Extreme THz nonlinearities in bulk and nanostructured semiconductors

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ABSTRACT

Phase-locked electromagnetic transients in the terahertz (THz) spectral domain have become a unique contact-free probe of the femtosecond dynamics of low-energy excitations in semiconductors. Access to their nonlinear response, however, has been limited by a shortage of sufficiently intense THz emitters. Here we introduce a novel high-field source for THz transients featuring peak amplitudes of up to 108 MV/cm. This facility allows us to explore the non-perturbative response of semiconductors to intense fields tailored with sub-cycle precision. In a first experiment intense transients drive Rabi-oscillations between excitonic states in Cu_2O, implying exciting perspectives for future THz quantum optics. At electric fields beyond 10 MV/cm, we observe the breakdown of the power expansion of the nonlinear polarization in bulk semiconductors. Furthermore, we employ the intense magnetic field components of our transients to coherently control spin waves in antiferromagnetically ordered solids. Finally, intersubband cavity polaritons in semiconductor microcavities are exploited to push light-matter coupling to an unprecedented ultrastrong and sub-cycle regime.

Keywords: High field terahertz; Coherent control; Extreme nonlinearities; Field resolved measurements

1. INTRODUCTION

Few-cycle light pulses in the elusive THz region of the electromagnetic spectrum have enabled new directions in fundamental science and technology[1]. This is mainly due to two unique benefits of THz optoelectronics: The possibility of generating inherently phase stable transients and the availability of field-sensitive detectors. Recently these technologies have been extended across an ultrabroad spectral window approaching the near infrared[2-5]. Thus, they are ideally applicable to resonant probing of low-energy elementary excitations in condensed matter on a sub-cycle time scale[6-11]. Actually, novel applications which have been envisioned for THz nonlinear optics and coherent control schemes, require enhanced field amplitudes[12-15]. Until recently this domain has been reserved to large-scale facilities such as synchrotrons and free-electron lasers.

Here we introduce a hybrid table-top laser system, which combines a flexible and compact Er:fiber laser with a high energy Ti:sapphire amplifier to provide unprecedented field amplitudes while maintaining the benefits of THz optoelectronics. Our setup generates few-cycle electromagnetic transients with inherently stable carrier-envelope phase (CEP) and center frequencies tunable from 1 to 107 THz. Peak electric fields of up to 108 MV/cm with maximum powers of 10^8 W and energies as large as 19 μJ pave the way to extreme nonlinear THz optics[16]. In the following sections we discuss this new laser system and four lead-off examples of extreme nonlinearities driven by the electric and magnetic fields of the transients, which go beyond the perturbative regime.

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2. NOVEL SOURCE OF HIGH FIELD THZ TRANSIENTS

Fig. 1 displays a schematic of our THz source. A low-noise four-branch Er:fiber system with all-fiber frequency conversion stages serves as a starting point. The output pulses from the first arm at a center wavelength of 1.55 μm is frequency doubled and seeded into a Ti:sapphire amplifier encompassing a regenerative cavity and a double-pass power stage (repetition rate: 1 kHz). A train of 5-mJ pulses of a duration of 100 fs featuring rms power fluctuations well below 0.2 % (integrated over 30 s) is obtained. Three schemes for the generation of intense and widely tunable THz fields from this pump as well as from the various fiber branches are implemented:

(*i*) Optical rectification of the 5-mJ pulses in a large-area ZnTe crystal provides transients with center frequencies around 1 THz. The single-cycle wave forms reach peak electric field amplitudes of up to 400 kV/cm and cover the low-frequency range from 0.1 to 3 THz.

(*ii*) In a second approach we obtain even more intense THz pulses. Two identical two-stage optical parametric amplifiers (OPAs) are pumped with the Ti:sapphire output pulses. A white-light continuum generated in a sapphire window serves as a shared seed for both amplifiers and ensures a high degree of mutual phase coherence. The signal components are superimposed for type-II or type-I difference frequency generation (DFG) in a GaSe or $AgGaS_2$ crystal. This way, we achieve pulse energies as large as 19 μJ. Since both OPAs share the same frequency comb as a seed, the carrier-envelope offset is expected to cancel in the DFG process (see next section and Refs. 17 and 18). The center frequencies are tunable up to 72 THz [16].

