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ULTRAFAST THz PHOTO-GALVANIC CARRIER TRANSPORT. EXTREME FIELD INDUCED REGIME

CARMINE SOMMA[1], KLAUS REIMANN[1], MICHAEL WOERNER[1], THOMAS ELSAESSER[1]
CHRISTOS FLYTZANIS[2]

[1] Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, 12489 Berlin, Germany
[2] Ecole Normale Supérieure, Laboratoire Pierre Aigrain, 75231 Paris Cedex 05, France
E-mail: christos.flytzanis@lpa.ens.fr

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Abstract. We review the photogalvanic effect (PGE) in poled media as a ratchet type directed transport of photogenerated carriers in the absence of other external bias. We demonstrate that besides the perturbative regime involving multiphoton absorption which has exclusively been addressed previously, the PGE can also proceed through field induced tunneling, also termed dynamic Zener tunneling. The demonstration was done in a high optical gap crystal, the ferroelectric $LiNbO_3$, with intense short THz pulses using two dimensional THz spectroscopy.

The photo-galvanic carrier transport is an intriguing effect both within the context of nonlinear optics and nano/micro-optoelectronics but also and foremost in connection with the basic issue of directed transport in statistical physics: it results [1–3] from off equilibrium carriers generated by intense coherent light in poled media, namely dielectrics lacking space inversion symmetry such as polar semiconductors, ferroelectrics or poled polymers, in the absence of any external bias. This endows the PGE with strikingly distinct features and advantages in respect to other conventional photovoltaic effects (PVE), inclusive photo-conduction, as it dispenses of the use of electrodes and allows for all-optical control of the carrier transport and motion, inclusive initialization, monitoring and reshaping, with ultrafast light pulses in a wide spectral range.

The occurrence of the photo-galvanic effect can be viewed [3] as a photo-driven manifestation of the ratchet type directed transport [4] that evolves under the compound breakdown of space and time inversion symmetry in poled media subsequent to off equilibrium generation of carriers with coherent light in the absence of other external bias. Still this transport proceeds differently for moderately intense and very high light fields, also termed the multiphoton (perturbative) and extreme field induced (non perturbative) regimes respectively, corresponding to distinct ways of the off equilibrium generation of the carriers.

Previous studies on the PGE concerned the multiphoton regime. Here we address the extreme field induced regime [7] and provide first evidence of its relation to the dynamic Zener effect [8] with THz fields in a poled medium.

The PGE effect has been extensively and exclusively studied [1, 2] in the moderately intense light intensities regime in the optical frequencies range above the absorption threshold of the poled dielectric using a perturbative approach: assuming the light matter (dipolar) interaction H_{EM} small with respect to the relevant electronic energies in the lattice potential V_L, or $H_{EM} << V_L$, the response of the carriers can be treated by an iterative-perturbative approach [5, 6] with due attention to the lack of spatial inversion symmetry and its impact on the dissipative/relaxation processes following the resonant photo-excitation/generation of the carriers. Because of the built-in spatial asymmetry the detailed balance assumption breaks down and the carrier scattering and relaxation patterns become anisotropic and proceed differently in the forward and backward directions with respect to the poling axis resulting in the directed photo-galvanic transport [2, 3]. At moderate light intensities the dominant term is the linear one which was found [1] to follow the Glass conjecture $j = \alpha GI$ for the photo-generated current density: α is the absorption coefficient the light intensity and G a third rank tensor related to the asymmetry in the medium and the associated driving tensorial force; this quadratic dependence on the electric field amplitude E has been accounted [5] within the perturbative approach of the carrier response and also extended to higher even orders involving multi-photon absorption and carrier generation.

Here we address [7] the regime of very intense light fields where H_{EM} becomes comparable to energies related to the lattice potential, or $H_{EM} = V_L$, and frequencies down in the THz range well below the band gap of the dielectric: assuming adiabatic regime the perturbation approach previously sketched breaks down as the light-matter interaction H_{EM} now drastically modifies the lattice potential and concomitantly the electronic band states as in the case of the Zener effect [9] with intense static electric fields: the valence band max and conduction band min move apart in a skew manner with increasing el-field intensity and the band gap shrinks and eventually vanishes and reverses allowing thus the carriers to tunnel across the bands predominantly in one direction specified by the relative direction of the applied field with respect to the poling axis.

