In summary, we have explained the "undulations" in terms of vibronic levels of acceptors and have thereby demonstrated that even shallow acceptor states in semiconductors are strongly modified by the dynamic Jahn-Teller effect. It thus becomes clear that any accurate theory of acceptor ground states—even in the effective mass approximation—must include this interaction.

The authors would like to thank Dr. K. W. H. Stevens and Dr. J. C. Slonczewski for many stimulating discussions of the Jahn-Teller effect of extended states, and Mr. G. D. Pettit and R. E. Fern for their expert assistance with the optical measurements.

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# Electric-Field-Dependent Magnetoresistance in Ferromagnetic Semiconductors

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(Received 17 December 1971)

Changes in the sign of the longitudinal magnetoresistance as a function of applied magnetic and electric fields are observed in single crystals of $p$-type $CdCr_2Se_4$. A phenomenological theory for the non-Ohmic behavior associated with spin-wave–carrier interaction is developed, and its comparison with the experimental results indicates the existence of spin-wave amplification by the drifting carriers. All the experimental results can be explained in terms of spin-wave–carrier interaction and spin-disorder scattering.

We have observed a variety of new phenomena in the magnetoresistance of a high-mobility ferromagnetic semiconductor. The study of the magnetoresistance as a function of magnetic and electric fields and as a function of the crystallographic orientation has yielded results that are in accord with expectations from spin-disorder scattering[1,2] and spin-wave–carrier interaction.[3,4] We propose a phenomenological theory for the non-Ohmic effects associated with the last interaction. The experimental results are found to be in qualitative agreement with this theory, and thus yield the first evidence for positive magnetoresistance that is associated with spin-wave amplification. A more detailed account of the analysis and the experimental results will be presented later.[5]

The material used for the measurements was

$p$-type $CdCr_2Se_4$, which is the only known high-mobility ferromagnetic semiconductor.[6] This material has a cubic spinel structure where the magnetic $Cr^{+3}$ ions are on the octahedral $B$ sites, i.e., they form a nearest-neighbor magnetic chain along the [110] direction.[7] It has been realized earlier[8] that the non-Ohmic behavior observed on *polycrystalline* samples of this material might be associated with the interaction of carriers and spin waves. However, the earlier experimental results, apart from not being self-consistent and different from sample to sample, do not represent the phenomena as they occur in *single* crystals and do not exhibit the expected behavior (see below). This is mainly because of the polycrystalline nature of these samples and heating effects associated with the low resistivity of the samples used.

For the present study *single crystals* doped with 0.4% Ag were grown.[9] The high resistivity of these crystals as well as the pulse technique used have assured the absence of heating effects. That heating did not occur was confirmed by observing exact reproducibility when increasing and decreasing the magnetic field several subsequent times. Since the sign of the charge carriers is not definitely known below 100°K,[2,6,10] and because the Curie temperature of $CdCr_2Se_4$ is $T_c = 130$°K, we shall report here on results obtained between 100 and 135°K.

In our pulse measurements the change of the current through the crystal due to the application of a magnetic field was differentially amplified. Using a box-car integrator the corresponding dc signal as a function of the applied magnetic field $H_0$ was displayed on a recorder. The voltage-pulse width was 35 $\mu$sec and the gate pulse was applied 25 $\mu$sec after the voltage-pulse application. It was found independently that steady-state conditions are established in less than 1 $\mu$sec after pulse application. Thus, all the results to be described below are associated with *steady-state* conditions.

