Near-resonance three-photon excitation profiles in xenon with segmented conical beams

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Multiphoton ionization profiles in xenon under three-photon excitation by azimuthally segmented conical laser beams have been studied and analyzed on the basis of the interference-based cooperative shift of the atomic resonance line. Segmentation of the conical wave front with various geometrical slits reduces the possible three-photon azimuthal sub-beam combinations in the third order process and a principal (or ''homogeneous'') Lorentzian component can be selected from the overall excitation profile. This Lorentzian is identical to the pressure-broadened atomic line shifted by the cooperative shift. A possibility of controlling the nonlinear response by spatial selection of interacting waves in conical beams is discussed.

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Multiphoton excitations of gas-phase atomic and molecular transitions are old and well studied subjects. A novel subset of the subject, also well studied, is that of linearly polarized three-photon pumping of optically allowed resonant transitions. The novelty is that at elevated concentrations a resonant excitation profile may become completely suppressed [1–3] or be shifted in wavelength (with identical shape and amplitude) depending on the geometry of the pump beam(s) used in a given multiphoton excitation scheme [4–7]. The atypical behavior of three-photon excitation derives from internally generated four-wave-mixing fields that act coherently with the imposed laser field(s) to produce the observed excitation profiles. It is the coherent sum of the two excitation pathways that produces a given profile. The excitation process becomes beam-geometry-dependent because the wave-mixing process is influenced by beam configuration(s).

In a recent paper, the unique beam geometries produced by focusing with axicon lenses were used to study three-photon-resonant-enhanced multiphoton ionization in xenon [8]. In this instance, the laser field can be described as a zero-order Bessel beam. The field distribution in a Bessel beam results from a superposition of infinitely many plane waves all inclined by a constant angle α toward the propagation axis. In the context of nonlinear optics, the Bessel beams produce noncollinear interactions of the driving fields and the angle α serves as an additional tunable parameter [9]. But in addition to the inclination angle α, which is the same for all components of a conical beam, there is also the azimuthal angle of a particular fundamental wave on the light cone. In a three-photon-induced process, the azimuthal angles of each of three interacting fundamental waves determine a given component of the nonlinear polarization of the medium and, in the present instance, the resultant third-harmonic (TH) field. A complete analysis of the three-photon-resonant ionization profiles for Bessel beams would require that all possible spatial combinations of interacting waves be taken into account in the third-order process, including the simultaneous treatment of direct three-photon excitation by the fundamental beam components and production of and excitation produced by the TH field. (TH production has been treated separately in [10,11].) A consistent treatment produces the well known wave-mixing interference effect associated with resonant three-photon linearly polarized pumping of optically allowed transitions.

Excitation profiles for three-photon pumping of the 6*s* level in Xe are quite different for Bessel as compared to Gaussian beams [8]. Though less tractable analytically, the profiles from Bessel beams were shown to be principally describable from a simple picture of plane waves, crossed at angle 2α, together with the characteristic feature of the cooperative line shifting associated with noncollinear three-photon excitation [5,7,12]. It was shown in Ref. [8] that at any gas pressure the location of the pressure-dependent, sharply-peaked ionization profiles produced by a Bessel beam with inclination angle α matches exactly the value of the cooperative shift for *two* plane waves crossing with crossing angle 2α. The pressure-dependent peaks of the strong ionization profiles registered with the use of Bessel beams correspond to the peak of the atomic line shifted by the cooperative shift [7,12].

In the present paper, we further demonstrate the underlying basis for resonant and near-resonant profiles from Bessel beams, and suggest possible implementation of the demonstrated features. Here we decompose a Bessel beam into various subbeams by segmenting the conical light front with different slit masks. By this technique, we select particular azimuthal components of a Bessel beam to produce selected spatial configurations of the fields used to drive near-resonant three-photon excitation in Xe. This procedure very nicely illustrates how the overall near-resonant three-photon excitation profiles of an optical transition are generated in Bessel beams. Again, the concept of the cooperative line shift is used for the analysis of the excitation profiles and its components in full-aperture and segmented conical beams. The present results give more direct insight into the complicated analytical problem associated with three-photon processes produced by axicon focusing.

