Magnetic quenching of time-reversed light in photorefractive diluted magnetic semiconductors

M. Dinu, I. Miotkowski, and D. D. Nolte
Physics Department, Purdue University, West Lafayette, Indiana 47907-1396
(Received 18 February 1998)

Magnetic fields selectively quench phase conjugation during photorefractive four-wave mixing experiments in the diluted magnetic semiconductor $Cd_{1-x}Mn_xTe$. Phase conjugation using circularly polarized light fields differentiates between the time-reversed and non-time-reversed components of the four-wave mixing signal. The experimental results establish the connection between the removal of time-reversal symmetry in the solid state by magnetic fields and the magnetic-field-induced quenching of time-reversed phase-conjugate light. [S0163-1829(98)08739-6]

I. INTRODUCTION

Phase conjugation is a nonlinear optical effect[1] involving coherent light beams that interact in a nonlinear optical material to generate a beam that exactly retraces the path and reconstructs the wave front of one of the incident waves, called the probe. This property suggests possible applications for phase conjugation such as beam cleanup in laser systems and distortion compensation for beam propagation in distorting media.[2–4] An often-used analogy between phase conjugation and the time reversal of light waves[5] is based on the wave-front reversal properties of phase conjugation. In the context of optics, invariance under time reversal produces symmetries in a range of optical processes, as for instance in inelastic light scattering.[6] Time reversal by phase conjugation has been shown to lead to the cancellation of the geometric Berry's phase.[5] However, the relationship between the time-reversal symmetry of a crystal Hamiltonian and macroscopic optical phenomena, in particular, phase conjugation, has only been tentatively explored. An avenue toward the clarification of the relationship between this fundamental physical property and phase conjugation is suggested by the removal of time-reversal symmetry and lifting of Kramers' degeneracy of a system upon the application of magnetic fields. The objective of this work is to assess the relationship between time reversal and phase-conjugate light by studying the influence of magnetic fields on phase conjugation.

Phase conjugation can take place through a variety of instantaneous electronic ($\chi^{(3)}$-type) nonlinear optical processes,[1,7] such as Raman and Brillouin scattering, the optical Kerr effect, two-photon absorption, or photon echoes. In contrast, the photorefractive effect,[8] which provides the mechanism for phase conjugation in the present work, is a noninstantaneous, low-intensity, and nonlocal optical nonlinearity. The photorefractive effect occurs during nonuniform illumination of a material and relies on the generation of a quasistatic space-charge field through the photoexcitation of charge carriers from deep-level defects and the subsequent transport and trapping of the carriers at deep carrier traps. The space-charge electric field, acting through the electro-optic effect, induces a refractive index grating in the material. Among the families of photorefractive materials (ferroelectric oxides, sillenites,[8] and the more recent photorefractive polymers and organic glasses[9,10]), photorefractive semiconductors constitute a category distinguished by fast response times and high sensitivity.[11]

For the study of the effects of magnetic fields on phase conjugation, diluted magnetic semiconductors stand out due to their pronounced magneto-optical properties. Diluted magnetic semiconductors are compound semiconductors (typically II-VI alloys), the cations of which are partially substituted with a transition-metal impurity, the most common being Mn^{2+}. The magnetic impurity can be incorporated in the lattice up to large alloy fractions while preserving the crystalline and band structure of the host semiconductor. As a consequence of the exchange interaction between the unpaired d-shell electrons of the Mn^{2+} ions and the s- and p-like electrons in the conduction and valence band of the host lattice, the presence of the Mn^{2+} ions leads to pronounced magneto-optical properties of the material. The most prominent of these properties are giant Zeeman splittings and giant Faraday rotations[12,13] that diluted magnetic semiconductors exhibit under moderate magnetic fields. The very large magneto-optical effects exhibited by these materials, for which CdMnTe has been most thoroughly characterized, make them an ideal material system for the study of magneto-photorefractive effects.

The effect of magnetic fields on photorefractive phenomena in diluted magnetic semiconductors has been studied previously in the case of two-wave mixing[14] and phase conjugation.[15] The use of linearly polarized beams highlighted the symmetry properties both of the electro-optic tensor (with cubic symmetry) and of the magneto-optical and photorefractive geometries. In the phase conjugation experiments, the presence of a magnetic field dramatically affected both the magnitude and the linear polarization of the phase conjugate signal.[15] Nevertheless, the experimental configuration did not permit a clear analysis of the effect of magnetic fields on the time-inversion properties of phase-conjugate light.

In this work we explore the effect of magnetic fields on phase conjugation using circularly polarized interacting beams, which are eigenmodes of propagation under magnetic fields. This experimental situation leads to the clarification of the role of magnetically induced nonreciprocity and its connection to time-reversal symmetry in phase conjugation. Section II describes a model of phase conjugation in the photorefractive diluted magnetic semiconductor CdMnTe. Section

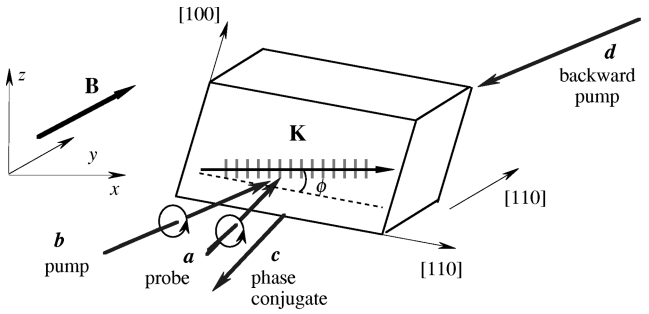

FIG. 1. The crystal orientation and beam interaction configuration for phase conjugation through four-wave mixing in a photorefractive cubic zinc-blende crystal. A magnetic field is applied along the propagation direction of the interacting beams in the longitudinal Faraday geometry.

