Hall effect in a three-dimensional percolation system

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We have studied the Hall effect in random mixtures of Al-Ge. The observed variation of the Hall coefficient over a wide range of concentration was found to be in full agreement with recent theoretical predictions for three-dimensional percolation systems. For the first time the critical behavior was studied near both percolation thresholds, namely, near the critical-volume concentrations of Al and of Ge. We observed a strong divergence of the Hall coefficient with exponent 3.8 ± 0.2 near the threshold of Al, and a much slower divergence with exponent 0.38 ± 0.05 close to the threshold of Ge.

The Hall effect in percolation systems near the transition threshold has been studied during the last ten years by several authors.[1-6] In a two-dimensional (2D) system, it is well understood that the Hall coefficient does not diverge at the threshold,[1-6] a prediction that has recently been confirmed experimentally.[7] In 3D systems, it has been predicted that the Hall coefficient diverges near the threshold as $(x-x_c)^{-g}$, values of the critical index g ranging from 0.9 to 8.3. Only some incomplete results are available,[8] and therefore the critical behavior of the Hall coefficient in 3D has not yet been fully established.

According to the theory of Bergman and Stroud[6] the effective Hall coefficient of a percolating *mixture* consisting of a good and a poor conductor is described by two terms:

$$R_e = AR_m(x-x_c)^{-g} + BR_i\left(\frac{\sigma_i}{\sigma_m}\right)^2 (x-x_c)^{-2t} \ . \quad (1)$$

$R_m(R_i)$ and $\sigma_m(\sigma_i)$ are the Hall coefficient and the conductivity of the good (poor) conductor, respectively. A and B are constants of order unity, and t is the conductivity critical index.

In this paper we report for the first time experimental results that confirm the existence of the two diverging terms in Eq. (1). Measurements of the Hall effect in an Al-Ge compound system allowed us to observe the variation of the effective Hall effect in agreement with the second term of Eq. (1). Furthermore, by chemical etching we have converted this compound into a one element (Ge) 3D percolation system, and measured its Hall coefficient. The results of these measurements are consistent with the first term in Eq. (1).

Equation (1) is based on an important result of Bergman, Kantor, Stroud, and Webman.[9] The Hall conductivities of two components λ_m, λ_i and that of the mixture λ_e in low field satisfy the relation

$$\frac{\lambda_e-\lambda_i}{\lambda_m-\lambda_i} = X\left(\frac{\sigma_i}{\sigma_m}\right) , \quad (2)$$

where X is a function only of the ratio of the Ohmic conductivity of the two components. Above x_c, the scaling function X is constant and the effective Hall conductivity diverges as $(x-x_c)^{\tau}$. The analogous scaling assumption for the Ohmic conductivity above x_c predicts a divergence of the Ohmic conductivity as $(x-x_c)^t$. Using the relation $R_k=\lambda_k/\sigma_k^2$ (for $k=m,i,e$) we can obtain Eq. (1), and also the relation $g=2t-\tau$. A more general expression for the second term of the right-hand side (rhs) of Eq. (1) is given by $BR_i(\sigma_i/\sigma_e)^2$, where σ_e is the effective conductivity of the medium. Equation (1) is obtained under the approximation $\sigma_e \simeq \sigma_m(x-x_c)^t$. Practically, this term becomes negligible when the ratio σ_i/σ_e is smaller than 10^{-3}. Then the effective Hall coefficient diverges more slowly than $(\sigma_i/\sigma_m)^2$ with the exponent $g=2t-\tau$.

Samples typically 2000 Å thick were prepared by vacuum coevaporation of Al and Ge on a hot substrate.[10] Figure 1 shows the diffraction pattern of the mixture which indicates that the Al and the Ge crystallines are randomly distributed. The crystalline size is of the order of a few hundred Å. Such grain size can be well described by classical percolation theory.[11]

The variation of the resistivity of the *mixture* with the Al volume fraction in the range 30%–60% is shown in Fig.

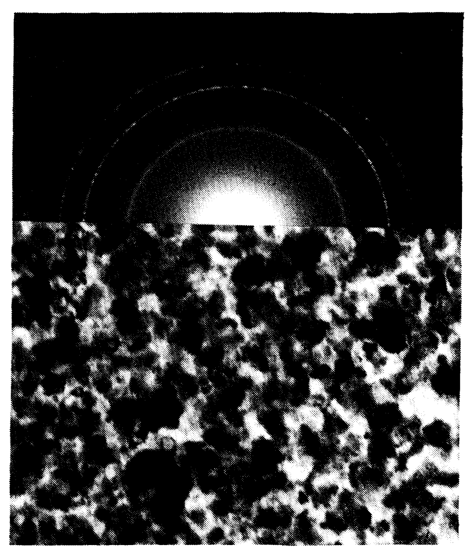

FIG. 1. The bright field and diffraction pattern micrographs of Al-Ge mixture.

