Two-color interference effect involving three-photon atomic excitation and four-wave mixing in crossed laser beams

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Through multiphoton ionization measurements, the polarization effects in destructive quantum interference under three-photon resonant excitation have been studied. Recent observations [V. Peet, Phys. Rev. A **74**, 033406 (2006)] have indicated that contrary to the well-known pattern of a total suppression of resonance excitation, the destructive interference becomes incomplete if three-photon transition is driven by crossed beams with orthogonal polarization planes. These observations have been tested for a more general case of two-color excitation and very similar polarization-dependent anomalies in the interference character have been registered. It has been shown that the destructive interference is modified and the resonance excitation does occur if two crossed laser beams have opposite circular polarizations. The pressure-induced evolution of the uncanceled ionization peaks has the ratio of blue shift to width close to 0.5 exactly as it is known for resonance ionization peaks registered under excitation by counterpropagating laser beams.

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The quantum interference effect between different optical harmonics was first reported in [1,2], where a very complete suppression of three-photon resonance enhanced multiphoton ionization (REMPI) in rare gases was observed. This cancellation effect was explained in terms of a destructive interference which occurs between three-photon coupling of the atomic states due to the laser field and one-photon coupling driven by sum-frequency field generated in the medium [3–12]. In conditions of strong resonant absorption of the sum-frequency field these excitation processes become equal in magnitude but opposite in phase so that the two excitation pathways interfere destructively and no excitation of the atomic state occurs above some certain threshold product of number density and oscillator strength. Under single-beam excitation, when all laser photons propagate in the same direction, the cancellation effect is able to suppress entirely any excitation of an odd-photon resonance which has a dipole-allowed transition back to the ground state. With counterpropagating laser beams the canceled atomic resonance reappears in REMPI spectra. This restored peak has a rather large pressure-induced blue shift proportional to the oscillator strength of the probed resonance [9,13]. Again, it results from an interplay between the two excitation pathways, where the wave-mixing field plays a significant role.

A very detailed theoretical treatment of the interference effects was developed and the general picture of this phenomenon in odd-photon resonant excitation was believed to be well analyzed and understood (see a review paper [12] and references therein). However, some observations were reported recently in experiments on REMPI of xenon in crossed laser beams [14]. When two linearly-polarized crossed beams with parallel polarizations were used for three-photon pumping of a dipole-allowed $\Delta J=1$ atomic transition the resonance-enhanced excitation pathway was canceled as expected. If, however, the pump beams had orthogonal polarization planes, a distinct ionization peak appeared in REMPI spectra under on-resonance excitation [14]. The evolution of this peak with pressure was independent of the crossing angle between two beams and was very similar to the evolution of the resonance ionization peak in counterpropagating laser beams. The resonance peak in cross-polarized beams was relatively weak and it means that the destructive interference still remained operative but the pattern of this interference differed significantly from that observed under ordinary single-beam excitation or excitation by crossed beams with parallel polarizations.

Recently it was reported that the resonance ionization enhancement occurs also if the two crossed beams have opposite circular polarizations [15]. The resonance ionization peak registered under such an excitation looks exactly as the peak in cross-polarized beams. The use of circularly polarized beams simplifies considerably the excitation problem since all single-beam excitation processes are eliminated: the sum-frequency generation for each of two circularly polarized beams vanishes and three-photon excitation of a $J=1$ atomic state by either left- or right-polarized light is forbidden. Three-photon excitation of a $\Delta J=1$ transition in this case is driven entirely due to combinations of two photons from one beam and one photon with opposite circularity from another beam.

In light of these observations, an extension of experimental data together with a considerable revision of the existing models of quantum interference effect are in order. In experiments of [14,15] the simplest single-color excitation mode was used, when three-photon excitation of the resonant transition was driven by two crossed beams from a single laser source. The one-photon coupling in this case is provided by an internally generated third harmonic of the fundamental laser light. It would be important to test the reported polarization anomalies in a more general situation, when the resonant transition is driven by two independent laser sources of frequencies ω_1 and ω_2. If these lasers are tuned to three-photon resonance at $2\omega_1+\omega_2$ or $\omega_1+2\omega_2$, then the generated sum-frequency field should interfere with three-photon exci-

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tation at resonance. The general pattern of a suppressed resonance ionization for such a two-color excitation by crossed pump beams is known from previous studies [9,11,16].

The present study is an addendum to the experimental findings reported in [14,15]. Here the polarization anomalies in quantum interference effect were tested in a more general case of two-color excitation by crossed laser beams. Special attention was paid to the use of circularly polarized beams, when all single-beam excitation processes are eliminated. The experimental procedure was similar to that used in previous studies [14,15], but modified for two-color excitation. Briefly, the output of an excimer XeCl laser was split and used to pump two dye lasers. The pulses of dye lasers had energies of 0.2–0.4 mJ, a pulse duration of 8–10 ns, and spectral width of about 10 pm full width at half maximum (FWHM). The parallel propagating laser beams were focused by a lens $f=75$ mm into a static gas cell. The crossing angle between two focused beams was about 15°. The cell was filled by xenon at a pressure of up to 2 bar. Photoelectrons resulting from the multiphoton ionization process were collected by a wire collector biased at +20 V. Ionization signal was amplified, digitized, and stored on a computer.

