Coherence and Resonance Effects in High-Order Harmonic Generation

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We present experimental measurements of harmonic generation in xenon using a 1064-nm 40-psec Nd-doped yttrium-aluminum-garnet laser. The harmonic yield is studied as a function of the position of the laser focus in the atomic beam. It shows regular oscillations whose period decreases with increasing harmonic order and which can be interpreted as phase-matching effects due to the tight focusing geometry. Moreover, the amplitudes of the oscillations vary rapidly with the laser intensity, a signature of resonance effects in the harmonic generation process.

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Recent experiments [1-3] have demonstrated the possibility for harmonic generation processes to reach very short wavelengths (≤ 20 nm) with reasonable conversion efficiencies. The nice spatial and temporal characteristics of the generated radiation as well as the extremely high brightness that can be achieved show that these processes will most probably lead to the development of useful vacuum-ultraviolet sources. However, there are still a few questions that need to be answered in order to fully understand the mechanisms governing harmonic generation in strong laser fields. This concerns mainly the collective aspect of harmonic conversion processes, namely, how can all the high harmonics be phase matched. For a low atomic density as is the case in the experiments, dispersion effects can be ignored in a first approximation. Phase matching is then essentially determined by focusing, which introduces a geometrical phase lag between the induced and driving fields [4]. Calculations of harmonic generation in xenon at 1064 nm have recently unraveled some of the questions [5]. An essential result was that phase matching of the high harmonics was found to be considerably enhanced in a strong-field regime as compared to the weak-field limit. This explained why the plateau observed in the harmonic emission from a single atom [6] is conserved when considering the response of the whole medium.

In this Letter, we present the first experimental evidence for phase-matching effects in strong-field high-order harmonic generation processes. This is done by studying the harmonic yield for a few harmonics in xenon at a given intensity, as a function of the position of the laser focus in the atomic beam. The harmonic signal shows oscillations whose period corresponds to the coherence length induced by the tight focusing of the beam. This spatial period decreases with increasing harmonic order and with decreasing laser confocal parameter. Moreover, we find that, although the oscillation pattern does not change significantly with the laser intensity, the amplitudes of the oscillations exhibit important variations, due to resonance effects in the harmonic production yield. The generation of high harmonics in strong laser fields results from a rather subtle interplay between (single-atom) resonance effects which occur at a given laser intensity (and therefore at a given space point) and interferences due to the loss of coherence induced by focusing.

Consider an incident Gaussian beam, propagating along the z axis, focused at $z=0$:

$$E_1(r,z)=\frac{bE_0}{(b^2+4z^2)^{1/2}}\exp\left(-\frac{k_1r^2b}{b^2+4z^2}-i\tan^{-1}(2z/b)+\frac{2ik_1r^2z}{b^2+4z^2}\right),$$

where b is the laser confocal parameter, which characterizes how tightly the beam is focused, E_0 the peak field strength, and k_1 the wave vector of the field. The polarization P_q induced by the incident field at the harmonic frequency $q\omega$ is defined by

$$P_q(r,z)=2\mathcal{N}(z)d_q(r,z)\exp\left(-iq\tan^{-1}(2z/b)+\frac{2iqk_1r^2z}{b^2+4z^2}\right),$$

where $\mathcal{N}(z)$ is the atomic density and $d_q(r,z)$ is the atomic dipole moment calculated for the field strength $|E_1(r,z)|$, and which we assume to have a constant phase [5]. For a medium with negligible dispersion, i.e., such that the refractive index can be considered to be independent of the frequency, we have $k_q \approx qk_1$. The phase difference between the polarization and the harmonic field that would propagate freely with the same confocal parameter as the incident field reduces to $(1-q)\times\tan^{-1}(2z/b)$. The harmonic field E_q that can be generated in the medium depends on the interferences between the source (driving) term P_q and the harmonic (driven) wave. The interference fringes will be separated by twice a coherence length approximately equal to $(b/2)\tan[\pi/(q-1)]$ close to the focus (slightly increasing away from it).

