Gouy Phase Shift for Few-Cycle Laser Pulses

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We measured for the first time the influence of the Gouy effect on focused few-cycle laser pulses. The carrier-envelope phase is shown to undergo a smooth variation over a few Rayleigh distances. This result is of critical importance for any application of ultrashort laser pulses, including high-harmonic and attosecond pulse generation, as well as phase-dependent effects.

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In recent years there has been increasing interest in the generation and application of short laser pulses. Pulse durations of less than 5 fs in the near-infrared (800 nm) are in fact routine since the introduction of the hollow-fiber pulse compression technique [1–3]. Since such laser pulses consist of merely a few field oscillations (few-cycle pulses), the actual time variation of the electric-field — and therefore all physical processes driven by the laser — depends on the phase of the carrier wave with respect to the envelope, the so-called carrier-envelope (CE) phase. Because of the high sensitivity of this quantity to small fluctuations in pulse intensity, cavity length, and temperature, ultrashort-pulse lasers normally deliver pulse trains with a randomly changing CE phase shot to shot. While phase stabilization of femtosecond oscillators is a well-established technique [4–7], the capability of stabilizing and controlling the phase of an amplified laser system was only recently demonstrated [8]. This allows precise electric-field shaping and offers exciting prospects including the reproducible generation and reliable measurement of isolated subfemtosecond pulses [9,10].

With the achievement of stabilization of the CE phase for amplified pulses, the question of determining the value of the phase had to be addressed. This problem was solved very recently [11]. However, an important issue of nonlinear processes driven with phase-stabilized few-cycle pulses is that they usually take place in a laser focus. It is known that an electromagnetic beam propagating through a focus experiences an additional π phase shift with respect to a plane wave. This phase anomaly was discovered by Gouy in 1890 [12] and has since been referred to as the Gouy phase shift. This phase change is, in fact, a general property of any focused wave and is also expected for sound waves [13]. Intuitive explanations of the observed anomaly have been proposed [14,15]. The experimental evidence of the Gouy phase shift relied for years on interferometric measurements [12,16]. More recently, direct observations of the polarity change of single-cycle terahertz pulses have been reported [17–19].

This phase shift has important consequences in the optical range of the electromagnetic spectrum. In laser cavities, the determination of the resonant frequencies depends on the Gouy phase [20]. Phase matching in high-order harmonic generation (HHG) is governed by the atomic response and the Gouy phase shift of the fundamental radiation [21–23]. More importantly, the Gouy phase strongly affects the spatial variation of the CE phase of ultrashort pulses in a laser focus, the subject of this Letter.

In principle, the Gouy phase shift of a TEM_{00} wave can be described by a simple formula depending on the focusing geometry and the wavelength,

$$\phi = -\arctan\left(\frac{z}{z_R(\lambda)}\right), \tag{1}$$

where the beam is traveling in the $+z$ direction and z_R is the Rayleigh distance (dependent on the wavelength λ). As few-cycle pulses consist of broad spectra with the wavelengths spanning about one octave, the different spectral components will experience the Gouy phase shift with different spatial scales. In particular, *blue* colors undergo a steeper phase change than *red* colors. It has been shown theoretically [24,25] that this leads to a difference of phase and group velocities in the neighborhood of the focus and therefore to CE phase slippage. For this reason, in general the CE phase variation cannot be described by Eq. (1). Indeed, the details of the phase change in the focus depend on the spatial profile of the laser beam and on the focusing geometry. In all cases, an overall π phase shift is expected between symmetric positions well distant from a spherical focus. Since the phase changes by π in the propagation through the focus, virtually all possible E-field shapes are available for the experiments. Thus, precise control of the spatial variation of the CE phase in the whole focal region is crucial to any kind of phase-dependent experiment.

In this Letter we present the first experimental determination of the evolution of the CE phase in the focus of

few-cycle laser pulses. Together with the ability to determine the value of the CE phase with high accuracy [11], this result constitutes the first full and unambiguous characterization of the electric field of the laser pulses in space and time within the paraxial approximation (i.e., neglecting the very small longitudinal component of the electric field).

