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Published in:
Physical Review Letters

DOI:
10.1103/PhysRevLett.77.1731

1996

Spatial Mode Control of High-Order Harmonics

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(Received 22 February 1996)

We demonstrate that the spatial mode of high-order harmonics can be continuously controlled. The control is achieved by spatially modulating the degree of elliptical polarization of the fundamental field using birefringent optics. A highly sensitive relationship between the efficiency of harmonic generation and the degree of laser elliptical polarization leads to atoms emitting harmonics mainly in regions of linear polarization. The harmonics are emitted as annular beams whose angles of divergence can be continuously varied. [S0031-9007(96)01027-7]

PACS numbers: 32.80.Rm, 42.65.Ky

Once a first understanding of a physical phenomenon is acquired, a natural and exciting development consists of designing ways to *control* it. This requires good understanding of the physical processes involved, and often sophisticated experimental designs. An area where this type of research is actively pursued is in physical chemistry, for the control of molecular fragmentation processes and chemical reactions. By using specially designed light fields, some dissociation paths can be selectively chosen and others eliminated [1]. The control of ionization, and of the angular distribution of electrons or fragments, by varying the phase shift between two light fields has been shown in several experiments [2].

The generation of high-order harmonics in strong laser fields has been studied for several years [3], and considerable progress has recently been made in the understanding of the physics involved. This progress is, to a large extent, due to the development of the quasiclassical interpretation [4]. In this approach, an electron initially in the ground state of an atom and exposed to an intense, low-frequency, linearly polarized electromagnetic field first tunnels through the barrier formed by the Coulomb potential and the laser field. When the field changes sign, the electron may be driven back towards the atomic potential and recombine to the ground state, giving rise to emission of high-energy harmonic photons.

This simple description of harmonic generation processes has led to a new experimental challenge, namely, how to control the harmonic emission in space or time. The single-atom emission can be controlled, if one can modify the relevant electron trajectories leading to the production of harmonics. A simple example of control of the single-atom response is to vary the degree of elliptical polarization of the laser field. According to the semiclassical picture, a small amount of laser ellipticity can modify the electron trajectory enough to make the electron miss the nucleus when it returns, thus preventing recombination and harmonic generation. Indeed, the harmonic conversion efficiency depends strongly on the laser ellipticity, as shown in several experiments [5–7] as well as in theoretical calculations [6,8,9]. An application of this effect, proposed by Corkum and coworkers [10], is to modulate in time the laser ellipticity, so as to restrict the harmonic emission to only one laser period at the maximum of the laser pulse, thus giving rise to an extremely short extreme ultraviolet pulse. The second possible form of control of harmonic generation concerns phase matching (propagation) in the nonlinear medium. The control of the phase variation of the nonlinear polarization in the medium could certainly improve the conversion efficiency and the spatial and temporal coherence properties of the emitted field [11,12].

In the present Letter, we demonstrate that by modulating the ellipticity of the laser in *space* we can control the spatial mode of the harmonics and obtain beam profiles ranging from Gaussian to annular, or even to several rings. This is achieved by using birefringent focusing optics, with axes at 45° to the laser polarization, and introducing a *variable phase shift* between the two components of the laser field with a Babinet compensator (effectively a birefringent material of variable thickness). The nature of the control is twofold: (1) From a microscopic point of view, we impose, for each atom at a given point in space, a particular laser ellipticity, and, therefore, a particular intensity and polarization for the harmonic light. (2) More interesting, from a macroscopic point of view, we control the volume where harmonics are generated, which, under certain conditions, allows us to modify and choose at will the spatial mode.

The experimental setup is shown in Fig. 1. The laser used was the terawatt laser of the Lund High-Power Laser Facility [13]. This is a titanium-sapphire laser which, for this experiment, was operated at a wavelength of 794 nm and a pulse duration of 150 fs. An energy of 8 mJ was sent into the vacuum chamber where the harmonics were produced. Special care was taken to optimize the spatial characteristics of the laser by spatial filtering.

The polarization of the laser beam was oriented at 45° to the axes of a Babinet compensator (BC), which consists of two (birefringent) quartz wedges, with the optical axes aligned parallel to each other, but perpendicular to the direction of the light propagation. By moving one of the wedges, the total thickness of the BC can be continuously

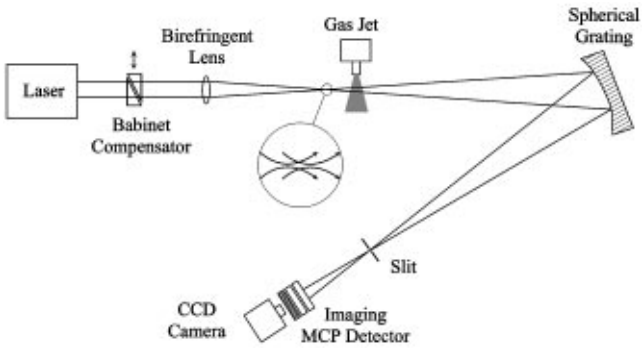

FIG. 1. Experimental setup.

varied, imparting a controllable phase delay between the polarization components aligned along the crystal axes.

