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Nonlinear Hall voltage in the hopping regime

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We report the observation of a nonlinear (quadratic) dependence of the Hall voltage V_H on magnetic field at small fields in the hopping regime. With increasing field the Hall coefficient, $R_H = V_H/IH$, of insulating compensated n-type CdSe increases approximately linearly, then remains roughly constant at a plateau value, and increases again at higher fields. No theory currently exists with which to compare this unanticipated behavior.

The existence of a Hall effect in insulating materials, where conduction occurs by hopping rather than by extended state conduction, was first considered in 1961 by Holstein,[1] who showed that quantum-mechanical interference between direct and indirect paths involving a third site can give rise to jump rates which depend on magnetic field and to a nonzero Hall voltage in the hopping regime. Subsequent theoretical work based on this concept has been carried out by a number of investigators.[2-7] The Hall coefficient is expected to be thermally activated, $R_H \propto \exp(KT^{-x})$, with a variable range, $R_{\mathrm{hop}} \propto T^{-x}$, with an exponent $x = \frac{1}{4}$ when the density of states which is constant near the Fermi energy,[6] and $x = \frac{1}{2}$ in the presence[6,7] of a parabolic "Coulomb gap" due to long-range Coulomb interactions.[8] Although Holstein[1] calculated a field-dependent transverse conductivity, σ_{xy}, no detailed theory is available that considers how the Hall coefficient R_H varies with field, and there has been no experimental evidence for such behavior at low fields.

Following earlier experimental work by Amitay and Pollak[9] and by Klein,[10] who failed to observe a Hall coefficient in insulating germanium and silicon with dopant concentrations 2 orders of magnitude below the critical concentration for the metal-insulator transition, Koon and Castner[11] have recently established the existence of a Hall effect in the hopping regime in Si:As samples with dopant concentrations much closer to the transition. Although they did not investigate the magnetic-field dependence in detail, they found a Hall coefficient which is different in different fields. Using appreciably lower magnetic fields, Roy *et al.*[12] measured a Hall coefficient for n-type CdSe between 1.3 and 4.2 K which was consistent with linear behavior within the accuracy of their experiment. In both instances, the Hall coefficient was consistent with variable range hopping of the form $R_H \propto \exp(KT^{-x})$ with $x \approx \frac{1}{4}$. In contrast, Tousson and Ovadyahu[13] found a Hall coefficient for In_2O_{3-x} films which is independent of temperature on both sides of the metal-insulator transition. For just-metallic films, they showed that the Hall coefficient is independent of field only for very small magnetic fields.

In this paper we report a nonlinear Hall voltage and a field-dependent Hall coefficient at small magnetic field in the hopping regime. Based on measurements over a broad range of temperature between 100 mK and 12 K for a series of insulating n-type CdSe samples, we also report a temperature dependence for the Hall coefficient of the form $R_H \propto \exp(KT^{-x})$ with $x \approx \frac{1}{2}$.

The compensated indium-doped CdSe specimens used for the present studies were obtained from the Polish Academy of Science, and are the same as those used for the resistance measurements published in Ref. 14. The differences, $N_D - N_A$, between donor and acceptor concentrations, henceforth referred to as the net dopant concentrations, were determined from the room temperature Hall coefficient. The samples designated as 1, 3, 4, and 5 were found to have net concentrations[15] 2.65, 2.30, 2.25, and 2.10×10^{17} cm^{-3}; from the Mott criterion and from the behavior of the conductivity at low temperatures,[14] the critical concentration is estimated to be about 3×10^{17} cm^{-3}. Measurements of the Hall coefficient were taken in two overlapping temperature ranges, in an Oxford Model 75 dilution refrigerator in magnetic fields to 9 T and temperatures down to about 100 mK (depending on the sample) and in a He^4 glass Dewar in fields to 4 T and temperatures up to 12 K. Standard low-frequency ac techniques were used, care was taken to minimize heating and thermoelectric effects, and magnetoresistive contributions were eliminated by reversing the direction of the magnetic field.

