is not possible to determine the new critical exponent $t_{M\perp}$.

The behavior of the transverse relative magnetoresistivity can be calculated by the following equation:

$$\frac{\Delta\rho_e}{\rho_e} = -\frac{\Delta\sigma_e}{\sigma_e} - \frac{\lambda_e^2}{\sigma_e^2} = -\frac{\Delta\sigma_M}{\sigma_M} - \frac{\lambda_M^2}{\sigma_M^2}(v - v_c)^{2t-g}. \tag{2}$$

Since the value of t is larger than g and $\lambda_M^2/\sigma_M^2 \ll \Delta\sigma_M/\sigma_M$ the second term in Eq. (2) is negligible. In this case the relative magnetoresistivity should exhibit a constant behavior since the magnetoconductivity $\Delta\sigma_e$ and the conductivity σ_e tend to zero with the same critical exponent t at the MIT. This prediction of the theory is not in agreement with the experimental $\Delta\rho/\rho$ data in Sn:Ar shown in Fig. 2. The relative magnetoresistivity $\Delta\rho/\rho$ as a function of the distance from the MIT tends to zero according to $\Delta\rho/\rho \propto (v - v_c)^{0.3}$ as a consequence of the critical behavior of the magnetoconductivity $\Delta\sigma$ and the conductivity σ, respectively. This discrepancy between experiment and theory is possibly due to the fact that the theory is made for random composite systems whereas the Sn:Ar mixture has a granular nature in which islands of crystalline Sn are surrounded by an Ar matrix

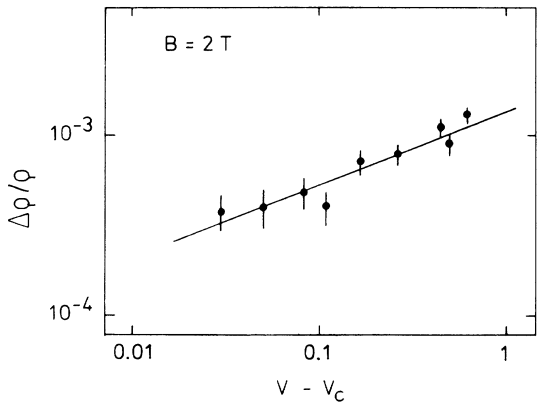

FIG. 2. Double-log plot of the relative magnetoresistivity $\Delta\rho/\rho$ as a function of the distance from the MIT at a magnetic field of 2 T and a temperature of 4.2 K. The solid line is a result of a least-squares fit to the data.

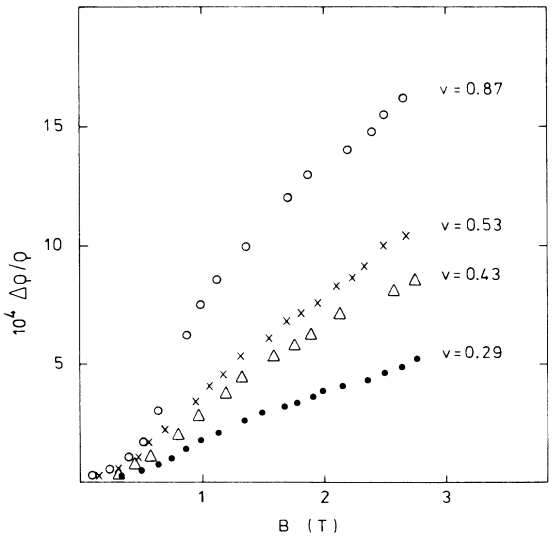

FIG. 3. Relative magnetoresistivity $\Delta\rho/\rho$ as a function of the external magnetic field at various Sn volume fractions v.

which is crystalline on a local scale but strongly disturbed.[12,13] This arrangement of the Sn clusters in the insulating matrix shows no self-duality, i.e., metallic and insulating regions can be distinguished by topological reasons in contradiction to the assumption made in theory. This may lead to a reduction of the critical exponent of the conductivity,[14] whereas the critical exponent of the magnetoconductivity seems to be unaffected.

The behavior of the relative magnetoresistivity $\Delta\rho/\rho$ as a function of the magnetic field for various Sn concentrations is shown in Fig. 3. Up to a magnetic field of 1 T the magnetoresistivity is proportional to B^2 which is the typical low-field behavior in metals.[15] At fields larger than 1 T there is a crossover to a linear slope $\Delta\rho/\rho$ as a function of B. This behavior cannot be explained in terms of a low-field ($\omega_c\tau \ll 1$) to high-field ($\omega_c\tau \gg 1$) crossover which is observed in pure metals[16] due to the influence of closed orbits[17] since the low-field condition is always fulfilled in Sn:Ar.

In conclusion we have shown that the relative magnetoresistivity $\Delta\rho/\rho$ scales with $\Delta\rho/\rho \propto (v - v_c)^{0.3}$ at the percolation threshold in Sn:Ar in contradiction to theory[8] which predicted a constant behavior of $\Delta\rho/\rho$. Consequently the critical exponent of the magnetoconductivity $\Delta\sigma$ is slightly higher than the conductivity exponent. This discrepancy between theory and experiment may be caused by the granular nature of Sn:Ar, i.e., the lack of self-duality. Therefore, more experimental work is needed, especially for systems where the third term in Eq. (1), containing the new exponent $t_{M\perp}$, is not negligible. This may be the case in mixtures of semiconducting materials with insulators.

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