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LETTER TO THE EDITOR

Multiple-harmonic conversion of 1064 nm radiation in rare gases

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Abstract. We report the observation of very-high-order odd harmonics of Nd:YAG laser radiation in rare gases at an intensity of about 10^{13} W cm^{-2}. Harmonic light as high as the 33rd harmonic in the XUV range (32.2 nm) is generated in argon. The key point is that the harmonic intensity falls slowly beyond the fifth harmonic as the order increases. Finally, a UV continuum, beginning at 350 nm and extending down towards the short wavelength region is apparent in xenon.

The non-linear interaction of an intense laser pulse with many-electron atoms leads to new interesting physical processes. Multiphoton ionisation of rare-gas atoms removes various numbers of electrons from the outer shell and produces multiply charged ions. Electron energy distributions (Lompré *et al* 1985, Johann *et al* 1986, Bucksbaum *et al* 1987, Petite *et al* 1987) and the production of multiply charged ions (L'Huillier *et al* 1982, 1983, Luk *et al* 1985) have previously been investigated in a collisionless environment at atomic densities of 10^{11}-10^{12} atoms/cm^3.

In order to provide a basis for understanding the physical processes involved, it is of interest to make further measurements, detecting the VUV and XUV radiation that may occur through fluorescence from excited multiply charged ions or through stimulated emission. Spectral analysis of the radiation generated is generally carried out using a spectrograph or a monochromator; the solid angle subtended by the aperture of such an instrument is small. As the overall collection efficiency is poor, it is necessary to increase the atomic density up to 10^{17}-10^{18} atoms/cm^3 in order to measure a significant radiation signal. Because of the high atomic density, the laser light no longer interacts with independent atoms but with a collective assembly of atoms characterised by a strong polarisability. Additional processes also occur, namely recombination processes, harmonic generation and, more generally, various parametric processes.

The purpose of the present letter is to describe harmonic generation in xenon, krypton and argon induced by the fundamental frequency of a Nd:YAG laser at 1064 nm. With the exception of a recent experiment on harmonic generation employing iodine-laser radiation (Wildenauer 1987) at 1315 nm, most of the experiments on rare gases reported in the literature use short primary-laser wavelengths: 266 nm (Reintjes *et al* 1978), 248 nm (Bokor *et al* 1983, McPherson *et al* 1987), 355 nm (Kung 1983) and 292-307 nm (Marinero *et al* 1983), in order to obtain the shortest wavelengths through harmonic generation. Therefore, very little is known about harmonic generation from infrared radiation at 1064 nm at a laser intensity of 10^{13}-10^{14} W cm^{-2} for which multiphoton ionisation of Xe, Kr and Ar is known to generate a fully ionised medium (L'Huillier *et al* 1983).

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The laser used in the present experiment is a commercial picosecond Nd:YAG laser (Quantel). The bandwidth limited 30 ps pulse is amplified up to 1 GW at a 10 Hz repetition rate. The laser pulse is then focused by a 200 mm focal length lens onto a gaseous target in a vacuum chamber pumped down to 10^{-8} Torr. The focused intensity distribution is determined from photometric measurements as in previous experiments (L'Huillier *et al* 1983, Lompré *et al* 1982). The gaseous target is generated by a pulsed gas jet from a 1 mm diameter nozzle whose valve opens only during a submillisecond interval synchronised with the incident 10 Hz Nd:YAG laser. The infrared laser pulse, directed perpendicularly onto the gas jet, is focused at 0.2 mm from the nozzle, where the high gas density region is narrowest and most sharply defined. An effective gas pressure of about 10 Torr is obtained at the focal point for a 150 Torr backing pressure. The calibration method based on determining the gas density at the focus of the laser has been described in detail elsewhere (Lompré *et al* 1988). The gas jet is projected directly onto the throat of an 800 l s^{-1} turbomolecular pump in order to maintain the background pressure at below 10^{-5} Torr during the entire duration of the experiment. The well confined gas beam geometry should substantially reduce reabsorption of the XUV radiation generated by the medium outside the interaction region. The XUV light generated at the focal point is analysed along the laser axis using a grazing incidence 275 line/mm gold-coated toroidal holographic grating blazed at 100 nm capable of analysing VUV radiation down to 50 nm. A second grating blazed at 15 nm is used to analyse XUV radiation below 50 nm. An EMI solar blind photomultiplier is used as a detector for the 115-355 nm range, while a windowless electron multiplier with CuBe dynodes is used for the 30-150 nm range.

