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Spin-to-orbital angular momentum transfer by second harmonic generation in thin dielectric films

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Abstract. We demonstrate spin-to-orbital angular momentum transfer in the bulk of nonlinear optical materials with a crystal symmetry that couples the longitudinal component of the pump field. Our predictions are experimentally confirmed with a thin film of gallium arsenide, which generates vortex beams of second-harmonic light when pumped with a CP Gaussian beam.

1 Introduction

Spin-orbit interactions [1] have been observed under different circumstances in linear optics: (i) conversion of spin angular momentum (SAM) to orbital angular momentum (OAM) in tightly focused beams propagating in isotropic and homogeneous media [2], (ii) spin Hall effect at dielectric interfaces illuminated with circularly polarized (CP) light [3], (iii) spin-orbit interactions for paraxial beams propagating along the optical axis of uniaxial homogeneous crystals [4] or anisotropic and inhomogeneous structures [5], (iv) quantum spin Hall effect of light due to spin-orbit interactions at metal-dielectric interfaces [6, 7]. Spin-orbit interactions has been experimentally observed in second harmonic generation from a BaB_2O_4 crystal pumped with CP beam, in which the interplay between linear spin–orbit coupling and nonlinear wave mixing is exploited to generate a variety of OAM states [8]. Interaction between OAM and SAM of light beams in three-wave mixing processes has been also theoretically investigated in the bulk of an isotropic chiral media [9].

2 Theory

The transverse field components of a Gaussian beam at its waist ($z = 0$) can be expressed as:

$$E_x^\omega = \alpha E_0 \,,\; E_y^\omega = j\beta E_0 \,,\; E_0 = e^{-\frac{x^2+y^2}{w^2}} \tag{1}$$

where w is the beam waist and α, β are free (complex) parameters that define the polarization state of the beam ($\hat{e} = \alpha\hat{x} + j\beta\hat{y}$). Maxwell's equations in absence of charges impose that $\nabla \cdot \vec{E} = 0$. It follows that the electric field component in the longitudinal z direction can be written, in the Fourier space, as [10]:

$$\tilde{E}_z^\omega = -k_z^{-1}(\alpha k_x + j\beta k_y)\hat{E}_0 \tag{2}$$

where k_x (k_y) is the spatial frequency in the x (y) direction and $k_z = \sqrt{k_0^2 - k_x^2 - k_y^2}$ the wavevector component along z, with k_0 the wavenumber in free space at the frequency ω. In the limit of a weakly divergent pump beam, $k_z \approx k_0$ and, with the transformations $jk_x \to \partial/\partial x$ and $jk_y \to \partial/\partial y$, the longitudinal field in real space can be written as:

$$E_z^\omega \approx \frac{1}{k_0}\left(j\alpha\frac{\partial E_0}{\partial x} - \beta\frac{\partial E_0}{\partial y}\right) = -j\frac{2E_0}{k_0 w^2}(\alpha x + j\beta y) \tag{3}$$

For CP beams ($\alpha = 1/\sqrt{2}, \beta = \pm 1/\sqrt{2}$), such field shows a donut-shaped amplitude $E_z^\omega \propto \sqrt{x^2+y^2}E_0(x,y)$ and acquires a helical phase distribution equal to $\tan^{-1}(\pm y/x)$. This linear spin-orbit effect is well known as handedness-dependent OAM transfer to nanoparticles illuminated by tightly-focused, CP Laguerre-Gaussian beams [2]. Here we present theoretical and experimental evidence of a new type of *nonlinear* spin-orbit interaction mediated by the longitudinal field, in which SAM to OAM transfer is achieved via three-wave mixing. This effect can be observed in crystals with a $\chi^{(2)}_{ijk}$ where at least one of the subscripts j and k is equal to z, i.e., if at least one of the interacting fields is the longitudinal field of the pump beam. One of such crystals is aluminum gallium arsenide (AlGaAs), whose only nonzero element of the nonlinear susceptibility is $\chi^{(2)}_{xyz}$ with all its index permutations (which are equal under Kleinmann symmetry). Therefore, let's consider second harmonic (SH) generation in a film of [100]-cut AlGaAs illuminated by a Gaussian pump beam carrying SAM at the fundamental frequency ω. Let the film

