Table II. Incident beam in the (110) direction.

0.484±0.020 0.461
$0.411 \pm 0.015$ $0.456$ $0.398 \pm 0.015$ $0.445$

<sup>&</sup>lt;sup>a</sup>Predicted from equipartition rule.

ticle Van de Graaff accelerator at Los Alamos. We wish to acknowledge the assistance of the P-9 staff at Los Alamos under J. L. McKibben and R. L. Henkel. We want to thank R. W. Healy for assistance in the experiment and for maintenance of the equipment. We are grateful to F. L. Vook and D. K. Brice, Sandia Laboratory, for aid in interpretation of the data.

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## FARADAY EFFECT IN MAGNETIZED SOLIDS\*

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In a recent Letter, Mitchell, Palik, and Wallis have reported measurements of the Faraday rotation in PbS. They find that the wavelength variation of the Faraday rotation below interband frequencies has a constant added to the usual  $\lambda^2$  dependence. This constant term varies approximately inversely proportional to the temperature. Mitchell, Palik, and Wallis explain this constant term as caused by the difference in some interband matrix elements

between electrons of different spins. In this Letter a more physical description of the cause of the effect is given. It is shown that the constant term comes from a polarization current produced by the variation in time of the spatial polarization of the electron wave functions,<sup>2</sup> and this constant term is expected to be proportional to the magnetization of the sample. The effect of collisions with the lattice is also estimated.

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The connection between the explanation given here and that given by Mitchell, Palik, and Wallis is made by a sum rule which relates the appropriate sum of interband terms to the spatial polarization of the conduction electrons' wave functions. Mitchell, Palik, and Wallis calculate the most important interband term which by the sum rule is related to the polarization contribution of the conduction electrons discussed here.

When the spin-orbit interaction is considered for metals, the wave function is not a simple product of spin and spatial parts. Bloch states are still the eigenfunctions, and for each value of wave vector k there are two different states which have equal and opposite average values of spin. For solids with a center of symmetry, the spin-orbit interaction causes a displacement of the electron-charge distribution in the unit cell for a given state, producing a polarization of electric charge which would not be there otherwise.2 In equilibrium, although each single state produces a polarization, the states with wave vectors  $\vec{k}$  and  $-\vec{k}$  and the same average value of spin have opposite polarization and are equally occupied, and there is no net polarization of the solid. If the solid is nonmagnetic so that both states at k are equally occupied, there is no spatial polarization even in nonequilibrium condition because the two states at k have equal but opposite spatial polarization. However, if the solid is magnetized so that the two states for a given k are not equally occupied, a net charge polarization of the solid will occur in nonequilibrium conditions. For example, consider the nonequilibrium situation corresponding to a rigid displacement of each state in k space by an equal amount. In this case there is a slice of states on the far side of the distribution of occupied states in the direction of the displacement whose polarization is not compensated because the states with the negative wave vectors are not occupied. If the displacement of the distribution of occupied states varies with time, the net charge polarization of the solid will also vary with time, giving rise to a displacement current familiar from Maxwell's equation. It has been shown that the usual quantum mechanical calculation can be interpreted in terms of this same polarization current in magnetized solids.3

Including the polarization current, the current contributed by an electron in a state  $\vec{k}$ 

can be written as3

$$\vec{\mathbf{J}}(\vec{\mathbf{k}}) = (e/\hbar)\nabla_{\vec{k}}E(\vec{\mathbf{k}}) + (d\vec{\mathbf{k}}/dt) \cdot \nabla_{\vec{k}}\vec{\mathbf{P}}(\vec{\mathbf{k}}), \qquad (1)$$

where

$$\vec{\mathbf{P}}(\vec{\mathbf{k}}) = e \int U_{\vec{\mathbf{k}}}^*(\vec{\mathbf{r}}) \vec{\mathbf{r}} U_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) d\tau_0$$