The longitudinal magnetoconductance of sample 4 (net dopant concentration 2.25×10^{17} cm^{-3}) is shown in Fig. 1 at various temperatures in magnetic fields to 9 T. We attribute the positive magnetoconductance in fields below about 2 T to the effect of a magnetic field on the quantum interference between different multiscattering paths, as calculated by Nguyen, Spivak, and Shklovskii,[16] Sivan, Entin-Wohlman, and Imry,[17] Schirmacher,[18] and others. The decrease at higher fields is thought to be due to field-induced modifications of the impurity wave functions, as discussed by Shklovskii and Efros.[19] As can be seen in the inset in Fig. 1, the low-field magnetoconductance is either linear or quadratic with magnetic field depending on the temperature. Detailed discussion of these and other results on the magnetoconductance will be published elsewhere.

For the four insulating n-type CdSe samples used in these experiments at all measured temperatures, the Hall voltage V_H was found to be a nonlinear function of the magnetic field at low fields. If a linear regime does exist, it is restricted to a narrow range at very small magnetic fields, where reliable measurements of the Hall voltage

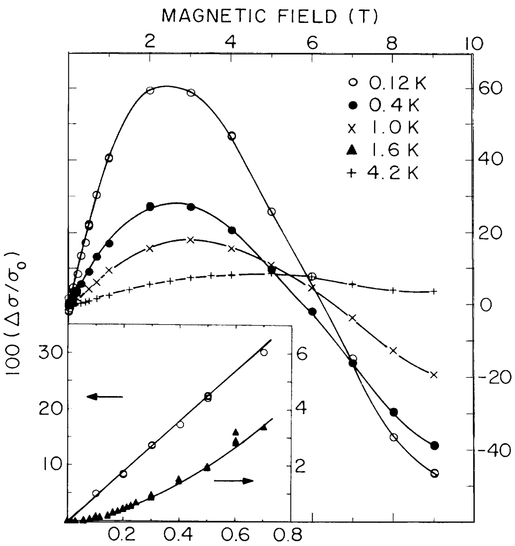

FIG. 1. The longitudinal magnetoconductance, $\Delta\sigma/\sigma_0 = [\sigma(H) - \sigma(0)]/\sigma(0)$, of compensated n-type CdSe with net dopant concentration $N_D - N_A = 2.25\times10^{17}$ cm^{-3} (sample 4) plotted as a function of magnetic field at various temperatures, as labeled. The lines are drawn to guide the eye. The inset shows the behavior in small magnetic field at two different temperatures.

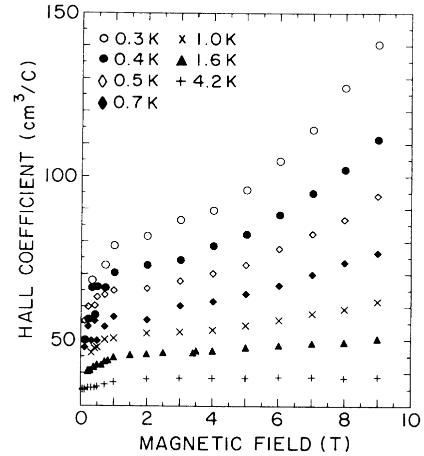

FIG. 2. The Hall coefficient R_H of compensated n-type CdSe with net dopant concentration $N_D - N_A = 2.25\times10^{17}$ cm^{-3} (sample 4) plotted as a function of magnetic field at various temperatures, as labeled.

are extremely difficult to obtain. The usual definition, $R_H = V_H/IH$, yields a field-dependent rather than a constant Hall coefficient, which is plotted as a function of the magnetic field at various temperatures for sample 4 in Fig. 2. The sign of the Hall coefficient is that expected for negative charge carriers. For all dopant concentrations and at all temperatures, the Hall coefficient increases approximately linearly with field, levels off, and increases again at higher field. As is true for the longitudinal magnetoconductance, we suggest that the behavior at large magnetic field is associated with a reduction in impurity wave-function overlap. In the remainder of this paper we will consider the behavior at low fields and the plateau region.