Because of the lack of inversion symmetry of few-cycle pulses, nonlinear photoionization of noble gases exhibits significant asymmetries in the spatial photoelectron distribution [26–29]. Thus, simultaneous photoelectron spectroscopy in the two opposite polarization directions is a possibility to retrieve the time variation of the electric field. This so-called stereo-ATI (above-threshold-ionization) scheme has proven to be the most efficient method of CE phase determination to date [11,30]. In our experiment we used a phase-stabilized laser amplifier system capable of producing few-cycle pulses [8]. The pulse energy was attenuated to 20 μJ and the beam focused with a $f/30$ geometry into a low-density xenon gas jet. Electrons emitted to the left and to the right are independently detected by two microchannel plates (Burle BiPolar TOF Detector, diameter 18 mm) located at a distance of 40 cm from the focus. With our focusing geometry, the electric field is expected to undergo the π phase shift within a range of a few millimeters. To reveal the influence of the Gouy phase, one has to detect selectively the electrons generated at a well-defined position of the focus. A pair of moveable slits perpendicular to the beam axis (z direction) and to the polarization axis allows the entire focal region to be scanned. The slit width is 250 μm, well below the Rayleigh range (≈ 1 mm), and, to achieve optimum spatial resolution in the z direction, they are placed at a distance of only 1 mm from the beam (Fig. 1). With this setup, the angular distribution of the emitted photoelectrons does not affect the phase resolution, which can be estimated to be ≈ 0.1 rad.

The photoelectron emission in the two opposite polarization directions measured with phase-stabilized pulses significantly differs in many aspects. On the one hand, the total count rate detected in the two arms of the spectrometer shows clear asymmetries; on the other hand, the difference of the two electron spectra can be used directly to retrieve the phase [11]. Both these features can be used to determine the Gouy CE phase shift. One possibility to observe the change of sign of the electric field in passing through the focus is to compare the electron yield in the two directions before and after the focus. For instance, if one detects dominant left emission before the focus, the opposite is expected after the focus. This can be verified with all possible CE phases simply by introducing a variable glass thickness in the beam path. At 760 nm, adding 52 μm of glass changes the phase by 2π without appreciably affecting the pulse duration. Such precise control is easily achieved by shifting a pair of wedges with a stepper motor (see Fig. 1). Figure 2 shows the

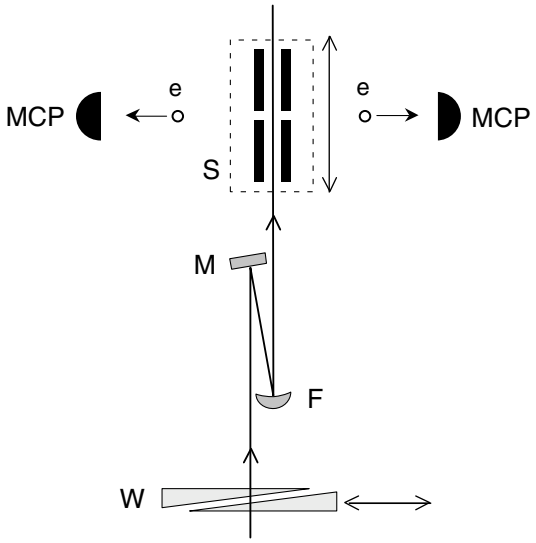

FIG. 1. Schematic of the experimental setup. W: pair of glass wedges to control the CE phase; M: flat mirror; F: focusing spherical mirror ($f = 250$ mm); MCP: microchannel plates; S: moveable slits (width = 250 μm). The angle of incidence on the focusing mirror is minimized to reduce astigmatism.

electron count rate asymmetry (left/right ratio) as a function of the glass thickness introduced. We made the measurement by moving the pair of slits to a distance of ≈ 2 mm before (dashed line) and after (solid line) the focus. The clear phase shift of π between the two curves represents a direct measurement of the Gouy CE phase shift in the focus.

