Probe of High-Order Harmonic Generation in a Hollow Waveguide Geometry using Counterpropagating Light

A. L. Lytle,[1,*] X. Zhang,[1] J. Peatross,[2] M. M. Murnane,[1] H. C. Kapteyn,[1] and O. Cohen[1]

[1]*JILA and Department of Physics University of Colorado at Boulder, Boulder, Colorado 80309 USA*

[2]*Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84602, USA*

(Received 19 September 2006; published 23 March 2007)

We use counterpropagating light to directly observe the coherent buildup of high harmonic generation in a hollow waveguide geometry. We measure, for the first time, coherence lengths for high photon energies that cannot be phase matched using conventional approaches. We also probe the transition through phase matching, the ionization level at which different harmonic orders are generated, and the change in the coherence length as the driving laser is depleted. These results directly prescribe the optimal structures or pulse trains required for implementing quasiphase matching.

DOI: 10.1103/PhysRevLett.98.123904 PACS numbers: 42.65.Ky, 52.38.−r

High-order harmonic generation (HHG) is a unique source of femtosecond-to-attosecond duration x-ray beams that can be highly spatially and temporally coherent [1]. This short wavelength light source has made possible new applications in ultrafast spectroscopy of atoms, molecules, and materials, as well as enabling new coherent imaging and attosecond dynamics measurements [2–6]. To date, most applications have used high harmonic light at relatively long wavelengths ($> \sim 10$ nm), due to the limited flux at shorter wavelengths. The main reason for the limited flux at short wavelengths is not the effective atomic nonlinear susceptibility, which scales only weakly with wavelength [7,8], but the fact that high-order harmonics are generated during the process of ionization. The large and dynamically varying index of refraction of the free electron plasma increases the phase velocity of the driving laser with respect to the generated high-order harmonics, preventing phase matched signal buildup for ionization levels greater than the critical level $\eta_c \approx 5\%$, 1%, and 0.5%, in Ar, Ne, and He, respectively [9–11]. Coherent buildup of the harmonic signal is then limited to a propagation distance over which the harmonic light advances out of phase by 180° relative to the driving laser—the coherence length (L_c). For conversion to shorter wavelengths where the driving laser intensity and gas ionization level must be high, L_c can be as short as tens of microns even for very low gas pressures of a few torr.

To compensate for this plasma-induced phase mismatch, past work demonstrated phase matching and quasiphase matching (QPM) employing waveguide geometries [9,12,13]. In the case of QPM, a periodically modulated waveguide was used to modulate the driving laser in both amplitude and phase. Even a small modulation of the driving laser ($\sim$1%) results in significant change in both the amplitude of the cutoff harmonics [14] and in the phase of all emitted harmonics. The amplitude modulation results from the positive linear dependence of the cutoff photon energy on the intensity of the driving laser [8,15,16]. Phase modulation of the high harmonics results from the fact that HHG is a nonlinear process with a finite, attosecond-duration time response that results in an "intrinsic" phase shift between the driving laser and the emitted light [15,17,18]. Thus, proper, periodic modulation of the laser intensity can suppress the cutoff harmonics in the out-of-phase coherence zones, as well as periodically shifting the phase to compensate for mismatch. Moderate enhancement of approximately an order of magnitude in signal can be obtained using a prefabricated waveguide of uniform period [12,13]. However, further optimization will require a detailed, preferably *in situ*, characterization of the coherence length under realistic conditions at ionization levels $> \eta_c$. These measurements will also need to take into account the inevitable laser loss, as well as group-velocity dispersion of the driving pulse that results in a continuous variation of the coherence length along the direction of propagation. Precise characterization, in combination with accurate simulation, will provide prescriptive information for the design of optimal waveguide structures and/or pulse trains for quasiphase matching [19].

In past work, the Peatross group [20–22] demonstrated that a weak counterpropagating pulse can be used to disrupt harmonic emission in the region where it overlaps with the forward propagating beam. A weak counterpropagating field induces a standing modulation in both amplitude and phase on the laser field with a periodicity corresponding to half the laser wavelength [23]. Both types of modulation translate into oscillations in the phase of the harmonic emission, the laser amplitude modulation having influence through the intrinsic phase. This modulation suppresses coherent buildup of the harmonic signal when the periodicity is much smaller than the coherence length of the conversion process. This past work was done in a free space focusing geometry using either a thin [20] or a "semi-infinite" [22] gas cell for the nonlinear medium. Counterpropagating pulses were used both to probe coherence lengths for harmonic buildup and to implement quasiphase matching under artificially poor phase-matching conditions, using a single pulse to suppress a single out-of-phase zone, in spectral regions that could be well phase matched.

