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Determining the carrier-envelope offset frequency of 5-fs pulses with extreme nonlinear optics in ZnO

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We excite ZnO samples with two-cycle optical pulses directly from a mode-locked oscillator with average powers of several tens of milliwatts. The emitted light reveals peaks at the carrier-envelope offset frequency f_ϕ and at $2f_\phi$ in the radio-frequency spectra. These peaks can still be detected in layers as thin as 350 nm—a step toward determining the carrier-envelope offset phase itself. © 2002 Optical Society of America

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The carrier-envelope offset (CEO) phase, ϕ, is the phase between the rapidly oscillating carrier wave of a laser pulse and its electric field envelope. The time derivative of ϕ is the CEO frequency, f_ϕ. Recently, several approaches to measuring the CEO frequency for pulses directly out of a mode-locked oscillator were reported.[1–4] Related work for amplified pulses[5–7] addressed the role of the CEO phase. The underlying basic idea[1–4] is to consider the interference contribution that results from the superposition of the fundamental pulse spectrum, which by definition has a phase 1ϕ, and its second (third) harmonic, which has a phase 2ϕ (3ϕ). The resulting beating with difference phase 1ϕ (2ϕ) can be used to determine the CEO frequency f_ϕ ($2f_\phi$; Ref. 8). To actually measure this interference for pulses directly derived from a mode-locked oscillator, researchers have used different approaches. For extremely short pulses, which cover one octave in the frequency spectrum, this term immediately occurs when the second harmonic is generated in a suitable crystal. In Ref. 4, a 1-mm-thick crystal and the complete laser output power were used to observe the f_ϕ peak. For somewhat longer pulses, the fundamental needs to be spectrally broadened to generate sufficient spectral overlap.[1,2] In such a setup the laser pulses are sent into a (photonic crystal) fiber for spectral broadening via self-phase modulation (SPM) before they are again focused onto a separate second-harmonic-generation (SHG) crystal, where the two contributions interfere.

Apart from being quite complex, such setups[1,2] have the inherent drawback that the CEO phase changes within the setup because of different group and phase velocities in the fiber, in the thick SHG crystal, and in the air in between. Ideally, one would like to generate the spectral components that are due to SPM and SHG in one medium, which is so thin that ϕ does not change within the medium, and still get a large beating signal. Ultimately, this would allow one not only to measure the CEO frequency but also to determine the CEO phase. A simplified measurement of the CEO frequency along these lines and the potential to determine the CEO phase itself is the subject of this work.

Our home-built laser system delivers 5-fs optical pulses at $f_r = 81$ MHz repetition frequency and average powers up to 230 mW. The spectrum covers the range from ~650-nm wavelength to 1050 nm, the center photon energy is $\hbar\omega_0 = 1.5$ eV. All pulse properties are very nearly similar to Ref. 9. These linearly polarized pulses are sent into a balanced Michelson interferometer that is actively stabilized by means of Pancharatnam's screw.[10] Using a reflective microscope objective with a numerical aperture (NA) of 0.5, we can tightly focus these pulses to an intensity profile that is roughly Gaussian with a measured 1-μm radius (the Rayleigh length is 2.9 μm). At the output of the interferometer, an average power of 8 mW in one arm corresponds to a peak intensity of $I_0 = 0.6 \times 10^{12}$ W/cm^2 (same definition as in Ref. 11). The maximum accessible intensity of $I = 2.04 \times I_0$ corresponds to an intensity of 5.0×10^{12} W/cm^2 for constructive interference of the two arms, equivalent to a peak of the electric field envelope of $\tilde{E}_0 = 6 \times 10^9$ V/m = 6 V/nm. Note that ZnO has no center of inversion and that it is birefringent. For the approximately 100-μm-thick single crystal, the crystallographic **c** axis lies in the plane of the platelet. For the 350-nm-thin epitaxial film grown on a sapphire substrate, **c** is perpendicular to the film. The light emitted into the forward direction is collected by a second reflective microscope objective (NA, 0.5) and spectrally filtered (3-mm Schott BG 39) for removal of the prominent fundamental laser spectrum and is sent into a 0.25-m focal-length grating spectrometer