III presents the experimental results, which are discussed in Sec. IV.

II. MAGNETO-PHOTOREFRACTIVE PHASE CONJUGATION IN CdMnTe

The physical mechanism studied here for the generation of phase conjugation is photorefractive four-wave mixing. Photorefractive phenomena are in general strongly dependent on crystal orientation and beam polarizations; these parameters must be fully specified. In the configuration shown in Fig. 1, two beams incident on the photorefractive zinc-blende crystal (probe **a** and forward pump **b**, shown as having negative helicity) write a photorefractive index grating. A third beam **d** (the backward propagating pump) diffracts off this index grating and generates the phase conjugate of the probe **a**. To satisfy the Bragg phase-matching condition for phase conjugation, the backward pump and the forward pump wave vectors must be antiparallel.

To derive the phase-conjugate beam amplitude, four interference gratings must in general be taken into account. However, in our situation the derivation of the phase-conjugate field is simplified considerably by several approximations that are easily satisfied in practice. First, the backpropagating pump is much weaker than the pump and the probe, making it possible to neglect all gratings apart from the transmission grating generated by the pump and the probe. Second, the energy transfer between the pump and the probe through two-wave mixing can also be neglected, because in our experiments it is usually only several percent. Therefore the modulation of the grating along the thickness of the crystal is constant. Third, the strength of the index grating does not vary along the y direction if the intensity at the back of the sample is above the saturation intensity.[8] Therefore the general problem of four coupled modes can be simplified to that of the backpropagating pump diffracting off the grating produced by the interference of the pump and the probe.

The presence of a magnetic field introduces an additional degree of complexity by producing circular birefringence in the crystal, through the Zeeman effect. We do not observe in the experiment, and therefore do not take into account in the model, any other influence of the magnetic field on the formation of the photorefractive grating. In the regime of relatively weak magnetic fields, where the giant Zeeman splittings are linear in the magnetization of the crystal,[16] the circular birefringence is also linear in the applied magnetic field:

$$n_- - n_+ \equiv 2\Delta n = 2V(\mathbf{B}\cdot\hat{\mathbf{k}})/k, \tag{1}$$

where $n_-(n_+)$ is the index of refraction for negative (positive) helicity light, $\mathbf{k}$ is the wave vector of light, and V is the Verdet constant of the material. When the field is along the direction of propagation of the pump and the probe, the projection of the magnetic field on either wave vector gives $\mathbf{B}\cdot\hat{\mathbf{k}} \simeq B$, because the internal half-angle between the two beams is usually small (less than 4°). In the presence of a magnetic field applied along the direction of propagation of light (in the so-called Faraday magneto-optical geometries[14,15]), the propagation eigenmodes of light are circularly polarized beams; therefore, in the following we will use a circular polarization representation for the interacting beams.

A. Four-wave mixing

The photorefractive effect is strongly dependent on the crystal orientation due to the tensorial nature of the electro-optic coefficient. For cubic zinc-blende crystals in the standard holographic cut (shown in Fig. 1), the anisotropic nature of two-wave beam coupling is described[17] by means of a Jones matrix M_ϕ. In the circular polarization representation it has the nonunitary Hermitian form

$$M_\phi \equiv \begin{pmatrix} M_\phi^{--} & M_\phi^{-+} \\ M_\phi^{+-} & M_\phi^{++} \end{pmatrix} = \begin{pmatrix} s_\phi/2 & 3c_\phi^2 s_\phi - s_\phi/2 + ic_\phi(3c_\phi^2-2) \\ 3c_\phi^2 s_\phi - s_\phi/2 - ic_\phi(3c_\phi^2-2) & s_\phi/2 \end{pmatrix}. \tag{2}$$

Here the components of the matrix are indicated by the superscripts, and we have used the abbreviations $s_\phi = \sin\phi$ and $c_\phi = \cos\phi$, where ϕ is the angle between the [110] crystalline axis and the direction of the space charge field $\mathbf{E}_{sc}$, which is in the direction of the grating vector $\mathbf{K}$. In the coordinate system depicted in Fig. 1, the space-charge electric field $\mathbf{E}_{sc}$ is oriented along the x axis and the evolution of the phase-conjugate beam **c** along the thickness of the crystal y is

$$\frac{d}{dy}\begin{pmatrix} c_- \\ c_+ \end{pmatrix} = \frac{ikn^3 r_{41}}{2\cos\theta_{in}} \frac{i\xi E_D}{1+E_D/E_q} \frac{(\mathbf{b}\cdot\mathbf{a}^*)}{I_a+I_b} M_\phi \begin{pmatrix} d_- \\ d_+ \end{pmatrix} \tag{3}$$

in the absence of a magnetic field.