Here  $E(\vec{k})$  is the energy of the state,  $\nabla_k$  is the del operator in  $\vec{k}$  space, e is the electronic charge, the wave function of the  $\vec{k}$  state is

$$\psi_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) = e^{i\vec{\mathbf{k}}\cdot\vec{\mathbf{r}}}U_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}),$$

 $\vec{r}$  is measured from a center of symmetry of the lattice, and  $U_{\vec{k}}(\vec{r})$  is the periodic part of the wave function. The second term on the right-hand side of (1) is the polarization contribution to the current given by the time derivative of the polarization. Neglecting collisions with the lattice, the acceleration of the  $\vec{k}$  states is given by

$$d\vec{k}/dt = e\vec{g} + e\vec{v} \times \vec{B}, \qquad (2)$$

where

$$\vec{B} = \vec{H} + 4\pi \vec{M}$$
,

 $\vec{\mathcal{E}}$  is the electric field,  $\vec{B}$  is the magnetic induction,  $\vec{\mathbf{v}} = \hbar^{-1} \nabla_k E(\vec{\mathbf{k}})$  is the velocity, and  $\vec{\mathbf{M}}$  is the magnetization per unit volume of the solid. If we neglect collisions with the lattice, then using the Boltzmann equation in conjunction with Eqs. (1) and (2) it is a standard problem to calculate the conductivity tensor for a solid. It follows from (2) that the distributionfunction distortion produced by external fields is the same as in the usual case of nonmagnetic metals. Differences come in evaluating the current produced by a given distortion of the distribution function from equilibrium. The magnetic metal has the additional polarization contribution to the current as given by (1). For example, consider the case for a completely magnetized solid with a spherical Fermi surface. For such a solid one expects that the spin-orbit interaction will produce a P(k) of the form

$$\vec{P}(\vec{k}) = A\vec{k} \times \vec{M}. \tag{3}$$

where A is a constant. Using the Boltzmann equation and calculating the current from (1), one finds with  $\vec{M}$  in the z direction and with electric fields varying in time with a radial frequency  $\omega$  that the off-diagonal conductivity

is given by

$$\sigma_{xy}(\omega) = -\frac{\omega_p^2 \omega c}{4\pi \omega^2} + \frac{P_0 \omega_p^2}{4\pi e v_o},\tag{4}$$

when  $\omega \gg \omega_c$ . Here

$$\omega_c = eB/m *C$$
,

$$\omega_p^2 = 4\pi n e^2/m^*,$$

 $P_0$  is the maximum value of P(k) for those electrons on the spherical shell with a magnitude of velocity  $v_0$ , n is the number of electrons per unit volume, and  $m^*$  is the effective mass of the electrons. The quantity  $P_0/v_0$  is independent of  $v_0$  for this model. When the solid is not completely magnetized, it can be shown that the conductivity is simply the sum of the contributions from each direction of magnetization of the electrons. Remembering that  $\vec{P}(\vec{k})$  changes sign with the magnetization of the electronic states, we find

$$\sigma_{xy}(\omega) = \frac{\omega_p^2 \omega_c}{4\pi\omega^2} + \frac{P_0 e}{\hbar k_0} (n_{\uparrow} - n_{\downarrow}), \tag{5}$$

where

$$\omega_{p}^{2} = 4\pi (n_{\uparrow} + n_{\downarrow})e^{2}/m^{*},$$

 $\hbar k_0 = m^* v_0$ , and  $n_{\uparrow}$  and  $n_{\downarrow}$  are the number of electrons per unit volume magnetized up and down, respectively. The form for  $\sigma_{\chi y}$  in (5) shows explicitly that when the solid is unmagnetized the polarization current is zero.