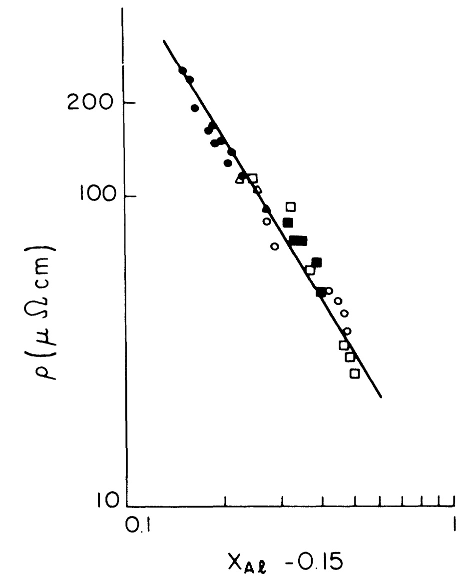

FIG. 2. Log-log plot of the effective resistivity ρ_e vs $x-0.15$ (here x is the Al vol% and $x_c=0.15$). The different symbols refer to different substrates.

2 on a log-log plot. Here we have chosen for x_c the bulk 3D value $x_{c3D}=15\%$.[12] The slope was calculated by a least-squares fit, giving $t=1.75\pm0.08$.

In the same samples we have performed Hall-effect measurements using the three-terminal method.[13] The sign of the Hall coefficient is positive, which indicates that the Ge has a dominant contribution in this range of con-

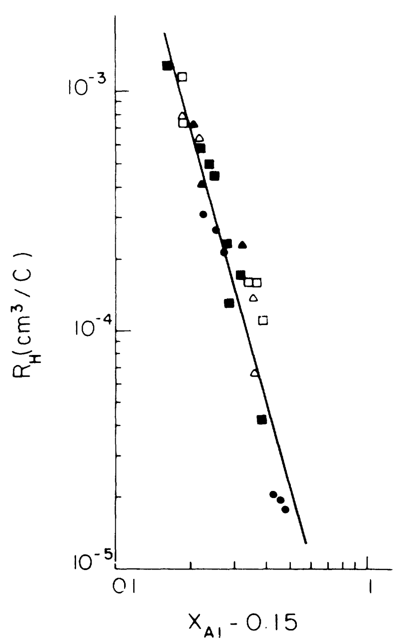

FIG. 3. Log-log plot of the effective Hall coefficient R_e vs $x-0.15$ (here x is the Al vol% and $x_x=0.15$). The different symbols refer to different substrates.

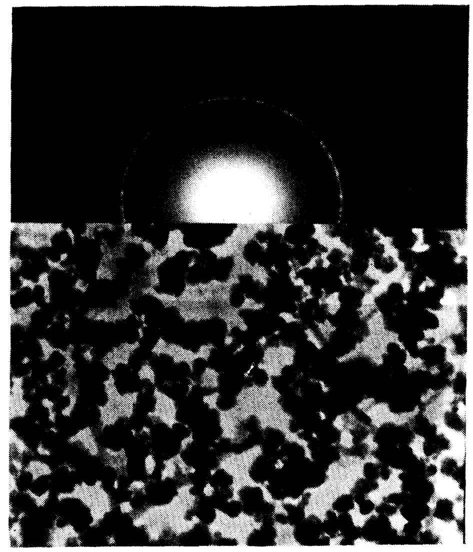

FIG. 4. The bright field and diffraction pattern of the Ge after the Al was etched out.

centrations. The log-log plot in Fig. 3 shows the volume fraction dependence of the effective Hall coefficient. From this plot we obtain a slope equal to 3.8 ± 0.2, which agrees with the exponent $2t$ in the second term of Eq. (1).

More generally, the ratio of the critical index of the effective conductivity of the mixture with that obtained from the effective Hall constant is approximately equal to 2. These results establish the existence of a term proportional to $(\sigma_e)^{-2}$ in the behavior of the Hall effect of the mixture.