(*iii*) Finally, we generate phase-stable THz transients with record-high center frequencies of up to 107 THz directly in the fiber laser. One amplifier branch is frequency shifted to generate tunable spectra in the wavelength range from 1.0 to 1.4 μm. For this purpose we employ dispersion-optimized highly nonlinear bulk germanosilicate fibers, which guarantee excellent stability. A second branch is operated at the fundamental wavelength of 1.55 μm. Both pulse trains are superimposed and focussed into a periodically poled $LiNbO_3$ crystal to generate THz transients via DFG[19]. The mutual phase jitter of both branches amounts to 43 attoseconds, which is less than one percent of the oscillation period of the carrier wave[20]. Therefore, CEP stability of the THz-pulses is expected also for this scheme. Due to the high repetition rate of 49 MHz and an average THz power of up to 1.4 mW, this mid- to near-infrared source is ideally suited for sensitive spectroscopy with field sensitive detection. In a later stage, the pulses may be further amplified in Ti:sapphire pumped parametric optics, similar to Ref. 21.

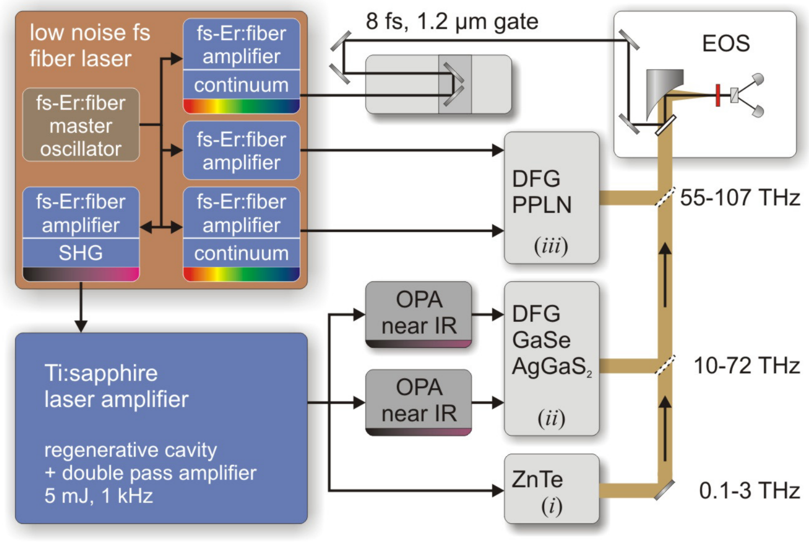

Figure 1. Schematic setup of the novel hybrid laser system with a four branch Er:fiber laser, a high-power Ti:sapphire amplifier, optical parametrical amplifiers (OPA), difference frequency stages (DFG), and a setup for electro-optic sampling (EOS). The accessible frequency ranges are indicated next to the THz-generation stages.

3. ULTRABROADBAND ELECTRO-OPTIC SAMPLING

The electric field traces of the THz transients from all three generation schemes are detected via electro-optic sampling with the pulses of a fourth Er:fiber branch. To this end, the 100-fs pulses from the Er:Fiber amplifier are coupled into a robust splice assembly of two nonlinear bulk fibers. The dispersion profiles of these fibers are numerically optimized for extremely broadband output spectra. Pulses as short as 7.8 fs with smooth spectra spanning the range from 0.9 µm to 1.4 µm are available after this compression stage[22]. Recently, we demonstrated even shorter pulses comprising only a single oscillation cycle of the carrier wave with a duration of 4.3 fs by coherently combining two such ultrabroadband spectra[23]. Pulses of this kind permit electro-optical gating even at the highest center frequency. The bandwidth is sufficient to resolve oscillation frequencies far above 100 THz directly in the time domain[5].

