THz emission from ultrafast optical orientation

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We show both theoretically and experimentally that the magnetization density accompanying ultrafast excitation of a semiconductor with circular polarized light varies rapidly enough to produce a detectable THz field.

A host of recent fundamental studies on the behavior of spins in semiconductors have been motivated by the idea of using the spin degree of freedom in information processing, the goal of a field that has come to be known as spintronics. Central to many of these investigations has been the optical injection of spin polarized electrons and holes into the band structure of a semiconductor. These spin polarized carriers are then available to be dragged by a bias voltage, for example, to regions of interest in any proposed device structure. Such optical injection of spin polarized carriers, also called optical orientation, can be affected by the absorption of circularly polarized light, which takes advantage of the selection rules to populate particular angular momentum states of the band structure.

In the analysis of most of these experiments it is assumed that the holes can be neglected, because in bulk semiconductors the hole spin lifetimes are typically much shorter than the electron spin lifetimes. This makes it difficult, if not impossible, to measure the optical orientation of hole spins in bulk semiconductors using usual techniques employed to study electron spin orientation. However, Hilton and Tang recently showed that pump-probe techniques could be used to probe the hole spin dynamics in bulk GaAs. They found a time of $\tau_h = 110 \text{ fs}$ for the hole spin relaxation time, which has spurred theoretical investigations. All such techniques measure optical orientation indirectly by monitoring, after excitation, the population in various bands, or its effect on luminescence or Faraday rotation.

In this manuscript we present an approach for the direct measurement and study of ultrafast optical orientation. While we focus on GaAs, the technique is applicable to a wide range of semiconductors. It relies on the magnetization that accompanies the injected spin density in optical orientation: Under ultrafast excitation this magnetization varies rapidly over a sub-picosecond timescale, and radiates in the terahertz (THz) regime. Such radiation has already been used to study a host of other ultrafast processes, and we show here that detailed measurement and analysis of the emitted THz electric field trace can be used to study the electron and hole spin dynamics.

To estimate the THz field strength radiated from ultrafast optical orientation, we compare the injected magnetization source $M(t)$ in optical orientation to the injected polarization source $P(t)$ in the shift current (also called the intrinsic photovoltaic effect). When a semiconductor is excited by photons with energies above the band gap, electrons and holes are injected at a rate that closely follows the temporal pulse profile $I(r, t)$, which is the intensity profile of the laser pulse in the sample. The carrier density injection rate is given by $\dot{n}(r, t) = \frac{\mu}{e} I(r, t)$, where $\omega$ refers to the carrier frequency of the laser, and $\xi_r$ is proportional to the absorption coefficient. The spatial coordinate $r$ accounts for the variation from the exponential decay into the surface, and the variation in intensity across the spot-size. The polarization and magnetization sources we consider both arise from the carrier injection, although they have different symmetry characteristics and underlying microscopic origins.

In III-V semiconductors, the shift current appears for excitation by certain linear polarizations of the field. In this effect the center of charge of the carriers moves as the carriers are promoted from the valence band to the conduction band, and a net current directly results,

\[ P_{\text{inj}}(t) = p \dot{n}(r, t), \]

where $p = ed$ is the average injected dipole moment per carrier. Here, $e$ is the electron charge and the displacement $d$ is on the order of a Bohr radius $a_B$. Detailed calculations show that for GaAs under excitation with linear polarized light along $[110]$ this distance $d$ is very close to the GaAs bond length, $d = 2.54 a_B$. Since the polarization $P_{\text{inj}}(t)$ rises on a subpicosecond timescale, it radiates in the THz regime. However, the polarization survives for much longer than a picosecond, and so its decay is not expected to radiate in the THz regime. Thus for the polarization that contributes to the far field radiation we have $\hat{P}(t) = \hat{P}_{\text{inj}}(t)$.

Optical orientation arises from circularly polarized excitation of carriers near the band edge. Because of spin-orbit splitting these carriers can have a net spin polarization, and accompanying this spin density is a magnetization density $M(r, t)$. For optically injected electrons, close to the conduction band edge, the injected magnetization density is given by

\[ M_{e, \text{inj}}(r, t) = \frac{\mu_e}{e} \dot{n}(r, t), \]
where $\tilde{\mu}_e$ is the mean magnetic moment of an injected electron. Unlike for free electrons – where the magnetic moment of an electron is simply $\mu = g_e \mu_B / h$, where $\mu_B$ is the Bohr magneton, $s = h/2$, and $g$ is the free electron $g$-factor, $g = 2.0023$ – the average magnetic moment of an injected electron can be written as $\tilde{\mu}_e = g^*_e \mu_B S / h$, where $g^*_e$ is an effective $g$-factor of an electron, and $S$ is the injected degree of spin polarization in units of $\hbar/2$.