As already pointed out, this term can dominate only if $\sigma_i/\sigma_e \geq 10^{-3}$. In order to measure the conductivity σ_i of the Ge, we etched out the Al from the mixture to obtain a one-element system containing only Ge. Figure 4 shows an Al-Ge sample after the Al was etched out. A comparison between the diffraction pattern of Fig. 1 and Fig. 4 shows that after the etching process the Al has indeed been eliminated. The resistivity of 3D Ge percolating

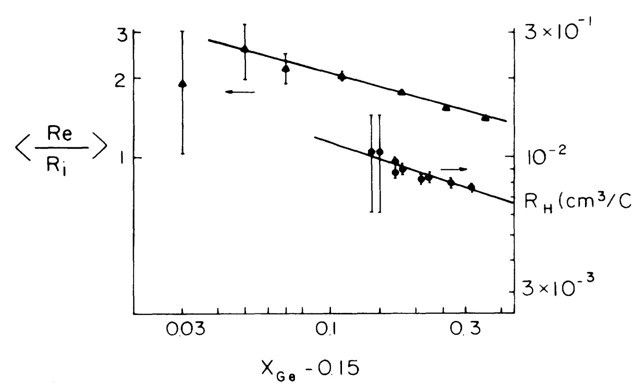

FIG. 5. Log-log plot of the Ge Hall coefficient vs $x-0.15$ (filled circles) (here x is the Ge vol%). The theoretical results from Ref. 9 are shown by filled triangles.

samples with 30–65 Ge vol% was measured. The lowest value of ρ_i obtained at $x=0.65$ is 1400 $\mu\Omega$ cm. Using the mean-field approximation we estimate the bulk value of $\sigma_i \simeq 1000\ \Omega\,\mathrm{cm}^{-1}$, indicating that the Ge is heavily doped. For $30\% < X_{\mathrm{Al}} < 40\%$ the condition on σ_i/σ_m is indeed fulfilled.

The formation of a one-element 3D system gives the opportunity to obtain the critical index g, since in this system the second term of Eq. (1) is equal to zero. The Hall effect measurements performed on these samples reveal a critical behavior of the Hall coefficient near x_c shown in Fig. 5. The slope was calculated by least-squares fit, giving $g=0.38\pm0.05$, and it is in the range of the values predicted by Bergman *et al.*[9] $g=0.29\pm0.05$. The value of R_i extracted from fitting the data to the first term in Eq. (1) is $R_i \simeq 1.3\times10^{-2}\ \mathrm{cm}^3\mathrm{C}$. [We assume that the coefficient A in the first term of Eq. (1) is equal to unity.]

Using the above values of R_i and σ_i, the measured values of R_e and σ_e, one obtains the value of the coefficient $B\simeq2$. Equation (1) is thus fully confirmed.

In conclusion, the relatively large conductivity of heavily doped Ge in an Al-Ge mixture and the possibility of etching out Al from the mixture allowed us to verify the behavior of the Hall coefficient of the mixture in a wide range of concentration. Equation (1) was confirmed in different limits. The critical index g was estimated and found to be compatible with that obtained by numerical simulations.

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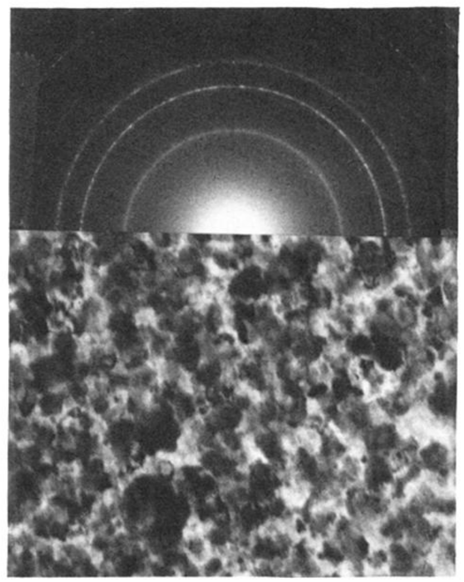

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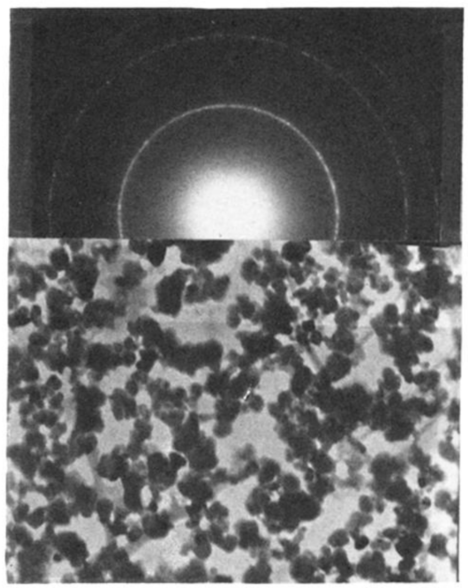

FIG. 4. The bright field and diffraction pattern of the Ge after the Al was etched out.