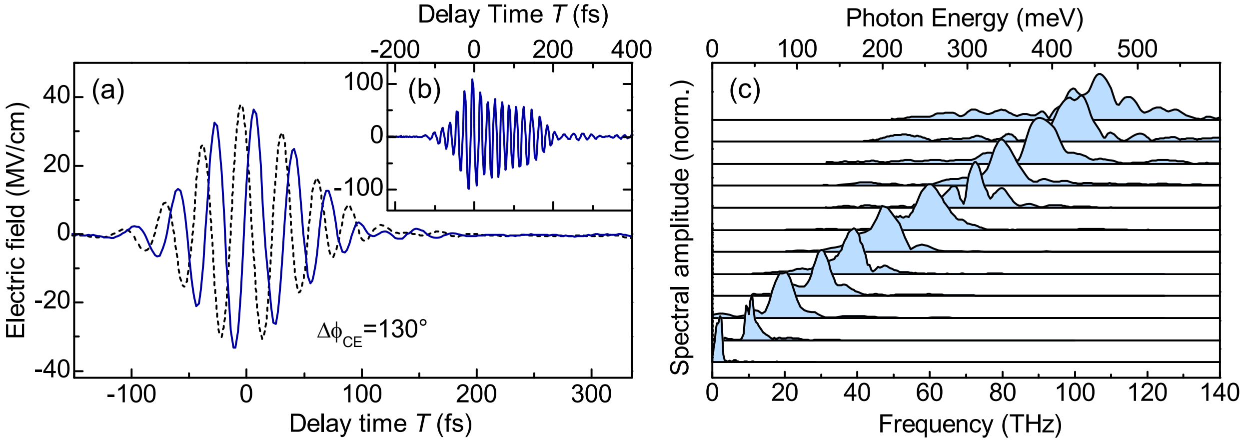

Figure 2. (a) Typical THz transient obtained from type-II difference frequency mixing in GaSe (blue line). The carrier envelope phase ϕ_{CE} is changed via the temporal overlap of the near infrared pump waves (dashed line). (b) THz transient from a thick emitter crystal, which reaches a peak field of 108 MV/cm. (c) Output spectra of the new hybrid laser source, obtained as Fourier transforms of the transients. The tuning range includes center frequencies from 1 THz up to 107 THz.

Fig. 2 (a) depicts the temporal trace of a typical high-field transient generated from scheme (*ii*). The fact, that it is possible to scan the field shape in a stroboscopic manner, is the ultimate proof of the phase stability of the OPA-based difference frequency technique. Moreover, by simply varying the temporal overlap of both pump pulses, the carrier envelope phase might be freely selected, as illustrated by the dashed curve. A proper selection of the generation crystal provides peak fields of up to 108 MV/cm, as shown in Fig. 2 (b). This value is easily calibrated by means of a thermal powermeter, since the average THz power reaches a value of 19 mW. These transients are by far the most intense THz pulses detected electro-optically to date[16].

Fourier transformation of transients from all three generation schemes yield phase and amplitude spectra which highlight the unprecedented bandwidth of more than 10 optical octaves with frequency components extending from 100 GHz to 140 THz [Fig. 2 (c)]. With photon energies spanning the entire range from 0.4 meV to 580 meV, many important resonances in condensed matter are directly accessible by intense THz pulses with full amplitude and phase resolution.

4. NON-PERTURBATIVE THZ NONLINEARITIES IN CONDENSED MATTER

Our transients reach electric fields as high as 1 V/Å which is comparable to the potential gradients within typical atoms. Therefore we expect to induce an extremely nonlinear response when applying these pulses to condensed matter. Electro-optic sampling provides direct access to amplitude and absolute phase information of the reemitted fields. In the following, we discuss four lead-off examples of non-perturbative nonlinearities in solids.

4.1 Coherent Control of Intra-Excitonic Transitions

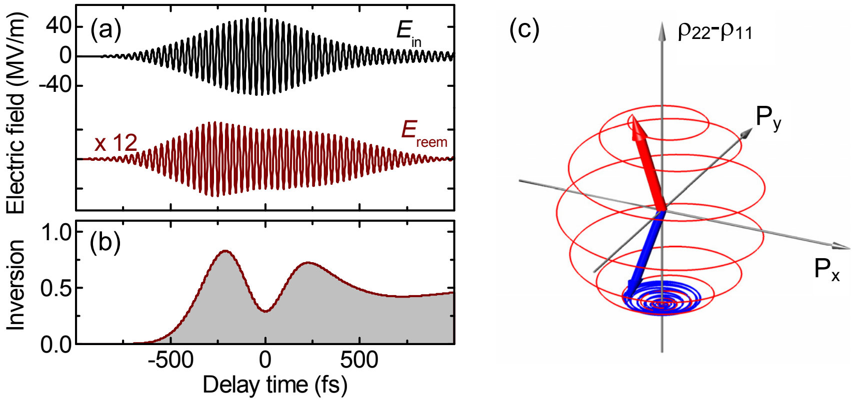

Figure 3. (a) THz transients resonantly drive the internal 1s-2p transition of paraexcitons in Cu_2O (black curve). The system reemits a field (red curve), which exhibits a non-monotonous envelope characteristic of up to two internal Rabi cycles. (b) Results of a microscopic theory confirm an oscillatory change of the population inversion of the 1s-2p two-level system. (c) Bloch sphere representation of the 1s-2p intra-excitonic two-level system.