Figure 1 shows a typical harmonic spectrum obtained for Xe in the 50-130 nm range with a windowless electron multiplier detector. This is a low-resolution scan and the harmonic line widths are solely dependent on the monochromator optics. All the odd harmonics between 9 and 21 are observed. The 17th, 19th and 21st harmonics also appear in the second order. The continuous background that appears in figure 1 is not an artifact, it disappears at lower pressure. It originates from a broad continuum

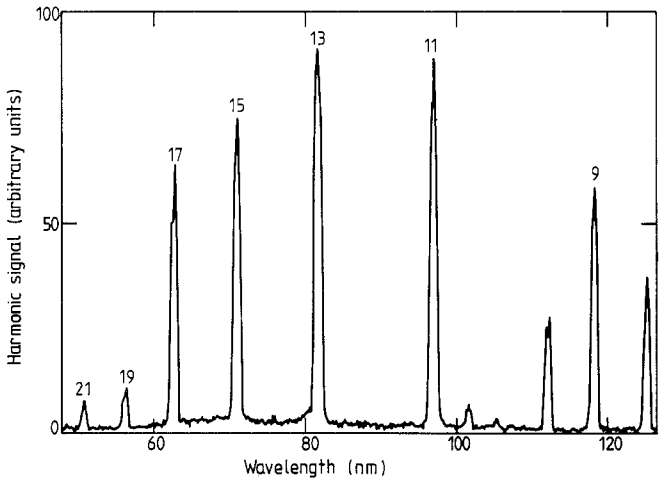

Figure 1. Harmonic spectrum obtained using a Xe gas jet showing all odd harmonics between 9 and 21. The peaks at 101, 112 and 125 nm are the second diffracted orders of the 21st, 19th and 17th harmonics respectively. The laser intensity was approximately 3×10^{13} W cm^{-2} and the Xe pressure at the focal point was about 10 Torr.

emission, which is intense for Xe, insignificant for Kr and non-existent for Ar. Finally, fluorescence from excited states, and, in particular, resonance lines from neutral atoms are also observed.

It is of interest to plot intensities of all the harmonic lines, including results obtained with the photomultiplier in the 115–355 nm range and with an electron multiplier in the 30–150 nm range. Figure 2 shows a plot of the relative intensity for harmonic generation in Xe, with corrections made for the spectral response of both the detector and the grating based on the characteristic curves given by the respective manufacturers. Whereas a decrement of three orders of magnitude is observed between the third and the fifth harmonics, the signal strength falls very slowly for any further increase in harmonic order.

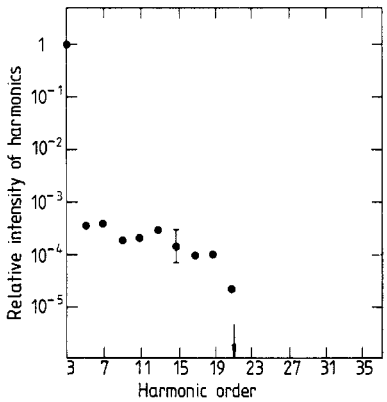

Figure 2. The relative intensity of the harmonics generated in Xe at a laser intensity of approximately 3×10^{13} W cm^{-2}. The typical error bar is shown for the 15th harmonic. The arrow indicates the highest harmonic order observed at 3×10^{13} W cm^{-2}.

As shown in figures 3 and 4, similar results have been obtained for Kr and Ar; however the harmonic spectrum extends up to the 29th order for Kr and up to the 33rd order for Ar corresponding to 32.2 nm radiation. In these three figures, the relative intensity of the harmonics is normalised to the third-harmonic signal strength for each of the rare gases. Figure 3 shows a plot of the relative intensities of the odd-order harmonics generated in Kr. At certain harmonic wavelengths, the generated XUV is partly absorbed, leading to modulations in the curve. This is the case for the 11th harmonic, which can photoexcite the Kr atom to a 4d or 4d′ state, and also for the 17th harmonic, which can photoexcite the Kr ion.

