Carrier-Envelope-Offset Phase Control of Ultrafast Optical Rectification in Resonantly Excited Semiconductors

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(Received 29 September 2006; published 19 April 2007)

Ultrashort pulse light-matter interactions in a semiconductor are investigated within the regime of resonant optical rectification. Using pulse envelope areas of around 1.5–3.5π, a single-shot dependence on carrier-envelope-offset phase (CEP) is demonstrated for 5 fs pulse durations. A characteristic *phase map* is predicted for several different frequency regimes using parameters for thin-film GaAs. We subsequently suggest a possible technique to extract the CEP, in both sign and amplitude, using a solid state detector.

DOI: 10.1103/PhysRevLett.98.167404 PACS numbers: 78.47.+p, 42.50.Md, 42.65.Re

It is well established in the field of ultrafast nonlinear optics (NLO) that the carrier-envelope-offset phase (CEP) can have a profound influence on the interaction between several-cycle laser pulses and a material [1]. Consequently, it is highly desired by the ultrafast optics community to be able to measure, and ultimately control, the absolute CEP of an ultrashort laser. In the regime of extreme NLO, the CEP influence from a several-cycle pulse has already been demonstrated in a number of areas, such as above-threshold ionization [2] and high-harmonic generation [3]. These phenomena require highly amplified pulses and although successful in obtaining the CEP, there can be some residual shifts due to various *higher-order* effects in the ionization physics and in the apparatus.

The *absolute* CEP is defined as the delay between the envelope maximum and the next peak of the carrier oscillation, and is quite different from the more common *relative* optical phase, namely, the phase between two different beams or pulses. In the solid state domain, proposals have been made for measuring the CEP using photoemission from a gold cathode [4]; however, the underlying mechanism of an observed phase shift is not clear and there remains geometrical averaging problems from the rough surfaces. Cundiff and co-workers have succeeded in measuring the relative CEP using quantum interference in a semiconductor [5], and used this effect to stabilize the phase evolution of a laser [6]. Though such self-referencing techniques are phase sensitive, they have yet to determine the absolute CEP of a single pulse, and some of the technical difficulties have been highlighted recently [7].

Several schemes for obtaining the CEP using semiconductors and *resonant* NLO have also been discussed recently. For example, the phenomenon of carrier-wave Rabi flopping [8] offers an alternative means to monitor phase-dependent propagation in semiconductor thin films [9], which exploits multiple Rabi flops (requiring $>4\pi$ area pulses [8]) that are sensitive to the carrier envelope phase. Unfortunately, no one has yet determined a value for the CEP using such a method. As has been reported previously, the optical signatures of carrier-wave Rabi flopping and the related second-harmonic-generation (SHG) in disguise of third-harmonic-generation [10] are complicated by a plethora of competing nonlinear effects that wash out any clear CEP signature [9,11]; so it seems unlikely that one can exploit such techniques to measure, or ''reverse engineer'', the CEP. In this Letter we introduce a new idea that considers several-cycle optically excited density oscillations in a direct-gap semiconductor film in the presence of an electrical bias. This results in phase-dependent signals that can be robust with respect to incident field strength (at least within a certain intensity range) while minimizing potential problems from long propagation and off-resonant excitation. Our results imply that full extraction of the CEP should be possible, *both in sign and amplitude*. Strategically, this scheme works at lower intensities than those required for carrier-wave Rabi flopping, and thus can work without the need for intensity calibration.

Since the pioneering experiments of Planken *et al.* [12], it is known that electrically biased semiconductors when optically excited with a short pulse will result in optical rectification, yielding electromagnetic transients typically found, or probed, at THz frequencies. Earlier related works on virtual photoconductivity by Yablonovitch *et al.* [13], and virtual transitions in quantum wells by Chemla *et al.* [14] and Yamanishi [15], have demonstrated that nonlinearities associated with the virtual transition of electrons and holes can result in very high speed modulation of the intraband current, even for bulk samples. For quantum wells and thin films, this effect is maximized by applying a bias field perpendicular to the in-plane charge carriers. Thus, the optically driven electric fields are determined from the dynamics of the interband polarization which can be extremely fast.

