Suppression of a four-photon resonance by four-wave mixing near an intermediate three-photon resonance

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We use resonance ionization involving two colors in single-pass and retroreflected geometries to study the suppression of excitation of a four-photon resonance by an interference involving four-wave sum-frequency generation occurring near an intermediate three-photon resonance. The $6p[\frac{1}{2}](J=0)$ state of xenon is excited by three blue photons plus one infrared photon. One beam, focused to $I_1 \simeq 7\times 10^9$ W/cm^2, is nearly three-photon resonant with the $6s[\frac{3}{2}](J=1)$ resonance; the second, unfocused beam ($I_2 \simeq 2\times 10^7$ W/cm^2) is tuned to maintain exact four-photon resonance with the $6p[\frac{1}{2}](J=1)$ state. One additional blue photon ionizes from the four-photon level. Third-harmonic light is generated by the first, nearly three-photon-resonant beam by four-wave sum-frequency mixing. One third-harmonic photon plus one photon from the second laser form a second excitation pathway to the four-photon level; destructive interference between the two pathways results in a reduction in the observed ionization. The degree of reduction depends on the xenon density and the detuning (both positive and negative) from three-photon resonance. This interference may be overcome by retroreflecting the nearly three-photon-resonant beam to provide an additional excitation pathway which generates no third harmonic. The study provides detailed information on the pressure-dependent range from exact resonance over which the familiar three-photon cancellation effect is operative; it shows that the interference occurs in high- and low-energy regions about three-photon resonance; and it establishes that interference is produced for large values of the phase mismatch even if the absorption of the interfering field is low. We compare our measurements of the reduction in ionization with predictions published by us elsewhere [M. G. Payne, J. C. Miller, R. C. Hart, and W. R. Garrett, Phys. Rev. A **44**, 7684 (1991)]; excellent agreement is observed. We also report the observation of suppression of pressure-induced wing absorption into the three-photon state observed in a classic "hybrid" resonance mode of excitation.

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I. INTRODUCTION

In a recent paper, Payne, Miller, Hart, and Garrett [1] have presented a fairly general theoretical treatment of resonant multicolor excitation of a four-photon transition under circumstances where the process is influenced by the presence of a nearly-three-photon-resonant intermediate state that can be three photon or one photon pumped from the ground state. The detailed study describes, for both focused and unfocused geometries, effects on four-photon excitation produced through an interference that is generated in the simultaneous coupling of the intermediate state by the laser field and the internally generated third harmonic of the laser field. A subset of the predicted effects has been verified in the present study.

The odd-photon interference effect has been the subject of numerous experimental and theoretical studies [2–8]. Studies of four-photon resonant multiphoton-ionization (MPI) and related effects conducted both by some of us [9,10] and by others [11–13] have produced a number of interesting results associated with a three-photon interference occurring at an intermediate three- and one-photon-allowed resonance. Prominent among these is the observation by Blasewicz *et al.* [9,11] of the production of both peaks and dips in the MPI signal as a nearly three-photon-resonant laser is tuned through exact four-photon resonance. In their experiment four-photon-resonant excitation was achieved by using two focused lasers of different colors; laser 1, was tuned near a three- and one-photon-allowed transition and the other, laser 2, was tuned to make $3\omega_1+\omega_2$ resonant with the four-photon level. Indeed, it was found that conditions could be modified in ways such that a peak in a MPI signal at four-photon resonance could be changed to a dip under different choices of gas pressures, laser detunings from three-photon resonance, and laser intensities. In Ref. [1] it was shown how these features, and others, are governed by the detailed behavior of the three-photon interference, which, in turn, is influenced primarily by the complex-phase mismatch Δk between the laser-1 field and its third harmonic. The phase mismatch $\Delta k=\Delta k_r+i\alpha$, where Δk_r is the real part of Δk and α is the complex absorptive part, which is one-half of the absorption coefficient for the third-harmonic field. A critically important consideration in this portion of the study was the absorption coefficient 2α for the third-harmonic field $3\omega_1$, which is composed of two parts; ordinary wing absorption on either wing of the resonance line (i.e., the high- or low-energy side of the absorption profile for $3\omega_1$), and two-photon absorption of $3\omega_1$ through two-photon excitation of $|2\rangle$ by absorption of a third harmonic and a laser-2 photon (ω_2). Hence α is a function of ω_2 intensity at resonance with $|2\rangle$. Thus the observations of Refs. [9,11] may be interpreted in terms of a combination of two-photon absorption of the third harmonic by the second, intense laser and a three-photon interference effect (i.e., an interference between the three-photon laser field and the third-harmonic field).