The weak-field Faraday rotation angle  $\theta$  per unit length of propagation when the solid is transparent is given by

$$\theta = -(2\pi/n_0 c)\sigma_{xy},\tag{6}$$

where  $n_0$  is the index of refraction of the solid for B=0. From (5) and (6) we see that in a magnetized solid the Faraday rotation as a function of wavelength or frequency has the form found for PbS. The constant term in the rotation comes from the polarization current. For paramagnetic solids such as PbS,

$$n_{\uparrow} - n_{\downarrow} = 2\chi(\tau)H/g\mu_{B},\tag{7}$$

where  $\chi(\tau)$  is the temperature-dependent magnetic susceptibility, g is the spectroscopic splitting factor, and  $\mu_B$  is the Bohr magneton. The magnetic susceptibility for noninteracting electrons at high enough temperatures such

that the electrons are nondegenerate and not near saturation is given by

$$\chi(\tau) \approx n g^2 \mu_{\rm B}^2 / 4k_{\rm B} T, \qquad (8)$$

where  $k_{\rm B}$  is Boltzmann's constant and  $n=n_{\rm A}+n_{\rm V}$ . This temperature dependence of  $\chi$  produces a  $T^{-1}$  dependence in the constant term of  $\theta$ , in agreement with measurements.

Using the measurements<sup>1</sup> on n-type PbS with  $n=3.5\times10^{17} \text{cm}^{-3}$  and the known value<sup>5</sup> of g=10, we find, using (5), (6), (7), and (8), that for the conduction band of PbS,

$$P_0/ek_0 = 1.1 \times 10^{-16} \text{ cm}^2.$$
 (9)

Using a value of  $k_0 = 2.3 \times 10^6 {\rm cm}^{-1}$  calculated for a degenerate electron gas with  $n = 3.5 \times 10^{17}$  cm<sup>-3</sup>, we find that  $P_0/e \approx 2.5 \times 10^{-10}$  cm. For electron densities of the order of metallic densities  $(10^{23} {\rm ~cm}^{-3})$ ,  $P_0/e \approx 10^{-8} {\rm ~cm}$ . The application of a model with a spherical Fermi surface to PbS is a good approximation, since both the valence and conduction bands in PbS are nondegenerate, nearly spherical, and parabolic in the vicinity of the band extrema.

To see if this value of  $P_0/ek_0$  is reasonable, we estimate theoretically its expected value to first order in the spin-orbit interaction. Following reference 2, we find

$$\frac{P(\vec{k})}{e} = \frac{2}{\hbar} \operatorname{Re} \sum_{n \neq n'} \frac{\langle n\vec{k} | \vec{r} | n'\vec{k} \rangle \langle n'\vec{k} | H' | n\vec{k} \rangle}{\omega_{nn'}(\vec{k})}, \quad (10)$$

where

$$H' = \hbar \{ [\vec{\sigma} \times \nabla V(\vec{\mathbf{r}})] \cdot \vec{\mathbf{p}} / 4m^2c^2 \},$$

the spin-orbit interaction. Here Dirac notation is used for the matrix elements between the zero-order Bloch states where the spin-orbit interactions are absent, and the integration is over a unit cell. The index n denotes the unoccupied bands, and n' denotes the occupied band. The energy difference between the states  $|n\vec{k}\rangle$  and  $|n'\vec{k}\rangle$  is  $\hbar\omega_{nn'}(\vec{k})$ . Also,  $\vec{\sigma}$  is the spin operator, P is the momentum operator, and  $V(\mathbf{r})$  is the periodic potential. A rough estimate can be obtained by replacing  $\omega_{nn}(\vec{k})$  by an average value  $\langle \omega_{nn'} \rangle$ . The sum over intermediate states n can be performed using closure (the term for n = n' is zero for crystals with a center of symmetry). Then assuming that  $V(\mathbf{r})$  is spherically symmetric in a unit cell,

we find approximately that

$$\frac{P_0}{e^k_0} \approx a^2 \frac{\omega_{\text{s.o.}}}{\langle \omega_{nn'} \rangle},\tag{10'}$$

where  $\hbar\omega_{\mathbf{S.O.}}$  is the energy separation of the atomic spin-orbit doublets and a is the atomic radius. For lead,  $a\approx 10^{-8}$  cm,  $\hbar\omega_{\mathbf{S.O.}}\approx 1$  eV and an average value for interband energies in PbS is about  $\hbar\langle\omega_{nn'}\rangle\approx 1$  eV. Thus, theoretically we expect

$$P_0/ek_0 \approx 10^{-16} \text{ cm}^2$$
,

in agreement with the experimental value given in (9).

