Observation of Light-Phase-Sensitive Photoemission from a Metal

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We demonstrate that multiphoton-induced photoelectron emission from a gold surface caused by low-energy (unamplified) 4-fs, 750-nm laser pulses is sensitive to the timing of electric field oscillations with respect to the pulse peak. This observation confirms recent theoretical predictions and opens the door to measuring the absolute value of the carrier-envelope phase difference of few-cycle light pulses with a solid-state detector.

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The interaction of light with matter is primarily affected by the frequency of the oscillating light fields. The timing of oscillation cycles within a light pulse does not play any role in these interactions until the duration of the pulse becomes comparable to the light oscillation period T_0. Processes driven by intense pulses of laser light that comprise merely a few wave cycles (henceforth few-cycle pulses) have been predicted to depend on the timing of the oscillations of the electric field $E_L(t)$ with respect to the pulse peak in addition to the carrier frequency $\omega_L = 2\pi/T_0$ and amplitude envelope $E_0(t)$ [1]. This timing can be quantified by the carrier-envelope phase difference φ in the carrier-envelope description, $E_L(t) = E_0(t)\cos(\omega_L t + \varphi)$, of the electric field of a light pulse. Recent experiments confirmed the sensitivity of atomic processes driven by strong, few-cycle laser fields to the carrier-envelope phase difference and the need for precisely controlling this parameter of ultrashort-pulsed light [2,3].

In femtosecond laser pulses delivered by mode-locked lasers φ is subject to a pulse-to-pulse shift $\Delta\varphi$ due to the presence of dispersive and nonlinear components in the laser cavity [4]. A frequency-domain technique [5,6] has recently allowed control of the evolution of φ, by stabilizing $\Delta\varphi$ in the multi-MHz pulse train of femtosecond lasers [7–10]. However, the actual value of φ still remained unknown. The recent generation of coherent soft x rays by amplified phase-stabilized few-cycle pulses provided access to φ, but only with a $\pm\pi$ ambiguity [3]. More recently, this ambiguity was removed by the simultaneous detection of strong-field-induced photoelectrons in opposite directions [11]. These measurements of φ have relied on high-energy ($\gg 1$ μJ) pulses available only at low repetition rates and on interactions with a gaseous medium. Meanwhile, theoretical predictions of φ-sensitive interactions in solids [12–14] have also been reported. In this Letter we present experimental evidence for the φ sensitivity of multiphoton-induced photoemission from a metal surface. This observation opens the way to the measurement of φ by a solid-state detector for the first time. The detector is sensitive at nanojoule energy levels, allowing phase measurements at the output of a laser oscillator.

Our experiments were performed with a laser system producing phase-stabilized, 4-fs, 3-nJ pulses at a carrier wavelength near 0.75 μm [15] and a repetition rate of $f_r \approx 24$ MHz. The system was described in detail in [16]; its principal building blocks are shown in Fig. 1. Phase stabilization was carried out with a small portion (15%) of the oscillator output by using the standard f-to-$2f$ interferometric method [4,9]. As a result, φ slips in the output pulse train by $\Delta\varphi = 2\pi f_{\mathrm{ref}}/f_r$ from one pulse to the next [9,17]. We set the frequency of the external sinusoidal reference signal $f_{\mathrm{ref}} = 1$ MHz, leading to a reproduction of the wave form in every 24th laser pulse [18].

Under these conditions φ evolves at a constant, well-controlled rate

$$\varphi(t) = \varphi_0 + 2\pi f_{\mathrm{ref}} t, \tag{1}$$

(where $t = n/f_r$ for the nth pulse), but the phase offset φ_0 is still unknown. The measurement of φ_0 requires an interaction that yields a physical measurable dependent on φ, with a dependence insensitive to other pulse parameters. This will exhibit a component modulated at f_{ref}, $S(t) = S_0\cos(2\pi f_{\mathrm{ref}} t + \theta)$. The measurement of the phase shift θ of the signal S with respect to the reference signal $R(t) = R_0\cos(2\pi f_{\mathrm{ref}} t)$ (see Fig. 2) along with the knowledge of the carrier-envelope phase difference that maximizes this physical measurable, $\varphi_{\max}$, yields φ_0 via the simple relation