The experimental arrangement in the present study was similar to that used in previous experiments with Bessel beams [8,13]. Briefly, the Bessel beam having inclination angle $\alpha = 17°$ was produced with the aid of a quartz axicon. The beam of a pulsed tunable dye laser was focused by the axicon into a gas cell filled with xenon. The laser was tuned in a spectral region near the three-photon 6*s* resonance of xenon. Resonantly enhanced ionization profiles were moni-

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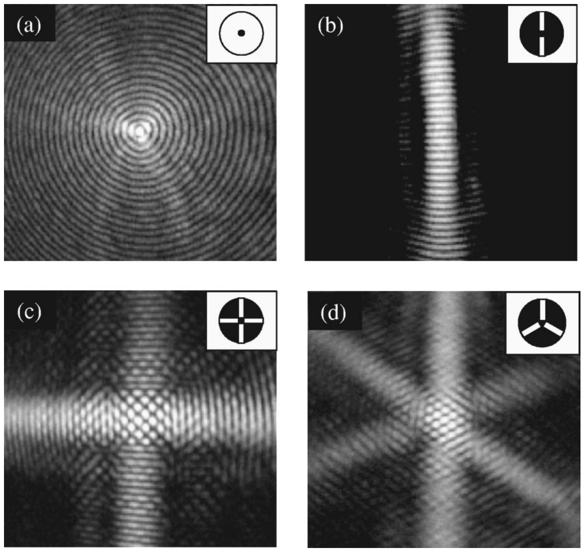

FIG. 1. CCD pictures of the focal regions: (a) full-aperture conical beam (Bessel beam); (b)–(d) segmented conical beams. The insets show the used slit masks.

tored, where the ionization signals were due primarily to generation and near-resonant absorption of TH photons in the target gas with subsequent ionization of excited atoms by laser light.

In the present study, we controlled the geometry of the driving field within the excitation volume through the azimuthal selection of interacting waves. Different slit masks were placed close to the entrance surface of the axicon (see masks depicted in the corner of each separate component of Fig. 1). The width of the slits was 0.6–0.8 mm. Selected radial slices of the laser beam were focused by the axicon forming segmented conical beams. As in the case of a full-aperture conical beam, all the components of the segmented beam were inclined by an angle α toward the propagation axis and crossed along this axis, but the composition as a function of azimuthal angle was controlled. As we see below, this provides a direct demonstration of how the line shapes are produced in near-resonant three-photon pumping with Bessel beams.

Figure 1 shows the focal regions of different conical beams used in the present experiments (in all cases the central part of the axicon was blocked by a 1–1.5-mm obstacle). The full-aperture laser beam focused by the axicon produced the known pattern of a zero-order Bessel beam. For segmented beams, this interference pattern was reduced to a pattern of a few crossed plane waves selected by the masks as shown in Fig. 1. In the description of a wave-mixing process from a full-aperture Bessel beam, all combinations of particular waves having any azimuthal angle ϕ from 0° to 360° on the light cone are superimposed. For our segmented beams, large parts of the field components are blocked and the input source is comprised of subbeams from a limited range of available azimuthal angles. Thus, for a segmented beam only some particular combinations of sources from the initial light cone remain valid, while others disappear. Note that the finite width of mask slits and diffraction on mask edges give some spreading in angles ϕ_i. This spreading is maximum for the inner edges of the slits and in our case it was up to $\pm 20°$. The influence of this spreading on measured ionization profiles will be considered below.

