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Terahertz driven amplification of coherent optical phonons in GaAs coupled to metallic dog-bone resonators

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Abstract. Two-dimensional terahertz spectroscopy on an AlAs/GaAs nanostructure covered by field-enhancing dog-bone resonators shows signatures of coherent optical phonon amplification. Amplification is due to stimulated phonon emission by a terahertz-driven electron current.

Amplification of sound in crystalline materials is a hot topic of current research. Recent work has concentrated on the stimulated emission of acoustic phonons in the GHz to sub-THz frequency range, e.g., of zone-folded acoustic phonons in semiconductor superlattices. In a recent experiment [1] we have demonstrated a gain coefficient of $g = 10000\ \mathrm{cm}^{-1}$ in the amplification process of such phonons persisting on a time-scale of several hundreds of picoseconds. A key concept of phonon amplification is the generation of a (quasi-stationary or time-averaged) carrier distribution which provides for phonons with wave vector **q** and energy $\hbar\omega_{\mathbf{q}}$ a population inversion within the distribution, i.e.,

$$\Sigma_{\mathbf{k}}\, [f_e(\mathbf{k}) - f_e(\mathbf{k}\text{-}\mathbf{q})]\ \delta[E(\mathbf{k}) - E(\mathbf{k}\text{-}\mathbf{q}) - \hbar\omega_{\mathbf{q}}] > 0.$$

In contrast to acoustic phonons, optical phonons display an almost constant dispersion for q vectors close to the Γ point and a much higher frequency [$\nu_{LO} = 9$ THz for longitudinal optical (LO) phonons in GaAs]. Creating a quasi-stationary electron (or hole) distribution with inversion for optical phonons is challenging because of the huge concomitant heat load in the system. This problem is circumvented when generating a terahertz-driven transient electron distribution displaying an inversion situation for optical phonons. Here, we apply terahertz fields enhanced by a metallic dog-bone resonator [2] to generate such a carrier distribution and amplify coherent optical phonons in GaAs within a short time

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period of 2 ps. The time evolution of the coupled phonon-carrier system is mapped with the help of two-color two-dimensional (2D) THz spectroscopy [3].

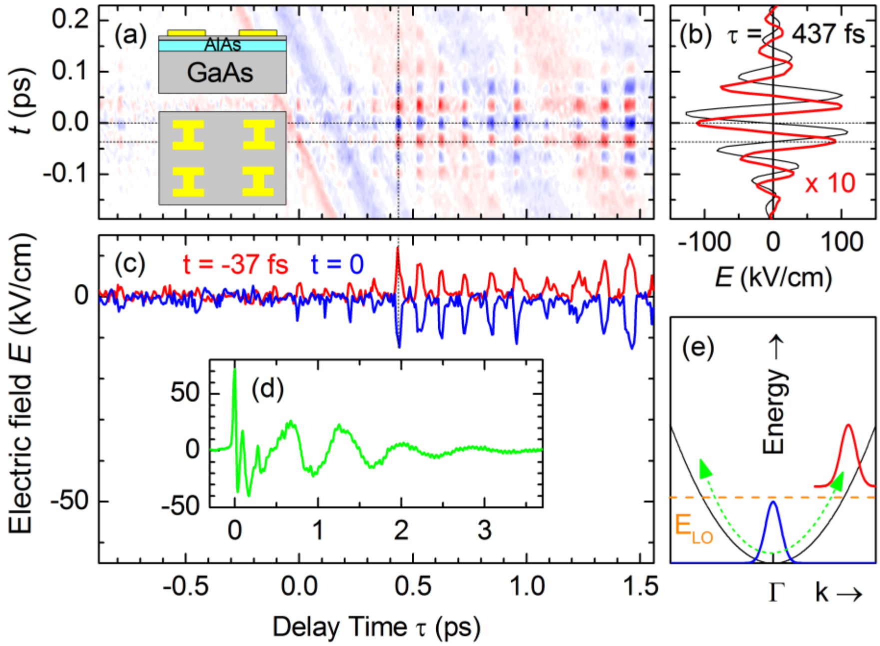

Fig 1. Two-dimensional (2D) nonlinear terahertz (THz) spectroscopy performed on an AlAs/GaAs nanostructure covered by a square array (periodicity 5 μm) of metallic dog-bone resonators as sketched in the inset. (a) Measured 2D scan of the nonlinearly emitted electric field $E_{NL}(t,\tau)=E_{both}(t,\tau)$ - 0.9 $E_{THz}(t,\tau)$ - $E_{MIR}(t)$ (in reflection geometry) as a function of real time t and delay time τ for applied THz [$E_{THz}(t,\tau=0)$ shown in panel (d)] and mid-infrared $E_{MIR}(t)$ pulses [black line in panel (b)]. For better orientation we subtracted only 90 percent of $E_{THz}(t,\tau)$ making its phase fronts visible. (b) Red curve: nonlinearly emitted field for τ = 437 fs [vertical dashed line in (a)]. (c) $E_{NL}(t = -37\text{ fs},\tau)$ (red line) and $E_{NL}(t = 0,\tau)$ (blue line) as a function of τ [horizontal dashed lines in (a)]. (e) LO phonon (energy: orange dashed line) amplification mechanism illustrated in the conduction band structure of GaAs (black parabola). Electrons (blue and red distribution functions) generated by interband tunneling are moved around in k space by the large vector potential $A(t,\tau) = \int_{-\infty}^{t} E_{THz}(s,\tau)\, ds$of the THz pulse thereby creating at some instants of time an inversion situation for the stimulated emission of optical phonons.

