Imaging the spin Hall effect of light inside semiconductors via absorption

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The opposite transverse shifts for the right and left circular polarization components of a 100 fs 820 nm linearly polarized pulse focused onto GaAs are observed *in situ* via absorption. A time-delayed normally incident probe pulse scanned across the excitation spot detects the differential circular dichroism associated with the pump-induced transfer of spin angular momentum from light to electrons. More generally, we show that for a nonnormally incident probe, one can observe the spin Hall effect for probe light through a variety of pump-induced changes to a material's optical properties. © 2009 Optical Society of America

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The spin Hall effect of light (SHEL) [1], so called because of its resemblance to the electronic spin Hall effect in solids, refers to the transverse splitting of a beam of finite lateral extent into its right (σ^+) and left (σ^-) circularly polarized components when it crosses an optical interface. Since the original proposal of this effect by Fedorov [2] and a demonstration by Imbert [3] in the case of total internal reflection, considerable theoretical [1,4–9] and experimental [9–11] works based on a variety of beam geometries and/or material structures have been reported. However, these studies consider only optical interfaces between transparent media, with absorption seen as hampering the ability to observe or utilize the SHEL, especially for an emergent beam on which measurements have typically been made. In this Letter, we show that the SHEL can be observed *in situ* by spatially resolved differential *absorption* measurements in a prototypical semiconductor, GaAs. A linearly polarized ultrashort pump pulse is tightly focused into an optically thin GaAs sample to generate transversely shifted ensembles of spin up (↑) and spin down (↓) electrons via the SHEL. Unlike nonresonant interactions, the transfer of light angular momentum to carriers in a semiconductor might lead to applications in areas such as spintronics. We observe the shift by scanning a time-delayed probe beam across the excitation region and measuring the differential pump-induced circular dichroism. Because the refractive index of GaAs ($n=3.6$) is much higher than that (1.5) of glass, the medium mainly used to study the SHEL to date—larger transverse shifts of up to 200 nm—are found, depending on the pump polarization and the angle of incidence. More generally, we show that the SHEL can be observed for a fixed beam geometry by varying the orientation of the sample and detecting any pump-induced transmission change in probe σ^+ and σ^- components.

A schematic of the pump and probe beam geometry is shown in Fig. 1(a). The sample is an 800-nm-thick [100]-oriented GaAs specimen (bandgap energy $=1.42$ eV at 295 K) mounted on a glass substrate. The $\lambda=820$ nm (1.50 eV) 100 fs pulses are obtained from a mode-locked Ti:sapphire laser of 76 MHz repetition rate. We define an $x-y-z$ Cartesian system with the $+z$ direction normal to the sample surface on the air side and the y direction parallel to the sample surface and transverse to the plane of incidence ($x-z$) of a pump beam. Pump pulses of 1.5 mW average power are focused onto the sample using an aspheric

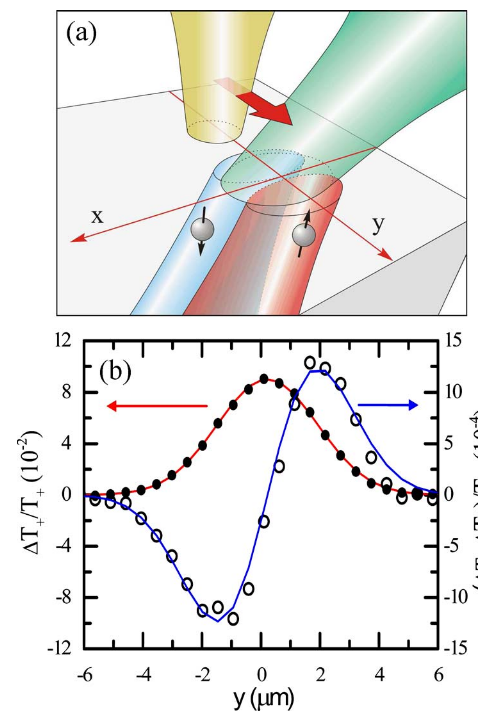

Fig. 1. (Color online) (a) Experimental geometry used in observing SHEL via absorption in GaAs. The pump beam incident in the $x-z$ plane is transversely split into σ^+ and σ^- components, generating separated spin up and down electron populations; these are probed by a normally incident beam across the excitation spot. (b) Measured normalized change in transmission (solid dots) of the σ^+ polarized component of the probe beam as it is scanned along the y direction across the excitation region of a p-polarized pump beam. Also shown (open dots) is the differential transmission for the circularly polarized components. The solid curves are fits based on a Gaussian function and first derivative of a Gaussian function.

lens with an NA, NA=0.16. For an energy absorption coefficient of $\sim 1.3\times 10^4\ \mathrm{cm}^{-1}$, the pump pulse resonantly generates an estimated peak carrier density of $2\times 10^{18}\ \mathrm{cm}^{-3}$. In the initial experiments, pump pulses are incident at an angle of $\theta_i = 55°$ relative to the normal, which is the largest deflection allowed by our experimental configuration. Linearly polarized probe pulses with $\leqslant 1$ mW average power are focused with an aspheric lens (NA=0.25) at normal incidence on the sample. A piezotransducer allows for the transverse scanning of the probe focus along the y direction, across the excitation spot. The probe light transmitted through the GaAs is collimated with a wide aperture lens (NA=0.3) and passed through a quarter-wave plate. A Wollaston prism in combination with two photodiodes serves as a polarization bridge to allow the extraction of the transmitted intensity, $T_\pm$, of σ^+ and σ^- probe beam components as well as their change $\Delta T_\pm$ owing to pump-induced circular dichroism.