The electric-field polarity reversal observed from before to after the focus does not describe the details of the phase slippage in the focus. Since many experiments take place over an extended area of the focus, e.g., HHG, it is

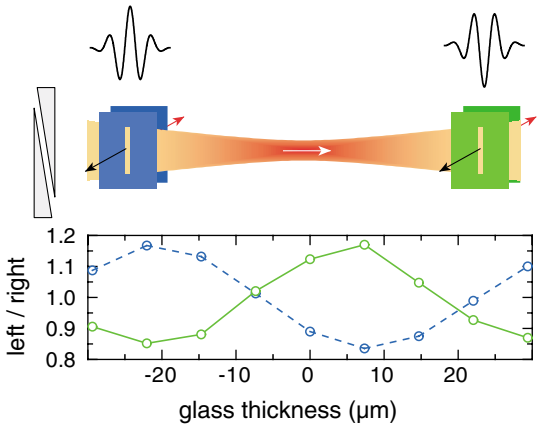

FIG. 2 (color). Varying the glass thickness in the beam path changes the CE phase of the pulses. The left/right ratio of the electron yield exhibits clear oscillations with a periodicity consistent with glass dispersion at the wavelength of the laser. The measurement was performed before (dashed line) and after (solid line) the focus. The π phase shift is due to the Gouy CE phase shift in passing through the focus.

essential to characterize the phase variation precisely and, in particular, to recognize possible anomalies in the behavior of the CE phase evolution. Indeed, the possible presence of a significant area of the focus with anomalous phase change has been suggested as an explanation of the observed enhancement of the HHG efficiency by using truncated Bessel beams [31].

In order to analyze in detail the phase variation within the focal range, we acquired electron spectra at several positions by moving the pair of slits. Figure 3 shows the detected left/right asymmetry as a function of the electron energy and of the CE phase for a few z positions. As in Fig. 2, the phase was changed by shifting the glass wedges. Different approaches are possible to retrieve the phase variation. The integrated electron yield provides a clear phase indication and can be used to show the overall phase shift in the focus (Fig. 2). However, scanning the focal range implies measuring at constantly changing

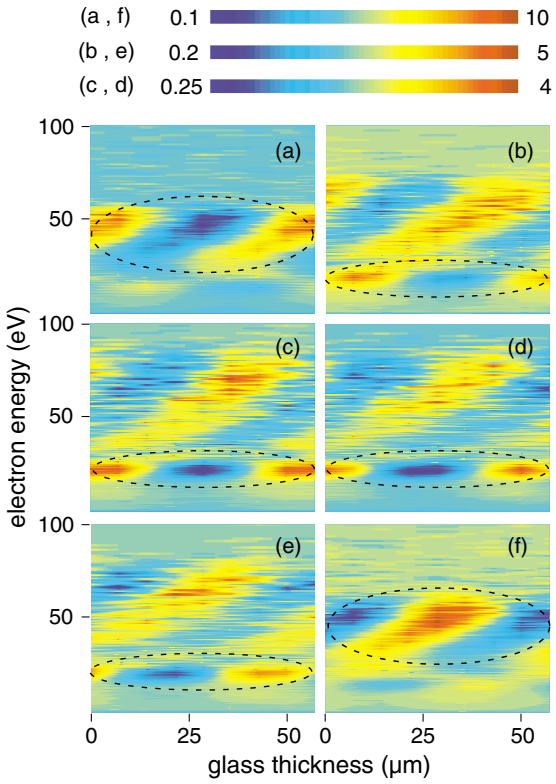

FIG. 3 (color). Left/right asymmetry maps (logarithmic scale) for different longitudinal positions as a function of the electron energy and glass thickness introduced. Reddish colors indicate dominant left emission; bluish colors dominant right emission. The maps (a)–(f) correspond, respectively, to the positions: $z = -1.75$ mm, $z = -1.0$ mm, $z = -0.25$ mm, $z = +0.25$ mm, $z = +1.0$ mm, $z = +1.75$ mm (positive values represent positions *after* the focus). The phase difference is determined by evaluating the shift of the characteristic structures (indicated by the dashed lines) of the asymmetry pattern. The extension of the electron yield to higher energies in the central part of the focus is due to the higher intensity.

intensities. Since it is predicted that the maximum of the integrated left/right asymmetry should occur at a phase that depends on the intensity [28], it is not easy to decouple the Gouy phase shift from the possible intensity-dependent phase shift. A better approach relies therefore on the evident spectral information of Fig. 3. Dark reddish and bluish colors correspond to spectral regions of strong asymmetry: Since these are easily distinguishable, they can be used to follow the phase evolution. Note that the measurements were made by approaching the focus from the outer part and by moving alternatively the slits before and after the focus. With this procedure, measurements of symmetric positions around the focus are consecutive, thus reducing detrimental effects from possible long-term phase drifts.