An alternative method for probing coherence length is the measurement of Maker fringes. This type of measurement was performed in HHG [24] using a gas cell of variable length to observe clear coherent oscillations in the signal resulting from the Guoy phase shift of a converging beam in a free space geometry. This converging beam geometry is not optimally phase matched, which is why coherent oscillations can be observed. The 25th harmonic order investigated in this experiment can exhibit good phase matching when the driving beam is diverging, where no coherent oscillations are observed. More generally, the free space focusing geometry used in past experiments provides limited means for controlling the phase-matching conditions, limiting the amount of information obtainable.

In this work, we combine the use of counterpropagating beams with a hollow waveguide geometry to demonstrate a general, *in situ* measurement of phase mismatch (i.e., coherence length) in an extended geometry of direct interest for quasiphase matching. This allows us, for the first time, to probe the coherence of many harmonic orders over extended propagation distances. In particular, we probe coherent buildup for higher harmonic orders that are generated at ionization levels greater than the critical ionization, η_c, where phase matching using conventional approaches is not possible. Also, by adjusting the gas pressure in the waveguide, we observe the transition through phase matching for lower harmonic orders. This is possible using the waveguide geometry, where phase matching can be controlled by pressure tuning [9]. From our measurements of the coherence length, we determine the ionization levels at which different harmonic orders are generated, and the changes in the coherence length as the driving laser is depleted through ionization, refraction, and other losses. This wealth of *in situ* information on phase-matching conditions provides new capabilities for advancing the science and technology of high harmonic generation. For example, these results have important implications for generating bright x-ray beams at short wavelengths using quasiphase matching. We can now design optimal structures for modulated-waveguide QPM and pulse sequences for QPM using shaped counterpropagating pulses [19]. Furthermore, these results show that the hollow waveguide geometry possesses exceptional coherence and a ''quasi-one-dimensional'' plane wave geometry in analogy to conventional fiber optics in the visible. The exceptional phase-matching conditions also confirm that this phase matched waveguide geometry is ideal for observing and manipulating ''single-atom'' intrinsic phase effects associated with rescattering, as we showed in earlier work using shaped pulses to optimize high-order harmonic generation [3,25,26].

The experimental setup is shown in Fig. 1. The output from a 1 kHz Ti:sapphire laser amplifier system (KMLabs Dragon™) is split into two beams: one has a duration of 25 fs while the other is stretched to approximately 2.5 ps

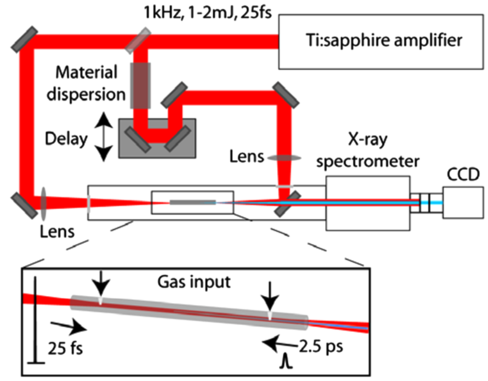

FIG. 1 (color online). Setup for *in situ* probing of the phase mismatch in HHG for harmonics generated at arbitrary ionization levels.

by propagation through 10 cm of SF18. The two beams are coupled into opposite ends of a 3.5 cm long, fused silica, hollow-core waveguide, with an inner diameter of 150 μm. In order to ensure a uniform pressure over most of the waveguide, the argon gas is introduced via two holes drilled into the waveguide 5 mm from either end, with the end sections providing a differential pumping region with an associated pressure ramp. The shorter pulse, with an energy of 300–500 μJ, drives the high harmonic generation process. The longer pulse is coupled into the waveguide by reflecting from a mirror with a 3 mm diameter hole drilled into the center to allow transmission of the harmonic light. The longer pulse has an energy of 150–250 μJ when coupled into the fiber, and is propagated via a delay line in order to vary the collision point of the two pulses in the waveguide. Note that the intensity of the (long) counterpropagating beam is too weak to independently ionize the atoms or generate high harmonics. The high harmonics are spectrally resolved using an imaging x-ray spectrometer (Hettrick Scientific), and detected with an x-ray sensitive CCD camera (Andor Technology). The fundamental light is rejected by two aluminum foil filters, each of 200 nm thickness. Modulations are observed by changing the delay of the counterpropagating pulse relative to the harmonic generating pulse, so that the position of collision of the two pulses moves along the propagation direction of the fiber. The collision point was moved from outside the fiber, through the 5 mm, differentially pumped rear section of the fiber, and into the longer, constant pressure region, where z is the position in mm relative to the exit of the fiber.

