RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 51, NUMBER 5 MAY 1995

Polarization of high-intensity high-harmonic generation

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(Received 30 November 1994)

We have discovered that the polarization of high harmonics generated by intense, short, elliptically polarized laser pulses is rotated with respect to the incident laser polarization. This strong-field effect is a test of models of intense laser-atom processes.

PACS number(s): 42.65.Ky, 32.80.Rm

I. INTRODUCTION

When intense, coherent light is focused into a gas, harmonics are generated at odd multiples of the incident frequency (see, e.g., the review [1] and references therein). For intensities above $\sim 10^{13}$ W/cm^2, the new phenomenon of very-high-harmonic generation (HHG) occurs. The intensities of generated harmonics no longer fall off rapidly with increasing order, and appear to form a plateau with a rather sharp cutoff deep in the vacuum ultraviolet.

Calculations based on perturbation theory break down in this intensity range, so that new physical models are required to describe the HHG phenomenon. Several different models have been proposed that successfully explain many features of the high-harmonic spectra [2–4]. Different models may be physically meaningful for different harmonic orders and pumping intensities. New experiments are required to test which of these models, if any, are valid.

Early experiments measured the harmonic intensity and the cutoff energy as functions of atomic species and density, laser intensity, and focusing geometry [1,5]. These have only a limited ability to differentiate between the models of HHG. New measurements involving more subtle aspects of HHG are needed to reveal the underlying physical mechanisms. In particular, several groups are now investigating HHG by elliptically polarized incident light [6], measuring harmonic conversion efficiency vs the ellipticity of the driving field, in order to test the predictions of the various models of HHG.

In this Rapid Communication, we report detailed measurements of the polarization of vacuum ultraviolet high harmonics generated with elliptically polarized incident light. These data offer previously unavailable information about the evolution of the electron wave function during harmonic generation. We feel that these experiments are a sensitive probe of the evolution of the radiating dipole at the moment high harmonics are generated. It is therefore possible that the quantum mechanical event corresponding to the classical acceleration of the electron, which is responsible for the radiation of the harmonics, can be resolved not only in magnitude, but also in direction. To our knowledge, no measurements of the polarization of odd-order harmonics generated in isotropic media by elliptically polarized light have ever been published.

II. EXPERIMENT

Our experimental apparatus is shown schematically in Fig. 1. Our light source (described in detail elsewhere [7]) is a Ti:sapphire oscillator–CPA system delivering a 10-Hz train of 785-nm, 1.5-mJ, 200-fs pulses. The incident light passes through a series of waveplates to produce pulses of arbitrary ellipticity, with major axes of the polarization ellipse at arbitrary angles to the horizontal. This light is focused into a 0.2-m focal length, normal incidence vacuum monochromator. At the entrance slit the laser pulse passes through a

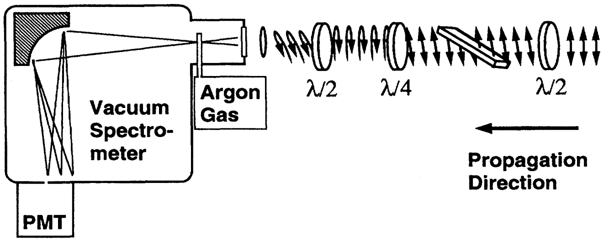

FIG. 1. Experimental schematic: The linearly polarized incident beam enters from the right. The polarization is rotated arbitrarily by a half-wave plate. The polarization linearity is improved by a thin film polarizer (TFP) which remains at the brewster angle while rotating around the laser beam axis. The light passes through a fixed quarter-wave plate; depending on the polarization direction before the quarter-wave plate, the light can be made to have any ellipticity. The polarization ellipse is then rotated 360° in small increments by another half-wave plate, after which the light is focused into the spectrometer. It focuses at the entrance slit location, where it passes through the gas target described in the text. The incident light and its harmonics are separated and refocused by a spherical surface grating. The grating can be rotated to deliver any harmonic to the exit slit and windowless photomultiplier tube (PMT).