In Eq. (3) $c_-(c_+)$ is the negative (positive) helicity component of the phase-conjugate beam. The refractive index is n, r_{41} is the electro-optic coefficient, θ_{in} is the internal half-angle between the pump and the probe, $E_D=2\pi k_B T/e\Lambda$ is the diffusion field, and E_q is the space-charge limiting field.[8] The factor ξ takes into account electron-hole competition;[18] in the single-carrier case $\xi=1$ for electrons and $\xi=-1$ for holes.

B. Magneto-phase-conjugation

Equation (3) gives the rate of change in the phase-conjugate beam amplitude at a given coordinate y along the thickness of the crystal. When the application of a magnetic field induces circular birefringence, the electric-field vectors of the positive and negative helicity components experience different phase delays, which can be described by appropriate Jones matrices. At the front face of the crystal the increment in the phase-conjugate amplitude is

$$\left.\frac{d}{dy}\begin{pmatrix} c_- \\ c_+ \end{pmatrix}\right|_{y=0} \propto \frac{(\mathbf{b}\cdot\mathbf{a}^*)}{I_a+I_b}\begin{pmatrix} \exp(in_-ky) & 0 \\ 0 & \exp(in_+ky) \end{pmatrix} M_\phi \begin{pmatrix} \exp[in_-k(L-y)] & 0 \\ 0 & \exp[in_+k(L-y)] \end{pmatrix}\left.\begin{pmatrix} d_- \\ d_+ \end{pmatrix}\right|_{y=L}, \tag{4}$$

where L is the thickness of the crystal and $n_-(n_+)$ is the index of refraction for the negative (positive) helicity. The dependence of the photorefractive grating amplitude on the pump and probe intensity can be dropped because the ratio between the pump and the probe intensity is constant in a first approximation. The phase-conjugate field at the front face of the crystal is obtained by integrating Eq. (4),

$$\begin{pmatrix} c_- \\ c_+ \end{pmatrix} = \int_0^L \frac{d}{dy}\left.\begin{pmatrix} c_- \\ c_+ \end{pmatrix}\right|_{y=0} dy, \tag{5}$$

to yield

$$\left.\begin{pmatrix} c_- \\ c_+ \end{pmatrix}\right|_{y=0} \propto \frac{(\mathbf{b}\cdot\mathbf{a}^*)}{I_a+I_b}\begin{pmatrix} M_\phi^{--}\exp(iVBL) & M_\phi^{-+}\mathrm{sinc}(VBL) \\ M_\phi^{+-}\mathrm{sinc}(VBL) & M_\phi^{++}\exp(-iVBL) \end{pmatrix}\left.\begin{pmatrix} d_- \\ d_+ \end{pmatrix}\right|_{y=L}, \tag{6}$$

with $\mathrm{sinc}(x)\equiv\sin x/x$. The intensities of the two polarization components at the front of the crystal are

$$|c_-|^2 \propto \left|\frac{s_\phi}{2}\right|^2 |d_-|^2 + \left|3c_\phi^2 s_\phi - \frac{s_\phi}{2} + ic_\phi(3c_\phi^2-2)\right|^2 \times \mathrm{sinc}^2(VBL)|d_+|^2 \tag{7a}$$

$$|c_+|^2 \propto \left|3c_\phi^2 s_\phi - \frac{s_\phi}{2} - ic_\phi(3c_\phi^2-2)\right|^2 \mathrm{sinc}^2(VBL)|d_-|^2 + \left|\frac{s_\phi}{2}\right|^2 |d_+|^2 \tag{7b}$$

In the preceding derivation we have not explicitly considered the effect of a finite absorption coefficient, nor of circular dichroism, on the phase-conjugate components. Both absorption and circular dichroism can be formally introduced into Eq. (4) by substituting

$$n_- \rightarrow n_- - i\alpha_-/2k \tag{8}$$

with a similar equation for the positive helicity. Absorption only leads to an overall $\exp(-\alpha_0 L)$ factor in the phase-conjugate components, and circular dichroism changes their dependence on the magnetic field. From the experiments, we can assess an upper limit on the circular dichroism $|\alpha_- - \alpha_+|/\alpha_0 \leqslant 0.05$ for a field $B=4T$. We will neglect absorption in the following sections, since its only effect is to scale all components of the phase-conjugate signal by a common factor.

The focus of our interest is how the phase-conjugate light depends on the applied magnetic field and on the crystal symmetry and orientation. For this purpose, we will make the distinction between the component of the phase-conjugate beam that has the same helicity as the probe, which we will call the ''time-reversed'' beam, and the component with the orthogonal helicity. This distinction is consistent with the terminology used for vector phase conjugation.[19,20]

The phase-conjugate beam depends on the crystal orientation through the angle ϕ, and on the magnetic field through the $\mathrm{sinc}^2(VBL)$ factor. Even in the case when the backward propagating pump has a well-defined circular polarization, the phase-conjugate beam contains polarizations of both helicities. In practical phase conjugation setups, there are two important situations, depending on whether the backward pump has the same or opposite circular polarization as the forward pump and probe.