Figure 2 shows the intensity of the 27th harmonic as a function of the collision position of the pulses within the waveguide, at several different pressures. This relatively low harmonic order was initially selected because it is generated at relatively low ionization levels and thus can be phase matched by pressure tuning in waveguides. At both low and high pressures, the phase mismatch is large, corresponding to short coherence lengths—approximately matching the length of the counterpropagating pulse

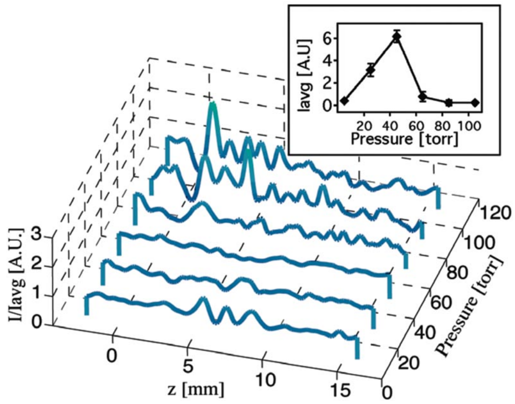

FIG. 2 (color). Observed normalized modulation intensity (I/I_{avg}) of the 27th harmonic as a function of gas pressure and position of the pulse collision point in the waveguide, with respect to the exit of the fiber. The coherence length increases as expected near the phase-matching pressure of 45 torr, leading to a disappearance of the modulations. The position in the waveguide at which the harmonic is generated also changes with pressure. (Inset) Average (unnormalized) harmonic output (I_{avg}) as a function of pressure, demonstrating phase matching at 45 torr.

(750 mm). As a result, several distinct modulations are observed. The coherence length associated with these modulations is equal to half the periodicity. At intermediate pressures around 40 torr, the phase mismatch decreases, resulting in a coherence length that is many times larger than the length of the counterpropagating pulse. Under this condition, the harmonic signal suppressed by the counterpropagating light is negligible compared the total output, and the modulations disappear. The presence of longer coherence lengths is further indicated by the observed large increase in the HHG flux (Fig. 2 inset).

The data shown in Fig. 2 also indicate the effect of the transparency of the gas medium on the observed output signal. The generated harmonic signal that contributes to the total output is limited by the absorption length of the gas. At low pressure, the modulations are most visible in the constant pressure section of the waveguide since the absorption length is long. For high pressure (short absorption length), the modulation in the harmonics is most pronounced in the end section of the waveguide used for differential pumping, where the pressure is lower and the absorption is less.

A measurement of the coherence length as a function of harmonic order provides information which allows us to extract the ionization levels at which different harmonic orders are generated. The coherence length is given by half the periodicity of the modulations observed in the output signal, such as those visible in Fig. 2 for high pressures. The phase mismatch between the driving laser and generated q^{th} harmonic order is given by

$$\Delta k = q\left\{\left(\frac{u_{11}^2\lambda_0}{4\pi a^2}\right) - P\left[(1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta(N_{\mathrm{atm}} r_e \lambda_0)\right]\right\}, \tag{1}$$