The effective $g$-factor can be calculated from Roth’s expression for the magnetic moment of a Bloch state in the $n$-th band [15],

$$\mu_n^z = -\frac{g \mu_B \sigma_n^z}{2} - \frac{2}{m} \sum_{s \neq n} \frac{p^x_n p^y_n - p^y_n p^x_n}{\epsilon_n - \epsilon_s},$$  

(3)

and defining the effective $g$-factor via $g^*_n = -2\mu_n / \mu_B$.

In Eq. (3), $\sigma_n^z$ is the expectation value of the Pauli spin matrix in the $n$-th band, $p^x_n$ is the $x$ component of the momentum matrix element between the bands $n$ and $s$, and $\epsilon_n$ is the energy of the $n$-th band. The $z$ direction is the direction along which the magnetization is aligned.

The appearance of $\sigma_n^z$ in the first term is required to generalize the more common $g$-factor expression [16] beyond $j = 1/2$ states. For optically injected electrons in GaAs, close to the conduction band edge, $g_e^* \approx -0.44$ at low temperatures [17].

This magnetization density $M_e(t)$ will vary on an ultrafast time-scale with the laser pulse, and it will radiate in the THz regime. The radiation will be magnetic-dipole like, in contrast to the electric-dipole like radiation from the shift current. Since the lifetime of the injected spins is known to be on the order of many picoseconds [2], their decay will not contribute to the THz and we have $\dot{M}_e(r, t) = \dot{M}_{e, \text{inj}}(r, t)$.

The far-field radiation from both magnetic and polarization sources is dependent on the second derivatives of the sources, $\dot{M}(t)$ and $\dot{P}(t)$. A simple estimate of the radiation from just the electron magnetization $M_e(t)$ can be made by comparing this source to the polarization induced by the shift current. Since both of these sources survive long enough that their decay profiles are not expected to radiate in the THz regime, it is mainly the generation (or turn on) of these sources that provides the THz radiation. The ratio of the peak strengths for the magnetization and polarization sources are directly comparable, since the different Fresnel coefficients governing the transmission of radiation for each type of source only determine the radiation patterns. So to first order we can estimate the strength of the radiation from the ratio $M_e(t) / P(t)$. Using Eq. (2) and (11) gives

$$\frac{\dot{M}}{P} = \frac{\mu_e}{p} = \frac{S h}{4(d/a_B)} \alpha,$$

(4)

where $\alpha$ is the fine structure constant. In GaAs optical orientation gives 50% spin polarization ($S = 0.5h/2$), so the ratio $M_e / P$ is roughly $1.6 \times 10^{-4}$. The THz fields from the shift current have been measured to be about 1500 V/m, so an absolute sensitivity of about 0.2 V/m is needed to measure the THz radiation from optical orientation of bulk GaAs electrons alone. This requirement is experimentally challenging [18] but would not be impossible. Note that this is because the THz field amplitude, rather than the intensity, is measured experimentally. Were the intensity measured, the signal from the magnetization effect would be down from that of the shift current by a factor of $(M_e / P)^2$.

The relatively short lifetime of the holes, however, suggests that the hole spins may radiate in the THz regime. Including the decay of the hole spins, the magnetization of the holes follows

$$\dot{M}_h(r, t) = \dot{M}_{h, \text{inj}}(r, t) - \frac{M_h(r, t)}{\tau_h},$$

(5)

where $\dot{M}_{h, \text{inj}}$ is the hole injection rate

$$\dot{M}_{h, \text{inj}}(r, t) = \tilde{\mu}_h \dot{n}(r, t),$$

(6)

analogous to the electron injection rate, but here $\tilde{\mu}_h$ is the mean magnetic moment of the injected holes.

On the one hand, the decay of the holes adds more temporal variation to the magnetization, and enhances the THz signal; on the other hand, since the decay lowers the overall magnitude of the magnetization, it is expected to decrease the THz signal. To include both effects, we have solved for the far-field radiation from an injected magnetization using a Green function formalism tailored to planar interfaces [18]. We find that for pulse widths of 100fs the effect of the hole spin decay ($\tau_h = 110$ fs) is to decrease the peak THz radiation from this source by roughly a factor of two. That is, the hole spins decay fast enough so that the decay process will contribute the THz radiation, but slow enough that the overall signal from it is not washed out.

