High-Order Harmonic Generation in Rare Gases with a 1-ps 1053-nm Laser

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We present experimental results of harmonic generation using a 1-ps 10^{15}-W/cm^2 Nd-glass laser. We see up to the 29th harmonic in Xe, 57th in Ar, and at least up to the 135th harmonic in Ne (160 eV), being then limited by our monochromator's resolution. The conversion efficiencies in Xe, Ar, Ne, and He are compared and different ways of optimizing the number of photons by defocusing the laser beam or increasing the density are discussed.

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Harmonic generation in gases depends on the individual response of each atom to the (strong) radiation field and on the collective emission from the medium. The single-atom response is governed by the characteristics of the laser light and in particular its frequency and intensity, and by the atom's nature, its ionization energy, and more generally its structure. The distribution of harmonics presents a characteristic behavior, with three distinct regions: a rapid (perturbative) decrease from the 3rd to the 7th or 9th harmonic, a long plateau where the harmonics have almost constant amplitudes (or very slightly decrease), and a rather sharp cutoff. Recently, Krause, Schafer, and Kulander [1] have extended the theoretical study of these processes [2] to much higher intensities, in the regime of tunneling ionization of atoms. They predict the possibility of producing many odd harmonics (e.g., more than one hundred) of an intense low-frequency laser focused into a gas target of light atoms or ions. The conversion efficiencies for these very-high-order processes are expected, however, to be very weak, making any experimental observation difficult.

In this strong field regime for the laser-atom interaction, the phase matching of the harmonics in the nonlinear medium is almost independent of the process order. The characteristic shape of the harmonic intensity distribution is only weakly affected by propagation effects [3]. However, the fact that the harmonics are phase matched and that the light emitted is coherent also means that the conversion efficiency increases rapidly with the atomic density (in general as the square power). Furthermore, for neutral systems, the length over which the harmonics can be built up coherently, and therefore the amplitude of the harmonic field that exits the medium increases with the laser confocal parameter. The conversion efficiency can be enhanced by using weak focusing optics. Thus, the variation of the parameters that affect the collective response from the medium allows one to optimize the number of photons produced and in some cases to compensate for a relatively weak emission from the atoms taken individually.

Although most of the experiments so far have been carried out at intensities below 10^{14} W/cm^2 [4], results obtained in the strong field regime begin to appear. Sarukura *et al.* reported observation of the 25th harmonic in He and Ne using a 500-fs 248-nm laser [5], a result interpreted [1, 6] as harmonic emission from the ions. Miyazaki and Sakai detected the 41st harmonic in He with a 800-fs 616-nm dye laser [7]. More recently, Macklin, Kmetec, and Gordon observed the generation of up to the 109th harmonic in Ne, using a 125-fs 806-nm Ti:sapphire laser [8], and Crane *et al.*, the generation of the 45th harmonic in He with the second harmonic (527 nm) of a 600-fs Nd-glass laser [9].

In the present work, we study high-order harmonic generation processes in the rare gases xenon, argon, neon, and helium using a 1-ps 1053-nm Nd-glass laser at intensities between 10^{14} and 10^{15} W/cm^2. The difference between the responses of the four rare gases (especially between Xe, Ar, and Ne) is spectacular and seems to be much more pronounced at this low frequency than with higher frequency lasers [5, 7]. The maximum orders observed in Xe and Ar are, respectively, the 29th and the 57th. In Ne and He, we see at least up to the 135th harmonic (160 eV), being then limited by the resolution of our spectrometer. We also show how one can adjust the atomic density and the position of the focus in the medium in order to increase the signal in He and Ne, which produce many harmonics but with a weak efficiency, or to obtain extremely high instantaneous powers and brightnesses in Xe and Ar which produce relatively fewer harmonics, but with a much higher conversion efficiency.

The laser system used in these experiments, which is based on chirped pulse amplification, has been described elsewhere [10]. The 1053-nm laser pulse from a continuous-wave mode-locked Nd-YLF oscillator is chirped into an optical fiber. A selected pulse gets amplified through a Nd-glass regenerative amplifier up to a few mJ. The laser is then split into two amplifier chains: the main "terawatt" chain which produces 1-J 1-ps pulses after compression at a repetition rate of 1 shot per min; and the so-called "gigawatt" chain, employed in these

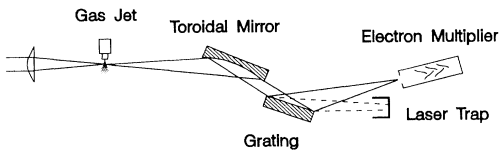

FIG. 1. Experimental setup.

harmonic generation experiments, which produces 30-mJ 1-ps pulses at a repetition rate of 1 shot every 10 s. The detection system is new and has been designed for these harmonic generation experiments. The interaction chamber is schematized in Fig. 1. The laser is focused by a $f = 300$ mm plano-convex lens just below the nozzle of a pulsed gas jet (the confocal parameter b is equal to about 3 mm). The light is analyzed on axis by a monochromator consisting of a toroidal mirror and a grating (Jobin Yvon) both coated with gold. The mirror, placed at 1 m from the laser focal spot to minimize the risks of damaging the optics with the incident laser light, refocuses the radiation onto a 100-μm output slit. The laser light reflected by the grating is trapped before being focused too much in order to avoid plasma generation on the walls of the chamber (see Fig. 1). The photons are detected by an electron multiplier. The spectral range covered by this system is approximately 7–60 nm. There is no entrance slit and no diaphragms, so that this spectrometer has a good detection efficiency.