The apparent saturation of the low-field component to a plateau value suggests that the field dependence at small fields may be due to quantum interference processes similar to those that determine the longitudinal magnetoconductance. According to current theory,[16–18] the longitudinal magnetotransport is governed by the flux through a "typical" area $A = (R_{\rm hop})^{3/2}\xi^{1/2}$ (where $R_{\rm hop}$ and ξ are the hopping length and the localization length, respectively), and saturation is expected approximately when the flux $\phi = BA$ becomes comparable with an elementary flux quantum ϕ_0. The temperature dependence of the resistivity[14] indicates that, depending on the sample and on the range of temperature, one finds either $R_{\rm hop} \propto T^{-1/4}$ (Mott hopping) or $R_{\rm hop} \propto T^{-1/2}$ (Efros-Shklovskii hopping[8]). One expects that saturation should occur at a magnetic field $H_{\rm sat} \propto T^{3/8}$ in the former case and $H_{\rm sat} \propto T^{3/4}$ in the latter. An estimate of $H_{\rm sat}$ obtained by tracking the "knee" of Fig. 2 yields $H_{\rm sat} \propto T^{\delta}$ with $\delta \approx 0.36$ for samples 3 and 4 and $\delta \approx 0.17$ in the case of sample 5, which is in apparent disagreement with these simple theoretical expectations. One should note, however, that the transverse magnetotransport is probably more complicated and not directly analogous to its longitudinal counterpart, and it is likely in any event that a full understanding will require analysis of the transverse conductivity, σ_{xy}, rather than of the Hall coefficient itself.

For a given sample, the Hall coefficient R_H at small magnetic fields is a function only of H/T, the ratio of field energy to thermal energy. This is shown in the inset in Fig. 3, where $[R_H(H,T) - R_H(0,T)]$ is plotted for sample 4 as a function of H/T. This also holds true for samples 3 and 5, but does not apply to sample 1, which has an

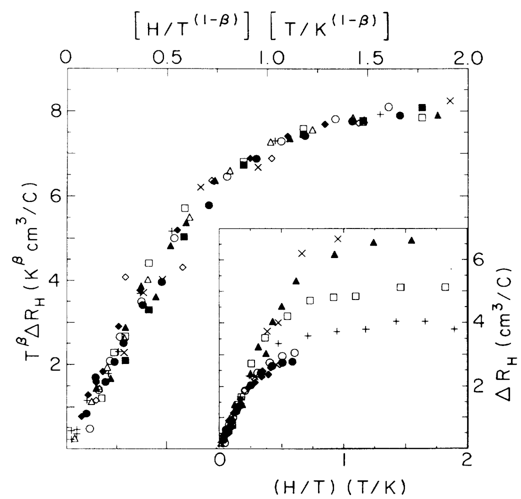

FIG. 3. Compensated n-type CdSe with net dopant concentration $N_D - N_A = 2.25\times10^{17}$ cm^{-3} (sample 4), $T^{\beta}\Delta R_H$ vs $H/T^{(1-\beta)}$ with $\Delta R_H = [R_H(H,T) - R_H(0,T)]$, and $\beta = 0.64$ [see Eq. (1)]. The plot of ΔR_H vs H/T shown in the inset illustrates that the magnetic field for the onset of the plateau region depends strongly on temperature. The symbols denote the following: ◇, 0.5 K; ×, 1.0 K; ▲, 1.6 K; □, 2.75 K; ■, 3.6 K; +, 4.2 K; ○, 5.0 K; ●, 6.0 K; △, 7.2 K; ◆, 8.8 K.

impurity concentration very close to the critical concentration, and for which R_H is proportional instead to $H/T^{0.6}$. We note that the dependence on H/T found for samples 3, 4, and 5 does not imply a simple dependence on the magnetization M, which is not a function of H/T in these materials but rather[20] of $T^{(1-\alpha)}f(H/T)$ (here $0<\alpha<1$ and α depends on dopant concentration). We also note that although the transverse and longitudinal components of the magnetotransport must ultimately be related, the magnetoconductance shown in Fig. 1 bears no simple relation to the Hall coefficient of the same sample shown in Fig. 2.

