BNL-65161

TWO-PHOTON IONIZATION AND THREE-PHOTON ABOVE-THRESHOLD IONIZATION OF ARGON

A. Bouhal[1], G. Hamoniaux[1], A. Mysyrowicz[1], A. Antonetti[1],
P. Breger[2], P. Agostini[2],
R. C. Constantinescu[3], H. G. Muller[3]
L. F. DiMauro[4]
[1]Laboratoire d'Optique Appliquée, ENSTA-Ecole-Polytechnique,
91125 Palaiseau, France
[2]Service des Photons, Atomes et Molécules
DSM-DRECAM CEA Saclay, 91191 Gif-sur-Yvette France
[3]FOM-Institute for Atomic and Molecular Physics Kruislaan 407, 1098 SJ
Amsterdam, The Netherlands
[4]Brookhaven National Laboratory, Chemistry Department
Upton 11973 NY

INTRODUCTION

Studies of nonlinear laser-matter interaction have been so far limited to wavelengths from the near Ultraviolet to Infrared, because of the low brightness of currently available sources outside this range. However nonlinear processes in the VUV/Soft Xray domain would initiate multiphoton innershell spectroscopy, XUV nonlinear optics and applications of such processes to metrology (for instance, autocorrelation measurements of ultrashort XUV pulses).

The probability of multiphoton transitions decreases rapidly with the number of photons involved. A typical two-photon bound-free transition has a rate of the order of .1 ps^{-1} at 10^{12} $W.cm^{-2}$ and scales as the square of the intensity. Such an intensity is therefore required to saturate a two-photon transition with a 100 fs pulse. This is difficult to achieve with the present state-of-the-art techniques for producing intense XUV pulses. To our knowledge, only two cases of such transitions have been reported so far. The first one is a two-photon ionization of argon by the third harmonic of a KrF laser[1]. Since the photon energy (15 eV) is just below the ionization energy (15.75 eV) for argon, the transition is quasi-resonant. The second case is a two-photon ionization of helium by the 9th harmonic of a Ti:S laser[2]. In the latter case an autocorrelation measurement of the harmonic pulse has been reported. In the present work, we report on two-photon ionization of argon at 133 nm (9.3 eV) from the third harmonic of a frequency doubled Ti:S laser, and a three-photon above-threshold ionization involving two 9.3 eV photons and one 3.1 eV photon.

SETUP

The output of a CPA Ti:Sapphire laser (800nm, 150fs, 30 mJ, 10 Hz) is frequency-doubled and focused to a 50 micron diameter spot by a 1 m focal length lens into a pulsed xenon jet whose backing pressure is 0.5 bar. The unconverted 800 nm radiation is absorbed in a BG38 color filter. The nearly flat-top spatial mode of the 400 nm light is converted into an annular profile by a circular disk blocking the center of the beam and an iris limiting the outer diameter. The disk is imaged on a pinhole (located at 90 cm downstream from the jet) by the lens. The pinhole stops the 400 nm beam while transmitting the harmonics produced in the jet. The efficiency of this simple filtering device is not perfect though: the forward scattering in the gas, the beam refraction caused by the free-electron density resulting from ionizing the jet and the diffraction of the disk cause leakage of fundamental radiation into the spectrometer. The amount of residual 400 nm light in the beam after the pinhole is estimated to be of the same order as that of a typical harmonic. The amount of 800 nm is completely negligible due to the combined transmission of the BG38 filter and the disk-pinhole assembly. This gives a comparable efficiency to other type of filtering schemes but with the advantage that the harmonic pulse is not affected by the filtering process: in particular, no pulse broadening results from the use of a diffraction grating or similar dispersive devices.

In general, multilayer coated spherical mirrors provide both frequency selectivity and focusing. However in the present experiment, we use a broad-band coated mirror with a reflectivity of about 30% for the low-order (3^{rd}-7^{th}) harmonics (133-57 nm) (see Fig.1). The focal length of the mirror is 35 mm and the focus position is externally adjustable.

The argon target gas is injected into interaction region of the spectrometer through a pulsed valve backed by a 0.5 bar pressure.