The birefringent quartz lens was designed with an average focal length of 40 cm. The difference between the ordinary and extraordinary refractive indices for the quartz is about 0.01 at 800 nm, leading to the formation of two focii with orthogonal polarization components, separated by 7 mm in the direction of propagation. The group delay times associated with the orthogonal polarization components are closely matched by designing the thickness of the BC to equal that of the birefringent lens, and aligning the optical axes of the two elements to be perpendicular. (In the following, we consider only the net phase difference introduced by the optics.) Also, on transmission through the combined optics, the B integral, i.e., the accumulated nonlinear phase, is maintained below 0.7.

The effect of the optics can be summarized as follows:

$$(E_0, E_0) \overset{\text{BC}}{\rightarrow} (E_0, E_0 e^{-i\varphi_{\text{BC}}}) \overset{\text{lens}}{\rightarrow} (E_1 e^{-i\varphi_1}, E_2 e^{-i(\varphi_2+\varphi_{\text{BC}})}), \tag{1}$$

where φ_{BC} is the net phase difference introduced by the BC, $E_1 = E_1(r, z)$ and $E_2 = E_2(r, z)$, the amplitudes of the two light components which differ from each other owing to the different focus positions. (r, z) denote the transverse and longitudinal coordinates. $\varphi_1 = \varphi_1(r, z)$, $\varphi_2 = \varphi_2(r, z)$ are the phases induced by focusing. Their expression for a Gaussian beam is $(i = 1, 2)$

$$\varphi_i(r, z) = -\tan^{-1}\left(\frac{2(z - z_i)}{b_i}\right) + \frac{2kr^2(z - z_i)}{b_i^2 + 4(z - z_i)^2}, \tag{2}$$

where b_i denotes the confocal parameter, z_i the focus for the ith component, and k the laser wave vector. The birefringent lens introduces a spatial modulation of the ellipticity that is derived mostly from the difference in the radial quadratic phase delay imparted along the two crystal axes.

Let us examine, for example, the polarization properties of the beam at $z_0 = (z_1 + z_2)/2$, i.e., between the two focii. Since $b_1 \approx b_2$, the amplitudes $E_1(r, z_0)$, $E_2(r, z_0)$ are equal, the ellipse is oriented at 45° to the lens axes, and the degree of ellipticity ϵ, defined as the ratio between the components of the field along the axes of the ellipse, is given by $\epsilon(r, z_0) = |\tan(\chi)|$, $|\chi| \leq \pi/4$, with $\sin(2\chi) = \sin(\varphi_1 - \varphi_2 - \varphi_{\text{BC}})$. A numerical evaluation of this formula shows that the ellipticity oscillates between 0 (linear polarization) and 1 (circular polarization). The ellipticity at the center depends on the phase offset introduced by the BC. In the transverse plane between the two focii, the ellipticity pattern consists of rings with a perfect contrast, i.e., the ellipticity varies from circular to linear. The rings become closer and closer as the distance from the axis increases.

These conclusions hold for the more general case ($z \neq z_0$), except that the contrast between the successive rings is not as good as when the two components have the same amplitudes. The ellipticity oscillates from 0 (linear polarization) to values typically between 0.5 and 0.9, depending on the transverse coordinate. These values are, however, significant when considering that high-order harmonic generation occurs efficiently only when the ellipticity is approximately between 0 and 0.2. The position of the rings where atoms strongly driven by the laser field generate harmonics can therefore be continuously controlled by moving the Babinet compensator. Can this control of the single-atom harmonic emission in space lead to control of the spatial mode of the macroscopic harmonic field? This depends on how much propagation effects will affect the harmonic profiles and requires, in particular, the following: (1) The ellipticity pattern in the transverse plane does not vary significantly over the length of the medium, (2) the spatial profile of the harmonics closely follows the distribution of the emitting dipoles in the medium, in other words, phase matching does not destroy the expected ring pattern, and (3) the control can be observed in the far field, requiring reasonable spatial coherence.