Results obtained with Ar are shown in figure 4. Here, the 13th harmonic is missing; a fact which can be explained by reabsorption of the 81.9 nm radiation by the neutral Ar in the photoexcitation of a 5d state. The high-order harmonic generation efficiency in Ar is more than one order of magnitude lower than that in Xe. A similar observation has previously been reported in the 90–105 nm range (Softley *et al* 1987). As seen in figures 2, 3 and 4, a sharp change occurs at approximately 6 eV. A similar behaviour has also been reported for harmonic generation induced in rare gases by an excimer laser at 248 nm (McPherson *et al* 1987). The shortest wavelength (35.5 nm) induced in Ar by a 248 nm laser pulse at an intensity of 10^{15}–10^{16} W cm^{-2} is roughly the same as that (32.2 nm) induced in the present experiment in Ar by a 1064 nm laser pulse at an intensity of only 10^{13}–10^{14} W cm^{-2}. Consequently, the efficiency for very short

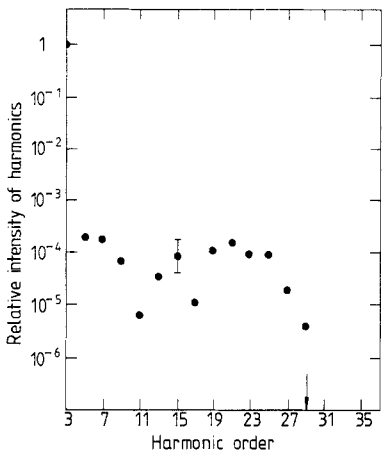

Figure 3. The relative intensity of the harmonics generated in Kr at a laser intensity of approximately 3×10^{13} W cm^{-2}. The arrow indicates the highest harmonic order observed at 3×10^{13} W cm^{-2}. The intensity of the 11th and 17th harmonics is reduced due to partial absorption through photoexcitation of Kr atoms and ions.

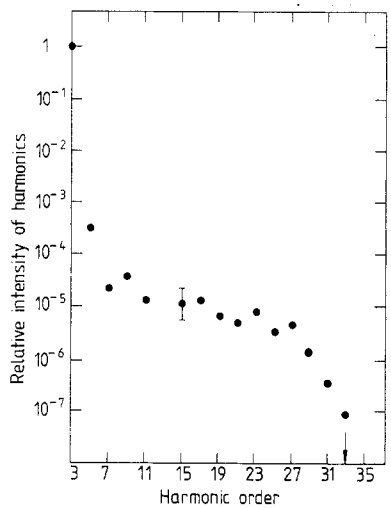

Figure 4. As figure 3 but for Ar. The 13th harmonic is missing due to a strong absorption of the 81.9 nm radiation in the photoexcitation of a 5d state.

wavelength harmonic generation appears to be much higher for a long primary-laser wavelength than for a short laser wavelength. This comparison emphasises the unexpected effect of primary-laser wavelength on the generation of very short wavelength radiation.

The third-harmonic generation of 354.8 nm from 1064 nm radiation exhibits positive-phase mismatch in Xe, Kr and Ar. As is well known, third-harmonic generation is forbidden in a strongly focusing geometry (laser beam confocal parameter much less than the length of the non-linear medium) and a positively dispersive isotopic medium (Bjorklund 1975). However, Kung has shown that by focusing a laser pulse close to the orifice of a gas jet, frequency tripling can take place for both positively and negatively dispersive media (Kung 1983). This is due to the fact that the strong focusing condition is no longer valid in a gas jet experiment. For example, in the present experiment, the confocal parameter is 4 mm, while the width of the gas jet at the focal point is estimated to be 1 mm. Odd-order harmonics observed in the present experiment can thus be explained.

Questions are now being asked about why very high odd-order harmonics are generated and why the harmonic signal strength falls very slowly for harmonic orders higher than the fifth. It should be pointed out that harmonics are only generated in a laser intensity range very close to that required for the multiphoton ionisation of the medium. As an example, the laser used in the present experiment delivers a maximum focused intensity of 10^{14} W cm^{-2}. At this intensity, no harmonic generation is observed in Ne, whereas multiphoton ionisation of Ne at 1064 nm requires a laser intensity of 3×10^{14} W cm^{-2} (L'Huillier *et al* 1983). To gain a better understanding of these processes, it would be useful to study the generation of high-order harmonics as a function of the multiphoton ionisation of the medium, and to determine whether structures induced in the ionisation continuum via multiphoton ionisation enhance

harmonic generation (Shore and Knight 1987). These kinds of experiments will be carried out in our laboratory in the near future.

The directional emission of harmonics along the laser axis over a very small angle is one of the most salient findings. As a consequence, multiple harmonic conversion of pulsed lasers in rare gases gives XUV radiation of substantially higher spectral brightness per pulse than other radiation sources in this spectral region. Therefore, such an XUV generator, which has the convenience of being a small laboratory-scale experiment source, could be efficiently used for the general purpose of XUV spectroscopy.

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