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connected to a CCD camera. Alternatively, we send the light through a combination of filters (3-mm Schott BG 39, 3-mm Schott GG 455, and Coherent 35-5263-000 480-nm cutoff interference filter for the 100-μm ZnO single crystal and Coherent 35-5289-000 500-nm cutoff interference filter for the 350-nm epitaxial film, respectively) onto a 50 Ω-terminated photomultiplier tube (Hamamatsu R 4332 bialkali photocathode), connected to a radio-frequency (rf) spectrum analyzer (Agilent PSA E4440A).

Figure 1 shows measured optical spectra in the spectral region energetically above the laser spectrum (which has its short-wavelength cutoff above 650 nm) and below the bandgap of ZnO of $E_g = 3.3$ eV for [Fig. 1(a)] low and [Figs. 1(b) and 1(c)] high excitation. All spectral components shown in Fig. 1 disappear if the ZnO crystal is moved out of focus. Polarization-dependent experiments under these conditions show that all these spectral components have the same linear polarization as the laser pulses. In Fig. 1(a), the light near the 390–470-nm wavelength is due to SHG, and the components above 500 nm are due to SPM. Interestingly, the independently measured interferometric autocorrelation of the laser pulses [with a thin β-barium borate SHG crystal; curve labeled IAC in Fig. 1(b)] is closely reproduced by the cut at the 395-nm wavelength [black curve in Fig. 1(c)]. This result indicates that the pulses are not severely broadened in the ZnO crystal as a result of, e.g., group-velocity dispersion. Furthermore, the spectral width of the SHG contribution indicates that phase-matching effects do not play a major role, which is not surprising considering the short Rayleigh range of the microscope objective. For higher intensities, the spectral overlap of SPM and SHG becomes immediately obvious from the spectra in Figs. 1(b) and 1(c), and a rich fine structure appears in this spectral region as a function of τ. Such measurements are crucial for comparison with theory (see discussion below). Feeding the spectral components in the interference region of Fig. 1 into the rf spectrum analyzer (Fig. 2), we find a large peak ($\geq$30 dB above noise level) at the carrier-envelope offset frequency f_ϕ as a result of the interference of the SPM and SHG and an additional smaller peak at $2f_\phi$ resulting from the interference of the SPM with the third harmonic.[12] To our knowledge a $2f_\phi$ peak has not been observed in any approach. The f_ϕ peak remains prominent for τ corresponding to two optical cycles (not shown), equivalent to a four-cycle pulse ($\approx$10-fs duration) at the output of the interferometer, showing that our approach would also work for longer pulses. Also, note that the average power at $\tau = 0$ corresponds to 64 mW, only a third of the total laser power. This average power potentially allows us to lock f_ϕ of the laser oscillator to some fixed frequency and use the main part of the laser power for further experiments. In the 350-nm-thin ZnO film, both the f_ϕ peak and the $2f_\phi$ peak are still visible. Interestingly, the $2f_\phi$ peak is even larger than the f_ϕ peak [Fig. 2(b)].

In conclusion, in our experiments we focused intense two-cycle optical pulses from a mode-locked laser oscillator onto ZnO and analyzed the emitted light with a rf spectrum analyzer. These experiments revealed two aspects.

(1) Using ~100-μm-thick ZnO single crystals, we found a large peak ($\geq$30 dB above noise level at 10-kHz bandwidth and 64-mW average power) at the CEO frequency, f_ϕ. The simplicity and robustness of the approach makes it attractive, e.g., for applications in frequency metrology[1,2] or for stabilization of the CEO frequency.