C. Same helicity back pump

When the three beams have the same (negative) helicity, $\mathbf{d}\equiv d_-$, the phase-conjugate components are

$$|c_-|^2 \propto |M_\phi^{--}|^2 |d_-|^2, \tag{9a}$$

$$|c_+|^2 \propto |M_\phi^{+-}|^2\ \mathrm{sinc}^2(VBL)|d_-|^2. \tag{9b}$$

The intensity of the component of the phase conjugate with the same polarization as the incident probe does not depend on the applied magnetic field, while the orthogonal

polarization component vanishes for several specific values of the magnetic field, when $VBL=(N+1/2)\pi$. This is a case where the ''non-time-reversed'' (opposite polarization) component of the phase-conjugate beam is quenched by the applied magnetic field. Although four-wave mixing generally does not preserve polarization in photorefractive crystals, in this arrangement, for $VBL=(N+1/2)\pi$, the phase-conjugate beam has a pure polarization state, which is the same as that of the incident probe.

D. Opposite helicity back pump

An important experimental situation is the one where the backward pump **d** is generated by the retroreflection of the pump **b**. If the incident beams **a** and **b** have negative helicity, then **d** has positive circular polarization upon reflection. Eqs. (7) then are

$$|c_-|^2 \propto \left| 3c_\phi^2 s_\phi - \frac{s_\phi}{2} + i c_\phi (3c_\phi^2 - 2) \right|^2 \times \mathrm{sinc}^2(VBL)|d_+|^2, \tag{10a}$$

$$|c_+|^2 \propto \left| \frac{s_\phi}{2} \right|^2 |d_+|^2. \tag{10b}$$

The applied magnetic field has no influence on the polarization component that is orthogonal to the incident polarization. On the other hand, the component of the phase-conjugate signal that has the same polarization as the incident probe (the ''time-reversed'' component of the phase conjugate) is quenched in the presence of the magnetic field for $VBL=(N+1/2)\pi$. Therefore, the magnetic field affects the components of the phase-conjugate beam in opposite manners depending on the helicity of the backward-propagating pump.

E. Reciprocity, Bragg matching, and time reversal

The effect of magnetic fields on phase-conjugate light is closely related to the nonreciprocal nature of magnetic circular birefringence. The influence of natural circular birefringence (optical activity) on phase conjugation by four-wave mixing has been the subject of some study,[21,22] but the link between either type of circular birefringence and reciprocity or time reversal has not been explicitly analyzed. Reciprocity imposes the relationship[23] on the scattering coefficient $S(\mathbf{k},\mathbf{k}')$ connecting an incident wave with wave vector $\mathbf{k}$ to a scattered wave $\mathbf{k}'$, given by

$$S(\mathbf{k},\mathbf{k}') = S(-\mathbf{k}', -\mathbf{k}), \tag{11}$$

which is a statement of the invariance of the scattering process under motion reversal, i.e., reversing the direction of propagation. Reciprocity is sufficient for the process of optical phase conjugation.

A simple example is the case of plane waves, which undergo reciprocal propagation in optically active media but nonreciprocal propagation in the presence of magnetic circular birefringence. The scattering matrix for the phase-conjugate wave at coordinate y [from Eq. (5)] is

$$S(\mathbf{k}_d,\mathbf{k}_c) = \begin{pmatrix} \exp(in_- k_c y) & 0 \\ 0 & \exp(in_+ k_c y) \end{pmatrix} M_\phi \begin{pmatrix} \exp[in_- k_d (L-y)] & 0 \\ 0 & \exp[in_+ k_d (L-y)] \end{pmatrix}, \tag{12}$$

which becomes

$$S(\mathbf{k}_d,\mathbf{k}_c) = \exp(in_0 kL) \begin{pmatrix} M_\phi^{--} \exp[i\Delta n(\mathbf{k})kL] & M_\phi^{-+} \exp[i\Delta n(\mathbf{k})k(2y-L)] \\ M_\phi^{+-} \exp[-i\Delta n(\mathbf{k})k(2y-L)] & M_\phi^{++} \exp[-i\Delta n(\mathbf{k})kL] \end{pmatrix} \tag{13}$$

in the paraxial approximation, after expansion ($\mathbf{k}\equiv\mathbf{k}_a$). Upon change of direction of propagation $\mathbf{k}\rightarrow-\mathbf{k}$, an externally applied magnetic field transforms the refractive index as $\Delta n(\mathbf{k})=-\Delta n(-\mathbf{k})$ from Eq. (1). Therefore the scattering matrix is not invariant under change of direction of propagation. The presence of magnetic fields leads to nonreciprocal generation of phase conjugate beams. This is in contrast with the case of natural optical activity, where the sign of $\Delta n(\mathbf{k})$ does not depend on the direction of propagation and which therefore satisfies the reciprocity relation.