Typical recorder traces are shown in Fig. 1. This figure demonstrates that when an electric field $E_z$ and a magnetic field $H_0$ are applied, the transverse magnetoresistance ($H_0 \perp E_z$) is negative, independent of crystallographic orientation, and its magnetic field dependence follows the magnetic field dependence of the magnetization, $M(H_0)$.[7] It was also found that the transverse magnetoresistance is independent of the electric field amplitude. All this behavior is that expected from spin-disorder scattering.[1,2] For the longitudinal magnetoresistance ($H_0 \parallel E_z$) below $T_c$, the behavior is quite different. As is shown in Fig. 1(a), the most striking feature of this magnetoresistance is its electric field dependence. This can be noticed even in the sublinear $I$-$V$ characteristics of the material when a magnetic field is applied.[5] The electric field dependence of the magnetoresistance, as obtained from the type of data shown in Fig. 1(a), is shown in Fig. 2 for $H = 5$ kG. In contrast with the transverse magnetoresistance, the

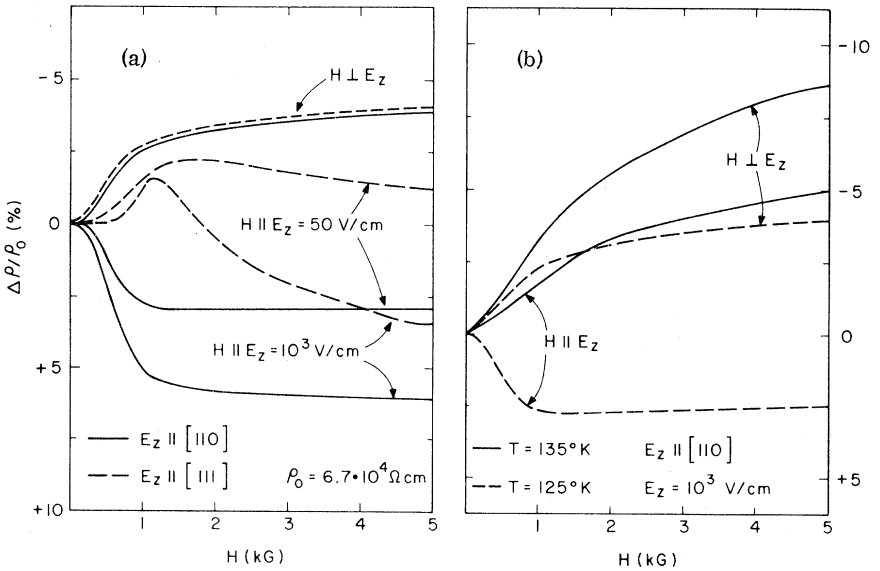


FIG. 1. Recorder traces of the pulsed magnetoresistance measurements of $Cd_{1-x}Ag_xCr_2Se_4$ ($x = 0.004$). (a) At 100°K; (b) above and below the Curie temperature.

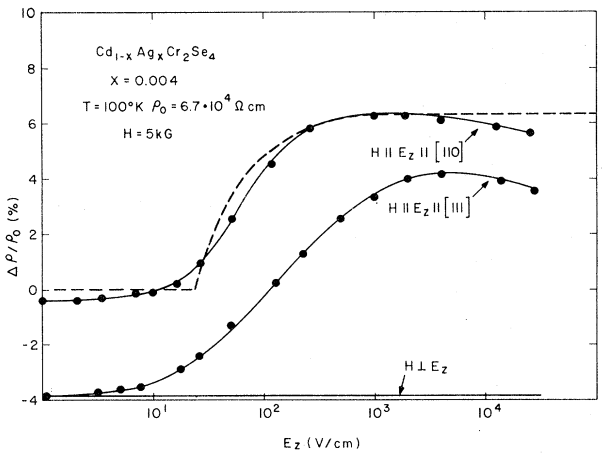


FIG. 2. The electric field dependence of the longitudinal magnetoresistance. The dashed curve represents the function $A(v_z - v_s)/v_z$, where $A$ and $m^* v_s/e\tau$ were determined by fitting the function to two experimental points for $v_z > v_s$ and taking $A = 0$ for $v_z < v_s$.

