Influence of Polarized Optical Pumping on the Ferromagnetism of $CdCr_2Se_4$

Norman Sanford[a]
Rensselaer Polytechnic Institute, Troy, New York 12181, and GTE Laboratories Inc., Waltham, Massachusetts 02155

and

R. W. Davies, A. Lempicki,[b] W. J. Miniscalco, and S. J. Nettel[c]
GTE Laboratories Inc., Waltham, Massachusetts 02155

(Received 11 February 1983)

This Letter reports on changes in magnetization when $CdCr_2Se_4$ is subject to optical pumping. The Faraday rotation of a second beam was used as a monitor. Circularly polarized pumping was found an order of magnitude more effective than unpolarized pumping. Models using magnetic coupling mechanics of first and second order in the exchange energy, respectively, account quite well for the observations.

PACS numbers: 78.20.Ls, 72.20.My, 75.30.E+, 75.50.Dd

The subject of coupling light to the magnetic system of a semiconductor and, in particular, the possibility of inducing a phase transition by illumination has been raised several times.

It was first Lems, Bongers, and Enz, and somewhat later Veselago *et al.* and Rudov and Veselago, who suggested that the magnetism of the ferromagnetic semiconductor $CdCr_2Se_4$ could be influenced by illumination.[1-3] A sensitive response of the magnetization to circularly polarized light in diamagnetic glasses was observed by van der Ziel *et al.*[4] Circularly polarized optical pumping was similarly employed by Genken *et al.*[5] to study the magnetic response of doped $CdCr_2Se_4$. Genken *et al.* attributed their results to domain effects. All of these investigators used pickup coils and very-high-intensity laser light to detect changes in magnetization. In a recent peper, Ayadi and Ferré report using Faraday rotation to monitor photoinduced relaxation of magnetism in the spin-glass cobalt aluminosilicate.[6] The use of Faraday rotation of a weak tunable probing beam in the presence or absence of a low-power cw monochromatic pump beam completely separated intrinsic effects from heating and other artifacts.

Polarized pumping was found to have an effect an order of magnitude larger than unpolarized pumping. As we shall see, this effect is attributable to first-order magnetic coupling as compared to second-order coupling with unpolarized pumping. Apart from their intrinsic interest, these observations contribute towards an understanding of the electronic structure and magnetic coupling mechanisms of the prototype magnetic semiconductor $CdCr_2Se_4$.

The single-crystal samples of $CdCr_2Se_4$ were polished on both sides to a thickness of 25 μm and mounted on thin fused silica substrates. The material was grown by both the vapor transport and the flux methods. Samples were mounted on a copper heat sink and cooled by flowing He gas in the bore of a split-coil superconducting magnet. The probe beam was produced by linearly polarizing the output of a lamp and monochromator. A rotating analyzer followed the sample, and synchronous detection using an in-phase and quadrature lock-in amplifier allowed the direct measurement of the rotation angle. The 1.06-μm pump was derived from a cw neodymium-doped yttrium aluminum garnet laser, and overlap of pump and probe was insured by directing both through an aperture on the sample surface. It was necessary to filter the scattered pump light from the probe by use of a double monochromator following the rotating analyzer. By synchronizing the two monochromator drives, Faraday rotation spectra could be measured both with and without pump. The absolute temperature was measured to within ± 2 K and was held constant to better than $\pm \frac{1}{2}$ K. All data presented here were taken at a constant magnetic field of 5 kG applied to insure that the sample was a single magnetic domain.

Figure 1 shows Faraday rotation in degrees as a function of probe wavelength at 43 K. The lower curve is without pumping; the upper includes the effects of a coaxial *unpolarized* pumping beam at 1.06 μm and approximately 1.6 W/cm^2. Scanning the probe through the pump wavelength swamps the detection optics and hence is not shown. In the absence of optical pumping we have

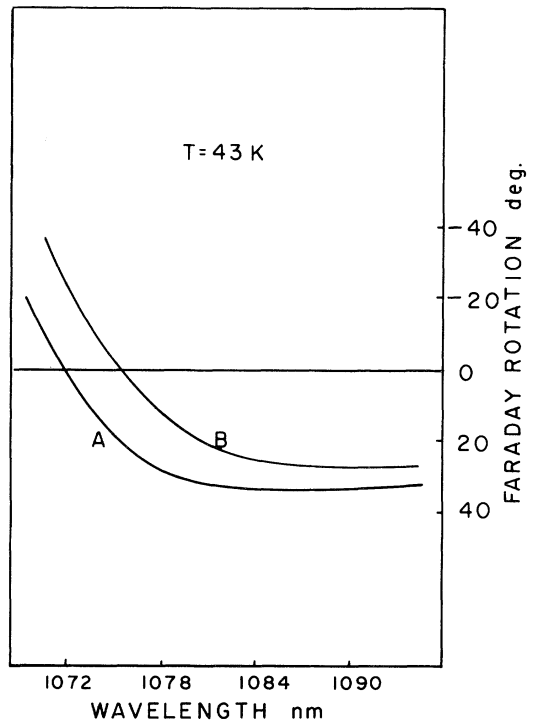

FIG. 1. Effect of unpolarized pumping on the Faraday rotation spectrum at 43 K. (a) Rotation spectrum in absence of pump excitation. (b) Rotation spectrum with coaxial pumping at a power density of 1.6 W cm^{-2}. Increased absorption at shorter wavelengths prevented resolution of rotation peak for this (50-μm-thick) sample.