$$\varphi_0 = \theta + \varphi_{\max}. \tag{2}$$

In this way, knowledge of $\varphi_{\max}$ (from theory) and

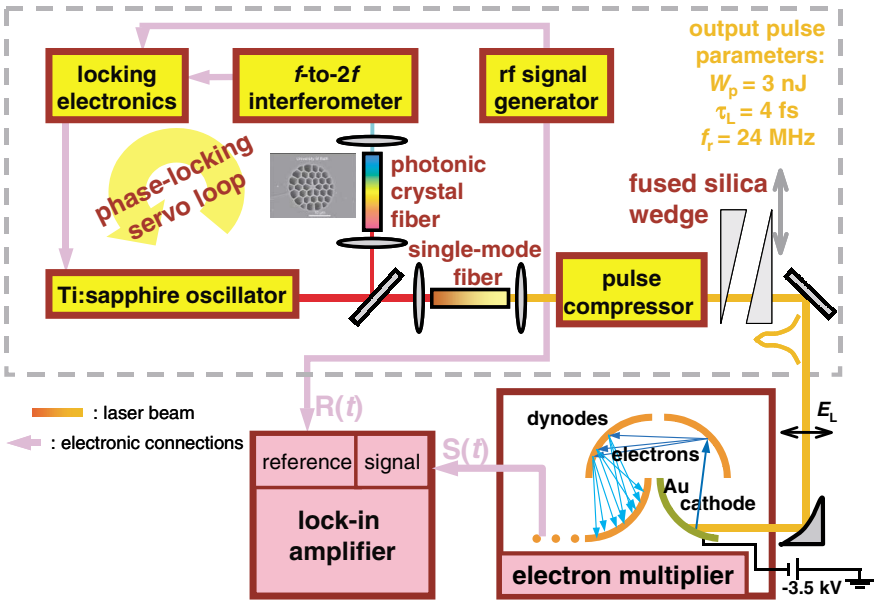

FIG. 1 (color). Schematic of the experiments. A 10-fs phase-controlled pulse train passes through a 1.5-mm-long single-mode fiber and a dispersive delay line consisting of ultrabroadband chirped mirrors to produce sub-5-fs pulses at a 24 MHz repetition rate. The carrier-envelope phase difference of the pulses can be shifted by known amounts by translation of one of a pair of thin fused-silica wedges. They are focused with an off-axis parabola onto a gold photocathode. The multiphoton-induced photocurrent is preamplified by an electron multiplier and selectively amplified by a lock-in amplifier triggered by the reference signal $R(t)$ at $f_{\mathrm{ref}} = 1$ MHz.

measurement of θ provides access to φ_0 and thus to the wave form of any of the pulses emitted by the laser (Fig. 2).

To exploit the recently predicted phase sensitivity of multiphoton-induced surface photoemission [14] for the generation of $S(t)$, we focused the laser beam onto a gold photocathode, which was integrated in a commercial electron multiplier tube (Hamamatsu R595). The laser beam impinged on the target at an angle of incidence of $\approx 70°$(to the surface normal). For peak intensities of 10^{12}–10^{13} W/cm^2 the time-averaged output current from the electron multiplier tube was found to follow a power-law scaling I_p^x, indicating that the observed electron emission is dominated by multi-photon-induced transitions [19]. The evaluated value of x between 3.0 and 3.5 is consistent with the 3-photon and 4-photon excitation channels implied by the 5-eV work function of gold and the broad laser spectrum extending over photon energies of 1.4–2.1 eV.

We searched for a modulation of the photoemission signal at f_{ref} by selectively amplifying spectral components of the photocurrent near f_{ref} with a lock-in amplifier referenced to $R(t)$. With a p-polarized laser beam carrying sub-5-fs pulses at the above intensity levels we have been able to reproducibly generate and observe this modulation. If the same pulses were delivered in an s-polarized beam the modulation disappeared completely. The modulation disappeared also when a photocurrent of similar magnitude was induced by 10-fs p-polarized laser pulses.

As nothing but φ was varying periodically at a frequency f_{ref} in the laser pulse train, the modulation $S(t) = S_0 \cos(2\pi f_{\mathrm{ref}} t + \theta)$ of the photocurrent observed with sub-5-fs pulses provides a clear indication of the phase sensitivity of the few-cycle-wave-induced nonlinear photoeffect. As a further check, we introduced a pair of thin fused silica wedges (see Fig. 1) in the laser beam and measured the variation of $S_0 \cos\theta$, which is directly displayed by our lock-in amplifier, as a function of the change ΔL in the path length through the plates. Relevant results, representative for a series of measurements, are depicted as triangles in Fig. 3. The most conspicuous feature of the data, the sinusoidal variation of $S_0 \cos\theta$ can be accounted for by θ varying linearly with the path length, $\theta = \theta_0 + \pi(\Delta L/L_{\mathrm{fit}})$. L_{fit} was evaluated as $L_{\mathrm{fit},A} = 20.3(+2.0/-1.5)$ μm and $L_{\mathrm{fit},B} = 19.3(+2.8/-1.9)$ μm from least-square fits (lines in Fig. 3) to the measured