Though similar, the present experiment examines the three-photon interference, and its effect on four-photon excitation, in a different regime than that of Blasewicz *et al.*, principally in that the second laser is several orders of magnitude less intense than in the Blasewicz study, thus making two-photon absorption of the third harmonic almost negligible. In this way we are able to examine three-photon interference induced mainly by a large real part of the phase mismatch Δk between the nearly three-photon-resonant laser and the associated third-harmonic field. This study accomplishes three goals in further elucidating details of the three-photon cancellation effect. First, it provides a quantitative measure of the range, about three-photon resonance, over which the cancellation effect is operative. The theoretical prediction of the dependence on the phase mismatch is confirmed. Second, the measurements show that the interference and the resultant cancellation of the four-photon excitation extends on the red side as well as the blue side of three-photon resonance. This point has also been made in the study of Elk, Lambropoulos, and Tang [13], and the effect was demonstrated at selected points on the high-energy side of the 6s state in Xe by Charalambdis *et al.* [12]. (In a single-color experiment, several *nf* levels can be accessed in four photons while maintaining nearly-three-photon resonance with the 6s.) Third, the present study shows that the cancellation can occur, as predicted, under circumstances where the absorption coefficient for the third harmonic is small as long as the real part of Δk is large in absolute value. That is, the absorption can be small, or even zero, and still the interference can be produced in a quantitatively predictable manner. This last point has been made before in work by Payne and Garrett [2,10,14,15]. However, others [3,6] have overstated the requirement for strong absorption of the generated field.

In Sec. II of this paper we extract results from Ref. [1] to show how the excitation of the four-photon level depends on the value of $|\Delta k b/2|$, the phase mismatch parameter in focused geometry with confocal parameter b. In Sec. III we describe the experimental conditions. Section IV reports the results and makes comparison with the predictions of Sec. II. The last section describes a related hybrid-resonance experiment, in which we demonstrate the suppression by the three-photon interference effect of wing absorption into the three-photon state.

II. THEORY

In this section we consider the description of four-photon-resonant ionization under the particular circumstances relevant to the present experimental study. We give expressions for the ionization to be expected from four-photon-resonant excitation both for unidirectional and counterpropagating focused-geometry excitation near the three-photon level; that is, we show results for the ionization with and without the three-photon-interference effect and consider the functional dependence of the ratio of ionization in these two cases on the three-photon detuning and the number density of the medium. This ratio quantifies the effectiveness of the interference in suppressing excitation as the intermediate three-photon coupling is detuned away from resonance. All three of the points mentioned above can be succinctly presented through a composite plot of data in terms of this parametrization.

In Ref. [1], MPI is considered with the pumping scheme shown in Fig. 1. Electric dipole transitions between the ground state $|0\rangle$ and state $|1\rangle$ are allowed in one and three photons, while $|2\rangle$ is coupled to the ground state in both two and four photons with state $|1\rangle$ serving as a nearly resonant intermediate state. Laser 1 of frequency ω_1 is focused with waist radius w, confocal parameter b and detuning from three-photon resonance $\delta_1=3\omega_1-(E_1-E_0)/\hbar$; its direction of propagation is chosen to be along the $+z$ axis. Laser 2 of frequency ω_2 is unfocused, temporally coincident with laser 1, and is assumed to be of uniform intensity over the focal region of beam 1; $\delta_2=3\omega_1+\omega_2-(E_2-E_0)/\hbar$ is the detuning from four-photon resonance.

