Resonantly enhanced multiphoton ionization of xenon in Bessel beams

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Resonantly enhanced multiphoton ionization of xenon near the three-photon $6s$ resonance has been studied under the excitation by the Bessel beams. It has been shown that the ionization spectra are significantly different in the cases of the Bessel and the Gaussian laser beams. This difference is caused by an internal third-harmonic field, which in the Bessel beam is generated in a wide spectral range due to the self-phase-matching mechanism. The cancellation of the $6s$ resonance in the Bessel beam has been demonstrated. Similar to the case of the Gaussian beams the canceled resonance appears in the spectra under the excitation by counterpropagating Bessel beams. The origin of some features in the ionization spectra is discussed.

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The output emission of lasers is usually transported in a form of Gaussian beams and by focusing these beams a very high light intensity can be obtained. For a tightly focused Gaussian beam, however, the beam size experiences large changes over a distance of propagation and the high-intensity region near the beam waist is rather small. Recently a new type of so-called diffraction-free beams has been of great interest. Diffraction-free beams give a high concentration of light energy into a small central spot, but the spot size remains nearly unchanged over a distance of propagation, which can be much longer than the Rayleigh length of a focused Gaussian beam. It allows one to produce very long and uniform excitation regions with high light intensity.

The amplitude profile of the simplest diffraction-free beam is given by the zero-order Bessel function of the first kind $J_0(r)$, thus these beams are termed as Bessel beams. The Bessel beams are sharply peaked near the optical axis without spreading over a long distance of propagation. Such beams can be produced, for example, when a parallel laser beam is focused by a conical lens (axicon). An axicon lens transforms a laser beam into a cone of plane waves behind the lens, where the interference pattern is the desired Bessel beam. Since 1970 this simple and effective method is used in experiments with extended excitation regions [1–4].

Both in a focused Gaussian and in a Bessel beam a high light intensity can be achieved. There is, however, a principal difference between these two kinds of beams. The Bessel beam can be viewed as a superposition of infinitely many plane waves whose wave vectors lie on a cone around the propagation axis. Thus, for any nonlinear process in the central lobe of the Bessel beam the interaction of individual waves, in general, is noncollinear in contrast to the focal region of a Gaussian beam, where a collinear interaction takes place. This principal difference results in a number of unusual effects observed with Bessel beams [4,5]. The resonantly enhanced multiphoton ionization (REMPI) of gases in noncollinear laser beams is known to be far more complicated and rich of effects [6–8] than the collinear case of plane waves or focused Gaussian beams. In conical beams like the Bessel ones the self-phase-matching mechanism [9,10] provides an efficient harmonic generation. Thus, the effects of an internally generated harmonic field in REMPI of gases for the Bessel beams can be pronounced in an essential different manner than in the case of the Gaussian beams. In the present work, we want to demonstrate some of these effects in the case of REMPI near the three-photon $6s$ resonance of xenon.

The experimental setup was described previously [11]. The REMPI of xenon both in the Bessel and the Gaussian beams was studied by the method of a selective laser-induced breakdown [11,12]. The breakdown of xenon in 1–6 bar (0.1–0.6 MPa) pressure range was initiated by a tunable dye laser pumped by an excimer XeCl laser. In the present experiments the dye laser was operated with the Coumarin-120 dye and the energy of the laser pulses was up to 4 mJ, pulse duration of about 15 ns, and the spectral width of 0.01 nm (FWHM). To produce a Bessel beam the laser output was focused into the gas cell by a quartz axicon with the cone angle of 120°. The axicon was mounted as one of the windows of the gas cell. Other four plain-parallel quartz windows of the cell allowed one to focus the laser beam into the target gas by spherical lenses and to detect the emission of laser-produced plasma.

Because of energy limitation of the used laser the experiments were carried out with relatively short Bessel beams. A stable laser spark with a length of about 1 mm was produced when the laser beam had the diameter of 2–3 mm on the axicon. The refracted beam had the cone half-angle of 17° at 440 nm and in the breakdown region the transverse intensity distribution had the form of $J_0(r)$ with the diameter of the central spot of about 10 μm. The maximum light intensity in the Bessel beam was about 5×10^{10} W/cm^2. The measurements in the focused Gaussian beams were carried out by using a spherical lens with $f=50$ mm. In this case the focused beam had the confocal parameter of about 1 mm and the length of the excitation region was nearly the same as the region of maximum light intensity in the Bessel beam.