The harmonics were produced in a 1 mm long pulsed jet of argon atoms. We observed an influence of the setting of the BC that was independent of the position of the jet on the propagation axis. However, the control was most evident and reproducible when the jet was placed a few millimeters after the two focii. This position has been shown to be the one that minimizes the phase variation of the nonlinear polarization along the propagation axis, leading to efficient harmonic generation and good spatial coherence [12]. It ensures that the three conditions mentioned above are fulfilled, in particular, by locating the gas jet several confocal parameters away from the focii, i.e., already in the far field of the laser. The emitted harmonics were separated by a normal-incidence spherical grating (see Fig. 1). No entrance slit was used. A 1 mm output slit placed in the image focal plane of the monochromator allowed us to isolate a given harmonic. A system of CsI-coated dual microchannel plates coupled to a phosphorus screen (Galileo VUV-25) was installed at about 20 cm from the output slit. The phosphorus screen, relayed by optical fibers, was read by a 16-bit charge coupled device camera.

Figure 2 shows a series of images of the angular distribution of the 13th harmonic for an increasing phase shift between the two orthogonal polarization components of the laser field, set by the BC. Consistent results were also

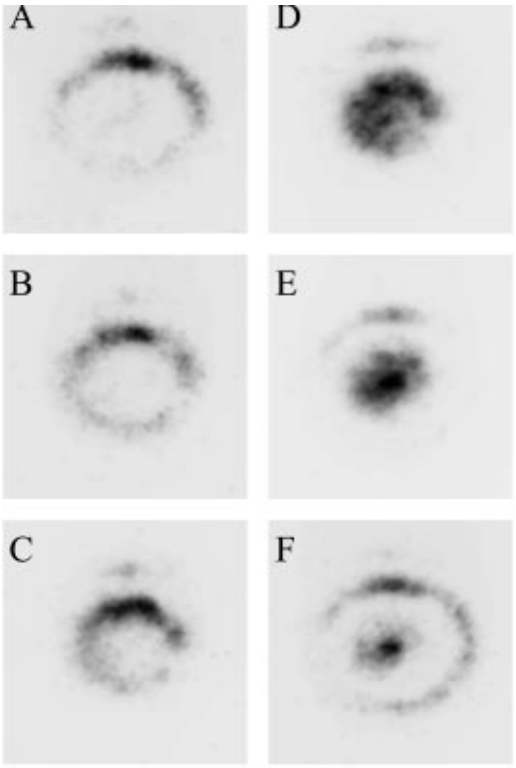

FIG. 2. Intensity profile of the 13th harmonic for several settings of the Babinet compensator.

obtained for other harmonics, ranging from the 7th to the 17th. The gas jet was located 8 mm after the second focus, and each image was acquired over typically 150 laser shots. The intensity profile in Fig. 2(A) consists mainly of a large ring. The diameter of this ring decreases as the Babinet setting is changed [Figs. 2(B) and 2(C)]. A second weak ring begins to appear. In Fig. 2(D), the angular profile is almost Gaussian-like, quite broad, with a weak external ring. In Figs. 2(E) and 2(F), the central profile becomes narrower, and the external ring stronger as well as narrower. Moving the Babinet compensator further, the profile becomes again like the one shown in Fig. 2(A).

In Fig. 3(a), we present profiles of the average radial intensity corresponding to the two images shown in Figs. 2(C) and 2(F). [The intensity $I(r, \theta)$ is averaged over the angle θ.] The difference between the two settings corresponds to a phase variation of $\approx \pi/2$. The dotted line shows the profile obtained without control, i.e., with a linear-polarized laser field (obtained by using a half-wave plate to orient the laser polarization parallel to one of the axes of the birefringent optics). This rather uniform profile appears as an envelope of the two "controlled" profiles.

A numerical simulation of this experiment is shown in Fig. 3(b). The theoretical method has been described in detail in [8] and applied to elliptically polarized laser fields. The single-atom dipole moments are calculated by solving the Schrödinger equation using a simple model valid in the tunneling regime, and the propagation equations are integrated within the slowly varying envelope approximation. Here, we apply this