With reference to Fig. 1, we note that states $|0\rangle$ and $|1\rangle$ are coupled both by a three-photon Rabi frequency $\Omega_{1,0}^{(3)}$ due to laser 1 and by a one-photon Rabi frequency $\Omega_{1,0}^{(1)}$ due to the internally generated third-harmonic field. An effective or net Rabi frequency $\Omega_{1,0}^{\text{eff}}$ is defined as

$$\Omega_{1,0}^{\text{eff}}=e^{-i\Delta k_r z}\Omega_{1,0}^{(3)}+\Omega_{1,0}^{(1)} \tag{2.1}$$

to describe this simultaneous, coherent coupling of $|0\rangle$ and $|1\rangle$ in one and three photons. In the present context, where δ_1 is not huge, the real part of Δk is determined almost entirely by the nearly resonant contribution. We

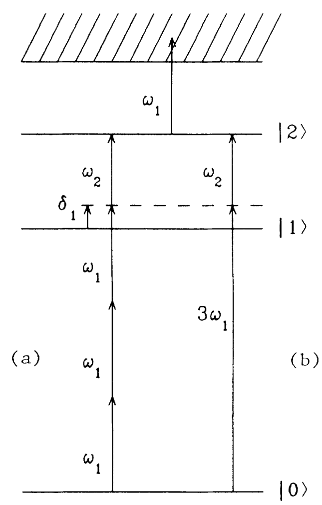

FIG. 1. Energy-level diagram showing (a) the excitation of $|2\rangle$ by three laser photons nearly resonant with $|1\rangle$ plus one additional photon of another color, and (b) the excitation of $|2\rangle$ by one third-harmonic photon plus one photon of another color. Interference between the two pathways to $|1\rangle$ can suppress the excitation of $|2\rangle$ and thus the MPI signal.

have

$$\Delta k_r \simeq -\frac{\kappa_{01}}{\delta_1} ,$$

where

$$\kappa_{01} = \frac{2\pi(3\omega_1)N|D_{01}|^2}{c\hbar}$$

is a parameter proportional to the number density N of the medium and the oscillator strength between $|0\rangle$ and $|1\rangle$.

From Ref. [1] (Secs. IV and V) we note that the effective Rabi frequency $\Omega_{1,0}^{\text{eff}}$ at time t and cylindrical coordinates ρ, z is given ultimately by

$$\Omega_{1,0}^{\text{eff}}(\rho,z,t) = e^{-i\Delta k_r z}\Omega_{1,0}^{(3)}(\rho,z,t)(1+iu)^2 \times \left[\frac{1}{(1+iu)^2} + i\frac{\Delta kb}{2}g(u,\Delta kb/2)\right] . \tag{2.2}$$

Here $u = 2z/b$, and the function

$$g(u,v) = \int_{-\infty}^{u} \frac{e^{iv(u-u')}}{(1+iu')^2}du' \tag{2.3}$$

is the familiar Gaussian diffraction integral for third-harmonic generation in a focused beam with position-independent absorption.

We assume that two additional photons of frequency ω_1 are required to ionize from $|1\rangle$. Following Ref. [1], this implies that the only significant contribution to the ionization is that which proceeds by way of the four-photon resonance; that is, we absorb $3\omega_1 + \omega_2$ to reach state $|2\rangle$, from which one additional photon of frequency ω_1 will cause ionization. In this case, if the temporal profile of the laser pulse is approximated as rectangular of length τ, the number of ions produced per pulse is

$$N_I = \frac{N\pi w^2 b\tau}{2}\left[\frac{\langle|\Omega_{2,1}^{(12)}(0,0,0)|^2\rangle}{\gamma_2}V(\delta_2)\right] \times \frac{|\bar{\Omega}_{1,0}^{(3)}(0,0,0)|^2}{\delta_1^2} \times \int_{-\infty}^{\infty}du\left|\frac{1}{(1+iu)^2} + i\left[\frac{\Delta kb}{2}\right]g(u,\Delta kb/2)\right|^2 . \tag{2.4}$$