Figure 1 shows the breakdown excitation spectra near the three-photon $6s$ resonance of xenon measured with the Bessel [Figs. 1(a) and 1(b)] and the Gaussian [Fig. 1(c)] laser beams. For the Gaussian beams the REMPI of xenon in this region is well studied in a broad range of excitation conditions [11–16]. The intense peaks in the spectrum result from the (4+1)-photon ionization via the excited $4f$ and $6p'$ states of xenon. In a dense gas the peaks have broad wings due to ionization via the excited molecular states of xenon

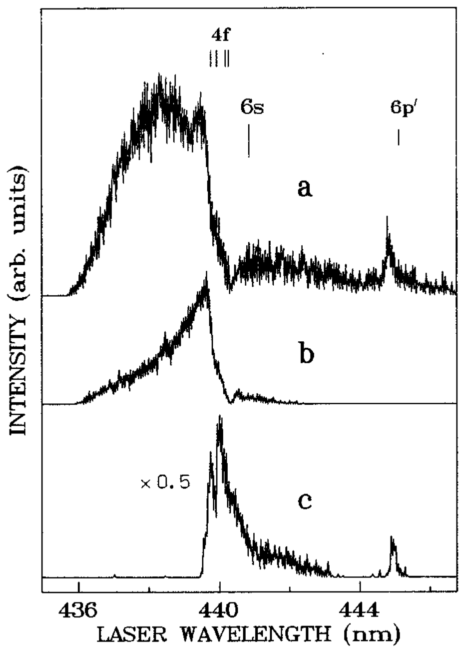

FIG. 1. Ionization profiles measured near the three-photon $6s$ resonance of xenon: **a,b,** Bessel beams at two different adjustments of the axicon (pulse energy, 2.4 mJ); **c**, Gaussian beam (pulse energy, 0.7 mJ). Xenon pressure, 3 bar.

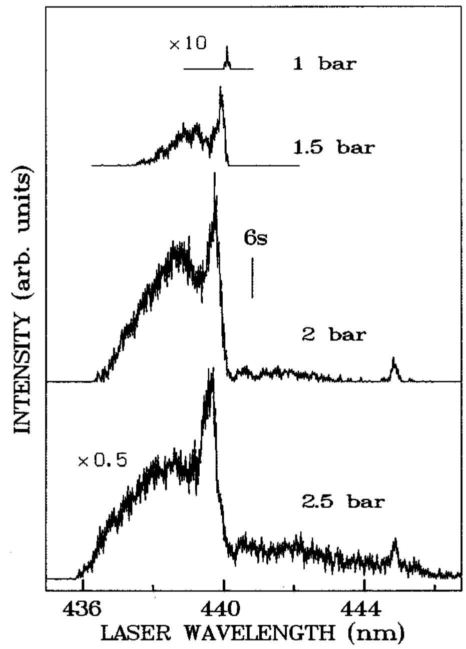

FIG. 2. Ionization profiles measured with the Bessel beam at different xenon pressures. Pulse energy, 2.6 mJ.

dimers. The $6s$ resonance in the spectrum is canceled as a result of a destructive interference between the one- and three-photon excitation pathways, where the one-photon process is due to the third-harmonic (TH) field [16,17].

In experiments with the axicon the quality of the produced Bessel beam was very sensitive to the quality of the initial laser beam and to the adjustment of the axicon with respect to this beam. Even small variations of these parameters changed the focusing geometry and transformed the ionization spectra. The general structure of the ionization spectra, however, remained very similar in all sets of experiments and all the spectra obtained with the Bessel beams differed significantly from those obtained with the Gaussian beams. Figure 1 shows two examples of the excitation profiles measured with two independent adjustments of the dye laser and of the axicon. The main feature of these spectra is a broadband at the high-energy side of the $6s$ resonance. At the position of the $4f$ resonances instead of the atomic peaks a more complicated structure is registered, which can have a form of a dip [Fig. 1(b)] or a form of an interference structure with a minimum and a maximum [Fig. 1(a)]. Such structures were clearly seen in all the measured spectra for the whole range of xenon pressure up to 6 bar.