The effect of nonreciprocity on the phase-conjugate beam can be analytically understood in terms of the Bragg condition that must be satisfied in four-wave mixing, $\mathbf{k_a}+\mathbf{k_c}=\mathbf{k_b}+\mathbf{k_d}$ (Fig. 2), so that the contributions to the phase-conjugate beam along the thickness of the crystal add constructively. Circular birefringence can modify the magnitudes of the in-

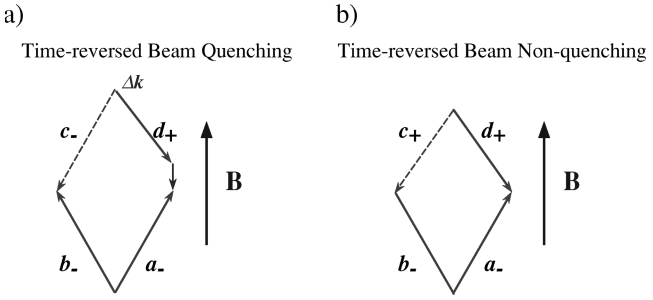

FIG. 2. A magnetic field can lead to the selective destruction of the Bragg phase-matching condition ($\mathbf{k_a}+\mathbf{k_b}=\mathbf{k_c}+\mathbf{k_d}$) in photorefractive phase conjugation: (a) for the time-reversed component of the phase conjugate $\mathbf{c}_-$ the circular birefringence induced by the magnetic field leads to a wave-vector mismatch $\Delta\mathbf{k}$ which gives rise to the characteristic sinc^2 dependence of its intensity with field, while (b) for the orthogonal polarization $\mathbf{c}_+$ the phase-matching condition is unaffected.

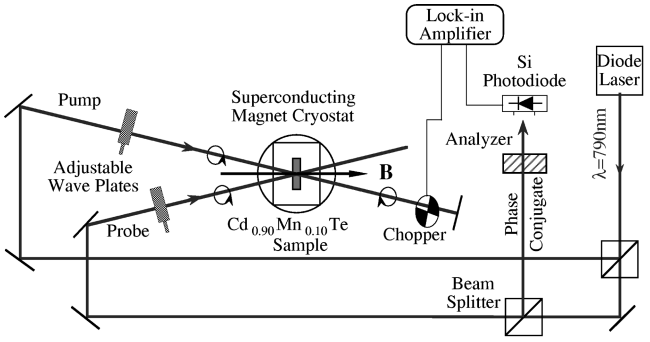

FIG. 3. The standard retroreflection experimental setup used in photorefractive phase conjugation through degenerate four-wave mixing. The circular polarization states of the interacting waves are indicated in the figure.

teracting wave vectors, leading to a wave-vector mismatch $\Delta\mathbf{k}$ that destroys the Bragg phase-matching condition and leads to the characteristic $\mathrm{sinc}^2(VBL)$ dependence of the diffracted beam intensity in Eq. (10).

The connection with time reversal comes about through the nonreciprocal nature of magnetic circular birefringence (the Faraday effect), which distinguishes it from natural circular birefringence (optical activity), which is a reciprocal effect. In contrast with the Bragg mismatch in uniaxial birefringent crystals, which can be canceled by tilting the pump beams or crystal, the Bragg mismatch due to magnetic-field-induced circular birefringence *cannot be compensated by angular tilt* while still preserving the direction of the phase-conjugate beam wave vector.

The analogy of phase conjugation with time-reversed light[7] is mathematically rigorous when the transformation of the scattering matrix under motion reversal is identical with the transformation under time reversal.[24] This is possible when the scatterer (composed of the scattering crystal plus external fields) is invariant under time reversal. The crystal Hamiltonian of an optically active crystal is symmetric under time reversal, and therefore motion reversal is always equivalent to time reversal. However, the crystal Hamiltonian of a paramagnetic crystal in a magnetic field is not symmetric under time reversal. Therefore, the transformations of the scattering matrix under motion reversal and time reversal are in general different. In this case, motion reversal is not equivalent to time reversal, except under specific conditions (crystal orientation and light polarizations) for which the transformations are accidentally degenerate (as in the case of the same helicity back pump described in Sec. II C).

In our experiments on four-wave mixing in CdMnTe crystals as a function of applied magnetic field, we have used a back pump that had the opposite helicity relative to the forward pump and signal polarizations, described in Sec. II D. In this configuration, and for the crystal orientations that we choose, the transformations under motion reversal and time reversal are not equivalent, and the phase-conjugate beam is continuously quenched with increasing magnetic field.

III. EXPERIMENT

We have investigated the effect of magnetic fields on phase-conjugate light in photorefractive four-wave mixing experiments using a standard retroreflection setup. The experimental arrangement is shown in Fig. 3. We used a nominally undoped CdMnTe crystal (10% Mn concentration), oriented in the standard holographic cut and with a thickness of $L=2.7$ mm. The coherent light source was a 45 mW diode laser emitting in the near infrared at a wavelength of 790 nm. This wavelength is close to the band gap of $Cd_{0.90}Mn_{0.10}Te$ (1.69 eV at the working temperature of 220 K), which ensures that we are in a regime where the magneto-optical effects are significant.

The photorefractive index grating is produced by the interference of the pump and the probe, which have negative helicity (right-handed circular polarization) upon incidence on the sample. The total intensity incident on the sample, corrected for all Fresnel losses, was $\approx 100\ \mathrm{mW/cm^2}$. The ratio of the probe and pump intensity was about 0.47, which gives a value $m=0.93$ for the modulation of the grating. The period of the interference grating was $\Lambda \equiv 2\pi/K = 2.5\ \mu\mathrm{m}$.