The output emission of dye lasers was polarized linearly in the direction normal to the common plane of propagation (s polarization). With the aid of zero-order quarter-wave plates the linear polarization of two beams was transformed into circular polarizations with opposite circularity. The cancellation effect was probed for the three-photon $6s[3/2]$ $J=1$ resonance of xenon ($\lambda_0=440.86$ nm). In two-color experiments, the emission wavelength λ_2 of one of the dye lasers was fixed near λ_0 and the second dye laser was tuned. Additionally, one-color reference experiments with unidirectional and counterpropagating laser beams were arranged to monitor the canceled $6s$ resonance. To provide the counterpropagating light, a plane mirror was placed near the output window of the gas cell. The mirror reflected the diverging laser beam back to the cell and a small fraction of the beam propagated again through the excitation region.

When excitation of a target medium is driven by two beams with arbitrary frequencies ω_1 and ω_2, the three-photon excitation of $J=1$ atomic states occurs when $\omega_1+2\omega_2=\omega_0$ or $2\omega_1+\omega_2=\omega_0$, where ω_0 is the resonance frequency. In experiments, both dye lasers were operated with the Coumarin-120 dye and the wavelength of one laser was fixed at λ_2 so that $\Delta\lambda=\lambda_2-\lambda_0 \ll \lambda_0$. When the wavelength of another laser λ_1 was tuned the excitation of the probed resonance occurred at two wavelengths $\lambda_1=\lambda_0-0.5\Delta\lambda$ and $\lambda_1=\lambda_0-2\Delta\lambda$.

Figure 1 shows multiphoton ionization spectra measured in different one- and two-color excitation modes. Figures 1(a) and 1(b) demonstrate the well-known cancellation of three-photon atomic resonance, when the resonance-enhanced ionization is absent under single-beam excitation [Fig. 1(a)], but a strong resonance ionization peak appears in the presence of weak counterpropagating light [Fig. 1(b)]. A very similar pattern of the cancellation effect persists also for two-color excitation by crossed beams in s polarization, where the resonance ionization peak is absent and only a weak and broad dispersion-like feature is registered near the canceled resonance [Fig. 1(c)]. This ionization feature is known for excitation by tightly focused beams and it can be found in many REMPI spectra measured near canceled three-photon atomic resonances in rare gases [2,14,15,17–21].

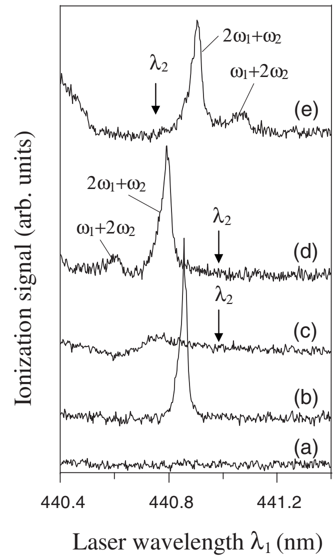

FIG. 1. Wavelength scans of the ionization signal near the $6s$ resonance of xenon: (a) single-beam excitation; (b) single-color excitation by counterpropagating beams; (c) two-color excitation by crossed s-polarized beams; (d), (e) two-color excitation by crossed beams with opposite circular polarizations. The excitation wavelength λ_2 for two-color profiles is marked by arrows. Xenon pressure is 0.5 bar.

The ionization spectra undergo significant changes if the excitation process is driven by beams with opposite circular polarizations [Figs. 1(d) and 1(e)]. In this case, two distinct ionization peaks appear in spectra. If the excitation wavelength λ_2 is set to the red from the resonance position, the two ionization peaks appear at the blue side of the resonance [Fig. 1(d)]. If the excitation wavelength λ_2 is tuned to the blue from the resonance position, the two ionization peaks are registered at the red side of the resonance [Fig. 1(e)]. The relative magnitude of these peaks is determined mainly by relative intensity and mutual overlap of pump beams in the interaction region. The position of two peaks in spectra is determined entirely by the detuning $\Delta\lambda$ exactly as it was considered above. In experiments, the wavelength λ_2 was tuned off the resonance position up to $\Delta\lambda=\pm 6$ nm. In all cases, the two ionization peaks were detected easily at corresponding positions $\lambda_1=\lambda_0-0.5\Delta\lambda$ and $\lambda_1=\lambda_0-2\Delta\lambda$. Very similar peaks were registered also if the two pump beams had linear polarization with orthogonal polarization planes as in experiments of [14]. All these observations confirm the existence of polarization effects in quantum interference between different optical harmonics under three-photon resonance excitation.