[This is essentially Eq. (5.3) of Ref. [1].] The quantities in the parentheses on the first line describe the enhancement due to the second laser coupling the fields due to the first laser to the four-photon resonance, while those in the second line of (2.4) come from $\Omega_{1,0}^{\text{eff}}$. The bar over the three-photon Rabi frequency indicates that the quantity is evaluated at the average power density appropriate to the particular time through the pulse. The arguments (0,0,0) in the Rabi frequencies indicate the quantities are evaluated at the focal point of laser 1 and at the peak of the average power. $V(\delta_2)$ describes the line shape if we should fix ω_1 (of bandwidth Γ_1) and tune ω_2 (of bandwidth Γ_2) across the four-photon resonance. The term $V(\delta_2)$ in (2.4) has the analytic form

$$V(\delta_2) = \sqrt{\pi}e^{-\delta_2^2/4\gamma_2^2}\left[1 - \frac{1-e^{-\Gamma_{I2}\tau}}{\Gamma_{I2}\tau}\right] + \frac{4\Gamma_{I2}}{\gamma_2}G(\delta_2/\gamma_2) , \tag{2.5}$$

where $\gamma_2 = (3\Gamma_1^2 + \Gamma_2^2)^{1/2}$ is the effective four-photon laser bandwidth, Γ_{I2} is the ionization rate out of $|2\rangle$ and

$$G(y) = -\tfrac{1}{2} + \frac{y}{4}e^{-y^2/4}\int_0^y dv\, e^{v^2/4} .$$

For reference we note that the first term on the right-hand side (rhs) of (2.5) represents absorption into $|2\rangle$ with subsequent ionization, while the second term, containing $G(\delta_2/\gamma_2)$, represents far-wing resonant enhancement by $|2\rangle$.

The behavior of the interference between the fields at ω_1 and $3\omega_1$ is contained in the integrand of (2.4). For the purpose of discussion, let the integrand in (2.4) be defined as M, where

$$M(2z/b,\Delta kb/2) \equiv M(u,v) = \left|\frac{1}{(1+i2z/b)^2} + i(\Delta kb/2)g(2z/b,\Delta kb/2)\right|^2 = \left|\frac{1}{(1+iu)^2} + ivg(u,v)\right|^2 . \tag{2.6}$$

The first term of M is the contribution due to laser 1, while the second represents the summed contributions of the third-harmonic field from all smaller values of $u = 2z/b$. The magnitude of $v = \Delta kb/2$ determines the degree of interference. For large values of $|\Delta kb/2|$, corresponding to small detunings, or to large number densities, or to large values of absorption, the second term in (2.6) will be similar in magnitude and opposite in sign to the first. To see this analytically, consider the regime of large $|\Delta kz|$. For $|v(1+iu)| > 15$, the function $g(u,v)$ of Eq. (2.3) can be approximated by the asymptotic series

$$g(u,v) \simeq -\frac{i}{(1+iu)}\sum_{j=1}^{J}\frac{j!}{(-v)^j(1+iu)^j} , \tag{2.7}$$

with J chosen so that $J < |v(1+iu)|/2$. Recalling that $v = (\Delta k_r + i\alpha)b/2 = (-\kappa_{01}/\delta_1 + i\alpha)b/2$, we may use (2.7) in (2.6) and write

$$M(u,v) \simeq \left|\frac{1}{(1+iu)^2} + \left[\frac{-1}{(1+iu)^2} + \frac{2}{v(1+iu)^3} - \frac{6}{v^2(1+iu)^4} + \cdots\right]\right|^2 . \tag{2.8}$$

The leading, v-independent term of the expansion represents the adiabatic part of the polarization at $3\omega_1$; it cancels the term due to three-photon pumping by the first laser. That is, the first term on the left is canceled by the first term of the series inside the parentheses in (2.8). For small α, the behavior of M is dominated by $\Delta k_r b/2$. As δ_1 approaches zero, the phase mismatch becomes larger and the remaining terms in the expansion approach zero; this is the three-photon-interference effect which causes $M(u,v)\rightarrow 0$ and $N_I \rightarrow 0$, thus canceling the excitation. As δ_1 is tuned away from three-photon resonance, the phase mismatch is lessened and the remaining terms in the expansion produce excitation. It is this dependence of the interference on the phase mismatch that we examine in the present experiment.