Figure 2(a) shows a spectrum obtained in neon at an intensity of about 10^{15} W/cm^2. Each point is an average of about three to five laser shots, with a strict selection in pulse energy and duration. The positions of the harmonics are indicated by the straight lines at the top of the figure. The width of the harmonics is constant, equal to the resolution of the monochromator (0.5 Å). Below 13 nm, the harmonics start overlapping, which results in an increase of the background level. The reflectance of gold, with which both the mirror and the grating are coated, decreases by more than 1 order of magnitude from 10 to 7 nm for the 11-deg incidence angle on the mirror and grating. Figure 2(b) shows the region from 12 to 8 nm, where we have divided the signal by the monochromator's response [11] and taken into account our detector's gain. There is no apparent cutoff, merely a steady slight decrease. The harmonics can be resolved until about the 135th harmonic (the highest order reported to date). In order to make sure that the observed light from 9 to 7 nm was coherent and due to harmonic generation, we checked carefully that the well resolved high harmonics (e.g., the 97th) and the signal in this 9–7 nm region had the same dependences both with pressure and with the laser intensity (i.e., same appearance and saturation intensities).

In Fig. 3, we compare the results obtained in xenon, argon, neon, and helium at an intensity at best focus equal to 1.5×10^{15} W/cm^2. In xenon, the intensity of

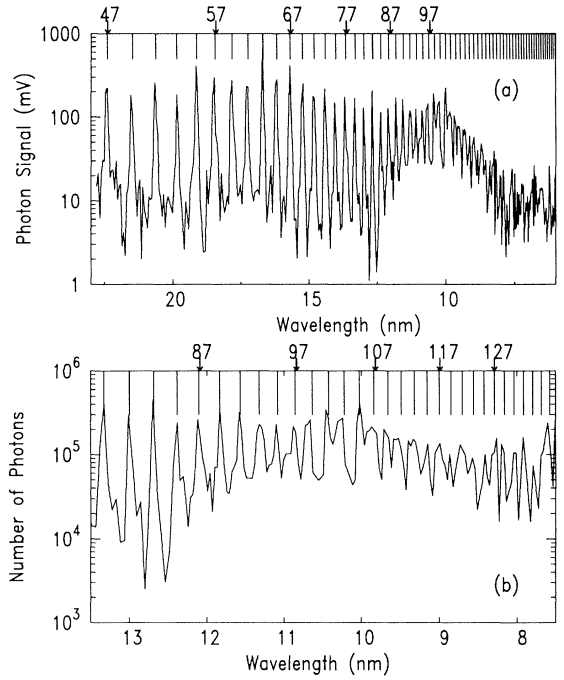

FIG. 2. Experimental spectrum obtained in neon at 40 Torr, 1.5×10^{15} W/cm^2. (a) Raw data; (b) data corrected from the spectrometer's response over 12–8 nm.

the harmonics is constant from the 5th harmonic [12] to the 23rd and then falls off abruptly over 4 orders of magnitude. In argon, there seem to be two successive plateaus, one up to the 27th harmonic, and the other from the 39th to the 49th harmonic [13]. In neon and helium, we see very long and quite similar plateaus, whose limits are experimental. Krause, Schafer, and Kulander [1] find that the energy W_p reached at the end of the

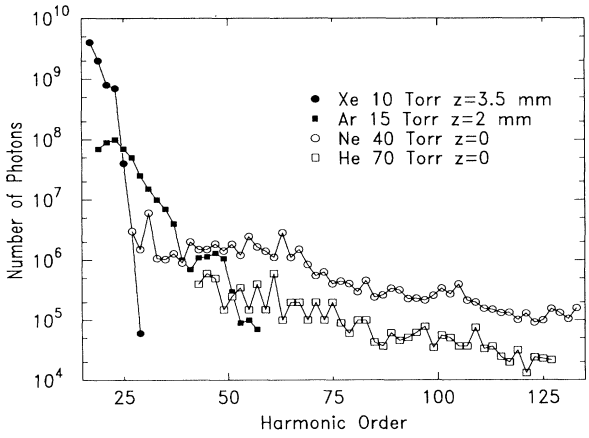

FIG. 3. Number of photons obtained per laser pulse in xenon, argon, neon, and helium as a function of the harmonic order.