The backward-propagating pump was produced by retroreflecting the pump beam transmitted through the sample, giving it positive helicity. The phase-conjugate beam generated by photorefractive four-wave mixing in the CdMnTe sample propagates back along the path of the probe and is detected using a silicon photodiode. The circular polarization components of the phase-conjugate signal are distinguished using an analyzer placed in front of the detector. The backpropagating pump beam was mechanically chopped and the detected phase-conjugate signal was measured using a lock-in amplifier referenced to the chopper frequency.

The limited coherence length of the diode laser (about 10 cm) ensures that the backpropagating pump is not coherent with the writing beams and therefore the mode-coupling equations in the previous section are valid. Also, due to the finite absorption of the sample ($\approx 6\ \mathrm{cm^{-1}}$) and to the Fresnel losses on the cryostat windows and sample surfaces, the backward pump is relatively weak and its effect (erasure) on the index grating can be neglected.

To apply magnetic fields on the crystal, the sample was placed in a superconducting magnet cryostat. The temperature of the CdMnTe crystal placed in the helium-flow sample chamber could be varied from liquid-helium temperatures to room temperature. The sample temperature was maintained at $T=220$ K throughout the experiments in order to maximize the strength of the photorefractive coupling by minimizing electron-hole competition. Also, the low temperature ensures the freeze-out of the free carriers thermally generated from shallow defects, which tend to erase the space-charge grating. These effects combine to produce a maximum photorefractive gain around the temperature $T=250$ K.[14]

To compare the measured dependence of the phase-conjugate intensity on the magnetic field with the predicted dependence, the Verdet constant was determined separately. Crossed linear polarizers were placed in the probe beam in front of and after the cryostat and the transmission of the sample was measured as a function of the applied magnetic field. This gave a Verdet constant $V=935$ deg/T cm at the operating temperature and wavelength.

The numerical value of the phase-conjugate reflectivity (defined as the ratio between the phase-conjugate beam and incident probe beam intensities) depends on the refractive index modulation of the photorefractive grating. Typically in our experimental situations $\Delta n \approx 1.5\times 10^{-6}$. The maximum

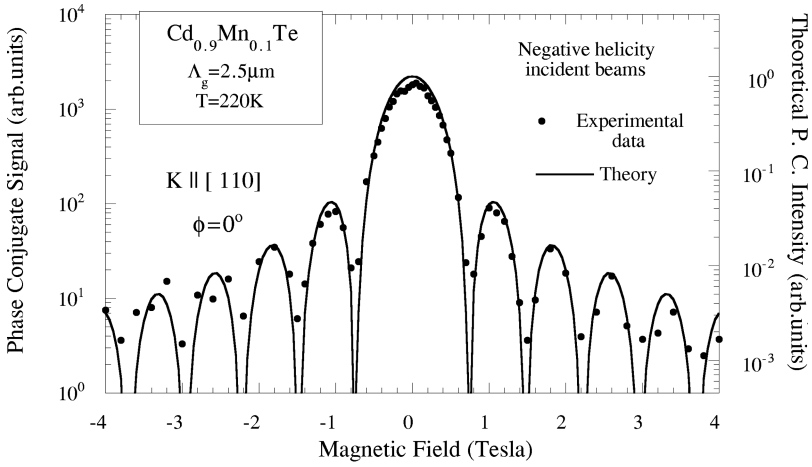

FIG. 4. The dependence of the phase conjugate intensity as a function of the magnetic field when the grating vector is along the [110] crystallographic orientation. The dots represent the experimental data while the continuous line is the calculated dependence within a multiplicative factor.

phase-conjugate reflectivity is correspondingly small, about $R=2\times10^{-6}$, which has been corrected for reflection losses. A measure of the accuracy of phase conjugation (wave-front reversal) is the fidelity of phase conjugation.[25,26] For complicated wave fronts, phase conjugation is generally only partial. In our experiments, phase conjugation fidelity could not be easily quantified due to the weak four-wave mixing signal. However, we verified the phase-conjugate nature of the diffracted beam by checking that the introduction of a mild phase aberrator (a long focal length lens) in the probe beam does not alter the magnitude of the signal.

We have investigated the dependence of the phase-conjugate beam intensity on crystal orientation and applied magnetic field for three different crystal orientations, defined by the direction of the grating vector **K** with respect to important crystallographic directions. The three orientations considered exhibit well-defined beam coupling properties for specific incident polarizations and are standard in photorefractive mixing experiments.

The first crystal orientation we examined had the grating vector **K** parallel to the [110] crystallographic axis. Equations (10) show that in this crystal orientation, where the angle $\phi=0°$, the phase conjugate consists entirely of the negative helicity component. The measured total phase-conjugate intensity is plotted as a function of the magnetic field in Fig. 4 on a logarithmic scale; the data are experimental and the curve is the calculated dependence of the phase conjugate on the magnetic field, within a multiplicative factor. The experimental data show that for this orientation the total phase-conjugate intensity is quenched by the magnetic field according to the predicted sinc^2 function from Eq. (10a).