Yet the relatively short lifetimes of holes, in bulk materials such as GaAs, prevents the study of their dynamics by many other techniques. Thus the hole $g$-factors, and their magnetic moments in general, have been largely neglected in bulk materials. In GaAs based nanostructures however, hole $g$-factors have been the subject of much recent attention [20]. It is typically found that in those systems that the holes have a $g$-factor many times larger than the electron $g$-factor. In this work however, we are interested in the optically injected magnetic moments $\tilde{\mu}_h$, rather than the $g$-factors themselves. The average magnetic moment $\tilde{\mu}_h$ will depend on the details of the populations injected into the heavy-hole and light-hole bands. We calculate directly the injected hole magnetization into these states, but for easy comparison to the electron magnetization, we quote our result as a hole $g$-factor $\tilde{g}^*_h$ relative to the injected electron spin density, so that

$$\tilde{\mu}_h = \frac{\tilde{g}^*_h g h S}{\tilde{g}^*_e h},$$

(7)
The tilde is to emphasize that we are not defining the $g$-factor relative to the hole spins. Following a similar approach as in [22] we use Fermi’s golden rule together with Roth’s formula Eq. (3) for the magnetization, and use a 30-band $k \cdot p$ band structure [21] to evaluate $M_{n,\text{inj}}$ directly. We find that $\tilde{g}_h^* = 14.1$, which implies that the hole magnetization injection is approximately 32 times larger than the electron magnetization injection and clearly dominates it [23].

Combining this with our calculations for the radiation emission (following [19]) we find that for GaAs the hole radiation should be of the order $3 \times 10^{-3}$, or about 1/300 times the shift current radiation.

Despite the relatively large expected signal from the holes, the ratio of THz emission from magnetization to shift current is still small, and one must choose an experimental geometry which minimizes the shift current. Differences in the spatial symmetry characteristics of shift current and magnetization sources are exploited to distinguish the weak magnetization source from the much stronger shift current source.

Fig. 1 illustrates the experimental setup for observing the transient THz radiation. An 80 MHz Ti:sapphire oscillator delivers 1 nJ, 100 fs pulses at 800 nm. For this wavelength, the injected kinetic energy of heavy (light) holes is 3.6 meV (9.5 meV), much less than the 300 meV needed to excite holes in the spin-orbit split-off band. The pulses are split into a pump beam for sample excitation and a probe beam to sample THz radiation via electro-optic sampling. The pump beam is incident onto an 100 $\mu$m thick (110) GaAs sample with a spot diameter of 25 $\mu$m, producing a peak incident intensity of 500 MW/cm$^2$ and a carrier density of $2 \times 10^{18}$ cm$^{-3}$ over an absorption depth of about 1 $\mu$m. Since the total magnetization or electric polarization is determined by total number of carriers excited, and not their density, there is no premium associated with beam focusing. The emitted THz radiation emitted along the surface normal direction from the sample back side is collected with a f = 50 mm, 90° off-axis parabolic and focused with the same type of mirror onto a 500 $\mu$m thick (110) ZnTe electro-optic sampling crystal. The 800 nm probe pulse is spatially overlapped with the THz field inside the ZnTe crystal and temporally scanned. The pump beam polarization is controlled by half- and quarter-wave plates (QWP) and a photo-elastic modulator (PEM). The half-wave plate (HWP) is oriented to provide light linearly polarized 45° from the $\hat{s}$ polarization vector (the vertical). The PEM operates in quarter-wave mode and modulates the polarization state at a frequency of 42 kHz, enabling lock-in detection techniques.

A major difficulty in measuring the emitted THz is that the injected magnetic dipoles are parallel to the laser beam propagation direction. For normal incidence excitation, this injects magnetic dipoles that are perpendicular to the surface. The component of the emitted radiation nearly parallel to the surface is small, and so there is little transmission. In the shift current effect however, the polarization is parallel to the surface, and this allows the radiated THz field to easily transmit through the surface. To enhance the emission of THz from the ultrafast optical orientation it is thus necessary to irradiate the sample at a high angle of incidence: we use an angle of incidence of 55°, exciting through an epoxy hemisphere (see Fig. 1b), to give an angle of refraction inside the GaAs, $\theta_R$, of about 20°. Even for this small angle it follows from the Maxwell saltus (boundary) conditions that the emitted THz radiation mainly propagates in the direction of the surface normal, because the induced electron-hole plasma is < 1 $\mu$m thick.