Proposal.—The aforementioned optical rectification regimes can be successfully described within the rotating-wave-approximation. In contrast, we wish to study the CEP influence of dipole fields created in a regime where the rotating-wave approximation obviously fails. A schematic of our proposed excitation scheme is shown in Fig. 1. A resonant nonlinear pulse excites a semiconductor film, which initiates carrier density oscillations [16]. These den-

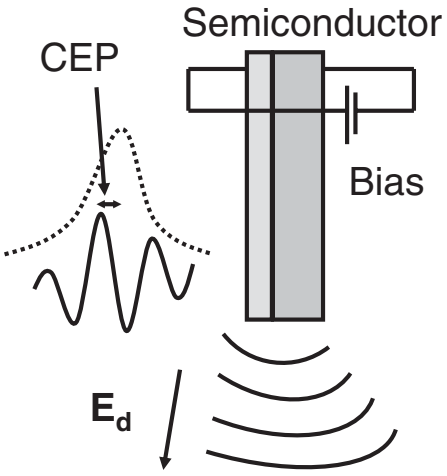

FIG. 1. Schematic of the light-matter excitation scheme. An ultrashort several-cycle optical pulse, with a specific phase (CEP), excites a large density of carriers in the semiconductor thin film. When electrically biased, even weakly, this results in the emission dipole-induced electric field propagating in the far field.

sity oscillations, when biased with an electric field, manifest in the emission of electromagnetic transients [12,17]. Similar schemes have been discussed previously for Rabi flopping [18] using much longer pulses (~150 fs) within the regime of the slowly-varying envelope approximations, where no phase dependence occurs. Here, we consider excitation of the semiconductor thin film with a several-cycle optical field of only 5 fs duration.

Theory.—To describe the ultrafast light-matter interactions we use a finite-difference time-domain (FDTD) method to solve the relevant Maxwell equations. We do not invoke any slowly varying envelope approximations for the phase and amplitude terms of the field. One has $\vec{\nabla} \times \vec{E} = -\mu_0 \partial \vec{H}/\partial t$, $\vec{\nabla} \times \vec{H} = \partial \vec{D}/\partial t$, $\vec{D} = \epsilon_0 \epsilon_r \vec{E} + \vec{P}_{nl}$, where $\vec{E}$ is the electric field, $\vec{H}$ is the magnetic field, $\vec{D}$ is the electric flux density, ϵ_r the relative electric permittivity, and $\vec{P}_{nl}$ the macroscopic nonlinear polarization. We model a single-shot excitation pulse $\vec{E}(t) = A\mathrm{sech}[-(t - t_0)/\tau_0]\sin(\omega t + \phi)\hat{\mathbf{n}}$, where A is the amplitude of the pulse, t_0 is the pulse offset, $\hat{\mathbf{n}}$ is a unit vector in the direction of the polarization, and $\tau_p = 2\mathrm{arcosh}(\sqrt{2})\tau_0$ is the full width at half maximum (FWHM) of the intensity profile (5 fs), in addition, $\omega = 1.24$ eV is the center frequency of the pulse (corresponding to a wavelength of around 1000 nm, which can be realized with a Ti:sapphire laser), and ϕ is related to the carrier envelope phase, CEP, though CEP $= \phi + \pi/2$ Since we perform calculations for 1D propagation only, we henceforth drop the vector notation with the electromagnetic fields.

The nonlinear polarization is modeled using semiconductor Bloch equations by accounting for two bands and the full region of wave vector space. Our approach is to include contributions from the semiconductor continuum states, excitation-induced interband relaxation, and a realistic (nonparabolic) band structure. This allows us to gain a qualitative insight into the essential underlying physics and focus on the important role of essential non-rotating-wave effects. Such a model is justified for several reasons. First, similar approaches have been used successfully to qualitatively model various experiments carried out for semiconductors in these ultrafast and highly nonlinear regimes [9,10,19]. Second, we have carried out calculations with and without excitons (within a first-order Hartree-Fock approximation [20]) and verified that the CEP dependence and emitted signals hardly change at all [21]. This stems from the fact that we are dealing with ultrabroadband nonperturbative field interactions that create a large density of electron-hole pairs ($>10^{19}$ cm^{-3}). Although our focus here is on introducing the qualitative physics of a new highly nonlinear excitation regime, we remark that at low densities, and for a quantitative understanding of the Coulomb correlations, nonperturbative Coulomb interactions can have an important influence on the short pulse dynamics [22].