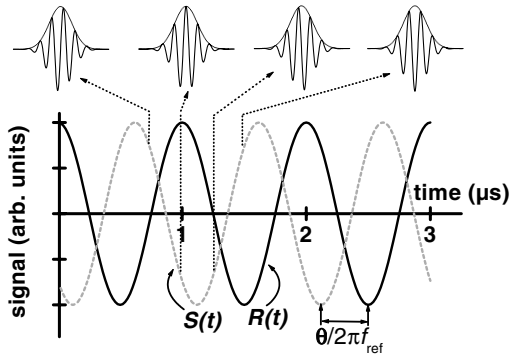

FIG. 2. The reference signal $R(t) = R_0 \cos(2\pi f_{\mathrm{ref}} t)$ along with the modulated signal component $S(t) = S_0 \cos(2\pi f_{\mathrm{ref}} t + \theta)$ of the photocurrent. The phase shift θ can be directly measured by phase-sensitive lock-in detection. With the carrier-envelope phase difference maximizing the number of extracted photoelectrons inferred from a numerical study and intuitive considerations: $\varphi_{\max} = -\pi/4$, the carrier-envelope phase difference can be determined in any pulse of the train as shown for a few representative cases assuming a somewhat idealized Gaussian pulse envelope. Knowledge of θ and $\varphi_{\max}$ creates an unambiguous connection between φ and the instantaneous phase of the reference signal, allowing the absolute value of the carrier-envelope phase difference to be referenced directly to $R(t)$.

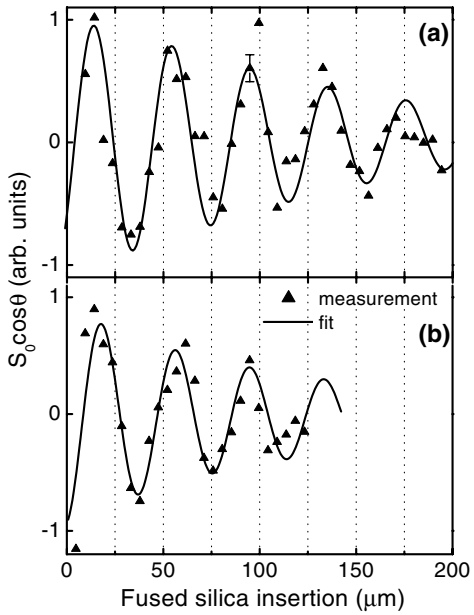

FIG. 3. In-phase component, $S_0 \cos\theta$, of the modulation of the photocurrent, $S(t)$, as a function of the change in path length through the fused silica glass wedges shown in Fig. 1. (a),(b) Photoemission signal recorded with pulses of a peak intensity of $I_p \approx 2 \times 10^{12}$ W/cm^2 and a duration (full width at half maximum) of $\tau_L = 4.5$ fs and 4.0 fs, respectively. The experimental data (triangles) are corrected for a constant (non-oscillating) phase offset of electronic origin. The lines are obtained by modeling the decrease of the photocurrent using the power law $S_0 \sim I_p^x$ with $x = 3.0$ and taking into account dispersive pulse broadening. Although the pulses broaden only by a few percent upon traveling a distance of a few tens of micrometers in fused silica, the resultant decrease in their peak intensity is sufficient to notably lower the photocurrent owing to the rapid I_p^x scaling discussed above. S_0 decays faster in (b) simply because the shorter pulse broadens more rapidly upon propagation.

data obtained in two independent measurements depicted in panels (a) and (b) of Fig. 3, respectively. The experimental conditions and modeling of the experiments are described in the caption of Fig. 3.

Propagation through any dispersive material causes a continuous shift in φ. This shift arises from the carrier wave and the amplitude envelope of the pulse traveling at different velocities, namely, with the phase and group velocity, respectively. The change in φ introduced by any added path length is imposed on all pulses of the pulse train, consequently the phase θ of any φ-induced modulation is shifted by the same amount. The propagation length giving rise to a π phase shift has been called the dephasing length L_{deph} [1]. It is determined by the wavelength dependence of the refractive index of the propagation medium and, for pulses approaching the single-cycle regime, by the spectral distribution of the pulse. For our experimental conditions, we computed $L_{\text{deph}} = 21.4$ μm. The excellent agreement of $L_{\text{fit},A}$ and $L_{\text{fit},B}$ with the calculated dephasing length provides compelling evidence for the modulation $S(t) = S_0 \cos(2\pi f_{\text{ref}} t + \theta)$ of the photocurrent being caused by the evolution of φ in the laser pulse train.

