Ultrafast Terahertz Response of Lithium Niobate in the Nonperturbative Regime

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Abstract: The response of a $LiNbO_3$ crystal to THz pulses in the nonperturbative regime is studied by two-dimensional spectroscopy. Phase-resolved detection allows for separating the THz bulk photovoltaic effect from other nonlinear contributions.

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Laser-driven sources for terahertz (THz) transients with very high electric field amplitudes allow for investigating the nonlinear response of condensed matter under conditions of a nonperturbative light-matter interaction where the coupling of electrons to the THz field represents their predominant coupling. Experiments in this regime have led to new insight into quantum coherent charge transport in semiconductors and carbon-based materials such as graphene [1]. Here, we study the nonlinear THz response of the well-known nonlinear material $LiNbO_3$ under nonperturbative conditions and separate the different contributions to the overall nonlinear response with the help of two-dimensional (2D) THz spectroscopy. Our data give the first evidence of a field-induced bulk photovoltaic effect in this material. While the conventional bulk photovoltaic effect manifests itself by the occurrence of a shift current (SC) of electrons generated by above bandgap photoexcitation [2], the strong THz field applied in our experiments enables Zener tunneling of electrons from the valence to the conduction band [3].

We perform collinear two-dimensional (2D) THz spectroscopy on an undoped single-domain $LiNbO_3$ crystal of 50 μm thickness. The ferroelectric c axis of the crystal is parallel to the electric field of the THz pulses. In our experiment, we use two strong phase-locked THz pulses A and B with a center frequency $\nu_0 = 2$ THz, generated by optical rectification of 800 nm pulses from a Ti:sapphire oscillator-amplifier system in two GaSe crystals. The electric-field transients transmitted through the sample are measured by electro-optical 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 when both pulses are present, and two for the single pulses. From these transients, the nonlinear electric field emitted by the sample is the difference $E_{NL}(t,\tau) = E_{AB}(t,\tau) - E_A(t,\tau) - E_B(t)$ (for further details of the experiment see [4]).

As shown in Fig. 1(b), the $LiNbO_3$ crystal emits a strong nonlinear electric field. 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 $E_{AB}(t,\tau)$ [shown in Fig. 1(a)], when the two THz pulses overlap, i.e., for $t = 0$ and $\tau = 0$. 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.

A 2D Fourier Transform (2DFT) of $E_{NL}(t,\tau)$ along the two time variables t and τ allows to separate the different terms of the nonlinear signal in the 2D frequency space spanned by ν_t, the detection frequency, and of ν_τ, the excitation frequency. Since electro-optic sampling yields the complete phase information, the 2DFT determines unambiguously real and imaginary parts of the nonlinear response in frequency space. As shown in Figs. 1(c) and (d), the 2DFT signal $E_{NL}(\nu_t,\nu_\tau)$ has contributions corresponding to second (2nd, at $\nu_t = \pm 2\nu_0$) and third (3rd, at $\nu_t = \pm 3\nu_0$) harmonic generation. Additionally, there are contributions at frequencies close to $\nu_t \approx 0$, labeled as shift current (SC) and as optical rectification (OR), and contributions at the fundamental THz frequency $\nu_t = \pm\nu_0$. The latter contribution (DP) is caused by the conversion of the fundamental into other frequencies.

$LiNbO_3$ displays a substantial second-order nonlinearity, allowing for nonlinear frequency conversion. Our THz frequencies (2 THz) are way below the band gap of $LiNbO_3$ and, thus, one expects purely real nonlinear susceptibilities

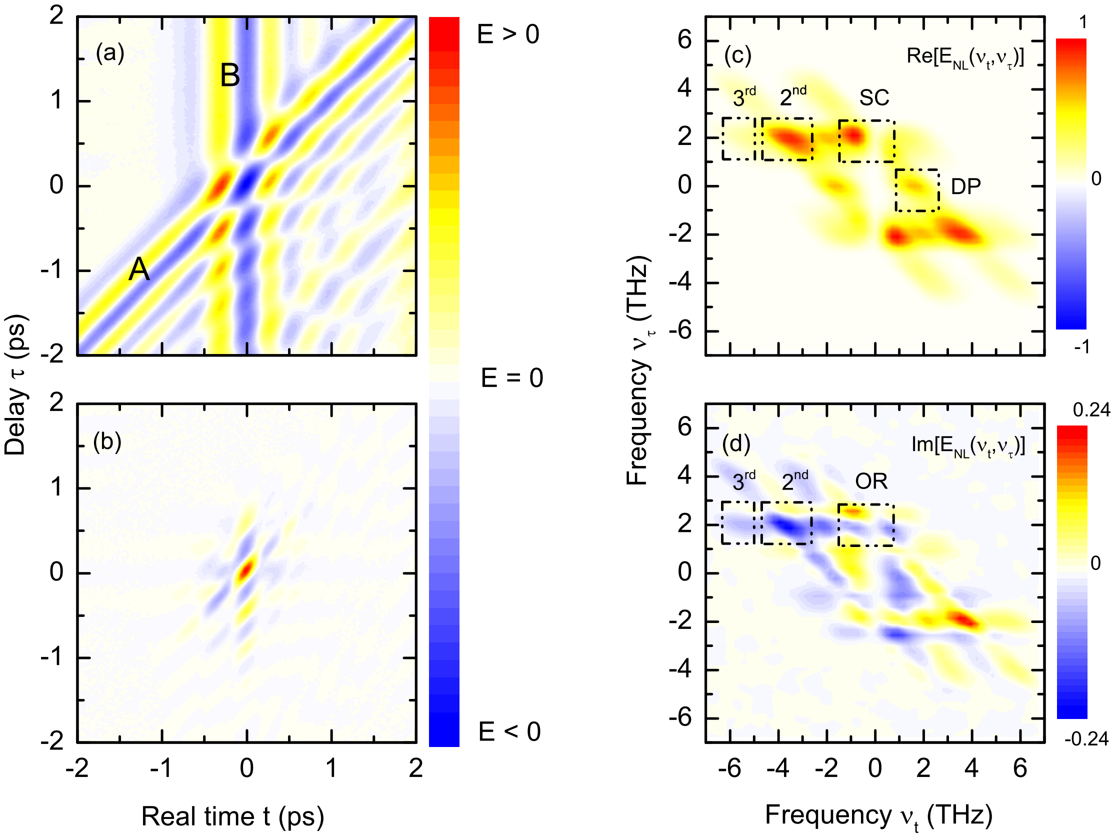

Fig. 1. (a) Electric field $E_{AB}(t,\tau)$ transmitted through the sample. (b) Nonlinear THz signal $E_{NL}(t,\tau)$ emitted by the sample. (c), (d) The real and the imaginary parts of the spectrum of the nonlinear signal $E_{NL}(t,\tau)$ show the signatures of the shift current (SC), the optical rectification (OR), the second (2^{nd}) and the third (3^{rd}) harmonics, and the pump depletion signal (DP).

and an imaginary part of the emitted nonlinear field only. However, the 2D spectra in Figs. 1(c) and (d) display a real part even larger than the imaginary part, a behavior well beyond nonresonant nonlinear optics. The results give evidence of a bulk photovoltaic effect, which underlies the observed the real part and is connected with the generation of free carriers in the crystal. Despite the low frequencies of the THz radiation, its very strong electric field induces Zener tunneling of electrons from the valence into the conduction band, i.e., generates a real shift current (SC) of electrons that gives rise to the observed new frequency components. 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.

References

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