We illustrate in a few simple cases in Fig. 1 how a harmonic field is generated in a nonlinear medium. Here, we take a 2-mm-long uniform medium and a 1.5-mm laser

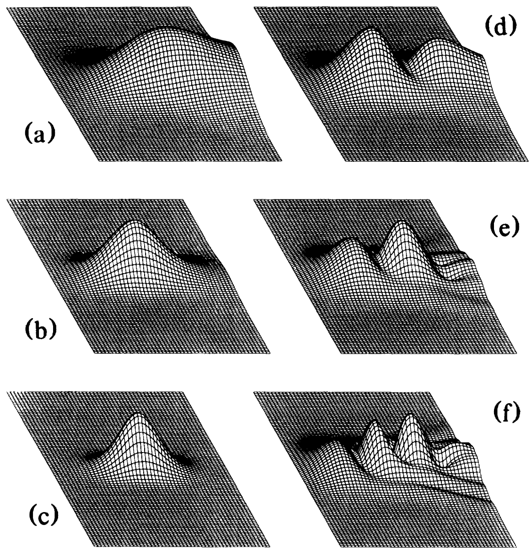

FIG. 1. Graphical representation of $|E_q(r,z)|$ in the nonlinear medium. The light propagates along the horizontal axis (z) from the left to the right. The first column shows the (a) third, (b) seventh, and (c) thirteenth harmonics calculated within the framework of perturbation theory. The second column shows the (d) seventh, (e) ninth, and (f) thirteenth harmonics calculated by assuming a nonlinear polarization varying as $|E_1|^5$.

confocal parameter. The first column [Figs. 1(a)-1(c)] shows the harmonic field E_q calculated in the weak-field limit for $q=3$, 7, and 13. In this approximation, the amplitude of the nonlinear polarization ($\propto |E_1|^q$) is essentially concentrated within *one* coherence length on either side of the focus, so that the interference pattern reduces to one oscillation. The second column [Figs. 1(d)-1(f)] shows the harmonic field E_q ($q=7,9,13$) calculated by assuming d_q to vary as $|E_1|^5$. This is approximately the average power law found for the single-atom response at high intensity [5,6]. For the high harmonics ($q \geq 7$), the polarization amplitude varies much more slowly in the nonlinear medium than in the weak-field limit, thus spanning *several* coherence lengths. Consequently, the figures exhibit interference effects whose period decreases with increasing process order. (Compare the number of waves displayed in the three cases.) As the effective order of nonlinearity taken to be 5 in Figs. 1(d)-1(f) decreases from the weak-field limit q, the amplitude over which the nonlinear polarization is distributed increases so that the oscillations due to phase matching grow and consequently the field that exits the medium becomes more intense.

The motivation for the experiments presented in this Letter was to show some evidence for these (nonperturbative) phase-matching oscillations. We use a 1064-nm 40-psec mode-locked Nd-doped yttrium-aluminum-garnet laser [2]. It is focused by a $f=200$ mm lens to an intensity of a few 10^{13} W cm^{-2}. We have measured the section at best focus to be 400 μm^2 ($\pm 20\%$), with a confocal parameter $b=1.5$ mm. The nonlinear medium is provided by a pulsed gas jet, with a maximum density of 5×10^{17} xenon atoms/cm^3. The vacuum-ultraviolet light is detected on axis by a monochromator consisting of a 275-line/mm gold-coated grazing-incidence toroidal grating and an electron multiplier. In order to probe any oscillations of the harmonic yield in the propagation direction, we move, by steps of 100 μm, the position of the focus in the atomic beam (by displacing the focusing lens

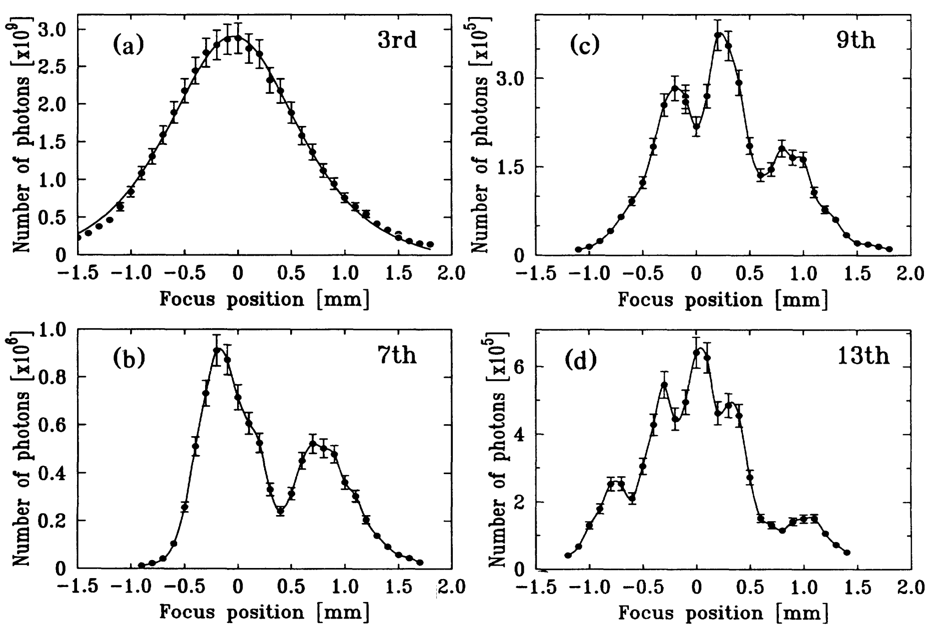

FIG. 2. (a) Third, (b) seventh, (c) ninth, and (d) thirteenth harmonics as a function of the position of the laser focus in the nonlinear medium. The laser propagates from the left to the right.

along the laser axis).