If the first, nearly three-photon-resonant, laser is reflected back on itself, an excitation pathway involving the absorption of two ω_1 photons traveling in one direction plus one ω_1 photon from the other direction (plus one from the second laser) is created; due to the radically different phase-matching condition, this pathway may be considered for our purposes here as lacking the interference effect. (See, however, Refs. [13–16].) With such counterpropagated excitation, one would see increased ionization. The ratio between the ionization with and without counterpropagation will depend on the degree of interference occurring in the uncounterpropagated case, which depends on the value of $\Delta kb/2$.

We define R as the ratio between ionization with the first laser counterpropagated and ionization with unidirectional excitation. Assuming $|\alpha| \ll |\Delta k_r|$ and using (2.5), we may write

$$R = \frac{\int_{-\infty}^{\infty} du \left[M + (F/3)\frac{1}{(1+u^2)^2} \right]}{\int_{-\infty}^{\infty} du M} = 1 + \frac{\pi F/6}{\int_{-\infty}^{\infty} du M(u, \Delta k_r b/2)} \ . \tag{2.9}$$

Here the quantity $(F/3)(1+u^2)^{-2}$ represents the additional ionization due to the counterpropagated excitation pathway. We assume the reflected beam to be refocused to have the same confocal parameter as the forward beam, with F the fraction of the forward energy reflected. The factor of $\frac{1}{3}$ results from the lack of correlation between the forward and reflected beams from our multimode laser.

III. EXPERIMENTAL DETAILS

We generate light at $\lambda_1 \simeq 440.8$ nm and $\lambda_2 \simeq 828.2$ nm with, respectively, a Lumonics model HD-300 dye laser using Coumarin 440 dye and a side-pumped Quanta-Ray PDL-1 dye laser using LDS-820 each pumped by the appropriate harmonic of a single Nd:YAG laser. Both lasers were linearly polarized, with the directions of polarization parallel. Pulse lengths were approximately 7 ns, and the laser bandwidths were approximately 0.2 and 1 cm^{-1}, giving an effective four-photon bandwidth γ_2 of about 1 cm^{-1}. The blue laser had a beam diameter of about 1 mm and a beam divergence of about 0.3 mrad. The lasers were calibrated in wavelength using the xenon $5p^6 \rightarrow 6s[\frac{3}{2}]$ and $6s[\frac{3}{2}] \rightarrow 6p[\frac{1}{2}]$ transitions for λ_1 and λ_2.

The blue beam was focused into the center of the 10-cm-long xenon cell with a lens with a focal length measured to be 5.5 cm; after passage through the cell, the light was recollimated with a 15-cm focal-length lens and retroreflected with a dichroic mirror. The 828-nm light from laser 2 was brought into the cell through the dichroic mirror along a direction almost opposite to that of the forward blue beam. It was necessary to introduce a slight angle between the blue and IR beams to allow the retroreflected blue beam to be blocked while still admitting the IR beam to the cell. The 15-cm lens that recollimated the blue beam plus a third lens formed a telescope for the IR beam, which was thus unfocused in its passage through the cell.

The cell contained both a wire-type proportional counter and a collection plate (mounted on the opposite side of the focal region from the wire) to which an electrometer could be connected. The most satisfactory results were obtained by placing +200 to +300 V on the wire and observing the positive ion current with the electrometer.