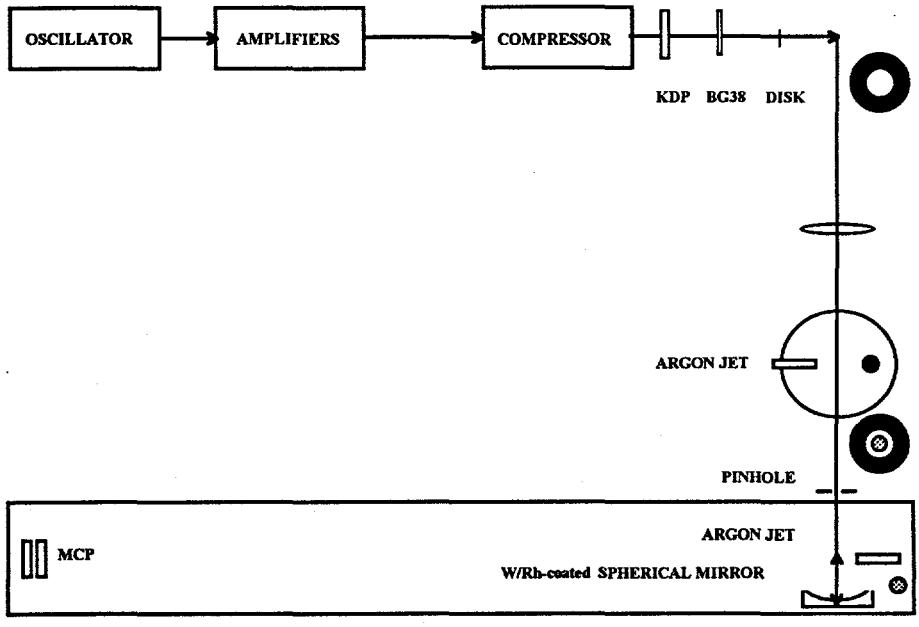

Fig. 1: Experimental setup. The beam cross-sections of the 400 nm (black) and harmonic (gray) light are shown at various positions.

DISCLAIMER

RESULTS

Figure 2 shows a typical electron energy spectrum of argon. Only two peaks are evident: a peak near 3 eV energy and a weaker peak at 6 eV. The first peak corresponds to a total energy of six 400 nm photons. It can therefore be assigned either to the absorption of a fifth-harmonic photon (80 nm) and one 400 nm photon or two third-harmonic photons. The total energy of the second peak corresponds to the absorption of seven 400 nm photons. It can be interpreted in three ways: (1) one-photon ionization from the seventh-harmonic, (2) one fifth-harmonic plus two 400 nm photons and (3) two third-harmonic plus one 400 nm photons.

Additional experimental information is shown in Fig. 3 as a variation of the two peak amplitudes as a function of the mirror focus position inside the spectrometer. Both peaks are strongly position-dependent, i.e. on the intensity of the radiation producing them, whatever wavelengths are involved in the ionization process.

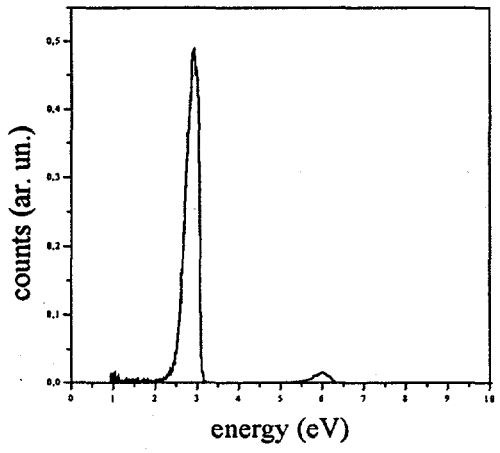

Fig. 2: Electron energy spectrum from ionization of argon by a combination of 133 nm, 80 nm, 57 nm and 400 nm photons (see discussion).

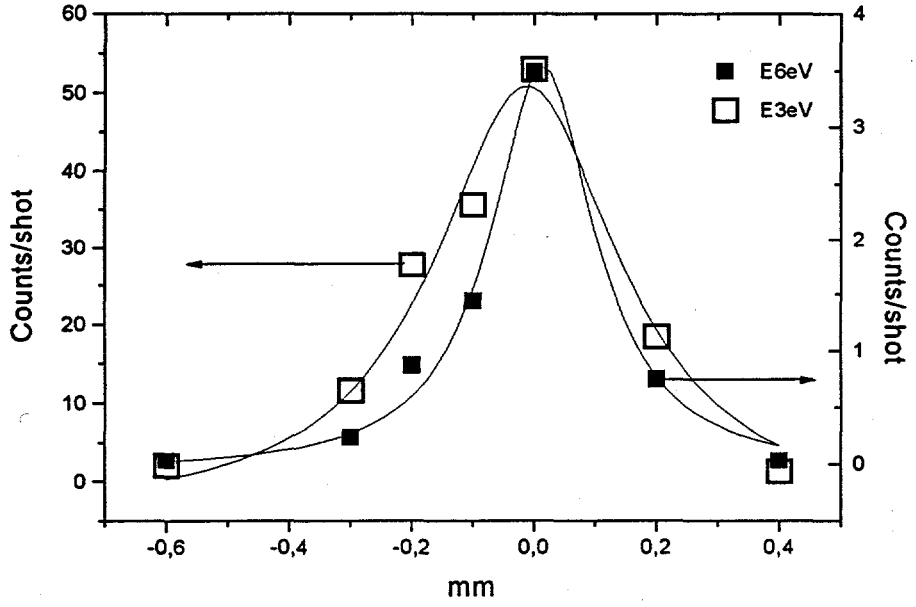

Fig. 3: Amplitudes of the two peaks in Fig. 2 versus the focus position inside the sensitivity zone of the spectrometer (squares). The solid lines are lorentzian fits to the data.