Following similar motivation given elsewhere [19], we employ a tight binding approximation to obtain the broadband electron and hole (full band) dispersions $\epsilon_n = t_n^0 - 2t_n^1[\cos(ak_x) + \cos(ak_y) + \cos(ak_z)]$, where $n = e, h$ corresponds to electrons and holes, respectively, and $-\pi/a \leq k_i \leq \pi/a$. We take sample parameters close to those for GaAs: $a = 0.56$ nm is the lattice constant, and t_n^0 and t_n^1 characterize the center of the bands and the width of the bands. The band gap is $E_g = 1.42$ eV and the effective electron and hole masses are $m_e = 0.069m_0$, and $m_h = 0.5m_0$, where m_0 is the free electron mass. These are used to obtain the band structure parameters $t_e^0 = \frac{1}{2}E_g + 3(\hbar^2/m_e a^2)$, $t_h^0 = \frac{1}{2}E_g + 3(\hbar^2/m_h a^2)$, $t_e^1 = \hbar^2/2m_e a^2$, and $t_h^1 = \hbar^2/2m_h a^2$. Since it is known that the nonlinear signals are dominated by the huge resonant enhancement at the band edge from the fundamental heavy hole and conduction band transitions, we employ a two-band approximation for the dynamical equations that follow. In addition, we also include important propagation effects and a background or surface SHG contribution to the polarization (observed also in those experiments).

The off-diagonal density matrices take the form

$$\dot{\rho}_{\vec{k}}^{eh} = -i(\epsilon_e + \epsilon_h)\rho_{\vec{k}}^{eh} - i(\rho_{\vec{k}}^{ee} + \rho_{\vec{k}}^{hh})\Omega_{\vec{k}} - \gamma_t \rho_{\vec{k}}^{eh}, \quad (1)$$

where $\gamma_t(t)$ is the excited-induced dephasing rate [19] whose value depends on the field-induced pair density: $N(t) = 2V^{-1}\sum_{\vec{k}} \rho_{\vec{k}}^{nn}(t)$, where V is the volume and the factor of 2 accounts for spin. For a typical calculation, the dephasing rates change from 1/300 fs to about 1/40 fs as the carrier density rapidly increases. The electron or hole occupations are obtained from $\dot{\rho}_{\vec{k}}^{ee/hh}(t) = -2\mathrm{Im}[\Omega_{\vec{k}}(\rho_{\vec{k}}^{eh})^*]$ where the Rabi frequency $\Omega_{\vec{k}} = d_{\vec{k}}E/\hbar$,

with the $\vec{k}$-dependent dipole moment $d_{\vec{k}} = d_0 E_g/(\epsilon_e + \epsilon_h)$, and $d_0 = 0.4e$ nm is the optical dipole moment. We have also included intraband scattering within a relaxation time approximation (density dependent using a similar form to γ_t above), but verified that it plays a negligible role for these 5 fs excitation studies. The macroscopic polarization that feeds back to Maxwell equations, which is entirely real, is obtained from $P_{nl}(t) = 2V^{-1}\sum_{\vec{k}} d_{\vec{k}}[\rho^{eh}_{\vec{k}}(\vec{k}, t) + \text{H.c.}] + P^{(2)}_{\text{surf}}(t)$, where H.c. denotes the Hermitian conjugate. To account for surface SHG contribution, we also add in an average nonlinear susceptibility of $\chi^{(2)} = 150$ pV/m [23], which we take to be frequency independent; adjusting this value has little qualitative effects on our findings but including the effect is important for creating interference contributions that ultimately enable one to get the CEP sign as well as its amplitude.