Maintaining a constant overlap between the forward and retroreflected blue beams was critical in obtaining repeatable results; a large, heavy mount for the retroreflection mirror proved to provide adequate stability for data taking runs of an hour or so. To optimize the overlap, laser 1 was tuned to the $6s[\frac{3}{2}]$ three-photon resonance and the mirror adjusted to maximize the MPI signal.

The photoionization cross section σ_I for the $6p[\frac{1}{2}]$ state of xenon is approximately 1.6×10^{-17} cm^2 for 440-nm light [17]. With the intensities employed, this transition is fully saturated, and so we expect an I_1^3 power dependence of the four-photon-resonant MPI signal as the intensity of the blue laser is varied. Employing only the proportional counter and a charge-sensitive preamp, we were able to observe an I_1^3 dependence only with I_1 so weak that the four-photon MPI signals were uselessly small; we attribute this behavior to a combination of space-charge effects limiting the electron mobility to the ion diffusion rate and the limited integration time of the (pulse-type) preamplifier. Employing the electrometer, which integrated the ion current over long times, allowed operation with both adequate signal levels and the proper I^3 power dependence. The experiment was performed with $I_1 \simeq 7\times 10^9$ W/cm^2 at the focus and $I_2 \simeq 1.5\times 10^7$ W/cm^2. Using $\Gamma_I = \sigma_I F$, with F the photon flux at 440 nm, we find $\Gamma_I \simeq 1.2$ cm^{-1}, which, in combination with our laser bandwidths, is in reasonable agreement with the observed full width at half maximum (FWHM) of the four-photon resonance of about 2 cm^{-1}.

To measure a value of R, we fixed λ_2 to place the four-photon resonance at a known detuning δ_1 from three-photon resonance. The blue laser was scanned across the four-photon peak with the output from the electrometer recorded on a chart recorder. We then blocked the retroreflected beam and repeated the scan. By using different values of δ_1 and P_{Xe} (the xenon pressure), we could map the dependence of R on $\Delta k_r b$. To guard against any systematic errors (due, for example, to a change in the overlap between forward and retroreflected beams), we varied detuning and pressure in an irregular fashion. Repeated values of R taken at a given value of $\Delta k_r b$ indicate that the repeatability of the measurements is better than 10%. Xenon pressures of 2 and 4 Torr were employed. R was measured on both sides of the three-photon resonance, with $|\delta_1|$ in the range 20–50 cm^{-1}. Reliable values for R could not be obtained for detunings too close to three-photon resonance, since as δ_1 decreases, the four-photon MPI signal without counterpropagation of the blue beam becomes comparable with the noise level, leading to large uncertainties in R. This was especially significant for the values taken on the red side of three-photon resonance, where, since the third-harmonic is very badly phase mismatched, the suppression is greater than at the same detuning on the blue side.

IV. RESULTS AND DISCUSSION

In Fig. 2 we show typical scans across the four-photon resonance. The scans labeled a and a' are taken without counterpropagation of the blue beam; that is, with the three-photon interference effect acting to suppress excitation of the four-photon level. Those labeled b and b' are taken with counterpropagation. The two pairs a,b and a',b' differ in the detuning from three-photon resonance, with the upper pair taken closer to resonance, thus showing more suppression in the uncounterpropagated scan.

In Fig. 3 we show the four-photon MPI signal as a function of δ_1 for both unidirectional and counterpropagated excitation. With the blue beam counterpropagated, we see approximately the expected δ_1^{-2} dependence due to nearly resonant enhancement by the three-photon level. Without counterpropagation, the MPI signal decreases drastically as we approach the three-photon resonance and thus approach nearer to complete cancellation between the two excitation pathways.