longitudinal magnetoresistance is found to increase significantly between 30 and 300 V/cm and to depend on the crystallographic orientation. To check whether the behavior of the longitudinal magnetoresistance is associated with the existence of long-range magnetic order, we have carried out the same measurements at $T = 125$°K $< T_c$ and $T = 135$°K $> T_c$. This was indeed confirmed as shown in Fig. 1(b). The results for $T = 135$°K were independent of the applied electric field (up to the highest measured field $E_z = 4 \times 10^4$ V/cm), while the results for $T = 125$°K were found to be qualitatively similar to those shown in Fig. 1(a) and Fig. 2 for $T = 100$°K. Measurements on the low-mobility[6,10,11] $n$-type crystals ($Cd_{1-x}In_xCr_2Se_4$, $x = 0.027$) have neither shown positive magnetoresistance nor did they indicate significant electric field dependence of $\Delta\rho/\rho_0$ down to 77°K (where the negative magnetoresistance[1,10] for $H = 5$ kG is 3%). All this, and the fact that no positive magnetoresistance was found for other low-mobility ferromagnetic semiconductors,[12] suggests strongly that the above observations are associated with the *high hole mobility* in the present material and with the existence of *long-range magnetic order*.

For the analysis, consider a chain of magnetic ions ordered along the $z$ axis, and that this one-dimensional ferromagnet is magnetized in the $z$ direction as a result of an applied dc magnetic field $H_0$. The thermally excited spin waves will then propagate with a velocity $v_s$ along the $z$ axis, while the associated circularly polarized magnetic and electric fields will be in the $x$–$y$ plane. If a dc electric field $E_z$ is applied, the carriers will drift with a velocity $v_z = e\tau E_z/m^*$, where $e$ is the electronic charge, $\tau$ is the carrier normal relaxation time, and $m^*$ is its effective mass.

Following the derivation for the interaction of electromagnetic plane waves with drifting electrons as given by Lampert,[13] one can easily show that for holes the circularly polarized spin-wave amplification (or attenuation) constant per unit time, under small signal conditions ($\alpha \ll \omega$), can be given by

$$\alpha = \frac{(\sigma/2\epsilon)(\mu/\mu_0)(v_s/c)^2(v_z/v_s - 1)}{1 + \omega^2\tau^2(1 - v_z/v_s + \omega_c/\omega)^2} . \qquad (1)$$

Here $\omega$ is the frequency of the circularly polarized electromagnetic field, $\sigma$ is the sample conductivity, $\epsilon$ is its dielectric constant, and $c$ is the light velocity *in the medium*. This result differs from that of Lampert (for electromagnetic plane waves) only in the inclusion of the $\omega_c/\omega$ term, where $\omega_c$ is the cyclotron frequency ($\omega_c = e\mu_0 H_0/m^*$), and in the consideration of the relative permeability of the medium, $\mu/\mu_0$ (taken by Lampert to be 1). Under the presence of spin waves this term is[4,14]

$$\mu/\mu_0 = 1 + \gamma M(H_0)(\omega_{sw} - \omega) \times [(\omega_{sw} - \omega)^2 + (\gamma\Delta H_k)^2]^{-1} ,$$

where $\gamma$ is the gyromagnetic ratio, $\omega_{sw}(k)$ is the spin-wave dispersion relation,[15] and $\Delta H_k$ is the linewidth.[14] Just as for the acoustoelectric effect[16] it is expected that the carrier interaction with the electromagnetic field of the frequency of maximum gain, $\omega_{max}$, will dominate the interaction. Considering for simplicity only the frequency dependence of $\mu/\mu_0$, we find $\omega_{max} = \omega_{sw}(k) - \gamma\Delta H_k$. In ferromagnets in general and in the material used for the experiments in particular,[7,14] we can correspondingly write $\mu/\mu_0 \approx M(H_0)/2\Delta H_k$.