Thus besides the perturbative regime sketched above that proceeds through single or multi-photon absorption and generation of carriers, in much higher light intensities and far lower photon energies than the absorption threshold, the photogalvanic carrier generation in the conduction band can proceed through THz field induced tunneling, dynamic (field induced) Zener tunneling, and set up the directed transport in the poled medium without the presence of an external bias.

The study of the PGE response of the well-known nonlinear material $LiNbO_3$ under non-perturbative conditions was performed using very intense ultrashort THz light pulses and the different contributions to the overall nonlinear response were separated and analyzed with the help of two-dimensional (2D) THz spectroscopy [10]. Laser-driven sources for terahertz (THz) transients with very high electric field amplitudes are particularly appropriate for the study of nonlinear processes in the extreme field effect regime and in addition provide outstanding all-optical means for the initialization, monitoring and following up of the processes. Our data give the first evidence of an extreme field-induced bulk photogalvanic effect in a poled dielectric at frequencies well below the band gap; the intense THz field, applied in our experiments, enables dynamic Zener tunneling of electrons from the valence to the conduction band. Furthermore the (2D) THz time resolved spectroscopy allows to determine phase and amplitude of the different contributions to the nonlinear response and identify their origin.

The study was performed on an undoped single-domain $LiNbO_3$ crystal of 50 mm thickness. The ferroelectric *c* (poling) axis of the crystal is parallel to the electric field of the THz pulses generated by optical rectification of 800 nm pulses from a Ti: sapphire oscillator-amplifier system in two GaSe crystals. In the 2D-THz spectroscopy, we use two strong phase-locked THz pulses A and B with a center frequency ν_0 = 2 THz. The field transients transmitted through the sample are measured by electro-optical (EO) sampling in a thin ZnTe crystal as a function of both the real time t and the delay τ between the two THz pulses. With two choppers synchronized to the pulse repetition rate, three transients are determined, one, E_{AB} (t,τ), when both pulses are present, and two, E_A (t,τ) and E_B (t), for the single pulses and the nonlinear electric field emitted by the sample E_{NL} $(t,\tau) = E_{AB}$ $(t,\tau)–E_A$ $(t,\tau)–E_B$ (t) is analyzed to extract the evolution of the photogalvanic transport in the $LiNbO_3$ crystal as highlighted in Figs. 1 and 2 below: for a sample of thickness d smaller than the THz wavelength, the nonlinear emitted terahertz field $E_{NL}(t,\tau)$ is proportional to the nonlinear current $J_{NL}(t,\tau)$ induced by the incident THz field.

As inferred from Fig. 1a the nonlinear signal $E_{NL}(t,\tau)$ reaches the highest value, about 20 kV/cm, and has the opposite direction of the total electric field $E_{AB}(t,\tau)$, when the two THz pulses overlap, *i.e.*, for $t = 0$ and $\tau = 0$. With the 2D Fourier Transform (2D-FT) of E_{NL} (t,τ) along the two time variables t and τ, the different terms of the nonlinear signal are separated in the 2D frequency space spanned by ν_t, the detection frequency, and of ν_τ, the excitation frequency and the real and imaginary parts of the nonlinear response in frequency space are unambiguously determined (Figs. 1, 2). As shown in Fig. 1c, d, the 2D-FT signal E_{NL} (ν_t,ν_τ) has contributions corresponding to second (2^{nd}, at $\nu_t = \pm 2\nu_0$) and third

(3rd, at $\nu_t = \pm 3\nu_0$) harmonic generation; there are also contributions at frequencies close to $\nu_t = \pm\nu_0$, labeled as shift current (SC) and as optical rectification (OR), and contributions (DP) at the fundamental $\nu_t = \pm\nu_0$, the latter caused by the conversion of the fundamental into other frequencies. Actually higher harmonics to arbitrary order should be present too but cannot be accessed with our present detection technique.