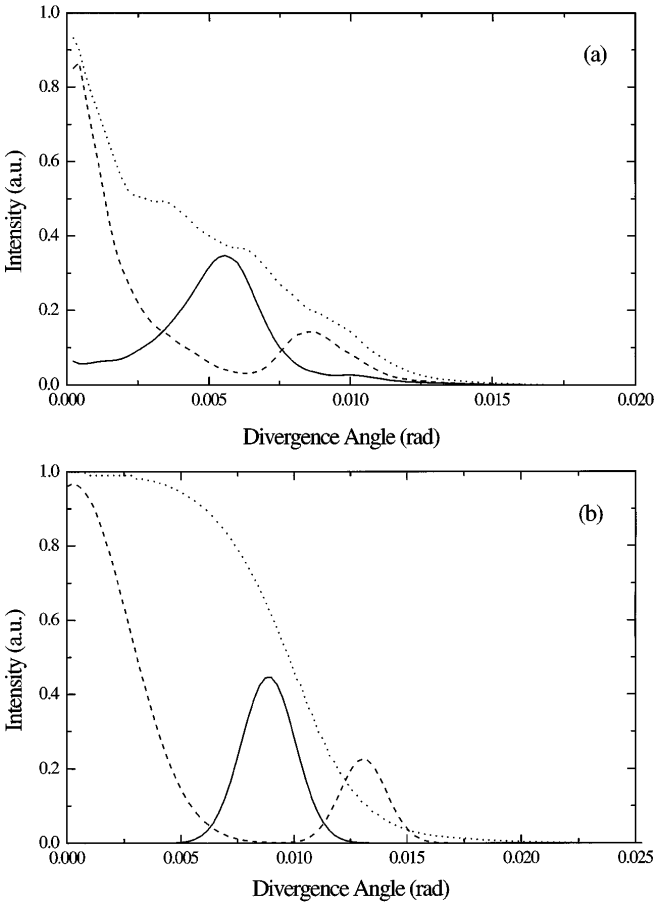

FIG. 3. Experimental (a) and theoretical (b) radial intensity profiles for the 13th harmonic for two settings of the Babinet compensator, in dashed and solid lines, corresponding to a phase difference of $\simeq \pi/2$. The dotted line shows the profile obtained without control, i.e., with a linear-polarized laser field.

approach to a laser field with a spatially varying degree of ellipticity as well as angle of rotation, inducing a strongly spatially varying nonlinear polarization. The results shown in Fig. 3(b) are in good agreement with the experimental data. The angular distributions are found to be extremely robust against peak intensity variations, more than in the linearly polarized case. The control of the spatial mode is not obtained to the detriment of the total conversion efficiency, which remains approximately the same.

Finally, we summarize in Fig. 4 the ensemble of data obtained by moving continuously one of the BC wedges over a distance of 5 mm (which corresponds to a relative phase variation of 11.1 rad). In Fig. 4(a), the laser polarization is at 45° to the axes of the birefringent optics, resulting in a radial intensity profile of the emitted harmonic that is repeated with a phase increase between the two orthogonal polarization components of π rad. In Fig. 4(b), the incident laser polarization is parallel to a crystal axis of the birefringent optics, leading to only one focus of a single polarization and therefore no radially varying ellipticity in the medium.

A temporal domain analogy of the spatial control investigated in this work is realized by producing a relative

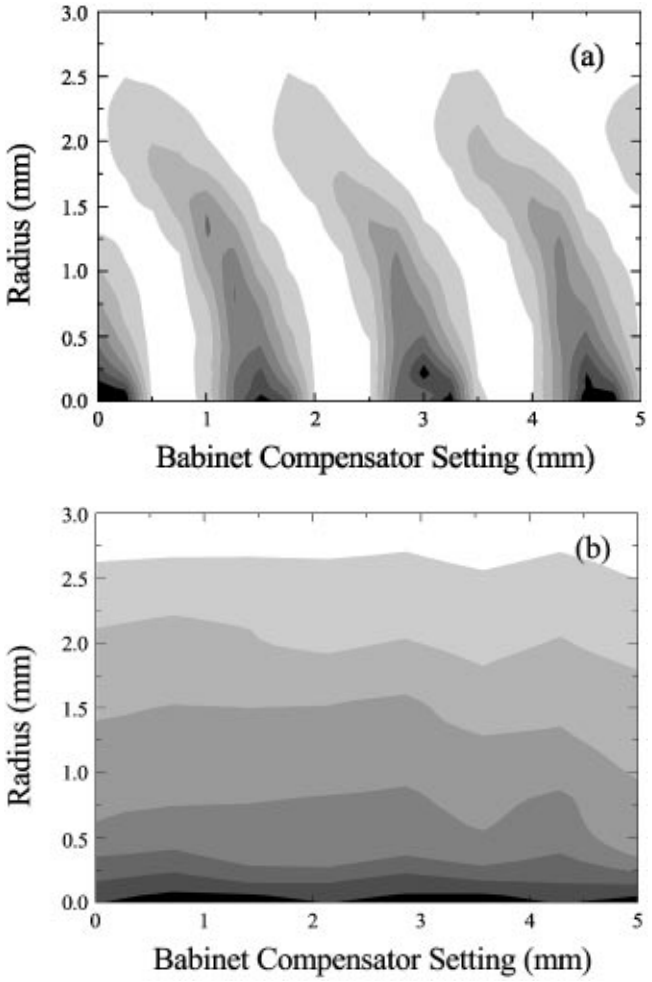

FIG. 4. Contour plot showing isointensity lines for the 13th harmonic as measured at the detector when the laser polarization is at 45° (a) and parallel (b) to one of the axes of the birefringent optics. The horizontal axis is the BC setting, the vertical axis is the transverse coordinate. The grey scale is proportional to the harmonic intensity.