where P is the pressure, η is the ionization level, λ_0 is the laser wavelength, r_e is the classical electron radius, a is the waveguide diameter, u_{11} is the lowest-order waveguide mode factor, N_{atm} is the number density of atoms at atmospheric pressure, and $\Delta\delta$ is the differential gas dispersion. Setting the waveguide term to zero, Eq. (1) can be solved for $\Delta k = 0$ to derive the critical level $\eta_c \approx 5\%$, 1%, and 0.5% in Ar, Ne, and He, respectively, above which phase matching is not possible. In the presence of a waveguide, phase matching occurs at a particular pressure for a given level of ionization $\eta < \eta_c$. For finite values of Δk (either for nonoptimal pressures or $\eta > \eta_c$), the coherence length (L_c) is given by $L_c = \pi/\Delta k$, and depends on the harmonic order as $1/q$ for fixed values of P and η. Figure 3(a) plots the measured coherence length at a fixed position in the fiber (near the exit of the constant pressure section) as a function of harmonic order at a pressure of 5 torr. Also shown are the calculated values of the coherence length as a function of harmonic order for three different ionization levels between 18% and 26%. From Fig. 3 it is apparent that higher harmonic orders are generated at increasingly higher ionization levels, because the coherence length falls faster than $1/q$. Knowledge of the ionization level at which a particular harmonic is generated enables us to determine at what time in the laser pulse a particular harmonic order

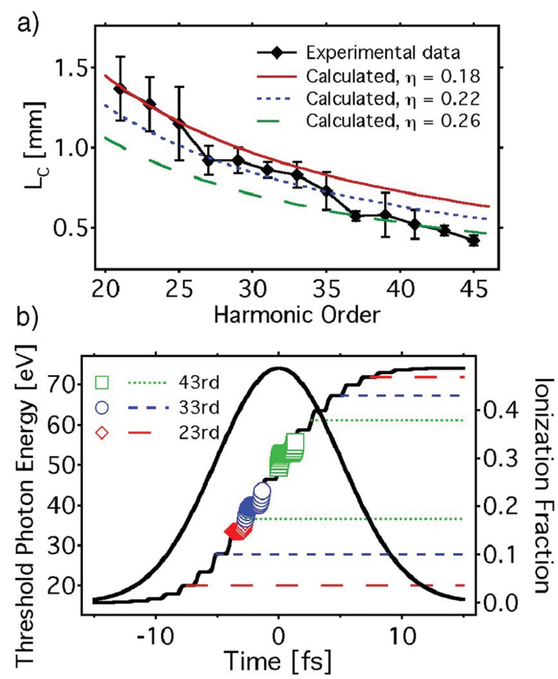

FIG. 3 (color). (a) Coherence length (L_c) vs harmonic order, measured at the exit of the center section of the waveguide at a gas pressure of 5 torr. L_c decreases faster than $1/q$ due to increasing ionization levels for high harmonic orders. The calculated values of L_c for fixed ionization levels are also plotted. (b) Position in the laser pulse at which various harmonics are generated based on the measurements shown in (a) and using ADK. The dotted lines show the energetically allowed generation range for the same harmonics, based on the cutoff rule.

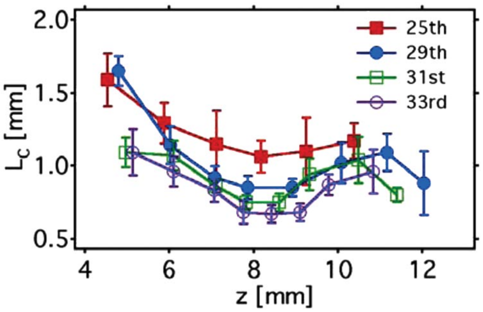

FIG. 4 (color). Measured coherence length as a function of position in the waveguide.

is generated most efficiently. Figure 3(b) shows the time during the pulse when three different harmonics are generated, using Ammosov-Delone-Krainov (ADK) [27] tunneling ionization rates combined with the measured coherence lengths from Fig. 3(a). It is worth noting that our ability to measure the coherence length depends on the duration of the counterpropagating pulse in comparison to the coherence length—ideally the pulse duration should correspond to a large fraction (50%–100%) of L_c to suppress emission from most of a single constructive or descructive zone. Longer pulses will suppress emission from adjacent zones, while shorter pulses will have a smaller effect on the signal output.

Figure 4 shows the coherence length measured as a function of position in the waveguide. For the lower harmonic orders, these data show a general decrease in L_c as the counterpropagating pulse moves further into the waveguide. This can be explained by the increasing loss of the driving laser energy towards the end of the waveguide. This results in a lower ionization fraction, reducing the phase mismatch and causing L_c to increase. The oscillations apparent in Fig. 4 are likely due to mode beating (present for the beams in both directions) between the EH_{11} and EH_{12} modes [28], which are expected in the relatively short waveguides of 3 cm that are used for these experiments.