DISCUSSION

One difficulty in assigning the electron peaks is the lack of direct information on the respective intensities at 400, 133, 80 and 57 nm (neglecting the remaining 800 nm). For the 400 nm radiation, to weak to be measured directly, we rely on the estimate discussed above yielding an intensity of 5×10^{13} W cm^{-2}. The number of photons at the third harmonic (133 nm) deduced from known third order susceptibilities[3] at 1054 nm (6.46×10^{-26} m/V^2) should be of the order of 10^{11} per pulse, assuming a fundamental intensity of 5×10^{14} W cm^{-2} and an atomic density of 10^{16} cm^{-3} in the harmonic jet. This value is supported by direct measurements[4] which also provide the relative conversion efficiencies for harmonics 3, 5 and 7 at three fundamental wavelengths (1055, 616 and 308 nm). Interpolating these results to 400 nm yields a ratio of the intensity at 133 nm to that at 80 nm of 10^3. The intensity at 57 nm should not exceed 10^{-6} of that at 133 nm from the same data. Using a mirror reflectivity of 30% and a pulse duration determined by perturbative scaling $\tau/\sqrt{2\sqrt{q}}$ (where τ is the pulse duration at 800 nm and q is the harmonic order), we estimate the intensities at 400, 133, 80 and 57 nm as 2×10^{13}, 10^{13}, 10^{10} and 10^7 W cm^{-2}, respectively. It follows that the two-photon ionization rate at 133 nm is more than 100 times larger than the rate corresponding to the absorption of one 80 nm and one 400 nm photon, assuming (in the absence of known values) the two generalized cross-sections equal. The same argument rules out higher-order processes, involving more than one 400 nm photon. We conclude that the 3eV peak is mostly due to the two-photon ionization at 133 nm with a very weak contribution from the (80 + 400) nm transition.

The stronger dependence of the 6 eV peak on the focus position rules out a direct, one-photon ionization from the seventh harmonic, which would be linear in intensity and therefore independent upon focusing, consistent with the above calculation. The most likely mechanism for this peak is an ATI transition involving two-133 nm photon + one excess-photon at 400 nm.

Since the signal-dependence on the focal position reflects essentially the 300 μm size of the spectrometer sensitivity zone, the intensity dependence of the electron count cannot be deduced from the data in Fig. 3. Let us just stress again that the observed FWHM is consistent with a diffraction-limited focusing of the 133 nm radiation and a peak intensity at 133 nm of about 10^{13} W.cm^{-2}.

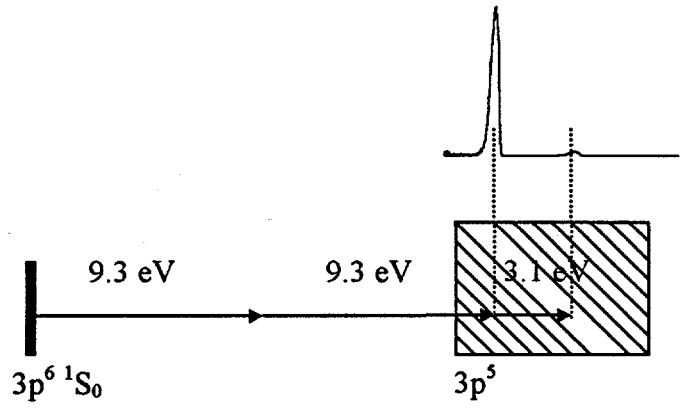

Fig. 4: Multiphoton transitions observed in argon.

In summary, the electron energy spectra produced by focusing low-order harmonics of 400 nm radiation in argon are consistent with the two and three-photon ionization transitions illustrated in Fig. 4. This result is one of the first multiphoton transitions reported in the VUV range and the first one involving an ATI transition. The peak intensity at the

higher harmonics is still too low, in the present state-of-the-art, to permit observation of nonlinear processes in the XUV domain. Both the conversion efficiencies and multilayer mirrors performance must be improved to obtain the intensity necessary to observe more interesting multiphoton innershell transitions. Nevertheless, it is already possible to perform autocorrelation measurements on large two-photon signals (like the 3 eV electron peak) as shown by Kobayashi e al[2].

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ACKNOWLEDGEMENT. This research was carried out at Brookhaven National Laboratory under contract DE-AC02-76CH00016 with the U.S. Department of Energy and supported by its Division of Chemical Sciences, office of Basic Energy Sciences.