It should be noted that for simplicity we have assumed a single velocity of the spin waves while all spin waves, for which $\omega_{sw}(k)/k \leq v_z$, can be amplified.[4] For the present steady-state conditions this seems a reasonable approximation since the spectrum of the thermally excited spin waves and the loss mechanisms are expected[16] to limit the amplified spin waves to a rather narrow-frequency band. Comparison[5] of the above $\alpha$ with the results obtained in Ref. 4 (where the dispersion relation is taken into account) shows agreement in the $\alpha$ dependence on $\sigma$ and $v_z$ for the noncoherent[4] case when $\min[\omega(k)/k] \ll v_z$. The fact that these conditions are likely[16] to prevail in the

steady state thus gives further support to the above approximation. Thus, $v_s$ here is an effective velocity of the spin waves in the above frequency band.

Using the general approach[16] first suggested by Weinreich,[17] we define an effective electric field $E_{eff} = 2\alpha W/env_s$, where $W$ is the energy density of the spin-wave system, and $n$ is the number of carriers. This field essentially describes the effective force $eE_{eff}$ exerted by (or on) the carrier on (or by) the wave. The force $eE_{eff}$ accelerates the carriers when $v_s > v_z$ and decelerates them when $v_z > v_s$. The relation holds for small-signal conditions,[16,17] i.e., when $|\Delta\rho/\rho_0| \ll 1$. Under these conditions we can further write $\Delta\rho/\rho_0 = E_{eff}/E_z = e\tau E_{eff}/m^* v_z$, and thus

$$\Delta\rho/\rho_0 = F(T)WM(H_0)(v_z - v_s)/v_z, \qquad (2)$$

where $F(T)$ is a function of the temperature $T$. As is immediately apparent from Eq. (2), positive magnetoresistance is expected to be observed under spin-wave amplification and negative magnetoresistance under spin-wave attenuation.

Since we are concerned here with measurements that were taken under steady-state conditions in the magnetoresistance, we should expect that the spin-wave energy density has reached its steady-state value $W_{ss}$, and that $W_{ss} \gg W_0$, where $W_0$ is the energy density of thermally excited spin waves.[3,15] Thus, for a given $H_0$ and $T$, we should have $A = F(T)W_{ss}M(H_0) = \text{const}$. This is unlike the case in the acoustoelectric effect, where current saturation does not always represent a steady state in the energy density of the acoustic waves.[16] The difference between the two phenomena is apparent since in the present case the carrier velocity exceeds by far the spin-wave velocity under steady-state conditions. The reasons for the difference emerge from the much weaker interaction and the transverse nature of the waves in the present case. Under spin-wave attenuation conditions on the other hand, $W < W_0$, and this cannot yield a significant negative magnetoresistance. Thus $A \to 0$. The negative magnetoresistance in the $E_z \| H \| [110]$ configuration for a very low electric field is rather due to spin-disorder scattering.

For the present material in the case of longitudinal magnetoresistance in the [110] direction, we approach the ideal configuration outlined above for spin-wave–carrier interaction. Comparison of Eq. (2) with the experimental results of Figs. 1(a) and 2 shows that *both* the $M(H_0)$ and the $(v_z - v_s)/v_z$ dependences of the predicted longitudinal magnetoresistance are indeed found. The last dependence is demonstrated by the dashed curve shown in Fig. 2. This curve is obtained by fitting the function $A(v_z - v_s)/v_z$ to two experimental points for which $v_z > v_s$. The fit yields $v_s m^*/e\tau = 25$ V/cm and $A = 6.3\%$. The first quantity is in accord with known velocities of spin waves[15] ($10^5 - 10^6$ cm/sec) and the mobility of holes in the present material[6] ($\sim 10^4$ cm$^2$/V sec). At present we cannot make an independent estimate of $A$ since too many experimental parameters are still unknown.[5] However, the qualitative fit is clearly apparent.