THz transients are especially well suited to couple to low-energy excitations of quasiparticles which may not even be accessible by other means for reasons such as vanishing interband dipole matrix elements. Electron-hole pairs in semiconductors which are bound by Coulomb-interactions, are an important example. They exhibit a level structure which is analogous to the hydrogen atom and have been discussed as potential candidates for Bose Einstein condensation (BEC) in solid-state systems.

Excitons that are relevant for BEC are usually long-lived and often exhibit weak if any coupling to visible and near-infrared light. Thus, it is a fundamental challenge to control these excitations by conventional optical techniques. The optically dark 1s paraexcitons in Cu_2O represent a prototypic example. Intense THz radiation turns out to be an elegant way not only to visualize these quasiparticles, but also to control their orbital degree of freedom: Multi-terahertz fields of the order of 0.4 MV/cm are used to promote 1s paraexcitons in Cu_2O into the 2p state, coherently. Fig. 3 (a) depicts an example of the nonlinear field response monitored by ultrabroadband electro-optic sampling. We identify up to two internal Rabi cycles, which is in quantitative agreement with a microscopic many-body theory[24,25]. The results point out a promising route for preparing ultracold exciton gases which may be eligible for potential Bose-Einstein condensation in semiconductors.

4.2 Extreme THz-nonlinearities in bulk semiconductors

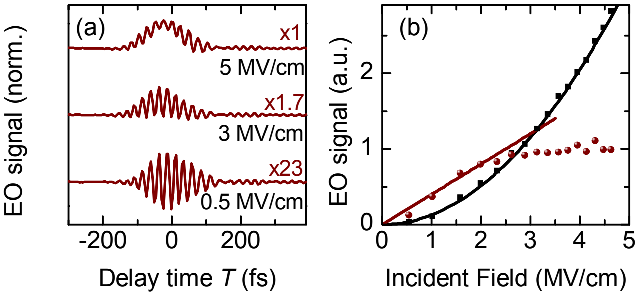

Figure 4. (a) Extremely nonlinear electro-optic sampling of transients with increasing peak fields in a 90-µm-thick GaSe detector crystal. The peak field is indicated next to each transient. (b) Field dependence of the unipolar (squares) and the rapidly oscillating components (circles) in (a).

In a second example, we investigate higher-order nonlinearities in bulk semiconductors. To this end, we increase the THz power yet further to reach peak intensities of 15 TW/cm^2 (see Ref. 16). The induced effects may be directly observed in the electro-optic sensor itself (see Fig. 4): Up to fields of 2 MV/cm, the electro-optic signal scales linearly with the THz amplitude [circles in Fig. 4(b)], as one would expect for a $\chi^{(2)}$ nonlinearity. However, for increasing THz field strengths, a unipolar component which is superimposed on the transient is clearly discernible in the time-domain data. The component which oscillates with the carrier frequency of the THz pulse saturates while the unipolar signal (squares) increases quadratically, which indicates $\chi^{(3)}$ and $\chi^{(4)}$ processes involving two and three THz photons, respectively. At intensities beyond 5 MV/cm, we start to observe THz induced continuum generation, interband luminescence, and ultimately THz radiation damage of the detector, indicating a catastrophic divergence of the power series expansion of the nonlinear polarization. More detailed investigations, which also include effects of crystalline anisotropy, are under way.

4.3 THz magnetooptics: Femtosecond coherent control of antiferromagnetic magnons

Nonlinear optics in the THz domain up to now has solely relied on the strong coupling of the electric field component to the charge degree of freedom. The magnetic field coupling to magnetic dipoles usually is weaker by a factor given by the fine structure constant $\alpha = 1/137$ and has been considered to be negligible. With our high-field source we explore, how intense single-cycle transients interact with magnetically ordered solids. We demonstrate for the first time, that the THz field couples directly to the spin degree of freedom.

