Third-harmonic generation in disguise of second-harmonic generation revisited: role of thin-film thickness and carrier-envelope phase

C. Van Vlack and S. Hughes

Department of Physics, Queen's University, Kingston, ON K7L 3N6 Canada

Received September 7, 2006; accepted October 9, 2006;
posted October 19, 2006 (Doc. ID 74830); published December 23, 2006

It has previously been reported that a peak at the spectral position of the second harmonic of an excitation laser can be generated in an inversion-symmetric medium in the regime of extreme nonlinear optics and that this peak may be exploited to measure the carrier-envelope phase of the excitation pulse. Here we revisit this phenomenon with regard to reverse engineering the carrier-envelope phase and demonstrate that the thin-film thickness and the incident field can have a drastic influence on pulse propagation, and so the reverse engineering would likely fail. © 2006 Optical Society of America

OCIS codes: 190.7110, 300.6470.

The coherent optical response of two-level atoms (TLAs)[1–3] and direct gap semiconductors[4] have been intensively studied over the past four decades. This has enabled researchers to explain a variety of interesting ultrafast nonlinear optical effects, such as Rabi flopping and self-induced transparency.[5–7] Virtually all of these phenomena have been successfully explained and understood by exploiting light–matter interaction theories that employ the slowly varying envelope approximations for the phase and amplitude terms of the excitation laser field. These studies are interesting not just in their own right but frequently lead to new applications in optical science and technology.

With the continued progress in ultrashort pulse generation, intensity envelopes containing a few optical cycles are now routinely available, and so the slowly varying interaction pictures can change. In such regimes, it becomes imperative to be able to measure, and also control, the carrier-envelope phase, CEP, defined as the delay between the envelope maximum and the next peak of the carrier oscillation. The electric field of interest can be written $E(t)=\tilde{E}(t)\sin(\omega_0 t+\mathrm{CEP}+\pi/2)$, where ω_0 is the frequency of the optical carrier and $\tilde{E}(t)$ is the familiar pulse envelope.

In the regime of extreme nonlinear optics, this envelope phase can have an influence on the light–matter interactions. One example is carrier-wave Rabi flopping,[8] which leads to the breakdown of the area theorem[3] and to phase-dependent Rabi oscillations. Although predicted for a TLA, this effect has been successfully observed in GaAs thin films.[9] Since the regime of carrier-wave Rabi flopping is dependent on the excitation phase, further analysis of this phenomenon has predicted a possible way of measuring the CEP.[10] Our experience implies that this is not a straightforward way of inferring the phase, because real measurements are complicated by a plethora of nonlinear propagation effects.

In a somewhat similar regime, Tritschler *et al.*[11] have exploited a new signal that arises from the third-harmonic generation (THG) in disguise of the second-harmonic generation (SHG) to map out an interesting phase diagram that may allow one to measure the CEP via a single-shot laser experiment. The origin of this intriguing effect is a nonrotating wave contribution that occurs when the broadband fundamental spectrum overlaps the third harmonic, resonantly enhanced at the transition energy of a TLA-like medium; although this signal appears near $2\omega_0$ it is typically different in spectral appearance from the usual SHG and is influenced in a significantly different way, especially through the CEP. Consequently, it appears in a much cleaner way than, e.g., via interference between a fundamental and overlapping third harmonic,[12] because the THG in disguise of SHG is essentially a resonant effect and has a different phase dependence. While the third-harmonic field appearing near an expected second harmonic is to be anticipated for short pulses, no such phase dependence has yet been reported with regard to measuring the phase, though the signal itself has been measured for ZnO thin films.[11] Motivated by this interesting prediction as a means to measure the CEP, and by the experiments, here we explore this effect in more detail (for this specific prediction) and show that if one is not careful to include important spatial propagation effects that are typically there for real samples, in general such phase-dependent effects may become washed out or easily misinterpreted. Our revisit to this problem is further motivated by the continued interest within the ultrafast optics community to be able to accurately measure and ultimately control the absolute phase.[13]