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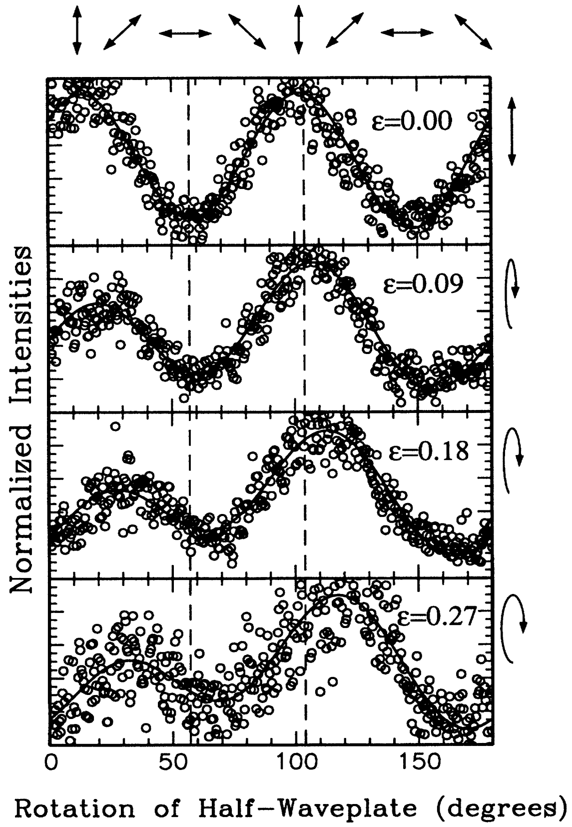

FIG. 2. Fifteenth harmonic in 9 torr of argon. Detected intensity vs rotation of driving field polarization ellipse. Directions of the major axes of the incident beam polarization ellipse are shown at the top. Zeros are suppressed, and the intensities are normalized to show equal modulation amplitude for each curve. All the curves were generated under identical circumstances, except for the ellipticity ε of the incident field (shown in the upper right corner of each plot). As the polarization ellipse of the incident light becomes closer to a circle ($|\varepsilon|\rightarrow 1$), the sinusoidal variation in the detected signal is shifted to the right. The circles represent data points; the solid curves are fits to the data.

hole in the side of a flattened hollow tube whose flattened surface is normal to the incident beam. The hole is drilled *in situ* by the same laser at higher power. The tube contains argon gas at densities of $1-3\times 10^{16}$ cm^3. Harmonics are separated and focused by an iridium coated spherical diffraction grating, which refocuses harmonics on an exit slit. Vacuum ultraviolet (vuv) radiation is detected by a windowless photomultiplier tube.

For all the observed harmonics, our grating exhibits a polarization-dependent diffraction efficiency. S-polarized light is favored over P-polarized light by as much as 2 to 1. We therefore use the grating to analyze the polarization of the harmonics. For each harmonic studied, we begin by generating harmonics with linearly polarized light, rotating the incident linear polarization 360° in small increments. The signal vs polarization angle is sinusoidal due to the polarization sensitivity of the grating. The maxima correspond to S polarized incident light. The ratio of maximum to minimum signal as the polarization is rotated is a measure of the polarization analyzer efficiency.

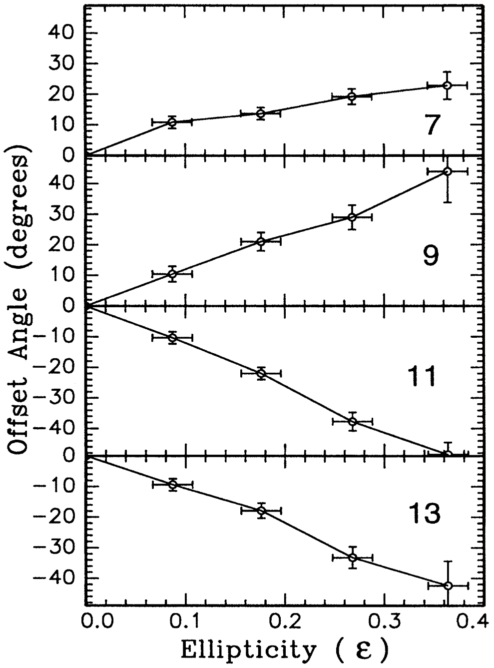

FIG. 3. Offset angle vs ellipticity for the 7th through the 13th harmonic in 9 torr of argon. The harmonic order is shown towards the right side of each plot. Note that the vertical scale changes sign for harmonic orders greater than 9.

When the incident light is elliptically polarized, the signal still varies sinusoidally as the incident polarization ellipse is rotated, but the maxima do not occur when the incident polarization ellipse is oriented with its major axis in the S direction (see Fig. 2). This suggests that the polarization axis of the harmonic is rotated relative to the polarization ellipse of the driving field. The angle between the incident and generated polarization ellipses can be obtained from the phase shift of the sinusoidal signal. The depth of modulation as the polarization rotates, when compared to the modulation for linear polarization, is a direct measure of the ellipticity of the harmonic. In this first Rapid Communication, we will concentrate on an analysis of the polarization offset. An improved signal to noise ratio in future measurements will enable us to analyze the ellipticities as well.