A particular probe pulse is delayed from a pump pulse by 2 ps, a time that is greater than the carrier cooling time of 500 fs [12] but less than the electron spin relaxation time of >30 ps [13]; any hole spin orientation in GaAs is expected to have completely decayed in $\leqslant 1$ ps [14]. Figure 1(b) displays the transmission changes experienced by a y-scanned linearly polarized probe pulse for a p-polarized pump pulse. Maximum fractional transmission changes of $\Delta T_\pm / T_\pm = 0.1$ are observed for spatially overlapped pump and probe pulses centered at $y=0$. Bleaching signals of this strength are expected, since the probe beam is coupled to electrons only ~ 90 meV above the conduction band edge. The $\Delta T_+/T_+$ data are represented well by a Gaussian function with a width (FWHM) of $4.2\pm 0.5\ \mu$m, which is consistent with the spatial convolution of the focused pump and probe spot sizes. This is close to the diffraction limit for our focusing optics. A pronounced dispersivelike curve is seen in Fig. 1(b) for the differential transmissivity, $[\Delta T_- - \Delta T_+]/T_+$, with a zero crossing at $y=0$. We assign this circular dichroism to a preferential spin orientation of electrons with σ^+ preferentially producing $\downarrow$ electrons while σ^- preferentially produces $\uparrow$ electrons. We have observed the circular dichroism for various probe delay times following pump excitation. As expected, the dichroism decays with a time constant of >30 ps, reflecting the net electron spin relaxation and diffusion. For our pump/probe wavelength, photons couple the conduction band to heavy and light hole bands but not the spin-orbit split-off band, for which the energy separation from the conduction band is 1.72 eV. In such a case, a net electron spin population is generated in the conduction band, resulting in an altered transmission for σ^+ and σ^- probe beam components through state filling effects [13]. Qualitatively, the results in Fig. 1(b) are indicative of $\uparrow$ electrons being preferentially generated for $y<0$, while $\downarrow$ electrons are centered at $y>0$. This spin separation occurs because of the SHEL-induced transverse separation of the σ^+ and σ^- pump beam components at the air–GaAs interface.

To quantify the displacement of the centers of the $\uparrow$ and the $\downarrow$ electron populations, we represent the $\Delta T_\pm / T_\pm$ data with 1D Gaussian functions $G(y\pm\delta/2)$ for $G(y)\propto \exp(-y^2/w^2)$ and $w \gg \delta$. Then $G(y+\delta/2) - G(y-\delta/2) \approx \delta \mathrm{d}G/\mathrm{d}y$ corresponds to the circular dichroism of σ^+ and σ^- probes as shown in Fig. 1(b). One then obtains

$$\delta = w\sqrt{\frac{e}{2}\frac{[\Delta T_-/T_- - \Delta T_+/T_+]_{y=w/\sqrt{2}}}{|\Delta T_+/T_+|_{y=0}}}, \qquad (1)$$

where, in the case of Fig. 1(b), $y=w/\sqrt{2}$ locates the maximum of the differential circular dichroism. This relation has been previously used to measure nanometer spatial separation of carriers and/or spins in time-resolved transmission experiments [15,16]. If one were to use the data from Fig. 1(b), an apparent separation of $\uparrow$ and $\downarrow$ electrons of $\delta = 37\pm 4$ nm would be deduced. However, this value is only a lower bound for the separation, since optical pumping of GaAs with circularly polarized light does not produce a pure spin state. For bulk GaAs the maximum theoretical degree of spin polarization is 0.5.

To obtain the actual SHEL-induced lateral shifts, Δy_{SH}, we conducted pump/probe experiments with cocircularly and countercircularly polarized pump beams with a fixed beam overlap. We measured the corresponding normalized transmission change, ρ [13], in the probe using a σ^+ probe and find $\rho = (\Delta T_-/T_- - \Delta T_+/T_+)/(\Delta T_+/T_+ + \Delta T_-/T_-) = 0.18\pm 0.02$, which is consistent with values reported in the literature [17]. Our experimental results therefore indicate an actual SHEL-induced transverse shift of $\Delta y_{\mathrm{SH}} = \delta/\rho = 205\pm 30$ nm. For p polarization, the theory predicts [4,9]

$$\Delta y_{\mathrm{SH}} = \lambda/\pi[\cos(\theta_r) - t_s/t_p \cos(\theta_i)]/\sin(\theta_i), \qquad (2)$$

where θ_r is the refracted angle of the transmitted light inside the sample and $t_{s,p}$ are the Fresnel transmission coefficients for the s- and p-polarized light. For our parameters, the theoretical prediction (175 nm) and the experimental value are in good agreement. The magnitude of Δy_{SH} is also considerably larger than that reported [9] for glass ($n=1.5$).