Keeping in mind that the experiments utilized ZnO thin films on sapphire,[11] we consider TLAs with a resonance energy close to the ZnO bandgap, namely, $E_g=3.3\,\mathrm{eV}$, which yields the required THG in disguise of SHG. We consider a several-cycle ultrashort pulse that excites a thin-film semiconductor mounted on a substrate with an in-general different dielectric constant. Figure 1 depicts a schematic of the pulse-sample excitation scenario. We follow the theoretical approach presented previously[8,14] (see also Fig. 1 of

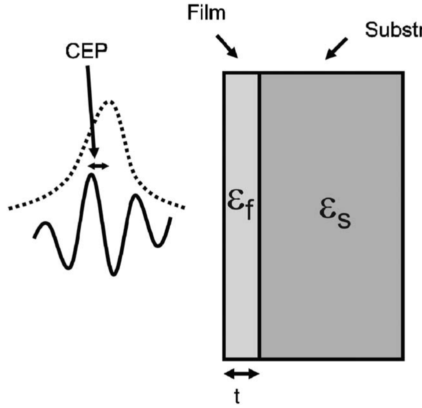

Fig. 1. Schematic of the light–matter geometry. An ultrashort several-cycle optical pulse comes in from the left and excites a nonlinear thin film of thickness t, where the film and substrate have optical dielectric constants, ϵ_f and ϵ_s, respectively.

Ref. 11), and adopt an ensemble of TLAs. The excitation pulse has a central energy $\hbar\omega_0 = 1.6$ eV, a FWHM intensity of $\tau = 5$ fs, and a sech envelope profile. For a ZnO thin film, the relevant (high-frequency) dielectric constant is $\epsilon_t = 4$,[15] while for sapphire it is $\epsilon_s = 3.1$. The polarization in the ZnO film is therefore dependent on a sum of linear and nonlinear optical source terms that are complicated by a possible back-reflection from the substrate due to the dielectric mismatch.

We point out that we are merely using ZnO-like parameters to connect to the typical experiments that have successfully been performed and to address more clearly the prediction that has been reported previously for a TLA material system. To come closer to real experiments carried out on semiconductors from a quantitative viewpoint, presumably one should include the full energy bands, complex many-body Coulomb interactions, and any real SHG, then follow a detailed systematic comparison with experiments. This will result in much richer and more complex signals, and is indeed closer to the technique used previously to explain the phase-averaged experimental results,[11] following a discussion of the basic mechanism and CEP dependence presented for a TLA ensemble. For now, we report the essential physics and added complications that arise already in trying to exploit such a regime of THG in disguise of SHG as a means to measure the CEP, even with a simple TLA-like nonlinear medium.

For thin-film samples, one must include both nonrotating wave effects and nonslowly varying envelope effects in space. The ultrafast dynamics of the nonlinear medium is calculated from the optical Bloch equations, namely, $\dot{\rho}_{12} = i\Omega n - (\Gamma + i\omega_g)\rho_{12}$ and $\dot{n} = i2\Omega(\rho_{12} - \rho_{12}^*)$, where n is the population difference between the excited and ground states, ρ_{12} is the off-diagonal element, and $\Omega = dE/\hbar$ is the Rabi frequency that includes the underlying optical carrier. These ultrafast dynamics act as the microscopic source for the macroscopic nonlinear polarization $P_{\mathrm{NL}} = 2Nd\,\mathrm{Re}[\rho_{12}]$, which couples self-consistently back to the propagating electromagnetic field. The Bloch equations are solved by a fourth-order Runge–Kutta method, and the Maxwell equations are solved by the finite-difference time-domain technique.[16] The off-diagonal relaxation time (dephasing) is set to $\Gamma^{-1} = 50$ fs (this value has little qualitative influence in any of our findings or conclusions), and the population decay rate is taken to be much longer than the excitation pulse and so plays no role for these ultrafast studies. The total density of TLAs is taken to be 6×10^{18} cm^{-3} and the dipole moment $d = 0.19e$nm. To include the background dielectric constant within the nonlinear film, the total polarization becomes $P = \epsilon_0\epsilon_f E + P_{\mathrm{NL}}$, while within the substrate $P = \epsilon_0\epsilon_s E$. We adopt scalar notation for the fields as only one-dimensional propagation is considered.