In other crystallographic directions in this material, such as the [111] direction, the simple nearest-neighbor magnetic chain model does not apply, and spin waves of another character are involved. Thus, a combination of this interaction and the spin-disorder scattering leads to a more complicated observation. The slight decrease of $\Delta\rho/\rho_0$ for very high fields might be due to the term $1 - v_z/v_s + \omega_c/\omega$ in Eq. (1) or due to the type of nonelectronic losses of the spin waves.[3,14,18]

The energy-loss mechanism responsible for the steady-state spin-wave energy density $W = W_{ss}$ must be due to the transfer of energy from the spin system to the lattice. When $W$ is small, magnon-magnon interactions are dominant in the present material[14]; but as the "temperature" of the spin system[3] becomes high enough compared to that of the lattice, a steady state is reached. The energy pumped by the carrier system into the spin system is further transferred to the lattice and the dominant frequency probably shifts from $\omega_{max}$ to some lower frequency.[16,18]

The authors are indebted to Dr. A. Rose for many helpful discussions. Comments made by Dr. S. B. Berger, Professor M. A. Lampert, and Dr. P. J. Wojtowicz are gratefully acknowledged.

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# Linearized Third-Derivative Spectroscopy with Depletion-Barrier Modulation

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Low-field (third-derivative) electroreflectance spectra taken on fully depleted space-charge regions are shown to be linear in the modulation potential and free from experimentally induced line-shape distortions due to modulation wave-form, dc bias, or barrier-potential effects. Using a metal-semiconductor (Schottky diode) configuration, accurate threshold energies of the $E_0'$ triplet of Ge are obtained. The observed spin-orbit–splitting energy of the valence band confirms that the highest transition also occurs at $\Gamma$.

We report that the quadratic scaling dependence of low-field electroreflectance (ER) spectra[1] upon the electric field can be combined with the square-root dependence of the electric field on the barrier potential in a fully depleted space-charge region to yield third-derivative[2] surface-barrier electroreflectance (SBER) spectra which are linear in the modulation *potential*, rather than the electric field, and therefore are rigorously free from modulation wave-form or dc bias effects. In contrast to high-field ER measurements,[3-5] we show that it is *not* necessary to use square-wave flat-band modulation in order to obtain quantitative spectra accurately representing crystal properties when the low-field depletion barrier conditions are satisfied. This result greatly simplifies experimental procedures and enables third-derivative spectroscopy to be extended to a wide range of materials and temperatures for which quantitative ER measurements are not presently feasible. Since these third-derivative spectra are theoretically the sharpest of all modulation spectra, very precise values of transition threshold energies and broadening parameters can be obtained when they are analyzed by previously developed low-field ER techniques.[6]

In a fully depleted space-charge region of a semiconductor, formed by, e.g., a $p$-$n$ junction,[7] metal-semiconductor barrier,[7] heterojunction,[7] or semiconductor-electrolyte[8] configuration, minority carriers which diffuse from the bulk and/or are thermally or optically excited in the space-charge region are extracted at the surface, so no inversion layer can form. The electric field $\mathcal{E}$ and the potential $\varphi$ are related at any point in the barrier by Poisson's equation[7]:

$$\mathcal{E}^2(\varphi) = -(2eN_D/\epsilon_0)(\varphi + kT/e), \tag{1}$$

where $N_D$ is the donor impurity concentration and $\epsilon_0$ is the static dielectric constant. At low fields, the field-induced change in the reflectivity, $\Delta R/R$, is a fourth-rank tensor of the form[1,2]

$$\Delta R/R = \mathcal{E}^2 L(\hbar\omega), \tag{2}$$

where the spectral line-shape function $L(\hbar\omega)$ is determined entirely by the energy band structure and its selection rules through the polarization of the incident light and the direction of the electric field. If $\varphi = V(t)$ is a time-dependent potential, the time-dependent part of $\Delta R(t)/R$ is linearly proportional to the time-dependent part of $V(t)$. The spectrum measured experimentally by phase-sensitive detection is therefore

$$(\Delta R/R)_{\text{expt}} = \Delta R_1/R = -(2eN_D V_1/\epsilon_0)L(\hbar\omega), \tag{3}$$

where $\Delta R_1$ and $V_1$ are the fundamental harmonic components of $\Delta R(t)$ and $V(t)$, respectively. The measured spectrum is completely independent of