(2) A peak at the CEO frequency is still observed with a 350-nm-thin ZnO film. This might pave the road for the next step: Assuming that the CEO frequency is stabilized to zero by whatever means, using the 350-nm ZnO film, one could in principle measure the CEO phase of the incident pulses itself, which would be the ultimate complete characterization of the laser pulses. To get there, two necessary conditions have to be fulfilled: (i) Neither the electric field envelope nor the CEO phase must change significantly within the measurement apparatus. The

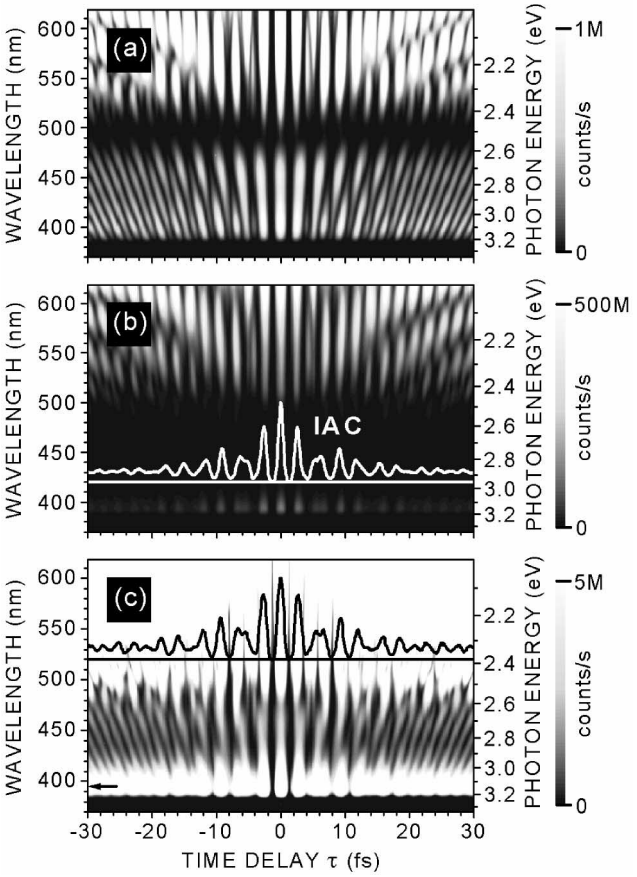

Fig. 1. Spectra of light emitted by the 100-μm-thick ZnO single crystal into the forward direction versus time delay τ of the Michelson interferometer, $\mathbf{E}\|\mathbf{c}$. (a) $I = 0.15 \times I_0$, (b) $I = 2.04 \times I_0$, and (c) same as (b) but with different saturation of the gray scale. The light intensities decay near the ZnO bandgap of $E_g = 3.3$ eV. The white curve in (b) labeled IAC is the interferometric autocorrelation measured independently with a β-barium borate crystal, and the black curve in (c) is a cut through the ZnO data at the 395-nm wavelength (indicated by the arrow). The optical spectra of the 350-nm-thin ZnO epitaxial layer are similar.