Finally, we must compute the field emission initiated by the bias field. The dipole field is $P_d = dN_{2D}(t)A$, where d is the bias-induced dipole moment and A is the area of the optical pulse on the sample, taken to be $0.8e$ nm and $\pi(5\ \mu\text{m})^2$, respectively; N_{2D} is the 2D sheet density within the thin film taken to be 20 nm in thickness with a relative electric permittivity $\epsilon_r = 10.9$, mounted on a sapphire substrate with $\epsilon_r = 3.1$; this corresponds closely to the experiments of Mücke *et al.* [9]. The form of the emitted dipole-induced field is $E_d = -\mu_0 \ddot{P}_d/4\pi r$, though the actual spatial dependence will depend on the geometry. For the purpose of estimating typical field amplitudes, we take $r = 1$ cm which results in electromagnetic transients with peak field strengths about 10 kV/cm, comparable to other studies [18].

Results.—In Fig. 2(a) we display the sheet density transients in the semiconductor as a function of time. Two different CEP values (near 0 and $\pi/2$) are considered that result in substantially different dynamics of the electron-hole pairs. The input electric field from air is taken to be 3 GV/cm, yielding pulse areas of about 3π within the sample (importantly, below the regime of carrier-wave Rabi flopping). We highlight that a very large density of electron-hole pairs is created, corresponding to volume densities of about 3×10^{19} cm^{-3}. On transmission through a sample, there is little difference on a transmitted field. This is clarified by the example shown in Fig. 2(b) that shows the incident and transmitted fields for two different phases (there is only a minute difference as highlighted in amplified spectral region shown by the inset). Next, we calculate the dipole-induced emitted field from the semiconductor sample and obtain rich broadband spectrum [see Fig. 2(c)], with a substantial phase (CEP) influence, especially near the energies of around 1.3 and 3.8 eV. It should be clear that the full nonlinear spectra appears since we have not made any envelope approximations. As mentioned earlier, observable differences can also be obtained in the transmitted field (even outside the optical rectification geometry), but only if one ramps up the incident field to such a degree that other competing effects wash out any clear CEP signature.

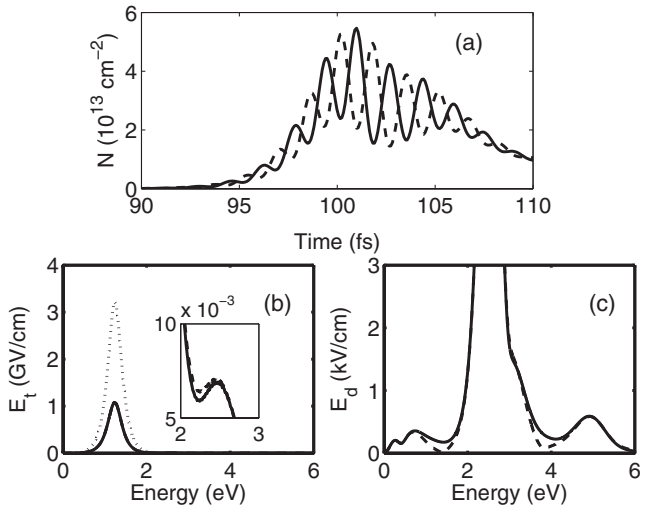

FIG. 2. (a) Electron-hole sheet density versus time for two different values of CEP (solid and dashed curve). (b) Incident field (dotted curve) and transmitted field using the same phases as above (solid and dashed curve); also shown as an inset is an amplified spectral region of the low intensity changes that have taken place. (c) Emitted field from the time dependent dipole, 1 cm away from the sample.