Figure 2 shows the ionization profiles measured with the Bessel beam at different xenon pressures. In these measurements the axicon was adjusted to get a maximum intensity of the peak on the band. Again, a broad ionization band dominates the spectra and an interference structure is registered on this band. The dip of this structure is located near the $4f$ resonances and its position is low-sensitive to the gas pressure. In contrast, with an elevated gas pressure the peak experiences a remarkable blueshift, which is approximately linear (0.26 nm/bar) with the pressure. Being extrapolated to the zero-pressure point, however, the positions of both the dip and the peak coincide near the position of the $4f$ resonances.

The origin of the broad ionization band in the spectra can be understood by taking into account the generation of a TH field in the Bessel beam. Similar ionization band caused by the TH field is well-known from the experiments with Gaussian beams [13,14]. In the Gaussian beams, however, the band near the resonance was observed at a much lower gas density. With an elevated pressure the band moves off the resonance because of the phase-matching requirements. In contrast, for a Bessel beam the self-phase-matching mechanism [9,10] provides automatically the phase matching in a negatively dispersive medium and an efficient generation of the TH field is possible for a wide range of refractive index variations.

The spectral region available for the TH generation in the Bessel beam can be estimated in the following way. The phase-matching condition $\Delta\mathbf{k}=\mathbf{0}$ along the propagation axis of a conical beam has the form [9]

$$3k(\omega)\cos\alpha = k(3\omega)\cos\beta, \tag{1}$$

where α and β are the cone half-angles of the fundamental Bessel beam and of the TH field, respectively. In our case $\alpha = 17°$ and the condition (1) is fulfilled for $n(\omega) \geqslant n(3\omega) \geqslant 0.956 n(\omega)$, where $n(\omega)$ and $n(3\omega)$ are the refractive indices for the pump frequency and the TH frequency, respectively. Within this region different combinations of the wave-vectors of individual waves allow one to fulfill the phase-matching condition (1) and to generate the TH photons. The TH light is cone-shaped with the cone half-angle ranging from 0 to α depending on the laser wavelength. Simple calculations of $n(\omega)$ and $n(3\omega)$ by using the Sellmeir formula have shown that in the case here the TH field at 2 bar of xenon can be generated within the spectral range from 440.4 nm to about 420 nm. Moreover, the width of this region is rather low-sensitive to the gas pressure. At the short-wavelength edge of the region $\beta=\alpha$ (collinear case) and the phase-matching condition has the form of that for the plane waves: $n(3\omega)=n(\omega)$. The intensity of the TH near this edge is expected to be low. At the long-wavelength edge $\beta=0$ and the TH light is peaked along the propagation axis of the Bessel beam. From the transverse phase-matching conditions [10] the generation of the TH field is expected to be most effective near the long-wavelength edge and it is just the region where the broad ionization band is registered in the spectra. Near the $6s$ resonance the TH light produces the ionization due to the one-photon absorption on the wing of pressure-broadened atomic line and on the molecular terms of xenon dimers. Both of these absorption processes populate the $6s$ state, which is ionized by further absorption of two laser photons.

For the axicons with other refraction angles the spectral region available for the TH generation, in general, will be changed. At a fixed gas pressure, however, the short-wavelength edge is independent on the used axicon. The position of the long-wavelength edge depends on the cone angle of the Bessel beam. If the cone angle of the used axicon is larger (i.e., the refraction angle is smaller) the long-wavelength edge will be moved toward the high-energy side of the spectrum. The region available for the TH generation in this case will be narrowed. An axicon with a larger refraction angle than that in our case will give a shift of the edge to the opposite direction. This shift, however, will be very small as toward the atomic line $n(3\omega)$ rapidly changes with the wavelength.