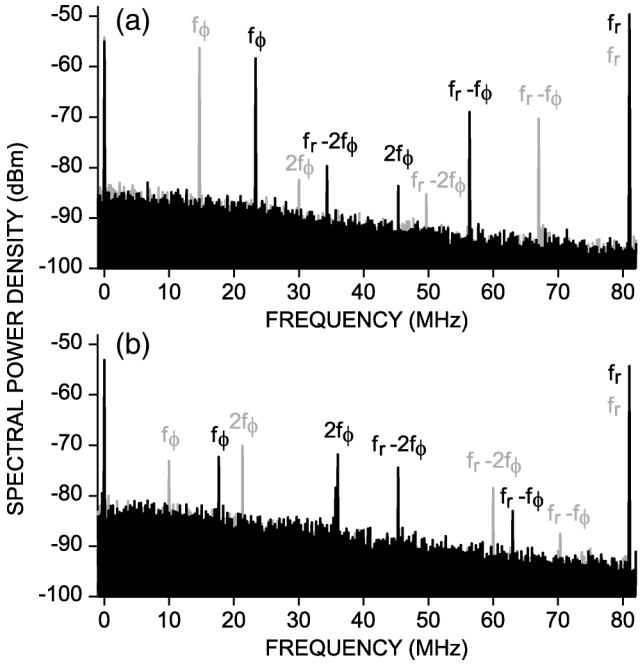

Fig. 2. rf spectra with 10-kHz resolution and video bandwidth, $\tau = 0$ fs. (a) 100-μm-thick ZnO single crystal corresponding to Fig. 1; 455–480-nm optical filter. (b) 350-nm-thin ZnO epitaxial layer; 455–500-nm optical filter. The peaks at repetition frequency f_r, carrier-envelope offset frequency f_ϕ, its second harmonic $2f_\phi$ and mixing products $(f_r - f_\phi)$ and $(f_r - 2f_\phi)$ are labeled. The black and gray data correspond to slightly different swivel mirror positions. When intracavity prism material (CaF_2) is removed, the f_ϕ and $2f_\phi$ ($f_r - f_\phi$ and $f_r - 2f_\phi$) peaks shift to the left (right).

latter condition is more stringent. The linear optical change of the CEO phase, $\delta\phi_{\mathrm{lin}}$, results from the different phase and group velocities[4] and can easily be calculated from the known Sellmeier coefficients of ZnO.[13] At $\lambda_0 = 2\pi c_0/\omega_0 = 826$ nm and $l = 350$ nm, $\delta\phi_{\mathrm{lin}}$ is merely 4.6% of 2π—a level of precision that might be sufficient for many applications. (ii) One needs a calibration or a detailed theory of how the measured beat signal is related to ϕ. Solving the Maxwell equations together with the known off-resonant values of $\chi^{(2)} = 6 \times 10^{-12}$ m/V (bulk) and $\chi^{(3)} = 7 \times 10^{-21}$ m^2/V^2 (bulk) for the actual ZnO sample geometry without further approximations, we can reproduce the behavior shown in Fig. 1 very well. Translating $\chi^{(3)}$ into a nonlinear refractive index of $n_2 = 5 \times 10^{-15}$ cm^2/W, the nonlinear shift of the CEO phase results as $\delta\phi_{\mathrm{NL}} = n_2 I 2\pi/\lambda_0 l = 1\%$ of 2π for $I = 5.0 \times 10^{12}$ W/cm^2 and $l = 350$ nm, even smaller than the linear optical shift. Unlike in the experiment, this off-resonant, perturbative treatment does not, however, deliver any significant peak at $2f_\phi$ in the rf spectra. This aspect is currently not resolved. Recently, a $2f_\phi$ peak was predicted theoretically[8] for resonant excitation of a semiconductor under the condition that the Rabi energy approaches the center photon energy, $\hbar\omega_0$, of the laser pulses, corresponding to the nonperturbative regime. In our ZnO experiments, the peak of the electric field envelope, $\tilde{E}_0 = 6 \times 10^9$ V/m, translates into a Rabi energy of $\hbar\Omega_{\mathrm{R}} = 1.1$ eV $\approx \hbar\omega_0 = 1.5$ eV. However, these resonant excitation conditions[8] cannot be directly applied to our ZnO experiments, in which intraband processes are expected to be important as well. Indeed, for the same electric field of $\tilde{E}_0 = 6 \times 10^9$ V/m, the ponderomotive electron energy of $\langle E_{\mathrm{kin}} \rangle = 1.27$ eV and the Bloch energy of $\hbar\Omega_{\mathrm{Bloch}} = 3$ eV are comparable to $\hbar\omega_0$ and $\hbar\Omega_{\mathrm{R}}$. Apart from being important for aspect (ii), this represents an exciting, unexplored, and challenging new regime of extreme nonlinear optics in solids in general.

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