phase delay between two orthogonal polarizations that varies in time instead of space. This can be achieved with a similar method to that already described, but using a flat birefringent crystal and an input pulse that is chirped in frequency. A linear chirp can be imparted to a laser pulse by normal material dispersion and is represented by a quadratic phase variation in time. The quadratic phase variation can be increased by the action of self-phase modulation, an effect normally taken to be deleterious. The pulse is split into two orthogonal polarizations, and a relative time delay is imparted with the use of the flat birefringent crystal. When the phase difference between the two polarizations is zero, linear polarization is produced. This enables a single ultrashort time window to be selected for harmonic generation at all radii. A high-quality spatial distribution is not required, as the spatial profiles associated with the orthogonal polarizations are identical and precisely overlaid. This technique could provide an alternative to the two-pulse method proposed by Corkum and coworkers [10] for the production of attosecond pulses.

In conclusion, our results emphasize the good understanding of the basic processes behind harmonic generation, allowing us to create controllable spatial variations in the nonlinear polarization and therefore providing a way to control the mode structure of the harmonics. In the present work, we use a birefringent lens inducing a strong radial distribution of the laser ellipticity and leading to annular beams with controllable radii. A wide variety of distributions could be formed by modulating the ellipticity of the laser fields with alternative birefringent optics. It should also be possible to use similar techniques with longer interaction lengths in order to optimize and control phase matching.

We acknowledge the support of the Swedish Natural Science Research Council and the EC Human Capital and Mobility Programme. Computer time for the numerical simulations was provided by the National Supercomputer Centre in Sweden, University of Linköping.

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[1] M. Shapiro, J. W. Hepburn, and P. Brumer, Chem. Phys. Lett. **149**, 451 (1988).

[2] C. E. Chen, Y. Y. Yin, and D. S. Elliott, Phys. Rev. Lett. **64**, 507 (1990); D. W. Schumacher, F. Weihe, H. G. Muller, and P. H. Bucksbaum, Phys. Rev. Lett. **73**, 1344 (1994); B. Sheehy, B. Walker, and L. F. DiMauro, Phys. Rev. Lett. **74**, 4799 (1995).

[3] See, for example, articles in the special issue on Short Wavelength Generation and High Intensity, J. Opt. Soc. Am. B January (1996), and references therein.

[4] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993); K. C. Kulander, K. J. Schafer, and J. L. Krause, in *Super-Intense Laser-Atom Physics*, edited by B. Piraux, A. L'Huillier, and K. Rzążewski, NATO ASI Ser. B, Vol. 316 (Plenum Press, New York, 1993), p. 95.

[5] K. S. Budil, P. Salières, A. L'Huillier, T. Ditmire, and M. D. Perry, Phys. Rev. A **48**, R3437 (1993).

[6] P. Dietrich, N. H. Burnett, M. Yu. Ivanov, and P. B. Corkum, Phys. Rev. A **50**, R3585 (1994).

[7] Y. Liang, M. V. Ammosov, and S. L. Chin, J. Phys. B **27**, 1296 (1994).

[8] P. Antoine, A. L'Huillier, M. Lewenstein, P. Salières, and B. Carré, Phys. Rev. A **53**, 1725 (1996).

[9] W. Becker, S. Long, and J. K. McIver, Phys. Rev. A **50**, 1540 (1994).

[10] M. Yu. Ivanov, P. B. Corkum, T. Zuo, and A. Bandrauk, Phys. Rev. Lett. **74**, 2933 (1995).

[11] J. Peatross, M. V. Fedorov, and K. C. Kulander, J. Opt. Soc. Am. B **12**, 863 (1995); J. E. Muffet, C.-G. Wahlström, and M. H. R. Hutchinson, J. Phys. B **27**, 5693 (1994).

[12] P. Salières, A. L'Huillier, and M. Lewenstein, Phys. Rev. Lett. **74**, 3376 (1995).

[13] S. Svanberg, J. Larsson, A. Persson, and C.-G. Wahlström, Phys. Scr. **49**, 187 (1994).