The component of the phase conjugate that can be considered to be time-reversed has negative helicity in this experiment, because circular polarization is preserved upon time reversal and we prepared the signal beam in a negative helicity state. Therefore in this orientation the phase-conjugate beam only contains the component that is time reversed with respect to the incident probe. For all other crystal orientations both polarization components of the phase conjugate are present.

For the grating vector oriented along the [111] crystallographic axis, the dependence of the phase-conjugate intensity on magnetic field is shown in Fig. 5. The measured total phase-conjugate intensity (dots) and the negative helicity component of the phase-conjugate beam (circles) are both plotted as a function of field. The component of the phase-conjugate beam having a polarization identical to that of the incident beams is again quenched by the magnetic field with the $\mathrm{sinc}^2(VBL)$ dependence. The positive helicity component of the phase conjugate is the field-independent baseline which persists up to high magnetic fields. This component

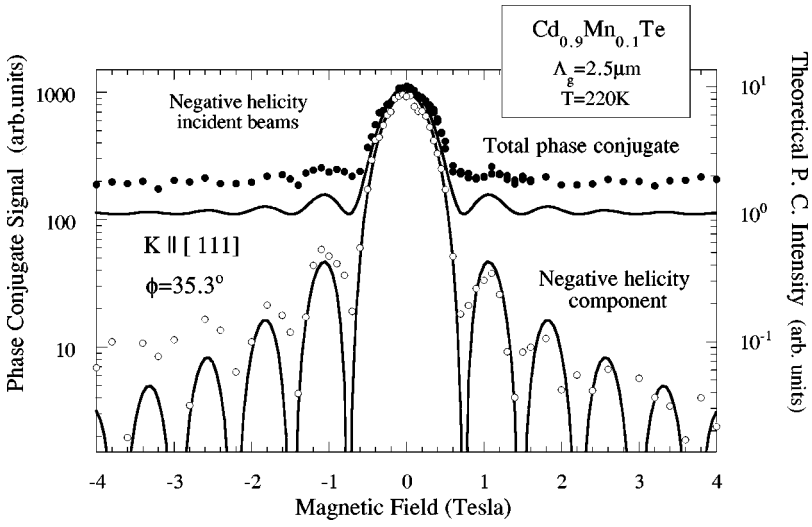

FIG. 5. The dependence of the phase conjugate as a function of the magnetic field when the grating vector is along the [111] crystallographic orientation. The points are the measured values of the phase-conjugate intensity (dots, total phase conjugate; circles, the negative helicity component) while the continuous lines are the calculated dependence for this orientation, up to a constant factor.

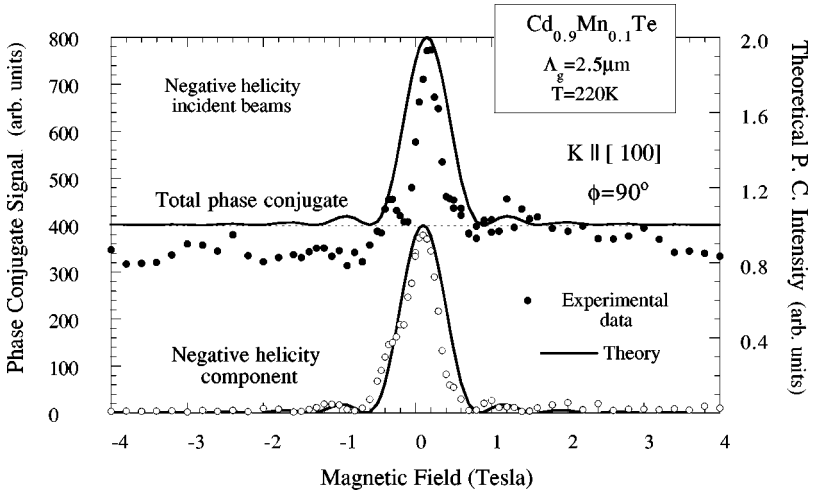

FIG. 6. The dependence of the phase conjugate as a function of the magnetic field when the grating vector is along the [100] crystallographic orientation. The measured phase-conjugate intensity (dots for the total phase conjugate; circles for the negative helicity component) is plotted together with the theoretical dependence.

remains constant and is unaffected by the presence of the magnetic field. The magnitude of the baseline in the predicted curve is smaller than in the experiment, perhaps because the ratio between the intensities of the two polarization components of the phase conjugate depends sensitively on the orientation angle ϕ. Therefore, the effect seen in the figure may be due to a small misorientation of the grating vector around the [111] direction.

This experimental condition represents a situation where an applied magnetic field only affects that component of the phase-conjugate light that constitutes the time-reversed replica of the incident probe, while the magnetic field has no effect on the orthogonal polarization component of the phase conjugate.

A further experimental verification that the time-reversed component is quenched under a magnetic field while the orthogonal polarization remains constant is depicted in the linear-scale plot in Fig. 6 for the case where the grating vector is along the [100] crystallographic direction. For this orientation the data have more scatter due to larger noise. The deviation of the total phase-conjugate signal from a flat baseline at high fields is attributed to drift during the measurement. The negative helicity (time-reversed) component is again quenched with the characteristic $\mathrm{sinc}^2(VBL)$ dependence, but the positive helicity component remains field independent up to high magnetic fields.