In Fig. 2, we plot the number of photons at the third, seventh, ninth, and thirteenth harmonic frequencies (in linear scale) as a function of the focus position. The zero on the horizontal scale indicates approximately the center of the atomic beam. The laser energy is kept constant for each series of measurements, with an allowed dispersion of $\pm 5\%$. The intensities are 1.9×10^{13} W cm^{-2} in Figs. 2(a)-2(c) and 2.5×10^{13} W cm^{-2} in Fig. 2(d). Each experimental point (solid circle in Fig. 2) is an average of 100 laser shots. We have indicated the statistical error bars. For the seventh, ninth, and thirteenth harmonics, the solid line is a spline interpolation of the data. For the third harmonic, it is the result of a perturbative calculation assuming a Lorentzian atomic density of width at half maximum of 0.8 mm and a Gaussian laser beam with a $b = 1.5$ mm confocal parameter.

The third and also the fifth harmonics behave according to what can be predicted from lowest-order perturbation theory, with a single maximum at the center of the medium. Oscillations begin to appear at the seventh harmonic. They are quite regular and their period decreases with increasing order: In Fig. 2, the seventh harmonic displays two oscillations, the ninth, three, and the thirteenth, five. The differences between two consecutive minima or maxima are, respectively, 0.8, 0.5, and 0.25 mm close to zero. These values are consistent with twice the geometrical coherence length for these processes, equal to 0.8, 0.6, and 0.4 mm, respectively. The smaller period obtained experimentally for the thirteenth harmonic might come from dispersion (e.g., a resonance in the atomic polarizability or the contribution of free electrons) or from other dephasing effects. Beyond the thirteenth harmonic, we cannot clearly resolve the structures because the period becomes too small and also because the length over which there is appreciable amplitude decreases. Apart from the third and fifth harmonics, the figures are completely different from what is obtained within the framework of lowest-order perturbation theory. Note that the number of waves observed in Figs. 1(d)-1(f) (obtained by assuming a 2-mm-long medium and the same focusing conditions as in the experiment) corresponds to the number of oscillations displayed in Fig. 2 over the same length. We performed a series of measurements in a different focusing geometry, with a $f = 300$ mm focal lens. Similar oscillations were observed, but with a period approximately doubled, as it should be for a more loosely focused geometry with a confocal parameter measured to be twice the previous one. This confirms that, in these experiments, phase matching is mainly influenced by the geometry and not much by dispersion.

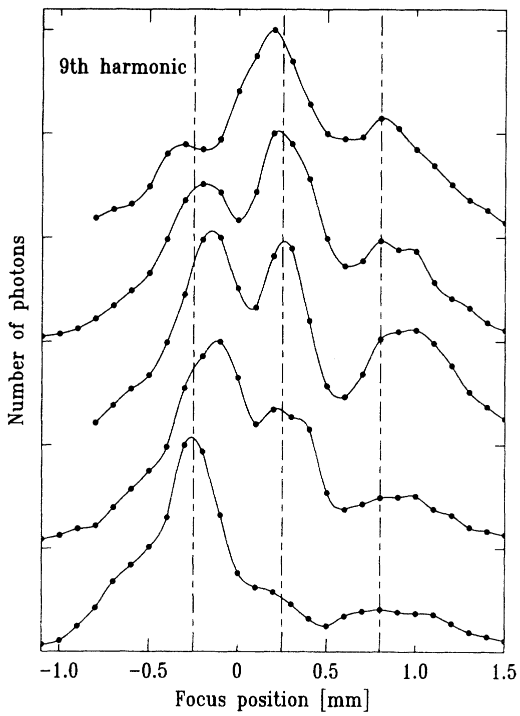

FIG. 3. Ninth harmonic as a function of the focus position. From the bottom to the top, the intensities are 1.4×10^{13}, 1.55×10^{13}, 1.7×10^{13}, 1.9×10^{13}, and 2.15×10^{13} W cm^{-2}.