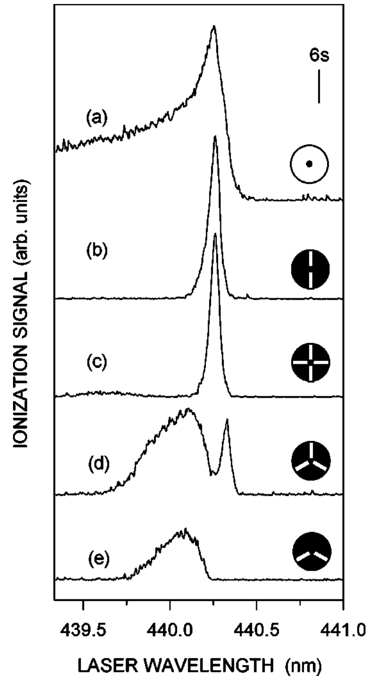

FIG. 2. TH excitation profiles: (a) full-aperture Bessel beam (laser pulse energy 0.5 mJ); (b) linear slit geometry (0.4 mJ); (c) two slits 90° (0.4 mJ); (d) three slits 120° (0.9 mJ); (e) two slits 120° (0.6 mJ). Xenon pressure 2 bar.

Results of experiments with segmented conical beams are summarized in Fig. 2. The upper trace shows results from the full Bessel beam (no mask). The shape of this profile is known from earlier studies [8,13]. For moderate values of gas pressure, the observed profiles have well-pronounced sharp maxima followed by a tail toward the blue side. The position of the maximum shifts linearly to shorter wavelength with increased gas pressure. As was shown in [8], the shift of this maximum from the three-photon 6*s* resonance matches exactly the value of the cooperative shift δ_c for two plane waves angled by 2α [7,12]. That is, the main contribution to the profile (the Lorentzian maximum) comes from excitation produced by planar two-beam combinations of fundamental waves entering the excitation zone from opposite sides of the axicon. This yields a Lorentzian profile at the predicted frequency for two beams crossing at angle 2α. This is essentially at the frequency where phase-matched TH production for two crossed beams occurs (see below). All other two- and three-beam combinations of interacting waves also contribute the excitation by the coherent sum of the fundamental and the generated TH field at frequencies off the main peak, and with much smaller volume over which generation occurs (smaller gain length). As a result, the TH profile gains a long tail on the blue side and some, though much smaller, broadening on the red side of the Lorentzian [8]. Note that no ionization peak is present at the 6*s* resonance (marked in Fig. 2). This cancellation results from the well-known wave-mixing interference between the one- and the three-photon excitation processes [1–3,14].

The other four traces in Fig. 2 show results with the geometrical masks indicated in the figure. We note that for a linear-slit mask [Fig. 2(b)] the ionization profile is reduced to a single Lorentzian-like peak at the position of the TH maximum in the initial unmasked Bessel beam. For other masks allowing more than one spatial combination of subbeams, additional spectral components can be recognized in the TH profiles. For two crossed slits [Fig. 2(c)], the same Lorentzian peak was registered together with an additional weak band on the short-wavelength side of the main peak. For the mask with three slits separated by 120° [Fig. 2(d)], the TH profile again has two components—a sharp Lorentzian peak and a blue band. The Lorentzian peak is located closer to the 6*s* resonance than the peak for one slit or for two slits at right angles. When any one of the three 120° slits was blocked, the Lorentzian peak disappeared and only the broad blue band remained in the spectrum [Fig. 2(e)].

Now we ask what can be learned from these results. Note first that a description of TH generation in conical beams requires consideration of two geometries of interacting subbeams. The first is the general case of photons from each of three interacting fundamental plane waves having different azimuthal angles ϕ_1, ϕ_2, and ϕ_3. The second is the two-beam case, where two fundamental photons come with the same azimuthal angles $\phi_1=\phi_2$ and the third photon comes from another part of the light cone. The collinear (single-beam) case $\phi_1=\phi_2=\phi_3$ is excluded as no resonant three-photon excitation and no phase-matched TH generation is produced by a single plane wave.