In conclusion, we combine the use of counterpropagating beams with a hollow waveguide geometry to demonstrate a general approach that in principle allows *in situ* measurement of the phase mismatch (i.e., coherence length) in HHG for any harmonic order, over long distances, at high ionization levels, and for arbitrary absorption. These measurements determine the ionization levels at which different harmonic orders are generated, the variation in coherence length as a function of propagation distance due to laser loss and modebeating, as well as the evolution of the coherence length as the HHG process is phase matched by pressure tuning. The long distances over which coherence is measured provides a dramatic demonstration of the high level of coherence of EUV beams generated in waveguides. In the future, by using the counterpropagating light to suppress emission from out-of-phase coherence zones, the EUV conversion efficiency can be significantly enhanced using quasiphase matching. The *in situ* coherence information directly prescribes how to design optimal structures and pulse sequences for such quasiphase matching schemes. Moreover, these measurements also provide an experimental description of the evolution of the laser driving pulse, both temporally and spatially, within plasma-filled waveguides. Finally, the combination of this probing method with phase retrieval or holographic techniques should enable complete reconstruction of the generated HHG field.

We gratefully acknowledge support for this work from the Department of Energy NNSA and the National Science Foundation. A. L. acknowledges support from a National Science Foundation Graduate Research program. This research made use of NSF Engineering Research Centers Shared Facilities supported by Grant no. EEC-0310717.

*Email address: lytle@colorado.edu

[1] H. C. Kapteyn, M. M. Murnane, and I. P. Christov, Phys. Today **58**, No. 03, 39 (2005).
[2] R. A. Bartels *et al.*, Science **297**, 376 (2002).
[3] R. Bartels *et al.*, Nature (London) **406**, 164 (2000).
[4] P. M. Paul *et al.*, Science **292**, 1689 (2001).
[5] M. Hentschel *et al.*, Nature (London) **414**, 509 (2001).
[6] L. Miaja-Avila *et al.*, Phys. Rev. Lett. **97**, 113604 (2006).
[7] J. J. Macklin, J. D. Kmetec, and C. L. Gordon, III, Phys. Rev. Lett. **70**, 766 (1993).
[8] K. C. Kulander, K. J. Schafer, and J. L. Krause, in *Super-Intense Laser-Atomphysics*, edited by B. Piraux, A. L'Huillier, and K. Rzazewski (Plenum, New York, 1993), Vol. 316, p. 95.
[9] A. Rundquist *et al.*, Science **280**, 1412 (1998).
[10] E. Constant *et al.*, Phys. Rev. Lett. **82**, 1668 (1999).
[11] C. G. Durfee *et al.*, Phys. Rev. Lett. **83**, 2187 (1999).
[12] E. A. Gibson *et al.*, Science **302**, 95 (2003).
[13] A. Paul *et al.*, Nature (London) **421**, 51 (2003).
[14] I. Christov, H. Kapteyn, and M. Murnane, Opt. Express **7**, 362 (2000).
[15] M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994).
[16] Z. H. Chang *et al.*, Phys. Rev. Lett. **79**, 2967 (1997).
[17] M. Lewenstein, P. Salieres, and A. L'Huillier, Phys. Rev. A **52**, 4747 (1995).
[18] Z. Chang *et al.*, Phys. Rev. A **58**, R30 (1998).
[19] X. Zhang *et al.*, Nature Phys. (to be published).
[20] S. L. Voronov *et al.*, Phys. Rev. Lett. **87**, 133902 (2001).
[21] J. Peatross, M. V. Federov, and K. C. Kulander, J. Opt. Soc. Am. B **12**, 863 (1995).
[22] J. Peatross *et al.*, J. Mod. Opt. **51**, 2675 (2004).
[23] J. Peatross, S. Voronov, and I. Prokopovich, Opt. Express **1**, 114 (1997).
[24] S. Kazamias *et al.*, Phys. Rev. Lett. **90**, 193901 (2003).
[25] R. A. Bartels *et al.*, Phys. Rev. A **70**, 043404 (2004).
[26] I. P. Christov *et al.*, Phys. Rev. Lett. **86**, 5458 (2001).
[27] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)].
[28] E. A. J. Marcatili and R. A. Schmeltzer, Bell Syst. Tech. J. **43**, 1783 (1964).