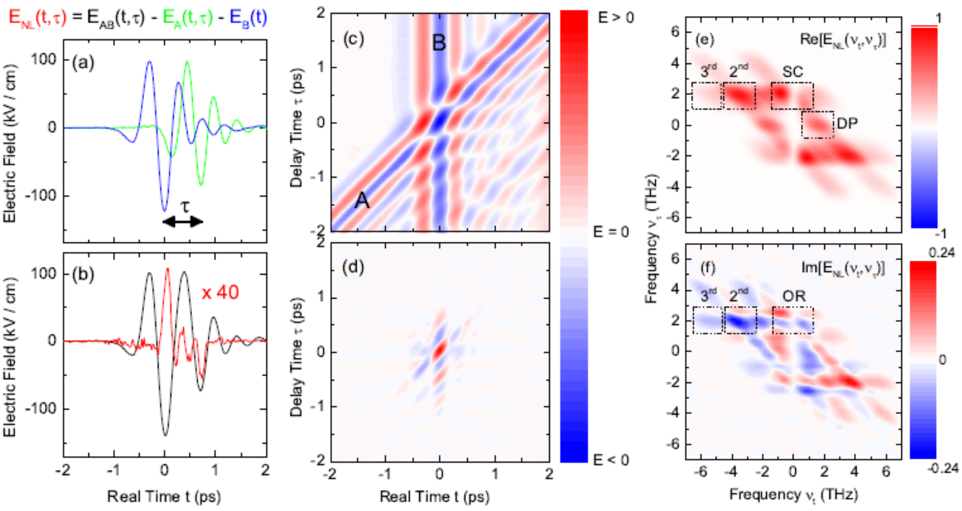

Fig. 1 – Two dimensional Fourier transform of the nonlinear current.

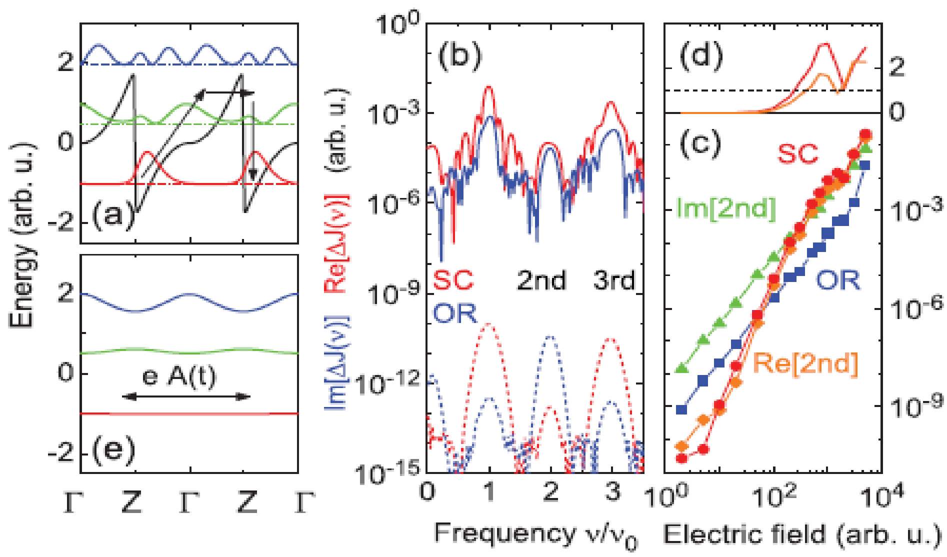

Fig. 2 – Contributions to the nonlinear electric field.

The $LiNbO_3$ crystal, being ferroelectric lacks space inversion symmetry and is known to possess high optical nonlinearities, in particular substantial second-order ones, allowing for efficient nonlinear frequency conversion and generation of harmonics. For incident and generated frequencies well within the transparency range of the crystal, as in our THz fields (2 THz), one expects [11] the fields generated by non-resonant nonlinear processes to emit with a phase $\pi/2$ over the incident one and only have an imaginary part, the corresponding susceptibilities being real. However, the 2D spectra in Fig. 1c, d display a real part even larger than the imaginary part which is an unambiguous signature of the generation of free carriers in the crystal and give evidence of a bulk photogalvanic transport without the involvement of direct photo-absorption: the strong electric THz-field induces dynamic Zener tunneling of electrons from the valence into the conduction band, which evolves differently in the forward and backward directions with respect to the poling axis, and sets up the observed shift current (SC) in line with a ratchet type process. The frequency spectrum of the SC reflects the dynamics of electron motion along the c axis of the $LiNbO_3$ crystal and contains components around zero frequency as well as harmonics up to – in principle – arbitrary order, in the present experimental configuration only accessed up to the third order.

Concluding we wish to stress again that our study provides evidence of an extreme field induced PGE in the transparency range of a dielectric well below the band gap, and relates it to the dynamic Zener tunneling in poled media. The process proceeds along the lines of a ratchet process. Further studies are needed to quantitatively assess certain features of the process such as the densities of the generated carriers, their velocities and the effective driving tensorial force, the analogue of the Glass coefficient in the perturbative regime.

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