Magneto-optic Nonlinearity Induced Charge Separation

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Abstract: We report the magneto-electric charge separation by optical nonlinearities driven jointly by electric and magnetic fields of light using an electric-field-induced second harmonic generation technique.
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Introduction

Ultrafast magneto-electric (ME) interactions have attracted a great deal of attention because they potentially enable novel ultrafast optical switching, sensing technology, energy conversion, terahertz emission, and ultrafast data storage. Most ME effects have been observed in multiferroic materials or in metamaterials [1,2]. In general, the interaction of the magnetic field of light with homogeneous materials may be ignored due to the low magnetic susceptibilities at high frequencies. Very recently, however, it has been reported that magnetic properties of homogeneous dielectric media can be controlled by optical nonlinearities driven jointly by electric and magnetic field components of light [3-6]. This nonlinear magneto-electric interaction also drives the bound electrons to move in curved trajectories even under non-relativistic conditions [3]. Such a curved motion of bound charge breaks temporal and spatial inversion symmetry. As a result, it produces several unforeseen physical phenomena such as longitudinally polarized second harmonic radiation, induced transverse magnetization at the optical frequency, and charge separation along the propagation direction. In this work, we focus on a study of the forward magneto-electric charge separation generated by a linear polarization femtosecond laser pulse.

Results and discussion

The sample for investigating the nonlinear ME charge separation (MECS) in this study is pentacene organic semiconductor. The pentacene thin film was prepared by thermal deposition on a glass substrate in a high vacuum. The sample was then kept in the inert gas environment during both storage and measurement to avoid photo-oxidation. To detect the MECS, we use a time-resolved second harmonic generation technique. The setup was utilized from a femtosecond amplifier laser system which delivers a pulse of 0.5 mJ at 10 kHz repetition rate. The bandwidth and therefore the pulse duration of the output laser pulse can be tuned using an Acousto-Optic Programmable Gain Control filter. The laser beam was split by two paths, the first one acts as a pump which as passed through a delay stage. A half-wave plate was used in the second path, the probe, to control the probe polarization. In the probe path, we placed a suitable band-pass filter after the sample allowing only the transmission of the second harmonic signal to be collected by a PMT. In a centro-symmetric medium, second harmonic generation is normally absent. However, the DC electric field from MECS interacts with the optical field from the probe, in a four-wave-mixing interaction so-called electric field induced second harmonic (EFISH) generation. By monitoring time-resolved EFISH, we are able to study MECS dynamics in homogeneous media. Since the MECS field or DC dipole moment points along the beam propagation direction (fig 1a), in order to maximize the EFISH signal, the probe field needs to be aligned in this direction. The crossbeam geometric pump-probe ideally satisfies this requirement. However, this geometry has a drawback that is its large spatial overlapping of the pump and the probe wave-fronts resulting in a poor temporal resolution. To circumvent it, we tilted the wave-fronts to 45° for both pump and probe pulses, as a result this method retrieves the temporal resolution which is compatible with the coaxial geometric pump-probe setup.

The polarizations of pump-induced MECS, $P^{(2)}(0)$, and four-wave mixing interaction between the MECS electric field and the probe field, $P^{(3)}(2\omega)$, are given by:

$$P^{(2)}(0) = \varepsilon \chi_{me} E(\omega) H(-\omega) \quad (1)$$

$$P^{(3)}(2\omega) = \varepsilon \chi^{(3)} E_{DC} E(\omega)_{pr}^2 \quad (2)$$

where the E_{DC} and $E(\omega)_{pr}$ are the electric fields caused by $P^{(2)}(0)$ and from the probe pulse, respectively. $E(\omega)$ and $H(\omega)$ are the electric and magnetic fields from the pump pulse.

Figure 1b shows the pump-induced EFISH signal as a function of pump-probe delay for several probe polarization angles. These SHG changes originate from MECS by the mean of optical nonlinearity acting together of the electric and magnetic components of light. Noted that there is no electronic excitation in pentacene since the pump and probe energies were set at 1.55 eV, well below the bandgap of pentacene. At 0^o polarization, where the probe electric field is aligned with the pump-induced electric field, E_{DC}, the change of SHG is largest. The change is negligible when probe polarization and E_{DC} are orthogonal, 90^o. At zero delay, the EFISH signal reaches the highest value and rapidly decays with a time constant of 0.5 ps. The previous studies have shown that the magneto-electric effects not only give rise to the longitudinal electric dipole but also generate transverse magnetization at the optical frequency [3-6]. Therefore, the EFISH decay time constant, which is slightly longer than the experimental time resolution, could be due to the magnetization relaxation of pump-induced magnetic dipole moments. Further research is needed to verify this assumption. MECS is generated by a nonlinear two-photon process as evidenced by the quadratic power dependence of the pump-induced SHG change, Fig. 1c.

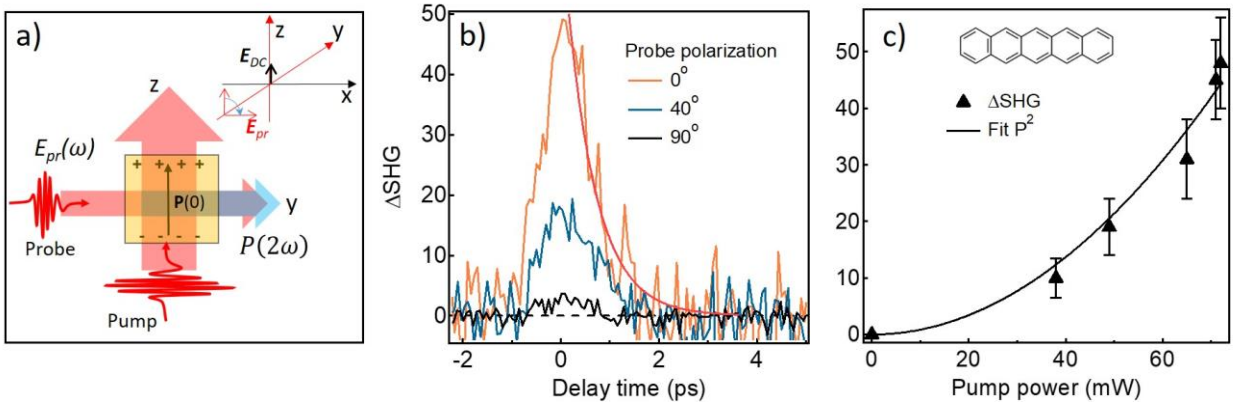

Figure 1. (a) Schematic of the cross beam geometry pump-probe experiment to detect magneto-electric charge separation. The pump pulse induces MECS resulting in a DC field or DC dipole, P(0), in a material (yellow square). The probe field interacts with that DC field in the four-wave-mixing process results in an EFISH signal, P(2ω). The probe pulse propagates in y direction with its polarization in the (x,z) plane. (b) Pump-induced EFISH (ΔSHG) signal from the pentacene thin film for three different probe polarization angles. 0^o and 90^o are corresponding to the polarization in z and x direction, respectively. The solid red curve is an exponential fit. (c) Quadratic pump power dependence of pump-induced SHG. The solid curve is a quadratic fit. Note that there is no electronic excitation in pentacene when the pump photon energy is set well below the bandgap. Inset: pentacene molecule structure.

Conclusions

We used an electric field induced second harmonic generation technique to demonstrate for the first time the existence of MECS at nonrelativistic intensities by magneto-electric nonlinearity. The charge separation in a polycrystalline pentacene thin film originates from a second order nonlinearity driven by dual electric and magnetic fields of light as proven by the quadratic dependence on the input intensity.

Acknowledgments

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