Figure 2 demonstrates the evolution of ionization peaks with pressure. Within the limits of experimental uncertainty, the shift Δ and the width Γ (FWHM) were linear with pressure (see the inset in Fig. 2) and a slope of 30–40 pm/bar for the shift and 70–80 pm/bar for the width were measured for the most intense peak at $2\omega_1+\omega_2$. Such a ratio of $\Delta/\Gamma \simeq 0.5$ is known for the canceled atomic resonances of xenon

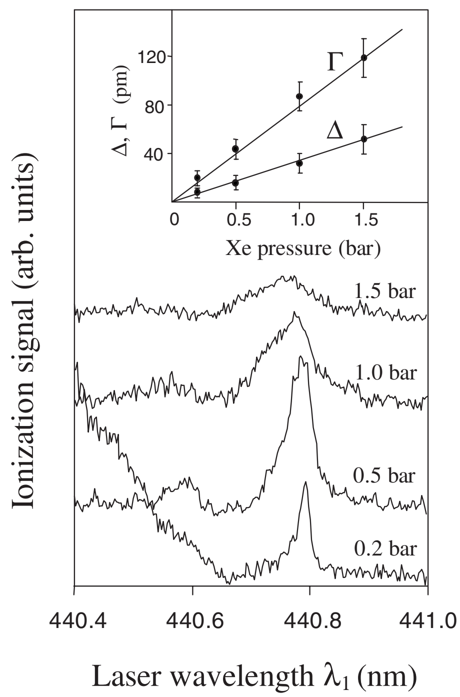

FIG. 2. Evolution of the resonance ionization peaks with pressure under excitation by crossed beams with opposite circular polarizations. Wavelength λ_2=441 nm. The intense band on the blue side of the profile at 0.2 bar is due to the phase-matched sum-frequency field $2\omega_1+\omega_2$ generation in the negatively-dispersive side of the 6s resonance. The inset shows plots of the blue shift Δ and width Γ of the ionization peak vs xenon pressure.

registered in counterpropagating beams [13]. Here again the resonance ionization peaks demonstrate their intriguing feature reported previously from experiments with cross-polarized beams [14]. Namely, the appearance of such peaks in angled beams looks exactly as it would be in the presence of weak counterpropagating light.

The experiments on two-color excitation of a resonance $\Delta J=1$ transition have confirmed and extended previous observations concerning the presence of polarization anomalies in quantum interference effect. So far, the physical interpretation and an adequate theoretical model of these anomalies are absent. Nevertheless, some general background of the "unusual" interference character can be derived from the following qualitative consideration. In previous theoretical studies the quantum interference effect in crossed beams was considered for the simplest case of beams linearly polarized in direction perpendicular to the propagation plane (s polarization). It corresponds to the usual choice of polarization in two-beam experiments on interference effect. For s-polarized beams both one- and three-photon couplings for an atomic $J=0$ to 1 transition follow the selection rule $\Delta m_J=0$ for the magnetic quantum number m_J, where the quantization axis is along the light polarization direction. In this case, the magnetic degeneracy of the upper $J=1$ state can be neglected [10] and, independent of the crossing angle between two beams, the whole excitation problem can be framed by a simple two-level model. Such an approach, however, becomes questionable for a more general case of crossed beams with different polarizations. If pump beams are cross polarized or they have opposite circular polarizations, the angular momentum considerations become important in analyzing the response of atomic system. Under three-photon excitation by angled circularly polarized beams the atom acquires angular momenta from two photons taken from one beam and a photon with opposite circularity from another beam. Noncollinear addition of angular momenta ± 2 and ∓ 1 gives the total-momentum eigenstates $|1,0\rangle$, $|1,+1\rangle$, and $|1,-1\rangle$ for the upper $J=1$ atomic state and this state enters the excitation problem as a coherent superposition of three degenerated sublevels $m_J=0,\pm 1$, where amplitudes of this superposition are dependent on the crossing angle between pump beams. Sum-frequency generation adds coherent one-photon couplings between the ground state $J=0$ and each of these sublevels. As a result, a multilevel excitation scheme with multiple interfering processes is realized. Experiments have shown that with circularly polarized beams the resonant excitation still remains suppressed but, in sharp contrast to the parallel polarizations of pump beams, the overall transition amplitude does not vanish and the resonance excitation is easily registered through resonance-enhanced multiphoton ionization. This effect has remained beyond the scope of existing models of the interference effect.

To conclude, the present study adds experimental information concerning polarizations effects in quantum interference effect. It has been shown that the destructive interference under three-photon resonant atomic excitation is modified significantly if three-photon excitation is driven by crossed laser beams with different polarizations. In particular, it has been demonstrated that the excitation of an atomic $\Delta J=1$ transition does occur under two-color excitation by circularly polarized beams. The pressure-induced evolution of the uncanceled ionization peaks has the ratio of blue shift to width close to 0.5 exactly as it is known for peaks registered under excitation by counterpropagating laser beams. Together with previous observations, these results accomplish a basic set of experimental data for further development and testing of theoretical models.

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