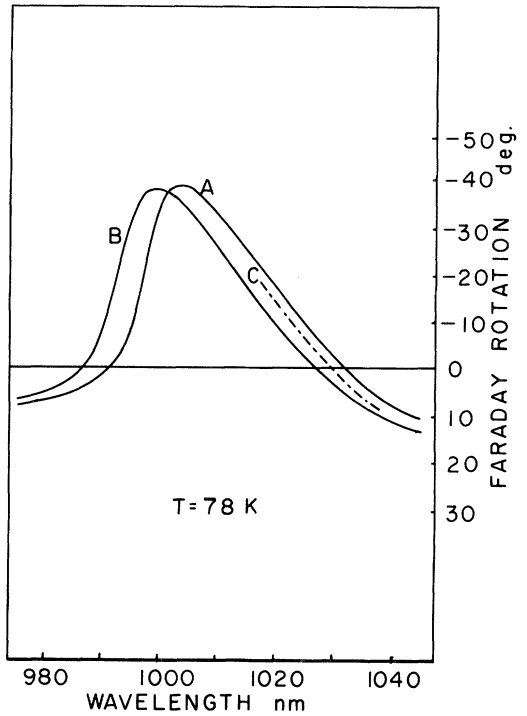

FIG. 2. Effect of circularly polarized pumping on the Faraday rotation spectrum at 78 K. (a) The displacement of the rotation spectrum to lower energy when right circularly polarized light is used as an optical pump. (b) At this temperature, left circularly polarized light at the same power level has the opposite effect. (c) Portion of rotation spectra in absence of optical pumping. For clarity only the (extremely quiet) low-energy zero crossing is shown. Increased absorption on the high-energy side of the rotation peak resulted in a rotation noise of that zero crossing equivalent to ± 5 deg. Sample thickness 25 μm, power intensity of pumping radiation 0.7 W cm^{-2}.

been able to obtain a set of Faraday rotation spectra taken at different temperatures which resemble those obtained by Kuse.[7] These latter data will be presented in a more complete follow-up article. They allow us to see that the shift to the red in Fig. 1 is produced by a change in magnetization equivalent to roughly a 5-K change of sample temperature. We note that such a red shift of the zero crossing corresponds to cooling, and precludes an explanation of the observed optomagnetism in terms of heating effects.

The pumping light corresponds to 1.17 eV. According to our present understanding of the electronic structure of the material based on photoluminescence,[8] photoemission,[9] band-structure calculations,[10] and other work,[11] this energy implies a transition from the Se p valence band to the upper Cr 3d band. The sensitive magnetic response at this frequency lends credence to our earlier conjecture that itinerant upper Cr 3d electrons are a main source of the magnetic coupling.[12] Indeed, one can make rough use of the published calculations of Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling to estimate that a shift of 5 K requires on the order of 10^{19} carriers/cm^3.[13] We estimate our carrier density from

$$n_e = Itd/\hbar\omega, \tag{1}$$

with the incoming radiation $I \approx 1.6$ W/cm^2, $t = 3 \times 10^{-3}$ sec, the absorption constant $d \approx 2200$ cm^{-1}, and $\hbar\omega = 1.17$ eV. The estimate of the carrier lifetime t is from measurements of the Faraday response under chopped optical pumping, and is in rough agreement with the response times reported by Lems, Bongers, and Enz.[1] With these numbers, we indeed obtain $n_e \approx 10^{19}$ carriers/cm^3.

Figure 2 shows the effect of *circularly polar-*

ized pumping on the Faraday spectrum at 78 K. Note that in this case the pump and probe do not interfere. Effects occur at remarkably low power levels. The observed shifts in the rotation spectra are in opposite directions depending upon the sense of the polarized pump. The origin of the offset is due to the exchange between selectively photoexcited electrons and the spin-aligned lower Cr $3d$ band, affecting the magnetization. In a saturated ferromagnetic state each of the excited electrons would see an aligned spin on all the Cr ions and interact strongly with them. Most of the photoexcited electrons will have their intrinsic spins aligned by spin-orbit interaction. If we regard the opposite circular polarizations of the pumping light as mainly populating the Cr $3d$ conduction band with oppositly aligned spins, we see that the magnetic energy of a magnetic site of spin $+\frac{3}{2}$ will, for each of the two pump polarizations, be given as