Having obtained θ from the phase-sensitive lock-in measurements, all we need for the determination of φ_0 is, according to (2), the phase maximizing the photoeffect, $\varphi_{\max}$. Multiphoton-induced electron emission was recently studied theoretically and found to exhibit a pronounced and robust φ sensitivity for pulse durations τ_L approaching T_0 [14]. The simulations reveal that the emitted charge per pulse has a maximum at $\varphi_{\max} \approx -\pi/4$ [20](sign convention: electric field of the p-polarized laser wave impinging at grazing incidence is positive if the field vector points into the material), implying a peak electric field pulling the electrons out of the irradiated surface some $T_0/8 \approx 0.3$ fs after the pulse peak.

This prediction is backed by a simple intuitive model. Our experiments have been performed in the perturbative regime of multiphoton absorption. Consequently, electrons are set free with low ($<\hbar\omega_L$) kinetic energy within a narrow ($\ll\tau_L$) time interval centered at the pulse peak. In a first approximation, the motion of the freed electrons is initially governed by the laser field, which is much stronger than the static electric field applied to extract and observe the photoelectrons. For carrier-envelope phases in the range of $-\pi/2 \leq \varphi < 0$, the electrons undergoing bound-free transition at the peak of the pulse envelope are initially pulled away from the surface and never return to it during the pulse. This range is further narrowed to a small interval around $\varphi = -\pi/4$ by the long range tail of the surface potential. For values of φ outside this "escape" range, the electrons freed at the pulse peak are pushed back into the metal either immediately at the instant of their birth or somewhat later, after performing a wiggle in the laser field. In both cases recapturing of the electrons by the metal is likely. For a few-cycle laser pulse multiphoton-induced bound-free transitions are distributed over a period shorter than T_0 at the pulse peak, preserving the sensitivity of center-of-gravity motion of the electron cloud to the carrier-envelope phase difference and resulting in maximum electron extraction efficiency near the center of the above-evaluated range of "favorable" phases, i.e., at around $\varphi_{\max} = -\pi/4$. For longer pulses, the broad temporal distribution of bound-free transitions extending over a period longer than $>T_0$ is expected to "blur" the φ sensitivity of the photoeffect.

The conclusions from this simple intuitive model are consistent with the predictions of [14] and supported by several experimental observations. These include the disappearance of the phase sensitivity of the multiphoton-induced photoeffect both for somewhat increased pulse durations and for s-polarized incident light. The latter observation is particularly enlightening; it supports the prediction of our intuitive model that not the bound-free transition of the photoelectrons but rather their subsequent laser-driven motion *normal to the surface* introduces the observed phase sensitivity. This consistent

picture is shadowed by a considerable discrepancy between the predicted and measured depth of phase-induced modulation of the photocurrent (several tens per cent versus less than one per cent). This discrepancy may have several reasons, including (i) the roughness of the surface of our evaporated gold sample, which tends to reduce the normal component of the local field on a microscopic scale, (ii) overlayer formation due to poor (10^{-5} mbar) vacuum conditions, and (iii) a jitter of the carrier-envelope phase difference [21] accumulated in the f-to-$2f$ interferometer and the pulse compressor (Fig. 1). In spite of all these contrast-reducing effects, the phase sensitivity survives to a sufficient extent in the sub-5-fs regime to allow determination of φ with an accuracy of better than $\pm\pi/4$, as can be inferred from the error bar in Fig. 3. We are confident that systematic studies and elimination of the above effects will result in substantial improvement of this accuracy in the near future.

With θ measured and $\varphi_{\max}$ deduced from an intuitive model backed by numerical simulations [14], φ_0 can now be determined from (2) and so can the carrier-envelope phase difference $\varphi_n = \varphi(t = n/f_r)$ of any pulse in the train emitted by the laser with a carrier-envelope phase difference evolution locked to a reference signal, as depicted in Fig. 2. As a result, provided that standard pulse (envelope) diagnostic tools are available, ultrashort light pulses from a femtosecond laser can be fully characterized—in terms of their electromagnetic wave form—for the first time. The resultant MHz-repetition-rate few-cycle light wave form synthesizer opens the door to controlling and probing condensed matter dynamics within the wave cycle of visible light, i.e., at a sub-fs time scale.

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