In our analysis of the data, we start with consideration of the two-beam geometry. This case is identical to the interaction of two crossed unfocused plane waves. It is known [7,12] that for two angled plane waves the excitation profile has the shape of a Lorentzian line shifted to the blue side of the expected atomic line position. The shift of this atomic resonance line profile is given by the analytic expression for the interference-based cooperative shift, and the Lorentzian shape of the profile is identical to the unshifted pressure-broadened atomic line [7,12]. Except for a small contribution due to collisional energy transfer, the position of the shifted peak coincides with the point where the real part of the phase mismatch, $\mathrm{Re}(\Delta k)$, between the driving field and generated TH field goes to zero [15]. For beams at crossing angles that are less than 100°–120°, the collisional term can be neglected, so the shifted peak appears essentially at the frequency where $\mathrm{Re}(\Delta k)=0$. (Note that this does not imply that $\Delta k=0$ since the propagation vector of the TH field may have a significant imaginary component.) Thus in the present context we simply refer to the strong Lorentzian peak, displaced from resonance by the cooperative shift, as being produced by phase-matched TH generation and subsequent absorption. For the full-aperture conical beam there is an infinite number of pairs of interacting waves having different crossing angles and, thus, different values of the cooperative shift. This leads to a large spreading of individual Lorentzians in the full Bessel-beam spectrum. The overall TH envelope for a Bessel beam builds up as a superposition of such Lorentzians [8]. This superposition of shifted lines has an interesting formal similarity with inhomogeneous broadening of spectral response in a system where individual homogeneous components get spectral spreading (e.g., due to Doppler effect) and superposition of these components produces the overall inhomogeneous spectral profile. With this analogy, the whole nonresonant-blue-side profile for a Bessel beam can be viewed as the result of ''beam-geometry broadening'' of the cooperative line, analogous to inhomogeneous broadening of an atomic line profile. This broadening arises due to an infinite number of possible combinations of interacting waves in Bessel beams, when every particular combination gives its own ''homogeneous'' contribution to the resulting excitation profile. Alternatively, the broadened profile can justifiably be ascribed to phase matched TH production in the negatively dispersive region on the blue side of the 6*s* resonance, since a detailed treatment reveals that this signal is completely dominated by the TH contribution to the coherent sum of transition amplitudes from the fundamental and TH fields [15].

Now consider segmentation of the input beam, which reduces the number of available combinations of interacting waves. This results in easily predictable changes in the Bessel-beam profile, as some combinations of the fundamental waves are blocked and the corresponding parts of the TH profile disappear. Again, this situation is in some sense analogous to hole burning in an inhomogeneous spectral profile. Here one can extract a selected ''homogeneous'' component from the profile of superimposed Lorentzians. But, because of axial symmetry of a full-aperture conical beam, it is impossible to form a ''homogeneous dip'' in the TH profile as none of the masks is able to block all spatial configurations of a given geometry. Nevertheless, for conical beams it is possible to separate some single configurations while blocking all others. In this case, the overall ionization profile should be reduced to a single ''homogeneous'' spectral component. This is just the case illustrated in Fig. 2(b), where a slit mask selected a single configuration of beam components. That is, beams entering the excitation zone from opposite sides of the light cone are selected. For this linear-slit mask, the TH generation proceeds via a planar combination of two interacting subwaves, where two photons of the fundamental are taken from one subbeam and the third photon comes from another subbeam from the opposite side of the axicon ($\phi_1=\phi_2$ and $\phi_3=\phi_1+180°$). Figure 2(b) shows that for a linear-slit mask a Lorentzian-like peak appears at the position of the TH maximum from the full Bessel beam of Fig. 2(a). The width of this very Lorentzian profile is proportional to xenon pressure [0.035 nm/bar full width at half maximum (FWHM)] and matches exactly the theoretical value for pressure-induced broadening of the atomic 6*s* resonance of xenon [4], and the shift is in agreement with [7,12].