Figures 3(a) and 3(f) correspond to the outer part of the focal range. The strong asymmetry in the high-energy part of the spectra (ATI *plateau*, dashed area) changes sign while passing through the focus, confirming the π phase shift already discussed (see Fig. 2). More interestingly, Figs. 3(b)–3(e) correspond to positions in the central part of the focus. The asymmetry in the plateau is partly smeared out, but another clear asymmetric area appears in the low-energy part of the spectra (20–25 eV, dashed area), just before the ATI plateau. Its nature is not entirely understood, but, being definitely a CE phase effect, it can be exploited to retrieve the phase variation in the focus. By moving in the direction of the beam propagation, i.e., from before to after the focus [Figs. 3(a)–3(f)], the characteristic pattern moves toward the left side of the graphs, i.e., toward decreasing glass thickness introduced. This is in full agreement with the intuitive physical origin of the Gouy CE phase shift. Indeed, in a dispersive medium like glass the phase velocity exceeds the group velocity. The same situation occurs in the propagation of the ultrashort pulse in the laser focus [24,25]. Thus, it is not surprising that, to observe similar features, less glass is needed after the focus with respect to a symmetric position before the focus. The Gouy CE phase shift determination can be performed simply by numerically evaluating the shift of the asymmetry pattern; converting the glass thickness shift obtained (μm) into phase difference (rad) is then straightforward.

Figure 4 shows the retrieved CE phase shift for the positions of Fig. 3 and for several others not shown there. For comparison, the Gouy phase of a Gaussian beam [Eq. (1)] with a $f/30$ focusing geometry is also shown (solid line). Note that the CE phase shift is *not* expected to follow the Gouy phase, the latter being a property of cw lasers. The beam in our experiments is a few-cycle pulse truncated Bessel beam, and the focusing geometry in our setup (see Fig. 1) inevitably introduces a slight astigmatism.

The pulses undergo the π phase shift within a few Rayleigh distances. Because of the rapid decrease of

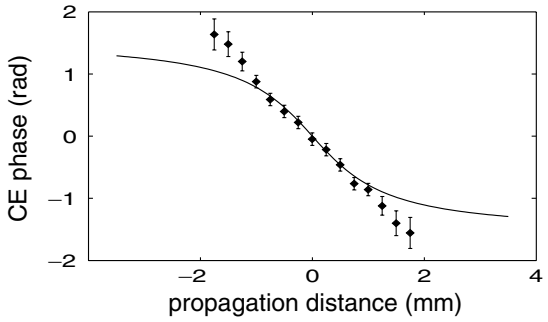

FIG. 4. Retrieved CE Gouy phase shift as a function of the propagation distance in the focus. The solid line is the Gouy phase of a cw Gaussian beam, shown for comparison. Note that, in the outer part of the focus, the electron count rate rapidly decreases, making detection of additional experimental points difficult.

electron yield at lower intensities, the measurements were stopped at a distance of ≈ 2 mm before and after the focus. This prevented observation of the expected area of the constant CE phase in the outer part of the focus. However, we point out that the region of interest for all experiments is entirely covered, and the estimated error for the experimental data is relatively low ($\lesssim 0.1$ rad). The phase changes smoothly with a constant slope and, what is particularly important for experiments, does not exhibit any wiggles or irregularities.

In conclusion, we have directly measured the CE phase shift of laser pulses evolving through a focus induced by the geometrical Gouy phase. This constitutes the first full characterization of phase-stabilized few-cycle optical pulses in space and time, an essential step for any application of such laser systems. Furthermore, these results provide access not only to the overall polarity reversal for pulses evolving through a focus [17–19], but also, for the first time to our knowledge, to the details of the phase variation *in* the focus.