Figure 2 shows some typical examples of the nonlinear THG in disguise of SHG that occurs for a fixed input field, taken to be weak enough to minimize higher-order effects but strong enough to produce a CEP dependence. The input electric field has an envelope area within the thin film of about $6-7\pi$ (Ref. 17) and thus is in the regime of carrier-wave Rabi flopping[8] ($>4\pi$) where phase-dependent effects are expected for several-cycle pulses. On the left panels [(a)–(c)] the transmitted signal associated with the SHG in disguise shows an obvious phase dependence when we assume equal dielectric constants for the film and substrate; a clear CEP dependence is obtained for all three propagation distances (50–200 nm) as a function of phase, in agreement with earlier predictions.[11] Note that we are considering an inversion-symmetric medium and there is no SHG, which would appear as a peak at an energy of 3.2 eV ($2\omega_0$); although not shown, no signal appears here. The gap or resonance energy of the nonlinear medium enhances the THG near 3.3 eV through phase-dependent light matter interactions. In principle then, it seems that such a phenomenon can indeed be exploited to measure CEP by straightforward reverse engineering.

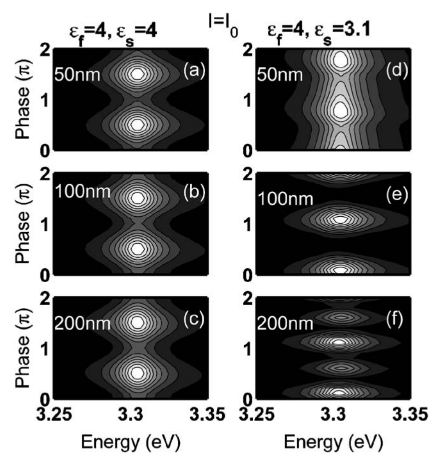

Fig. 2. Contour plot of the transmitted THG in disguise of SHG near the resonance energy of the TLA ensemble (3.3 eV). Panels (a)–(f) depict various film thicknesses of the nonlinear medium for two different substrate optical constants and a fixed input intensity from a 5 fs pulse with a central energy at 1.6 eV. The phase shown is the CEP $-\pi/2$.

Next, a more typical case is considered with different dielectric constants between ZnO film and a sapphire substrate. In Figs. 2(d)–2(f) there is now a profound influence mediated by the dielectric mismatch. It is seen that the influence of nonlinear film thickness is substantial, resulting in no obvious trend with phase and in multiple peaks for the thicker sample. If one then applied the phase diagram as shown in Figs. 2(a)–2(c), clearly one would obtain the incorrect CEP as the peak positions move depending on various factors, e.g., the nonlinear film thickness.

On further analysis of varying thicknesses, dielectric mismatches, and input intensities, we conclude that great care would be needed to interpret any sensible CEP trend with the resulting data. In Figs. 3(a) and 3(b) we show further evidence that the peaks can also move and multiply (more than two peaks per 2π phase scan) in general as a function of incident intensity; additionally, the phase dependence can completely wash out during these two cases. We also carry out calculations with the dielectric mismatch [Figs. 3(c) and 3(d)], and now coincidentally come back to a CEP dependence that is very similar to the original calculations (i.e., a peak when the phase is $\pi/2$, $3\pi/2$), but only for a certain incident field and for a particular film thickness [Fig. 3(c)]. With this set of calculations, there seems no obvious dependence of the CEP for nonlinear thin films mounted on a substrate with a dielectric mismatch. Moreover, even with identical substrate and film dielectric constants, the influence of the THG signal in disguise of SHG can change or disappear completely.