$$E = (\mu B \pm \tfrac{3}{2} J' f), \tag{2}$$

where J' is the first-order intra-Cr exchange, f is the fraction of chromium sites with excited electrons, μ is the magnetic moment of a localized Cr magnetic spin, and B is the internal magnetic field of the specimen. The internal magnetic field B contains the indirect interaction between localized chromium spins. We are suggesting that this last interaction takes place via RKKY, an interaction of second order in the Coulomb exchange.[12,13] For a given pair of localized spins this coupling is small compared to J', a first-order exchange energy. As a result, the fractional population of pumped-excited carriers per chromium site, f, is large enough for the coupling between the photoexcited spins and a chromium magnetic spin, $\frac{3}{2} J'f$ in Eq. (2), to become appreciable compared to the coupling with the internal field, the μB term.

It is Eq. (2) which must be used in conjunction with the Curie-Weiss magnetization curves[14] to compute a new self-consistent field B. With $f = 1.8 \times 10^{-3}$ [see Eq. (1)], $J' = 0.42$ eV (from the temperature dependence of the absorption edge),[15] we find a pump-induced increment in B which, further, is of a magnitude equally well produced by a temperature change of 5.1 K. Referring now to the temperature-dependent rotation spectra obtained without pumping, which, as mentioned, we have also recorded, we find a shift of +1 nm for every 1.1 K of cooling. This translates into a change of 3.2 K for the observed shift of either polarized pumping peak from the center, as shown in Fig. 2, in satisfactory agreement with the calculated 5.1-K increment. The main uncertainty is our knowledge of f.

As we depart from 78 K, where the rotation peaks are close to the fixed pump frequency, the symmetry in shifts for right- and left-polarized pumping gradually disappears because of the temperature dependence of the energy splitting between the subbands. Lastly, going back to Fig. 1, we must admit from the foregoing that the shift in wavelength of the zero crossing for unpolarized pumping could have been the result as much of unequal excitation to the two upper Cr subbands as of the true optomagnetic effect we have suggested. In conclusion, we draw attention to our verification that the observed optomagnetism is not a trivial effect due to heating, to the fact that with circular polarization it can be observed at remarkably low pumping levels, and to the likelihood that extension to higher temperatures could provide a controlled phase transition.

We are indebted to D. Guenther and B. C. McCollum for crystal growth. Helpful and stimulating discussions with E. J. Johnson, T. Collins, and J. Kafalas are gratefully acknowledged. This work was supported in part by the Air Force Office of Scientific Research under Contract No. F49620-79-C-0182.

[a]Present address: Sperry Research Center, Sudbury, Massachusetts 01776.

[b]New address: Optical Diagnostic Systems, Inc., Wayland, Massachusetts 01778.

[c]Permanent address: Rensselaer Polytechnic Institute, Troy, New York 12181.

[1]W. Lems, P. J. Bongers, and U. Enz, Phys. Rev. Lett. 21, 1643 (1968).

[2]V. G. Veselago, E. S. Vigeleva, G. I. Vinogradova, and V. T. Kalinnikov, V. E. Makhotkin, Pis'ma Zh. Eksp. Teor. Fiz. 15, 316 (1972) [JETP Lett. 15, 223 (1972)].

[3]S. G. Rudov and V. G. Veselago, Fiz. Tverd. Tela 21, 3250 (1979) [Sov. Phys. Solid State 21, 1875 (1979)].

[4]J. P. van der Ziel, P. S. Pershan, and L. D. Malmstrom, Phys. Rev. Lett. 15, 190 (1965).

[5]G. M. Genkin, Yu. N. Nozdrin, I. D. Tokman, and V. N. Shastin, Pis'ma Zh. Eksp. Teor. Fiz. 35, 162 (1982) [JETP Lett. 35, 199 (1982)].

[6]M. Ayadi and J. Ferré, Phys. Rev. Lett. 50, 274 (1983).

[7]D. Kuse, IBM J. Res. Dev. 14, 315 (1970).

[8]S. S. Yao, F. Pellegrino, R. R. Alfano, W. J. Mini-

scalco, and A. Lempicki, Phys. Rev. Lett. 46, 558 (1981).

[9]W. J. Miniscalco, B. C. McCollum, N. G. Stoffel, and G. Margaritondo, Phys. Rev. B 25, 2947 (1982).

[10]T. Kambara, T. Oguchi, and K. I. Gondaira, J. Phys. C 13, 1493 (1980).

[11]M. Heritier and P. Lederer, J. Phys. (Paris), Colloq. 41, C5-93 (1980).

[12]N. Sanford and S. J. Nettel, J. Appl. Phys. 52, 3542 (1981).

[13]D. C. Mattis, *The Theory of Magnetism* (Harper and Row, New York, 1965).

[14]C. Kittel, *Solid State Physics* (Wiley, New York, 1976), Chap. 15.

[15]G. Harbeke and H. Pinch, Phys. Rev. Lett. 17, 1090 (1966).