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[1] M. Nisoli, S. De Silvestri, and O. Svelto, Appl. Phys. Lett. **68**, 2793 (1996).
[2] M. Nisoli, S. De Silvestri, O. Svelto, R. Szipöcs, K. Ferencz, Ch. Spielmann, S. Sartania, and F. Krausz, Opt. Lett. **22**, 522 (1997).
[3] G. Steinmeyer, D. H. Sutter, L. Gallman, N. Matuschek, and U. Keller, Science **286**, 1507 (1999).
[4] J. Reichert, R. Holzwarth, Th. Udem, and T.W. Hänsch, Opt. Commun. **172**, 59 (1999).
[5] H. R. Telle, G. Steinmeyer, A. E. Dunlop, J. Stenger, D. H. Sutter, and U. Keller, Appl. Phys. B **69**, 327 (1999).
[6] 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).
[7] A. Apolonski, A. Poppe, G. Tempea, Ch. Spielmann, Th. Udem, R. Holzwarth, T.W. Hänsch, and F. Krausz, Phys. Rev. Lett. **85**, 740 (2000).
[8] A. Baltuška, Th. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, Ch. Gohle, R. Holzwarth, V. S. Yakovlev, A. Scrinzi, T.W. Hänsch, and F. Krausz, Nature (London) **421**, 611 (2003).
[9] M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) **414**, 509 (2001).
[10] R. Kienberger, E. Goulielmakis, M. Uiberacker, A. Baltuška, V. Yakovlev, F. Bammer, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) **427**, 817 (2004).
[11] G. G. Paulus, F. Lindner, H. Walther, A. Baltuška, E. Goulielmakis, M. Lezius, and F. Krausz, Phys. Rev. Lett. **91**, 253004 (2003).
[12] L. G. Gouy, C.R. Acad. Sci. Paris **110**, 1251 (1890).
[13] N. C. R. Holme, B. C. Daly, M. T. Myaing, and T. B. Norris, Appl. Phys. Lett. **83**, 392 (2003).
[14] R.W. Boyd, J. Opt. Soc. Am. **70**, 877 (1980).
[15] S. Feng and H. G. Winful, Opt. Lett. **26**, 485 (2001).
[16] C. R. Carpenter, Am. J. Phys. **27**, 98 (1958).
[17] A. B. Ruffin, J. V. Rudd, J. F. Whitaker, S. Feng, and H. G. Winful, Phys. Rev. Lett. **83**, 3410 (1999).
[18] P. Kužel, M. A. Khazan, and J. Kroupa, J. Opt. Soc. Am. B **16**, 1795 (1999).
[19] R.W. McGowan, R. A. Cheville, and D. Grischkowsky, Appl. Phys. Lett. **76**, 670 (2000).
[20] A. E. Siegman, *Lasers* (University Science Books, Sausalito, 1986).
[21] Ph. Balcou and A. L'Huillier, Phys. Rev. A **47**, 1447 (1993).
[22] M. Lewenstein, P. Salières, and A. L'Huillier, Phys. Rev. A **52**, 4747 (1995).
[23] F. Lindner, W. Stremme, M. G. Schätzel, F. Grasbon, G. G. Paulus, H. Walther, R. Hartmann, and L. Strüder, Phys. Rev. A **68**, 013814 (2003).
[24] Z. L. Horváth, J. Vinkó, Zs. Bor, and D. von der Linde, Appl. Phys. B **63**, 481 (1996).
[25] M. A. Porras, Phys. Rev. E **65**, 026606 (2002).
[26] P. Dietrich, F. Krausz, and P. Corkum, Opt. Lett. **25**, 16 (2000).
[27] I. P. Christov, Appl. Phys. B **70**, 459 (2000).
[28] S. Chelkowski and A. Bandrauk, Phys. Rev. A **65**, 061802 (2002).
[29] D. Milošević, G. G. Paulus, and W. Becker, Opt. Express **11**, 1418 (2003).
[30] G. G. Paulus, F. Grasbon, H. Walther, P. Villoresi, M. Nisoli, S. Stagira, E. Priori, and S. De Silvestri, Nature (London) **414**, 182 (2001).
[31] M. Nisoli, E. Priori, G. Sansone, S. Stagira, G. Cerullo, S. De Silvestri, C. Altucci, R. Bruzzese, C. de Lisio, P. Villoresi, L. Poletto, M. Pascolini, and G. Tondello, Phys. Rev. Lett. **88**, 033902 (2002).