In summary, we have revisited the prediction of THG in disguise of SHG in the regime of extreme nonlinear optics, and explored such a mechanism from the viewpoint of reverse engineering the CEP. We find that for nonlinear films and typical dielectric constants for materials where such an effect has been observed and predicted there will, unfortunately, be no clear dependence of the phase, though the effect itself is certainly there. Perhaps with a detailed theory-experimental analysis there might be some merit in attempting to extract the CEP, though it would be far from trivial, even for the simple case of a TLA ensemble. Further experiments in this exciting field and more detailed theoretical analysis may clarify some of these nontrivial dependences.

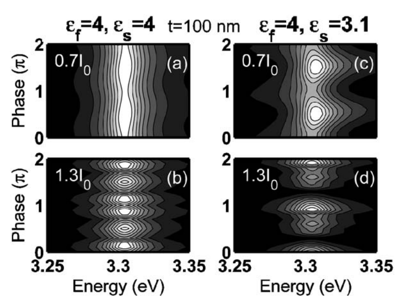

Fig. 3. Contour plots (a)–(d) show the transmitted nonlinear field near the energy gap for different input intensities and two different substrate optical constants; the thickness of the nonlinear film is fixed at 100 nm.

We thank A. Ishizawa and H. Nakano for ongoing discussions about this work and for sharing recent experimental results. We also thank M. Wegener for useful comments. Funding is gratefully acknowledged from National Sciences and Engineering Research Council and the Canada Foundation for Innovation. S. Hughes's e-mail address is shughes@physics.queensu.ca.

References

1. G. L. Lamb, Jr., Rev. Mod. Phys. **43**, 99 (1971).
2. S. L. McCall and E. L. Hahn, Phys. Rev. Lett. **18**, 908 (1967).
3. L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, 1995).
4. H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors*, 4th ed. (World Scientific, 2004).
5. I. I. Rabi, Phys. Rev. **51**, 652 (1937).
6. I. I. Rabi, J. R. Zacharias, S. Millman, and P. Kusch, Phys. Rev. **53**, 318 (1938).
7. S. T. Cundiff, A. Knorr, J. Feldmann, S. W. Koch, E. O. Göbel, and H. Nickel, Phys. Rev. Lett. **73**, 1178 (1994).
8. S. Hughes, Phys. Rev. Lett. **81**, 3363 (1998).
9. O. D. Mücke, T. Tritschler, M. Wegener, U. Morgner, and F. X. Kärtner, Phys. Rev. Lett. **87**, 057401 (2001).
10. O. D. Mücke, T. Tritschler, M. Wegener, U. Morgner, and F. X. Kärtner, Phys. Rev. Lett. **89**, 127401 (2002).
11. T. Tritschler, O. D. Mücke, M. Wegener, U. Morgner, and F. X. Kärtner, Phys. Rev. Lett. **90**, 217404 (2003).
12. See, e.g., M. Mehendale, S. A. Mitchell, J.-P. Likforman, D. M. Villeneuve, and P. B. Corkum, Opt. Lett. **25**, 1672 (2000).
13. See, e.g., D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, Science **288**, 635 (2000).
14. R. W. Ziolkowski, J. M. Arnold, and D. M. Gogny, Phys. Rev. A **52**, 3082 (1995).
15. See, e.g., K. Postava, H. Sueki, M. Aoyama, T. Yamaguchi, Ch. Ino, Y. Igasaki, and M. Horie, J. Appl. Phys. **87**, 7820 (2000).
16. A. Taflove and S. C. Hagness, *Computational Electrodynamics: The Finite-Difference Time-Domain Method*, 3rd ed. (Artech House, 2005).
17. This pulse area corresponds to (is created with) a peak incident electric field of 35 GV/m in vacuum.