We now discuss the relation of the polarization current given here to the more usual way of calculating these effects, such as that by Mitchell, Palik, and Wallis. The contribution of the polarization current to  $\sigma_{\chi y}$  is, as are all other contributions to  $\sigma$ , affected by inter-

band effects. The Boltzmann equation normally does not include such interband effects, and its results are valid, as in this Letter, only for frequencies  $\omega \ll \omega_0$  where  $\omega_0$  is the smallest interband frequency. In the case of interest here,  $\omega_0 \gg \omega \gg \omega_{\mathcal{C}}$ , the usual expressions for  $\sigma_{\chi y}$  have the form

$$\sigma_{xy} = \frac{A}{\omega^2} + \sum_{n \neq n'} \frac{B_{nn'}}{\omega_{nn'}^2 - \omega^2}.$$
 (11)

The first term on the right-hand side corresponds to the first term on the right-hand side of (4). The second term on the right-hand side corresponds to the second term on the right-hand side of (4) and is a sum over interband matrix elements which becomes frequency independent when  $\omega \ll \omega_0$ . In this limit, and letting the magnetic field be zero, this term becomes equal to<sup>3,6</sup>

$$\sum_{n \neq n'} \frac{B_{nn'}}{\omega_{nn'}^2} = \frac{ie^2}{\hbar V} \sum_{k} \sum_{n \neq n'} (\langle n'k \mid y \mid nk \rangle \langle nk \mid x \mid n'k \rangle - \langle n'k \mid x \mid nk \rangle \langle nk \mid y \mid n'k \rangle), \tag{11'}$$

where V is the volume of the solid and now the states  $\langle n'k |$  and  $\langle nk |$  include all spin-orbit effects. Mitchell, Palik, and Wallis approximately calculate this interband term, which includes all possible transitions such as ones involving a spin flip. From the fact that [x,y]=0, it follows immediately that the sum in (11'), when the term n=n' is added, is zero, and, from this sum rule, (11') is equal to minus the term with n=n'. This term with n=n' depends only on the properties of the occupied states and, as is shown in reference 3, gives the polarization-current contribution to  $\sigma_{XV}$ .

We now estimate the effects of collisions with the lattice. It is not sufficient to assume simply a single relaxation time for a magnetized solid. The simplest reasonable model requires two relaxation times. Because of the same spin-orbit interaction which polarizes the electron wave functions, the scattering to the "right" of an electron in a k state will be different than that to the "left." This anisotropic scattering is the cause of the large anomalous Hall effect in magnetized metals. The need for two relaxation times in the simplest case can also be

seen in the following manner. Consider a rigid displacement of the occupied distribution function in k space. A single relaxation time would mean that the rigid displacement would relax without a change in shape. However, because of the anisotropic scattering, more electrons would scatter to the "right," say, than to the "left," and the displacement would change shape and get a bulge to the "right." It takes at least two relaxation times to describe this. We pick the following simple model to describe this. Denote the rigid displacement by a distribution function  $\varphi_{\mathbf{1}}$ , and the "bulging to the right" distortion by  $\varphi_2$ . If at t=0 we start with only  $\varphi_1$ , then we assume that the distribution function  $\varphi$  varies in time 2.s

$$\varphi = \varphi_1 \exp(-t/\tau_1) + \varphi_2 [\exp(-t/\tau_2) - \exp(-t/\tau_1)].$$
 (12)