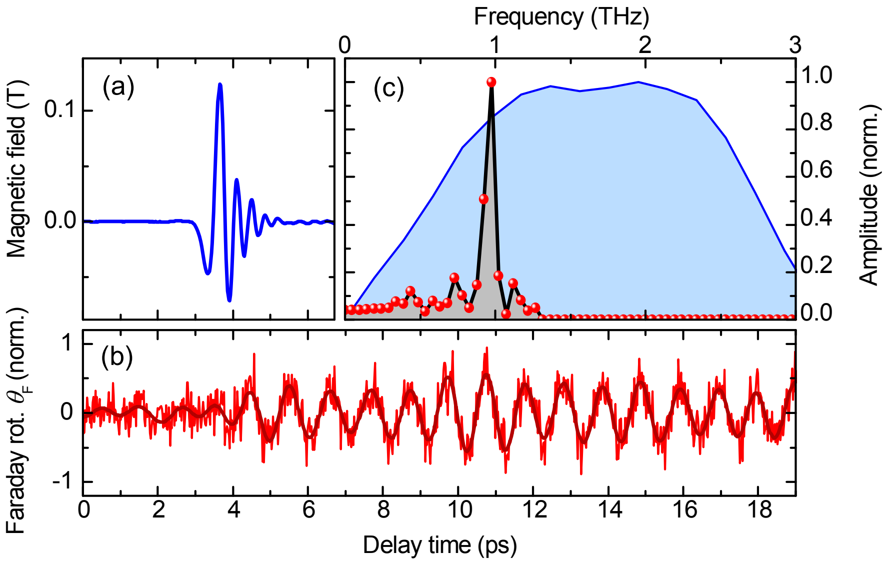

Figure 5. (a) Magnetic component of the THz transient incident on the NiO sample. (b) Faraday rotation of an 8-fs probe pulse copropagating through the antiferromagnetic sample. (c) Fourier transform of the excitation transient (blue) and the spin wave oscillations from (b).

For our experiment we choose the prototypic antiferromagnet NiO, which we expose to single-cycle transients as depicted in Fig. 5 (a). The peak electric field of 0.4 MV/cm corresponds to a magnetic field amplitude of 0.3 Tesla. The spectrum of our excitation pulse is depicted in Fig. 5 (c) and spans a frequency interval from 0.1 to 3 THz. An 8-fs near-infrared pulse probes any dynamics in the solid which is induced by the THz pulse. To our surprise, the strong electric field component leaves no measurable trace in the sample. Especially, there is no signature of any thermal heating of the electron-system. Instead, we observe a coherent magnon which is excited exclusively via direct magnetic Zeeman interaction of the transient with the spins. It manifests itself as a periodic Faraday-rotation of the probe pulse after the THz-excitation as depicted in Fig. 5 (b). These collective spin oscillations exhibit a frequency of 1 THz and a long dephasing time of 39 ps, corresponding to a narrow line in the Fourier transformation [Fig. 5 (c)]. This is the first time resolved observation of a magnon in an antiferromagnet with vanishing net-magnetisation. In a further step a phase-locked sequence of THz magnetic transients allows us to coherently switch the magnon oscillation on and off, on a single-cycle time scale (not shown here, see Ref. 26). Our experiment opens up a new route to directly access the spins in essentially all kinds of magnetically ordered solids. It may also inspire novel applications in next-generation high-speed memory devices and spintronics.

Beyond applications in semiconductors, we are currently also exploring the utility of our high-field THz facility in nonlinear optics of molecules and atoms. Due to the large ponderomotive potential, strong THz fields interacting with atoms may in principle be an attractive option for the generation of extremely high-order harmonics.

4.4 Ultrastrong light-matter coupling: Switching on a sub-cycle timescale

A fundamentally different class of THz nonlinear optics is addressed in the final set of experiments: ultrastrong light-matter coupling with the vacuum field of a microcavity rather than with an intense coherent external field. To enhance the interaction of light with electronic excitations, semiconductor microcavities spatially confine a photon field to the location of a semiconductor based light emitter. The confinement allows the electronic system to absorb and spontaneously reemit a photon repeatedly before dissipation sets in. This situation gives rise to mixed light-matter modes, so-called cavity polaritons. Recent efforts concentrate on the frontier of ultrastrong coupling, where the rate of photon exchange, i.e. the vacuum Rabi frequency, becomes comparable with the oscillation period of light itself[27,28,29]. Unconventional quantum electro-dynamics (QED) phenomena have been suggested for situations, where the interaction strength is modulated non-adiabatically[30]. While a well defined confinement has been achieved in all three spatial dimensions, control in the fourth dimension – time – has been barely developed.