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be in the plane $z = 0$, at the waist of the pump beam, where the transverse field can be written as in Eq. 1, with $\alpha = \sqrt{2}/2$ and $\beta = \pm\sqrt{2}/2$. Let its thickness be negligible or much smaller than a wavelength, so that the fields can be approximated as constant within the film and the induced SH source is a nonlinearly polarized sheet located at $z = 0$. The spectral components of the induced polarization on the nonlinear sheet at the SH frequency can be written as: $\tilde{P}_x^{2\omega_0} \propto \tilde{E}_y^{\omega_0} * \tilde{E}_z^{\omega_0}$, $\tilde{P}_y^{2\omega_0} \propto \tilde{E}_x^{\omega_0} * \tilde{E}_z^{\omega_0}$, $\tilde{P}_z^{2\omega_0} \propto \tilde{E}_x^{\omega_0} * \tilde{E}_y^{\omega_0}$, ($*$ stands for convolution). The transverse components of the SH electric field emitted by such sources can be retrieved via the spectral Green function approach outlined in [11]. For left CP ($\alpha = \sqrt{2}/2$ and $\beta = \sqrt{2}/2$), we thus get:

$$\begin{aligned} E_x^{2\omega}(z = 0^+) &\propto \frac{2E_0^2}{kw^2}\,[-3x + j\,y] \\ E_y^{2\omega}(z = 0^+) &\propto \frac{2E_0^2}{kw^2}\,[-y + j\,x] \end{aligned} \tag{4}$$

at $z = 0^+$, i.e., in forward SH generation, and

$$\begin{aligned} E_x^{2\omega}(z = 0^-) &\propto \frac{2E_0^2}{kw^2}\,[x + j\,y] \\ E_y^{2\omega}(z = 0^-) &\propto \frac{2E_0^2}{kw^2}\,[3y + j\,x] \end{aligned} \tag{5}$$

at $z = 0^-$, i.e., in in backward SH generation. The y component of the forward SH field and the x component of the backward SH field are endowed with OAM with charge $\ell = \mp 1$, respectively. For a pump with right CP, the dual situation occurs, with the forward and backward SH fields carrying OAM with charge $\ell = \pm 1$, respectively.

3 Experiment

Besides being supported by full-wave numerical simulations, the above findings have been confirmed by a SH generation experiment performed on a thin film of AlGaAs on sapphire. The latter was obtained by a planar structure, grown by molecular beam epitaxy, consisting of a high-quality 400nm-thick $Al_{0.18}Ga_{0.82}As$ layer deposited on a (100) GaAs substrate. A 500nm-thick $Al_{0.8}Ga_{0.2}As$ sacrificial layer was inserted before the GaAs epitaxial growth. The sample was then glued on a sapphire host substrate with a flip-chip process. The growth substrate and the sacrificial layer were then removed by mechanical and selective chemical etching, leaving only the 400 nm thick AlGaAs as the transparent mirrorflat-surface film required for the measurements (i.e. with both linear and nonlinear absorption coefficients $\alpha = \beta = 0$ at the pump wavelength). A linearly polarized pump beam was provided by an Amplitude/APE optical parametric amplifier delivering 160 fs pulses at a wavelength $\lambda = 1550$ nm. By varying systematically its polarization with a quarter-wave plate, it was focused on the nonlinear film at an intensity of a few GW/cm^2 by a microscope objective with f=1.5cm and NA=0.16, while the forward-generated SH light was collected by another microscope objective f=3.1mm and NA=0.68. The polarization states of both pump and SH beams were determined by retrieving their full set of Stokes parameters via the method proposed by B. Schaefer et al. [12]. Unless the pump is CP, a two-lobe SH far field is imaged on the back focal plane as expected for symmetry reasons. When the pump is CP, the superposed intensity image and polarization map are in striking agreement with the numerical predictions, as shown in Fig. 1. Our experiment was completed by the acquisition of the conversion efficiency ($P_{2\omega}/P_\omega \approx 10^{-7}$, two orders of magnitude less than the corresponding efficiency obtained with an AlGaAs metasurface) [13] and the typical fork-shaped phase map of the vortex beam, which was obtained as the interference between the output beam and a reference beam at 775 nm obtained by frequency doubling the pump on a standard BBO crystal and synchronizing it with a delay line.

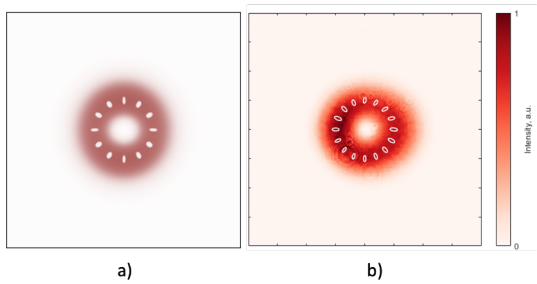

Figure 1. SH back-focal-plane intensity and polarization maps for a CP pump beam: numerical (a) and measured (b) results.

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