The relaxation time for  $\varphi_1$  is  $\tau_1$ , and  $\tau_2 > \tau_1$  such that as time progresses  $\varphi_2$  builds up with respect to  $\varphi_1$ . It is now straightforward to calculate  $\sigma_{\chi y}$  from the assumptions implied by (12), and we obtain for the spherical mod-

el considered previously

$$\sigma_{xy} = -\frac{\omega_{p}^{2}\omega_{c}}{4\pi(\omega + i/\tau_{1})^{2}} + \frac{\left[\sigma_{xx}(0) - \omega_{p}^{2}\tau_{1}/4\pi\right]\omega_{c}}{(\tau_{2} - \tau_{1})} \left[\frac{1}{(\omega + i/\tau_{1})^{2}} - \frac{1}{(\omega + i/\tau_{2})^{2}}\right] - \frac{\sigma_{xy}^{M}}{(\omega\tau_{2} + i)(\omega\tau_{1} + i)} + \frac{P_{0}\omega_{p}^{2}\omega}{4\pi e v_{0}} \left[\frac{\tau_{1}}{\omega\tau_{1} + i} + \frac{i\left[\tau_{1} - \sigma_{xx}(0)4\pi/\omega_{p}^{2}\right]}{(\omega\tau_{1} + i)(\omega\tau_{2} + i)}\right].$$
(13)

Here  $\sigma_{\chi \nu}^{M}$  is that part of the dc value of  $\sigma_{\chi \nu}$ that is proportional to the magnetization of the sample, and  $\sigma_{\chi\chi}(0)$  is the dc value of the diagonal conductivity for H=0. In the derivation of (13) it was assumed that  $\varphi_2$  can also be represented by a rigid displacement of the equilibrium distribution function, but rotated with respect to  $\varphi_1$ . It is clear that the frequency dependence of  $\sigma_{xy}$  when relaxation times are important is quite complicated even for this greatly simplified model. Present experimental measurements<sup>7</sup> give some indications of such complexities, though the frequency range where relaxation effects are important has not yet been thoroughly investigated. A recent phenomenological theory<sup>8</sup> for calculating  $\sigma_{xy}$ for magnetized conductors includes only the term in (13) proportional to  $\sigma_{\chi V} M$ .

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their measurements before publication.

## REACTION He<sup>3</sup>(p, n)3p AT $E_p = 14.1$ MeV\*

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In a recent Letter, Ajdačić et al. have reported data on the zero-degree proton spectrum from the reaction  $H^3(n,p)3n$ . They report a breakup cross section much larger than the prediction of Gammel and MacKellar, and their data suggest the existence of a bound trineutron  $(n^3)$ . Since the existence of a bound trineutron is in disagreement with current estimates of light nuclei binding energies, we have investigated the reaction  $He^3(p,n)3p$ , seeking evidence for a strong three-nucleon interaction. (To establish the equivalence of these charge-conjugate reactions one does not have to assume charge independence of nuclear forces but needs only to invoke charge symmetry—a feature of

the two-body force which has been clearly demonstrated. 4)

Using the Livermore 90-in. variable-energy cyclotron-neutron time-of-flight facility,<sup>5</sup> we have attempted to measure the neutron spectra from the reaction  $\mathrm{He^3}(p,n)3p$  from 3° to 90° in 15° steps with collimated 10-m flight paths. The gas targets were 10-cm-long cells (one atm pressure) with  $\frac{1}{4}$ -mil Ta entrance and exit windows. We report here the 3° spectra for 14.1-MeV proton bombardment of  $\mathrm{He^3}$ ,  $\mathrm{He^4}$ , and  $\mathrm{H^2}$ . The time spectra are shown in Fig. 1. Because 14-MeV proton bombardment of  $\mathrm{He^4}$  produces no neutrons ( $E_{\mathrm{threshold}} > 20$  MeV), it serves as an excellent background

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