As is evident from the inset in Fig. 3, the field at which the plateau is reached depends on temperature. For low and intermediate fields the data for all except sample 1 are found to fit the empirical expression

$$R_H(H,T)=R_H(0,T)+T^{-\beta}F(H/T^{(1-\beta)}) , \qquad (1)$$

where F is a "universal" function for a given sample, with $\beta=0.65\pm0.05$, 0.65 ± 0.05, and 0.83 ± 0.05 for samples 3, 4, and 5, respectively. To illustrate this behavior, Fig. 3 shows $T^{\beta}[R_H(H,T)-R_H(0,T)]$ vs $H/T^{(1-\beta)}$ for sample 4. We caution, however, that although Eq. (1) describes the data in the range of experimental parameters of our studies, this may be an approximation to a different functional form over a broader range.

To study the temperature dependence of the Hall coefficient, it is not clear in the absence of theoretical guidance whether one should use its zero-field extrapolation or some other choice such as the plateau value. Within our experimental accuracy, either choice yields an exponentially activated variable-range-hopping form, $R_H \propto \exp(KT^{-x})$, with an exponent $x>\frac{1}{4}$ for samples 3-5. Figure 4 shows the Hall coefficient extrapolated[21] to zero magnetic field, $R_H(0,T)$, plotted on a logarithmic scale as a function of $T^{-1/2}$. The residuals for fits of the data between 100 mK and 1.6 K to the variable-range-hopping form using exponents $x=\frac{1}{4}$, $\frac{1}{2}$, and 1 are shown in insets (A), (B), and (C), respectively. As in the case of the longitudinal resistivities[14] in the same temperature range, the exponent x for samples 3-5 is about $\frac{1}{2}$, while $x=\frac{1}{4}$ fits well for sample 1 (not shown), which is quite close to the transition.

To summarize, we report the first observation in the hopping regime of a Hall voltage which exhibits a nonlinear dependence on magnetic field at small fields. The Hall coefficient, defined by $R_H=V_H/IH$, increases approximately linearly at low fields to a plateau, and then increases again at high fields. Except very near the metal-insulator transition, R_H is consistent with Eq. (1) over the range of temperature and magnetic field of our experiments. As is true for the longitudinal resistivity, the Hall coefficient is consistent with a variable-range-hopping form, $R_H \propto \exp(KT^{-x})$, with $x>\frac{1}{4}$ for samples not very near the transition.

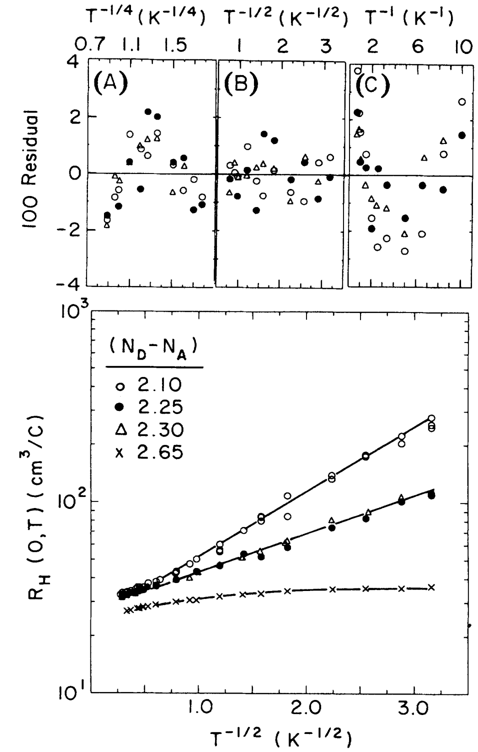

FIG. 4. The Hall coefficient extrapolated to zero field, $R_H(H=0,T)$, plotted on a logarithmic scale as a function of $T^{-1/2}$ for four samples with net dopant concentrations labeled in units of 10^{17} cm^{-3}. The insets (A), (B), and (C) show the residual of $\ln(R_H)$ obtained by fitting the data between 100 mK and 1.6 K to the expression $\ln R_H \propto T^{-x}$ with $x=\frac{1}{4}$, $\frac{1}{2}$, and 1, respectively. The symbols denote the following samples: △, 3; ●, 4; ○, 5.

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