plateau obeys the rule $W_p = I_p + 3U_p$, where I_p denotes the ionization energy and $U_p = e^2E^2/4m\omega^2$ (e, m are the electron's charge and mass; E, ω the field's strength and frequency) the ponderomotive energy at or below the saturation intensity (I_s) for the atomic system considered (i.e., the intensity at which the atoms get ionized). This energy of $3U_p$ corresponds to the maximum energy that an electron, born at rest in a laser field, can gain [14, 15], so that W_p can be understood as the (maximum) energy that the system can absorb and reemit under the form of energetic photons. Our experimental results are remarkably consistent with this simple rule. We find 27 and 62 eV for the maximum energy of the plateau in Xe and Ar, to be compared with W_p= 25 and 60 eV for the saturation intensities, respectively, of 7×10^{13} W/cm^2 and 1.5×10^{14} W/cm^2 in our experimental conditions [16]. In Ne and He, the rule gives W_p = 170 and 230 eV (I_s = 5 and $7\times$ 10^{14} W/cm^2). These values are higher than the 160 eV corresponding to the 135th harmonic, which reinforces our feeling that the observed light is coherent, due to higher-order harmonic generation.

We adjusted the pressure from 10 Torr in xenon to 70 Torr in helium in order to optimize the signal-to-noise ratio and to increase the harmonic emission in Ne and He. The number of photons obtained is found to vary approximately as the square of the atomic density over the range investigated. A more spectacular way of increasing the conversion efficiency is to use a weaker focusing geometry or simply here to defocus the laser beam in the medium. The idea is that the strength of the field that leaves the medium depends on the length $L_{\rm coh}$ over which it can be built up coherently [3, 12]. For neutral systems, this length depends on the laser focusing geometry and on the process order. It increases away from the focus (one gets closer to a plane wave approximation) until $L_{\rm coh}(z) \approx \pi b(1+4z^2/b^2)/2(q-1)$ becomes larger than $L/2$, L denoting the medium's length and z the coordinate on the propagation axis. To observe a significant effect, the intensity at best focus must be much higher than the saturation intensity, which was the case in these experiments only in Xe and Ar. We estimate our best focus intensity to be equal to 1.5×10^{15} W/cm^2, i.e., 2, 3, 10, and 20 times the saturation intensity in He, Ne, Ar, and Xe, respectively. In Fig. 4, the numbers of photons for the 17th harmonic in xenon and the 25th harmonic in argon are plotted as a function of the position of the focus (in mm) in the jet. The maximum emission is obtained at $z = \pm$ 3.5 mm in Xe and at $z = \pm$ 2 mm in Ar (i.e., $I \approx 3I_s$). The number of photons increases by a factor of 10 in Ar and of 50 in Xe as the laser is defocused. Thus we get quite high numbers of photons, almost 10^{10} photons per pulse for the 17th harmonic in xenon.

We did not see any clear evidence for harmonic emission from Xe^+ or Ar^+ ions, though we certainly had enough laser intensity. We performed phase matching calculations in a plasma environment and concluded that

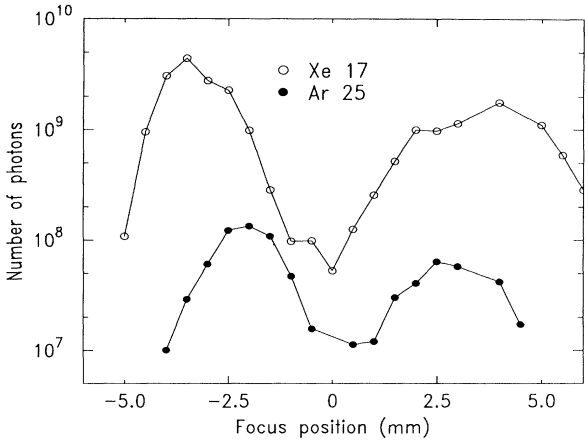

FIG. 4. 17th and 25th harmonics in Xe and Ar as a function of the position of the focus in the jet. The laser propagates from the left to the right.

the decrease in intensity resulting from the effect of free electrons would not be more than 1 order of magnitude, so that these harmonics should be produced with a reasonable efficiency. However, at intensities such that Xe^+ or Ar^+ would generate harmonics, the ionized medium produces an intense plasma light which hinders the observation of these harmonics, even at pressures as low as 5 Torr. Temporal resolution or a more collimated detection geometry which would reduce the isotropically emitted plasma light relative to the harmonic light are needed for detecting harmonic generation from ions in our experimental conditions.

In conclusion, we have observed coherent light emission by optical harmonic generation in gases at energies at least up to 160 eV. The harmonic emission is directional and of short pulse duration (shorter than the pump pulse, i.e., a few hundreds of femtoseconds in this experiment) [12]. The instantaneous power generated, e.g., at 20 eV (the 17th harmonic in Xe) reaches about 30 kW, with a conversion efficiency of 10^{-6}. The instantaneous brightness is 10^{22} photons/Å s, a number that is several orders of magnitude higher than that obtained with conventional light sources in this spectral domain. We have shown how similar brightnesses could be reached in the 100–200 eV range, and even at higher energies (400 eV) from ionic systems [1], by using more powerful and weakly focused lasers and by increasing the atomic density. These high-order harmonic generation processes will undoubtly lead to the development of bright, short-pulse, and versatile sources of coherent radiation in the vacuum ultraviolet, soft x-ray ranges.

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