The inset of Fig. 1(a) shows the array of metallic dog-bone resonators (MDRs) on a layered GaAs/AlAs/GaAs semiconductor structure (thicknesses 3 nm / 40 nm / 300 μm). A numerical simulation of THz fields in this geometry gives an up to 20-fold enhancement of the electric field amplitude in the GaAs part of the sample at the frequency of the GaAs LO phonon. In the experiments, we performed fully phase-resolved 2D THz spectroscopy, using a THz and a midinfrared pulse, which interact with the sample in a reflection geometry. As shown in Fig. 1(d), the pump pulses consist of a short 9 THz component with a peak amplitude on the order of 50 kV/cm and a weaker trailing 1.5 THz component extending into the picosecond range. The first component is resonant to the optical phonons in GaAs. Because of the low frequency, the corresponding vector potential $A(t,\tau) = \int_{-\infty}^{t} E_{THz}(s,\tau)\, ds$ of the THz pulse drives both "real" and "virtual" carriers in k space over a considerable fraction of the GaAs Brillouin zone [4]. The MDR-enhanced electric field strength within the GaAs part of the sample reaches values larger than 1 MV/cm thereby creating "real" electron-hole pairs by interband tunneling [5,6]. The second midinfrared pulse [black line in Fig.~1(b)] is centered at 15 THz, i.e., off-resonant to both the optical phonons and the fundamental GaAs bandgap around 400 THz.

Fig. 1(a) displays the measured 2D scan of the nonlinearly emitted electric field defined by $E_{NL}(t,\tau) = E_{both}(t,\tau) - E_{THz}(t,\tau) - E_{MIR}(t)$ as a function of the real time t and the delay time τ between the THz pump and midinfrared probe pulses. To make the phase fronts of the 9 THz pulse visible, we plot $E_{NL}(t,\tau) - 0.1\ E_{THz}(t,\tau)$. We observe exclusively a spectrally unstructured THz pump-midinfrared probe signal while a midinfrared pump-THz probe signal is absent. The nonlinearly emitted midinfrared field [red line in panel (b), derived from a cut along the vertical dashed line in panel (a)] is a 90° phase-shifted replica of the mid-infrared pulses with amplitudes of more than 10 kV/cm. In panel (c) we plot $E_{NL}(t = -37\text{ fs},\tau)$ (red line) and $E_{NL}(t = 0,\tau)$ (blue line) as a function of the delay time τ for two different real times [horizontal dashed lines in (a)]. A 90° phase-shift of $E_{NL}(t,\tau)$ to earlier times corresponds to a refractive index change of $\Delta n/n \approx -2\%$. Such an index change agrees well with the generation of "virtual" electron-hole pairs by the electric field of the THz pulse and the field from the coherently excited optical phonons. In other words, the midinfrared pulse experiences a quasi-instantaneous Kerr nonlinearity $\Delta n \propto [E_{LO}(t,\tau) + E_{THz}(t,\tau)]^2$ with $E_{LO}(t,\tau)$ being the internal electric field caused by the optical phonon polarization and $E_{THz}(t,\tau)$ the field of the THz pulse.

Most interesting is the fact that $E_{NL}(t,\tau)$ plotted in panel (c) displays oscillations with the LO phonon frequency and an amplitude that increases substantially with increasing delay time, well beyond the duration of the 9 THz component of the first pulse. In parallel, the width of the individual oscillation cycle increases, resulting in a pronounced increase of the area under a single cycle. This rise of $E_{LO}(t,\tau)$ is a direct hallmark of amplification of the initially launched optical phonons.

In Fig. 1(e) we illustrate the LO phonon amplification mechanism with the help of a schematic conduction band structure of GaAs (black parabola). Electrons (blue and red distribution functions) generated by interband tunneling [5,6] are driven in k space by the large vector potential $A(t,\tau)$ of the THz pulse [4]. For a cycle-averaged motion with electron energies well above the threshold of phonon emission (LO phonon energy: orange dashed line) the corresponding electron distribution in the Γ valley of GaAs shows at the majority of instants of time an inversion situation for the stimulated emission of optical phonons. The coherent phonons kicked off by the initial part of the pump pulse (panel d) thus experience amplification within the period of large amplitudes of the THz vector potential. This novel amplification scheme holds potential for applications in phononics.

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