The SHEL effect has also been observed for s-polarized excitation pulses. We find a reduced lateral separation of σ^+ and σ^- components compared with p-polarized pump pulses, as expected from the theory [9].

The theory predicts that the transverse shift of σ^+ and σ^- components strongly depends on θ_i. Our present experimental configuration relies on pump and probe beams being independently focused with optics of large NA. The geometrical limitations of the present experimental setup inhibit significant alteration of θ_i alone. Instead, we rotate the GaAs in the plane of incidence through an angle $(\theta_i)_{\mathrm{pr}}$, essentially varying the angle of incidence of probe beam as well as the pump beam (see the inset of Fig. 2), while keeping the angle between the pump and the probe beam propagation directions fixed at 55°. The measured apparent displacement (δ) deduced from the

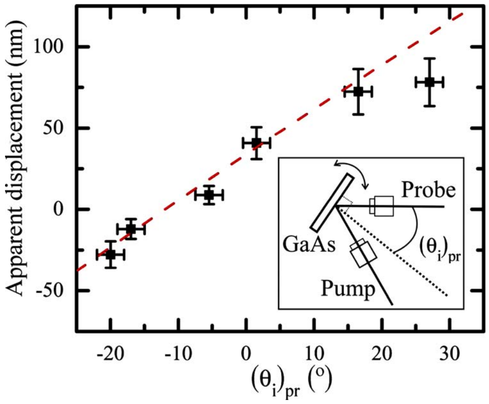

Fig. 2. (Color online) Apparent displacement for the SHEL measured as a function of probe incidence angle, $(\theta_i)_{\rm pr}$, while keeping a fixed angle (55°) between the pump and probe beam as indicated in the inset. The dashed line represents a theoretical prediction based on Eq. (2).

circular dichroism as above is shown in Fig. 2 as a function of $(\theta_i)_{\rm pr}$. Surprisingly, this apparent displacement is seen to strongly increase with $(\theta_i)_{\rm pr}$. To understand this trend, one has to consider the SHEL for *both* the pump and the probe pulses. While the spatial separation of σ^+ and σ^- components of the pump beam, $(\Delta y_{\rm SH})_{\rm pu}$, decreases with increasing $(\theta_i)_{\rm pr}$, a displacement of opposite sign is introduced in the probe pulse (since the angles of incidence of pump and probe beams differ in sign). As discussed in the context of Fig. 1(b), the circular dichroism detected in the pump/probe experiment is limited by the experimental degree of spin polarization of our optical orientation method. In contrast, a SHEL modified probe pulse essentially acts as a *pair* of spatially separated probe pulses with σ^+ and σ^- polarizations. These components are fully distinguishable by the polarization optics used in our detection scheme. As a result, a transverse displacement between the σ^+ and the σ^- components of the probe beam provides a direct observation of the SHEL, which corresponds to a detected signal $\sim 1/\rho$ times larger than for a same transverse displacement in the pump beam. By considering an arbitrary displacement between the σ^+ and σ^- components of the probe *and* the pump pulses, we can relate the apparent displacements obtained experimentally to the actual SHEL-induced beam displacements through $\delta = \rho(\Delta y_{\rm SH})_{\rm pu} - (\Delta y_{\rm SH})_{\rm pr}$.

The dashed line in Fig. 2 represents the theoretical prediction for δ based on Eq. (2). It is seen to reproduce the general trend of the experimental data related to the value of δ extracted from the data through Eq. (1). The dichroism induced by the probe beam is opposite to that induced by the pump beam when both pump and probe angles of incidence have the same sign $[(\theta_i)_{\rm pr} < 0]$. This leads to negative values of the apparent displacement in Fig. 2.

To consider more generally the effectiveness of this *in situ* detection scheme, we consider in particular the case of a normally incident pump beam and an off-normal probe beam. The pump beam, which does not experience a SHEL shift on entering the second medium, establishes an effective partial aperture via, e.g., bleaching, through which the transmissions of the separated σ^+ and σ^- components of the probe beams are altered and subsequently detected independently. Note that in this case a pump-induced spin-polarized carrier population is not a necessary requirement for the detection of the SHEL-induced displacement. Any pump-induced change acting equally on the probe σ^+ and σ^- components will lead to a direct observation of the SHEL experienced by the probe beam. Indeed, this concept may be extended to nonlinear optical interactions between the pump and probe beams.

In summary, using pump/probe techniques we have demonstrated a direct *in situ* method to quantitatively analyze the SHEL at the interface between a transparent medium and a semiconductor via transfer of the spin angular momentum from the light beam to electrons. It is also noted that any mechanism whereby a pump beam modifies an off-normal incidence probe beam can be used to detect the SHEL in the probe, since the probe is split into its σ^+ and σ^- components. This opens up the possibility of observing SHEL effects in a wide class of materials.

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