The most interesting aspect of these measurements was brought about by the study of how the structures were changing with the laser intensity. In Fig. 3, we show the number of ninth harmonic photons as a function of the position of the focus at several intensities varying from 1.4×10^{13} to 2.15×10^{13} W cm^{-2} before ionization becomes significant. The curves have been normalized to have the same maximum value. The oscillation pattern remains quite similar for the different intensities, with approximately the same positions for the minima and maxima (the latter are indicated by the dot-dashed lines in the figure). However, the relative amplitudes of the maxima and of the minima vary quite rapidly over a very small range in intensity. Note that they would be independent of intensity if the (single-atom) harmonic emission rate were following a simple I^p power law, p denoting an effective order of nonlinearity and I the laser intensity. Therefore, we think that these rapid variations are the signature of resonance effects in the harmonic production yield. As shown experimentally a few years ago in the case of ionization processes [7], excited states can shift into resonance during the rise time of the laser pulse, or at different locations of the interaction volume, thus enhancing (locally) the ionization probability or here, the harmonic generation rate [8]. Consider, for simplicity, what happens on the propagation axis. A resonance in the atomic harmonic emission rate occurs at a given intensity and therefore in two positions symmetric relative to the focus. The effect of the resonance on the total harmonic yield will be dramatically enhanced or reduced, depending on whether the wavelets emitted at these two positions add constructively or destructively. This interference depends on the distance between the two positions

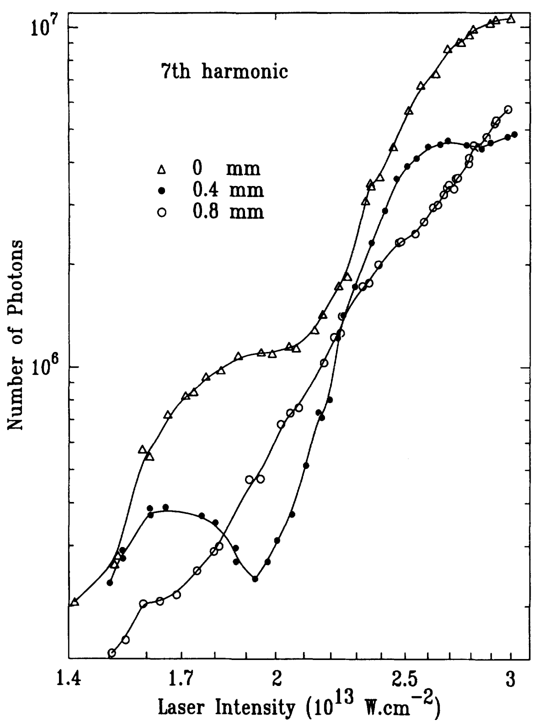

FIG. 4. Seventh harmonic as a function of the laser intensity for different positions of the focus.

(relative to the coherence length) and consequently on the laser peak intensity. One should therefore expect strong modulations as the intensity is varied.

As a further consequence, the harmonic yield as a function of intensity should be extremely dependent on the position of the focus in the medium. We show in Fig. 4 the variation of the seventh harmonic with the laser intensity for three positions separated by 0.4 mm. The curves are indeed quite different, going from a rather smooth behavior (open circles), to a much more accented one (solid circles), with a pronounced dip around 1.9×10^{13} W cm^{-2}. Note that the smoothest curve is obtained when the focus is close to one edge of the atomic beam, so that the interference effects mentioned above should be less important.

As is often the case in nonlinear optics, macroscopic and microscopic responses are closely linked. It is the strong-field (resonance) effects in the harmonic generation yield that allow the polarization to be less concentrated in the medium than in the weak-field limit, thus compensating for the loss of coherence induced by focusing. Conversely, the observation of these resonances depends crucially—and in a nontrivial fashion because of the interference effects—on the macroscopic parameters of the interaction.

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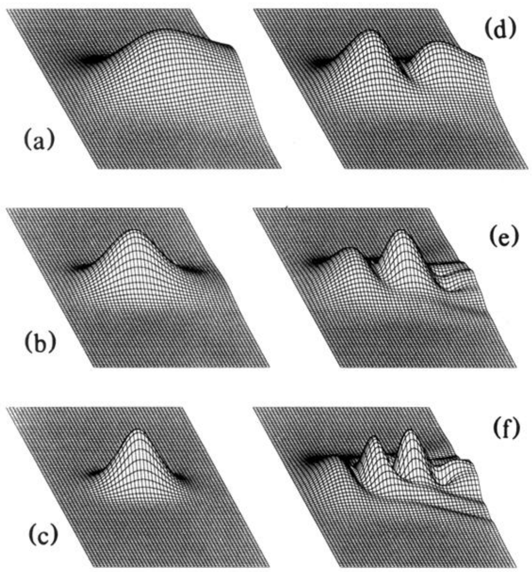

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