We define $\beta$ as the angle between the QWP’s fast axis and $\hat{s}$. The shift current and magnetization signals both depend on $\beta$ in a characteristic and unique way, so we focus on that variable to distinguish between the two THz sources. If we define $\gamma$ as the angle between the [001] crystal axis (lying in the plane of the sample) and $\hat{s}$, the internal THz field emitted by the two types of sources are given by

$$\Delta E_{\text{Thz}}^{\text{shift}} = C e d \cos (\theta_R) (2 \sin \gamma - 3 \sin^3 \gamma) \cos (2 \beta), \quad (8)$$

$$\Delta E_{\text{Thz}}^{\text{mag}} = C \mu_{\text{eff}} \sin (\theta_R) \sin^2 (2 \beta), \quad (9)$$

where the proportionality constant $C$ includes universal constants and Fresnel reflection and transmission coefficients. The effective magnetic moment $\mu_{\text{eff}}$ describes the sum of electron and hole magnetic moments, although we expect it to be dominated by hole term. It
is evident that the shift current (magnetization) signal is maximized (minimized) when $\beta = 0^\circ$ and minimized (maximized) when $\beta = 45^\circ$. This is because when $\cos(2\beta) = 0$ ($\sin(2\beta) = 0$), the light polarization is modulating between $+45^\circ$ and $-45^\circ$ linear (right and left circular) states. We choose $\gamma = 0.02\text{rad } (1.2^\circ)$, for which $2\sin\gamma - 3\sin^3\gamma \approx 2\gamma$, in order to provide a small shift current source with which to compare the magnetization signal.

Fig. 2b illustrates the expected variation of the THz field strength from the shift and magnetization sources, taking the ratio of the source strengths to be 0.015. Other possible sources of THz radiation were neglected. Optical rectification is small relative to the shift current effect above the band gap, and shares the same symmetry dependence. Sources of THz radiation related to Dember fields have no dependence on crystal orientation or pump polarization. Fig. 2b shows results from experimental measurements of the THz field strength as a function of $\beta$. The THz field strength is defined as the peak-to-peak value of the observed THz trace, although several other definitions of signal strength were used and each resulted in a similar pattern as a function of $\beta$.

The deviation of the expected sinusoidal pattern created by the shift current alone is attributed to the signal from the magnetization source. The zero crossings of the Fourier series fit to the data of Fig. 2b are separated by more than $90^\circ$. This behavior indicates the presence of an additional THz source with a $\beta$-dependence as shown in Fig. 2a. The difference between maximum and minimum signal values recorded at $\beta = 90^\circ$ and $0^\circ$ respectively was $0.05 \pm 0.14 \mu \text{V}$. This difference would have had to be on the order of $1 \mu \text{V}$ to attribute the data’s deviation to a uniform vertical shift. Experimental data indicates that the relative strengths of the magnetization and shift current sources have a ratio 0.017, which is within a factor of six of the theoretical value of the hole contribution. More work is required to confirm that the offset in Fig. 2b is mainly due to the magnetization source. Given all the possible sources of systematic error, we consider this extremely good agreement in the first study of this effect.

Increasing the THz emission from the spin injection can be achieved in a few ways: The THz generation depends on the injection rate of spin polarized carriers, and so using a shorter pulse will give a larger injection rate and increase the THz signal. One could also use materials with a larger $g$-factor. Materials with $g$-factors near 50, such as InAs, have been reported [24]. Finally, some materials also allow for a higher degree of spin polarization $S$ to be achieved; in strained GaAs or the II-VI wurtzite materials, for example, optical orientation can give an almost 100% polarized electron spin population.

In conclusion, we have investigated the possibility of measuring THz radiation from the ultrafast optical orientation of electrons and holes. This THz radiation is due to a rapidly-varying magnetic source, and we have shown that it is large enough to be detected against the more usual THz signal from electric current sources. Measurements of the THz radiation from spin-injection can complement the wide range of experiments on spin polarization decay already existing in the literature, providing a direct measurement of the injected spin dynamics of both electrons and holes in semiconductors.

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