III. RESULTS

Using a nonlinear least-squares (NLS) fitting routine, we determined the offset of the signal variation (i.e., the polarization rotation angle) for the various ellipticities and harmonic orders. These values convert directly to offset angles. The results of one set of data appear in Fig. 3. We define ellipticity ε as the ratio of the two axes of the polarization ellipse, so that $\varepsilon=0$ corresponds to linear polarization, and $\varepsilon=1$ corresponds to circular polarization. The sign of the ellipticity and that of the offset angle are defined according to standard convention: When looking at the target, positive ellipticity means the electric field vector precesses clockwise; positive offset angles increase counterclockwise.

If the incident light is linearly polarized, we observe harmonics polarized linearly in the same plane, as required by

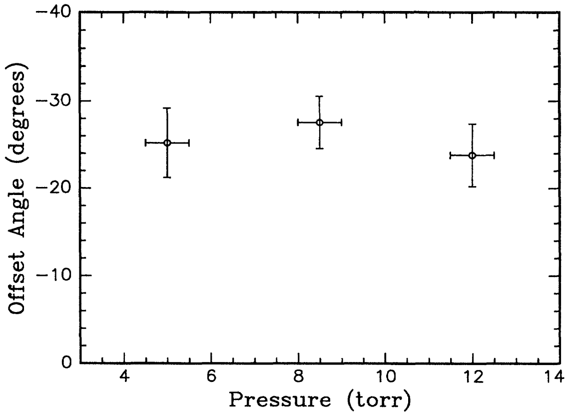

FIG. 4. Offset angle vs argon gas target pressure, 15th harmonic. The incident ellipticity is 0.19.

symmetry. The absence of rotation for HHG signals produced by linearly polarized light eliminates one important class of systematic errors that might mimic our results.

The offset angle is an odd function of the ellipticity, i.e., the polarization axis tilts in the opposite sense when the handedness of the ellipticity is reversed. Since this is a necessary consequence of parity and time-reversal invariance, it provides another systematic error check in our experiment.

For argon gas driven by 785-nm radiation, the offset increases monotonically in magnitude with increasing ellipticity of the driving field. The offset angle is positive for the 7th and 9th harmonics, and negative for subsequent harmonics from 11th to 17th order. However, initial measurements show both the sign and magnitude of the offset to be dependent on the wavelength of the driving field and on the choice of nonlinear medium. Future reports will describe this in more detail.

We also performed measurements of the offset angle vs target gas density for a constant ellipticity of the driving field (see Fig. 4). There was no measurable dependence on density across the range of pressures used.

The polarization rotation reported here may be a property of the single-atom response to a strong driving field, so that the harmonic radiation from each atom has a polarization rotated with respect to the incident laser. Alternatively, the harmonic polarization may rotate as it propagates through the gas, because of laser-induced circular birefringence. The latter has been observed in at least two forms: as self-rotation of the polarization ellipse [8], or as rotation of the polarization (usually linear) of a weak probe beam in the presence of a strong circularly polarized beam [9]. In both cases, however, one should expect a linear dependence of the rotation angle on the gas density, which is clearly absent in our experiments. We conclude that this effect is due to the single-atom response to a strong laser field.

Recently, a few groups have begun to perform calculations that can model elliptical incident polarization and resolve the polarization of the harmonics [3,10,12]. Although preliminary results do show some variation in the harmonics' polarization with changes in the incident ellipticity, a conclusive comparison with models of HHG will have to wait until the theories are better able to account for elliptical driving fields. We note that our results offer some support for the semiclassical, two-step model of HHG [2]. In this model, the atom is tunnel-ionized near the peak of the field [13], and an electron wave packet is launched into the continuum with approximately zero kinetic energy. When the field direction reverses, the wave packet may be driven back towards the atomic core. In this picture, rescattering of the wave packet from the core is responsible for HHG. This process should depend on the polarization state of the incident light. For elliptically polarized laser light, overall HHG efficiency is reduced because of the reduced probability that electrons are driven back towards the atomic core. This agrees with recent experiments [6], although for symmetry reasons qualitatively similar behavior is expected in harmonic generation in isotropic media, in any intensity regime; in the first report of harmonic generation in gas [11], the authors give a detailed description of this and other effects. For those electrons that do reencounter the core, the azimuthal symmetry of the scattering event is broken, allowing the observed offset angle between the polarization of the incident field and the harmonics. Further calculations might help to establish the connection between HHG and semiclassical rescattering models.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation, through the Center for Ultrafast Optical Science, under Grant No. FTC PHY 8920108. Peter Shkolnikov was supported by AFOSR.

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