IV. CONCLUSIONS

In this paper, we have explored the analogy between optical phase conjugation and time-reversed light and have shown that it is valid only when the transformations of the scattering matrix describing four-wave mixing are identical under motion reversal and time reversal. The success of this analogy over the past two decades has rested on the invariance of most optical crystals (excluding magnetic crystals) to time inversion. In the presence of finite crystal magnetization the crystal is no longer invariant under time reversal, and the analogy between optical phase conjugation and time-reversed light breaks down, except under accidental conditions.

We have experimentally demonstrated the breakdown of the time-reversal analogy through the application of a magnetic field on a diluted magnetic semiconductor during four-wave mixing in a Faraday geometry using circularly polarized light. Rather than exhibiting a discontinuous violation of the time reversal analogy, the time-reversed signal is quenched as a continuous function of applied field that decreases second order in the magnetic field for small field strengths. The quenching of the time-reversed light can be described as a continuous Bragg misalignment with increasing magnetic field, but differs in a fundamental way from Bragg misalignment in optically active crystals. The Bragg misalignment caused by magnetic fields cannot be corrected by rigid rotation of the crystal or pump beams through small angles, and therefore represents a fundamental modification of the phase-conjugate signal caused by the magnetic field.

ACKNOWLEDGMENT

D. D. Nolte acknowledges support by National Science Foundation Grant No. ECS-9708230.

[1] A. Yariv and R. A. Fisher, in *Optical Phase Conjugation*, edited by R. A. Fisher (Academic, New York, 1983).
[2] A. Yariv, D. Fekete, and D. M. Pepper, Opt. Lett. **4**, 52 (1979).
[3] S. Watanabe, T. Chikama, G. Ishikawa, T. Terahara, and H. Kuwahara, IEEE Photonics Technol. Lett. **5**, 1241 (1993).
[4] H. Bruesselbach, D. C. Jones, D. A. Rockwell, R. C. Lind, and G. Vogel, J. Opt. Soc. Am. B **12**, 1434 (1995).
[5] W. R. Tompkin, M. S. Malcuit, R. W. Boyd, and R. Y. Chiao, J. Opt. Soc. Am. B **7**, 230 (1990).
[6] A. K. Ramdas and S. Rodriguez, J. Phys. Colloq. **42**, 887 (1981).
[7] B. Y. Zeldovich, V. I. Popovichev, V. V. Ragulskii, and F. S. Faizullov, Zh. Eksp. Teor. Fiz. Pis'ma Res. **15**, 160 (1972) [JETP Lett. **15**, 109 (1972)].
[8] G. C. Valley and J. F. Lam, in *Photorefractive Materials and Their Applications I*, edited by P. Günter and J.-P. Huignard (Springer-Verlag, Berlin, 1988).
[9] Stephen Ducharme, J. C. Scott, R. J. Twieg, and W. E. Moerner, Phys. Rev. Lett. **66**, 1846 (1991).

[10] P. M. Lundquist, R. Wortmann, C. Geletneky, R. J. Twieg, M. Jurich, V. Y. Lee, C. R. Moylan, and D. M. Burland, Science **274**, 1182 (1996).
[11] A. M. Glass and J. Strait, in *Photorefractive Materials and Their Applications I*, edited by P. Günter and J.-P. Huignard (Springer-Verlag, Berlin, 1988).
[12] R. L. Aggarwal, S. N. Jasperson, P. Becla, and J. K. Furdyna, Phys. Rev. B **34**, 5894 (1986).
[13] D. U. Bartholomew, J. K. Furdyna, and A. K. Ramdas, Phys. Rev. B **34**, 6943 (1986).
[14] R. S. Rana, Eunsoon Oh, K. Chua, A. K. Ramdas, and D. D. Nolte, Phys. Rev. B **49**, 7941 (1994).
[15] R. S. Rana, M. Dinu, I. Miotkowski, and D. D. Nolte, Opt. Lett. **20**, 1238 (1995).
[16] J. K. Furdyna, J. Appl. Phys. **64**, R29 (1988).
[17] J. Strait, J. D. Reed, and N. V. Kukhtarev, Opt. Lett. **15**, 209 (1990).
[18] F. P. Strohkendl, J. M. C. Jonathan, and R. W. Hellwarth, Opt. Lett. **11**, 312 (1986).
[19] A. Brignon, P. Sillard, and J.-P. Huignard, Appl. Phys. B: Lasers Opt. **63**, 537 (1996).
[20] M. J. Damzen, S. Camacho-Lopez, and R. P. M. Green, Phys. Rev. Lett. **76**, 2894 (1996).
[21] T. Sawada and K. Ujihara, IEEE J. Quantum Electron. **QE-25**, 1937 (1989).
[22] T. J. Hall, A. K. Powell, and C. Stace, Opt. Commun. **75**, 159 (1990).
[23] Claire Gu and Pochi Yeh, Opt. Lett. **16**, 455 (1991).
[24] R. G. Sachs, *The Physics of Time Reversal* (The University of Chicago Press, Chicago, 1987).
[25] J. Falk, Opt. Lett. **7**, 620 (1982).
[26] Claire Gu and Pochi Yeh, Opt. Commun. **107**, 353 (1994).