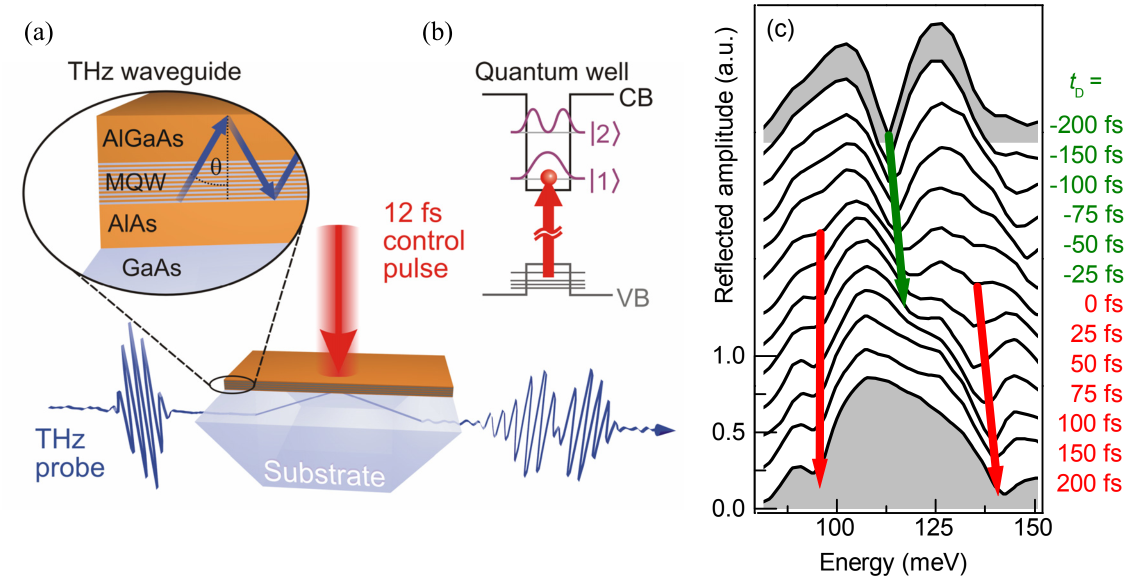

Figure 6. (a) Schematic of the femtosecond optical switch: A planar AlGaAs-waveguide based on total internal reflection encases 50 undoped GaAs quantum wells separated by AlGaAs barriers. Near infrared 12-fs control pulses populate level |1⟩, as depicted in the energy level diagram in (b). The buildup of cavity polaritons is monitored by a THz probe pulse reflected off the cavity. (c) THz reflectance spectra given for various delay times t_D between the near infrared pump and the THz probe pulse. The spectra reveal the non-adiabatic switch-on dynamics of ultrastrongly coupled cavity polaritons. Green arrow: bare cavity resonance, red arrows: intersubband cavity polariton resonances.

In our experiment, we demonstrate an all-optical switch to tune from weak to ultrastrong light-matter interaction and turn on maximum coupling on the femtosecond scale[31]. Our resonator comprises a planar AlAs step index waveguide, containing undoped GaAs quantum wells which serve as the light emitter [Fig. 6 (a)]. Switching takes place by photoinjecting electrons into the lowest electronic subband |1⟩ of the quantum wells with a 12-fs near-infrared laser pulse

[Fig. 6 (b)]. This process activates a mid-infrared transition to the next higher subband $|2\rangle$ which may efficiently couple to TM polarized waveguide modes propagating at an angle of 65°. Ultrashort multi-terahertz pulses trace the formation of light-matter interaction as a function of the delay time t_D following the control pulse [Fig. 6 (c)]. Within less than a cycle of light, the system changes abruptly from a bare microcavity to an ultrastrongly coupled cavity polariton system. In this regime, a novel class of extremely non-adiabatic phenomena becomes observable: We monitor directly, by detection of the reemitted terahertz field amplitude, how a coherent photon population converts to cavity polaritons during the ultrafast switching event. This system forms a first promising laboratory for unprecedented sub-cycle QED effects and represents an efficient room-temperature switching device at the ultimate speed.

5. CONCLUSIONS

We have introduced a novel source of phase-locked few-cycle transients in the intriguing THz and mid-infrared range with peak intensities outperforming even large-scale facilities such as free-electron lasers, by several orders of magnitude. Simultaneously, the tuning range marks a new record for table-top CEP-stable THz systems. Even the highest frequency pulses are gated electro-optically giving direct access to the temporal trace of the oscillating electromagnetic field. This light source paves the way to extremely nonlinear THz optics and coherent control mediated either by electric or magnetic field coupling to charge and spin degrees of freedom. The specific examples of coherent control of intra-excitonic transitions, collective spin oscillations, non-perturbative nonlinearities in bulk semiconductors and non-adiabatic control of light-matter coupling may illustrate some of the excitement about the emerging field of extremely nonlinear THz optics: Not only does it open unforeseen vistas in condensed matter physics, but it also facilitates a new kind of THz quantum optics and sub-cycle quantum electrodynamics.

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