Although we have carried out detailed calculations for a two-band semiconductor model, the underlying physics behind the CEP signals can be qualitatively understood from simple NLO optics arguments. For any inhomogeneously broadened system that yields optical rectification, extremely short pulses with a center frequency ω_0 can create dipole fields at ω_0^{dc} (usual THz range, which stems from the broad bandwidth of the excitation pulse [12]), and $n2\omega_0$, with n a positive integer (written in terms of the familiar perturbative NLO field expansion). These broadband generated fields overlap in frequency at the spectral wings, e.g., between ω_0^{dc} and 2ω, and between 2ω and 4ω, and so on. Within our chosen energy scale, these overlapping spectral regions (expected to be near 1.2 and 3.6 eV) indeed closely correspond to the observed CEP-dependent features.

To motivate experimental measurements, we explore the CEP dependence in more detail by seeking out a *phase map* that depicts the emitted spectra, spanning 0–6 eV as a function of phase. We carry out a systematic investigation for different incident fields to explore the robustness of any phase trends with respect to incident field, since this usually destroys CEP signatures in resonant NLO. Figures 3(a)–3(c) display a clear CEP map that is qualitatively maintained for all three incident fields. In general, we find that a similar CEP dependence is obtained for pulse envelope areas of about 1.5–3.5π (0.25 to 1.4 I_0), but for higher

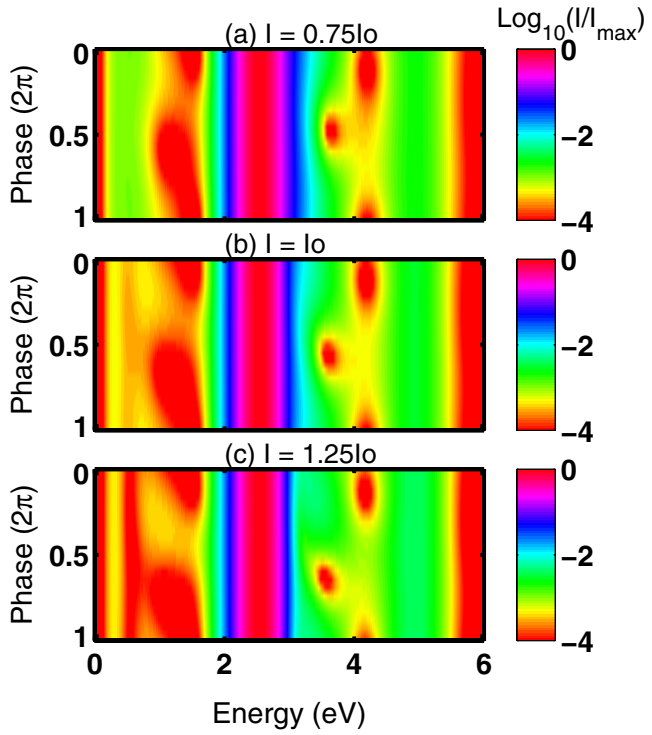

FIG. 3 (color online). (a–c) Contour "phase maps" of the emitted field 1 cm from the semiconductor, for several different incident intensities.

intensities one runs into problems from carrier-wave Rabi flopping (additional nonlinearities), while for lower intensities the nonperturbative response is too small to facilitate any CEP influence. Interestingly, besides the usual optical rectification signals at lower energies, we obtain very clear CEP signatures near 1.6 eV and also 4 eV, whose characteristic shape suggests reasonable determination of the CEP in both sign and amplitude. We also rehighlight the important point that without the surface or background SHG, the phase maps would only allow determination of the absolute CEP (not its sign); so this interference is essential for creating the CEP asymmetry.

In conclusion, we have introduced a light-matter interaction regime with several-cycle pulses and thin-film semiconductors, to predict a clear dependence on the CEP in resonant nonlinear optics. In contrast to other proposed techniques, our idea exploits optical rectification geometry in the regime of resonant nonlinear optics, works at lower pulse intensities, requires little intensity calibration, and minimizes propagation problems with thicker samples. Phase maps have been presented for GaAs thin films.

This work was supported by Natural Sciences and Engineering Research Council of Canada and the Canadian Foundation for Innovation. We thank S. T. Cundiff, T. B. Norris, P. Corkum, J. E. Sipe, M. Wegener, H. Nakano, A. Ishizawa, J. M. Fraser, and M. M. Dignam for stimulating discussions.

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