An explanation of the origin of an interference structure on the band is more difficult. This structure arises when the TH band matches the position of the four-photon resonances. Similar situation in the case of Gaussian beams leads to the appearance of either a dip or a peak on the ionization band [13]. In the Bessel beams, however, both a dip and a peak are registered simultaneously near the $4f$ resonances. The dip is slightly shifted to the red from the $4f$ resonances and its position is low-sensitive to the gas pressure. The peak, however, undergoes a significant shift with the gas pressure. At a relatively low pressure the peak matches the position of the $4f$ resonances, but with an increased pressure the peak shifts to the blue side off the resonances. Tentatively, the appearance of such spectral features can be caused by an interference of even-photon excitation processes. In the presence of a TH field there are two pathways to excite the $4f$ states. The first one is the absorption of four laser photons and the second one is the absorption of a TH photon plus one laser photon. These two excitation pathways can interfere and the corresponding structure arises in the ionization profile. Similar but less pronounced interference structures near the $4f$ resonances were predicted for a two-color ionization of xenon [18]. Note again that in the Bessel beams a noncollinear interaction of both the TH and the fundamental fields takes place and it can result in some unusual features in the ionization spectra.

Under the excitation by the Bessel beams the three-photon $6s$ resonance in spectra is absent. Similar cancellation effect is well-known for the Gaussian beams where it is caused by a destructive interference between one- and three-photon excitation pathways under on-resonance excitation. Obviously, similar effect is responsible for the cancellation of the $6s$ resonance in the Bessel beams. A common method to observe the canceled resonances is based on the use of counterpropagating laser beams. In this case the destructive interference is incomplete and the corresponding resonance appears in the spectrum. To produce a counterpropagating excitation geometry with the Bessel beams a pair of identical axicons was used. An additional axicon was mounted inside the gas cell opposite to the first one (see the inset in Fig. 3). The flat surface of the second axicon was coated by aluminum to produce a return mirror. In this arrangement the Bessel beam after the first axicon was retroreflected on itself by the second one.

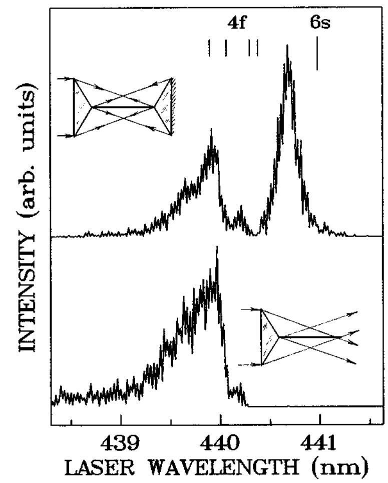

FIG. 3. Cancellation of the $6s$ resonance of xenon in the Bessel beams. Upper trace, counterpropagating Bessel beams; lower trace, unidirectional Bessel beam. Xenon pressure, 2.5 bar; pulse energy, 2 mJ. The insets show the excitation geometries.

Figure 3 shows the ionization profiles measured in unidirectional and counterpropagating Bessel beams. As it could be expected, under the excitation by counterpropagating beams an intense $6s$ resonance appears in the spectrum. This resonance is blueshifted from the unperturbed position due to the ac Stark effect and the pressure-induced shift. In the breakdown experiments the ac Stark shift of the resonances provides a probe of light intensity in the focal region [11]. In the case here for retroreflected excitation geometry the light intensity of 9×10^{10} W/cm^2 has been determined from the ac Stark shift of the $6s$ resonance.

The ionization band to the blue from $6s$ in counterpropagating beams is suppressed despite the increased light intensity. This suppression results from the weakened TH field under such an excitation. In the case of a complete overlap of the countepropagating beams a standing wave is formed, where no TH field can be generated. In our case the spatial and time overlap of both Bessel beams is incomplete, therefore a partial suppression of the TH field takes place.

In summary, we have demonstrated that the REMPI spectra of xenon undergo significant transformations when the Bessel laser beams are used instead of the Gaussian ones. These transformations result from the principal difference of these kinds of beams, where either a collinear (Gaussian beam) or a noncollinear (Bessel beam) interaction of individual waves takes place. Due to the self-phase-matching mechanism the harmonic field near the atomic line in a Bessel beam can be generated in a much broader spectral range than in the case of a Gaussian beam. The harmonic photons produce additional excitation pathways and transform the ionization spectra. For the three-photon excitation of the atomic resonance the effect of an internally-generated TH field in both types of beams is pronounced in the same manner. In a unidirectional excitation geometry the resonance is canceled both in a Bessel and a Gaussian beam. In counterpropagating Bessel beams the resonance appears in the spectra similar to the counterpropagating Gaussian beams.

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