For two crossed slits [Fig. 2(c)], the same Lorentzian peak was observed together with an additional weak band on the blue side of the main peak. For this case there are three possible combinations of subbeams. The first combination is realized for each set of two slits similar to the case of a linear slit geometry. These combinations produce the same Lorentzian peak as in Fig. 2(b). In a second possible combination, two photons come from the opposite sides of a slit and the third photon comes from another slit ($\phi_2=\phi_1+180°$, $\phi_3=\phi_1+90°$). As it was shown in [8], such a nonplanar three-beam combination is equivalent to a planar two-beam combination giving the main TH peak. The last is a two-beam combination, where two photons come from one side of a slit and the third photon comes from another slit ($\phi_1=\phi_2$, $\phi_3=\phi_1+90°$). This particular combination is responsible for the appearance of the weak TH band to the blue of the main peak.

For two crossed unfocused beams, the location of the pure Lorentzian component in the spectrum is determined by the applicable value of the frequency shift δ_c given by [6,7,12]

$$\delta_c=\Delta_0\left(-1.11+\frac{9}{1-\cos\theta}\right)\simeq\frac{9\Delta_0}{1-\cos\theta}=\frac{9\Delta_0}{2\sin^2\theta/2}, \qquad (1)$$

where $\Delta_0=\pi N_0 F_{01}e^2/2m\omega_3$, N_0 is the gas density, F_{01} is the oscillator strength, ω_3 is the TH angular frequency, and θ is the angle between two waves. In the present context the small pressure-induced term $-1.11\Delta_0$ [12] can be omitted in the following considerations. From simple considerations of the conical interaction geometry, it is easy to see that two particular waves with azimuth angles ϕ_1 and ϕ_2 will cross at an angle θ given by

$$\sin\frac{\theta}{2}=\sin\frac{\phi}{2}\sin\alpha, \qquad (2)$$

where $\phi=\phi_2-\phi_1$. Using this expression, Eq. (1) can be written as

$$\delta_c=\frac{9\Delta_0}{2\sin^2(\theta/2)}=\frac{9\Delta_0}{2\sin^2(\phi/2)\sin^2\alpha}=\frac{\delta_0}{\sin^2(\phi/2)}, \qquad (3)$$

where $\delta_0=9\Delta_0/2\sin^2\alpha$ determines the position of the main peak with $\theta=2\alpha$. Thus, the shifted position of the Lorentzian peak for any given pair of subbeams can be determined in units of δ_0. For instance, for two beams crossing at $\phi=90°$ the shift is $\delta_c=2\delta_0$. This point corresponds exactly to the maximum of the blue band registered with two crossed perpendicular slits [Fig. 2(c)]. Similarly, for $\phi=120°$, $\delta_c=\frac{4}{3}\delta_0$. Again, this point can be recognized in our spectra as the maximum of the band registered with two or three slits separated by 120° [Figs. 2(d) or 2(e)].

For any single two-beam combination, the TH profile should have a Lorentzian shape with width independent of crossing angle. However, the spectral profiles produced by beams crossing at 90° and 120° were registered as broadbands instead of narrow Lorentzians (see Fig. 2). This effect results from the angular spreading of the individual beams, which was up to $\pm 20°$ in our case. The sensitivity of δ_c to the angular spreading $d\phi$ can be estimated by $d(\delta_c)/d\phi$ as

$$d(\delta_c)=-\frac{\delta_0}{\sin^2(\phi/2)\tan(\phi/2)}d\phi=-\frac{\delta_c}{\tan(\phi/2)}d\phi. \qquad (4)$$

This expression shows that the larger the value of ϕ, the less sensitive is $d(\delta_c)$ to $d\phi$, and that $d(\delta_c)\to 0$ at $\phi\to 180°$. For the pure Lorentzian peak produced from a linear-slit geometry, where $\delta_c=\delta_0$, the angular spreading of $\pm 20°$ leads to a negligible (about 3%) broadening of this peak. By contrast, for beams crossing at 120° the same spreading leads to an asymmetrical broadening of the corresponding Lorentzian by $0.2\delta_0$ to the red and by $0.37\delta_0$ to the blue. For beams crossing at 90° the broadening is further increased, being about $0.5\delta_0$ and δ_0 for the red and the blue sides, respectively. As a result, instead of narrow Lorentzians, broader bands were registered for crossing angles 90° and 120°.

For the general case of three nonplanar interacting waves from the axicon, we determine the cooperative shift to be given by the expression

$$\delta_c=\Delta_0\left(-1.11+\frac{18}{\sin^2\alpha[3-\cos(\phi_1-\phi_2)-\cos(\phi_1-\phi_3)-\cos(\phi_2-\phi_3)]}\right) \qquad (5)$$

or, assuming $\phi_1=0$ and neglecting the small constant term,

$$\delta_c=4\delta_0/[3-\cos\phi_2-\cos\phi_3-\cos(\phi_2-\phi_3)]. \qquad (6)$$

For three beams separated by 120° this expression gives the value $\delta_c=\frac{8}{9}\delta_0$ for the shift. Among all possible two- and three-beam combinations from segmented conical beams, this is the minimum value of the cooperative shift. The corresponding peak was registered in experiments with the three-slit mask [see Fig. 2(d)]. As expected, this peak had a Lorentzian profile with the same width of 0.035 nm/bar (FWHM). It should be noted that for this symmetrical three-beam configuration the generated TH field is directed along the axis of the fundamental conical beam. For such a case, the general expression for the TH generation in Bessel beams [10,11] gives zero TH intensity. However, the experiments with segmented conical beams have shown remarkable TH production for such symmetrical configuration. In full-aperture Bessel beams the contribution from this particular combination gives a discernible broadening of the red wing of the main TH peak.

We note that the integrated intensities of the experimental peaks in Fig. 2(d) were found to have a ratio of very close to 6:1 at any gas pressure. This ratio corresponds exactly to the numbers of possible combinations of photons producing those peaks: there is only one way to combine one photon from each of the three beams to produce the sharp peak, but there are six different 2+1 combinations (i.e., two photons from one beam and one from another) that produce the broader TH profile. Thus, since the excitation volume (gain length) is the same for all combinations, one should register peaks with 6:1 integral intensities, as we observed.

The sensitivity of the three-beam δ_c to the angular spreading $d\phi$ can again be estimated by $d(\delta_c)/d\phi$:

$$d(\delta_c) = -\frac{4\delta_0\{[\sin\phi_2 + \sin(\phi_2-\phi_3)]d\phi_2 + [\sin\phi_3 - \sin(\phi_2-\phi_3)]d\phi_3\}}{[3-\cos\phi_2 - \cos\phi_3 - \cos(\phi_2-\phi_3)]^2}. \quad (7)$$

For the symmetrical three-beam configuration $d(\delta_c)\rightarrow 0$ when $\phi_2 \rightarrow 120°$ and $\phi_3 \rightarrow 240°$. In our case of $\pm 20°$ spreading, the corresponding Lorentzian peak gets only a minor (about 5%) broadening toward the shorter wavelength.

To conclude, the use of conical excitation geometry together with apodization of the entrance beam allows one to realize multibeam excitation schemes of a given spatial geometry and to vary the nonlinear response of the medium in a controlled manner. For a given nonlinear process, the full-aperture Bessel beam realizes all possible configurations of interacting waves on the fundamental light cone. With the aid of simple geometrical masks, a segmented conical beam can be produced, where particular configurations of the fundamental waves are selected. This leads to a transformation of the overall nonlinear response according to the phase-matching requirements for a selected configuration. Thus, for nonlinear optics of Bessel beams the apodization of the entrance beam, or the azimuthal selection of interacting waves, gives a simple and reliable way to vary and to control the nonlinear response.

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