In Fig. 4 we show our data plotted against the predicted behavior from Eq. (2.9) in the form $(R-1)/F$ versus $-\Delta k_r b/2$. Here we use for F the fraction of the intensity of the forward beam at the focus of the reflected beam;

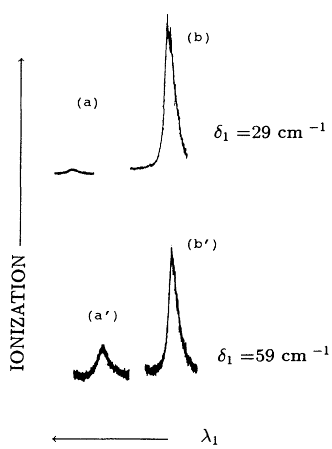

FIG. 2. MPI signal as the first laser is scanned across the four-photon resonance. Scans labeled a and a' are taken without retroreflection of the nearly three-photon-resonant beam, while in b and b' the retroreflected excitation overcomes the three-photon interference. The upper and lower pairs are for blue-side detunings from three-photon resonance of 29 and 59 cm^{-1}, respectively, with $P_{\mathrm{Xe}}=4$ Torr. A smaller detuning produces more suppression.

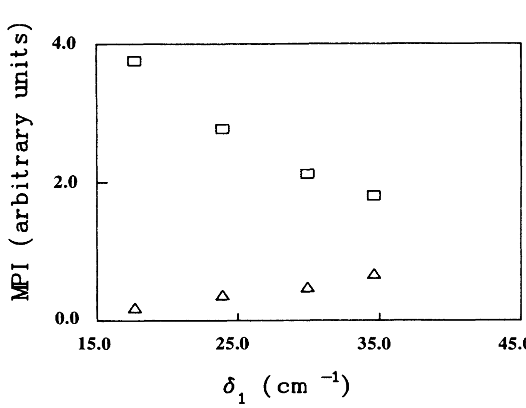

FIG. 3. Four-photon MPI as a function of detuning δ_1 from three-photon resonance. The upper curve (squares) shows MPI with the nearly three-photon-resonant beam retroreflected; it shows an increase in ionization as we tune closer to the enhancing three-photon resonance. The lower curve (triangles), taken with the beam not retroreflected, shows the result of the three-photon-interference effect; the ionization decreases as we approach resonance. Both curves are at the same laser intensities and detection sensitivities.

since the reflected beam is focused with a lens with a focal length three times that of the lens used in the forward beam, we expect $F=0.1$. To find $\Delta k_r b$, we treat our laser as if it had a diffraction-limited Gaussian beam; using $f_{01}=0.269$ for the oscillator strength of the $5p^6 \rightarrow 6s[\frac{3}{2}]$ transition [18], we find

$$-\frac{\Delta k_r b}{2}=102.9\frac{P_{\mathrm{Xe}}}{\delta_1} ,$$

with P_{Xe} in Torr and δ_1 in cm^{-1}. The solid line in Fig. 4 is the result of evaluating Eq. (2.9) numerically using techniques outlined in Ref. [1].

To achieve the fit shown, we have multiplied the experimental values of $\Delta k_r b$ by 1.25 and the observed values of $(R-1)/F$ by 0.6. The adjustment to $\Delta k_r b$ is insignificant, given that we model the focused laser beam

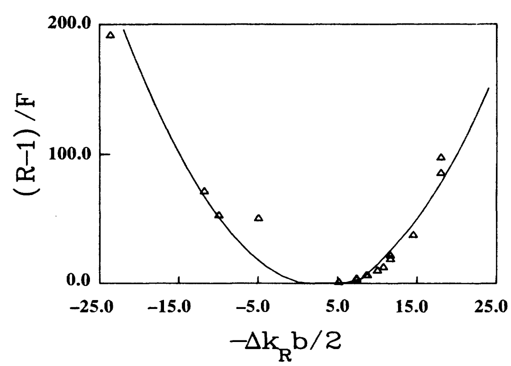

FIG. 4. Suppression of four-photon MPI vs phase mismatch. R is the ratio of MPI with and without counterpropagation of the nearly three-photon-resonant beam. F is the fraction of the forward intensity in the reflected beam. $-\Delta k_r b/2 =102.9 P_{\mathrm{Xe}}(\mathrm{Torr})/\delta_1(\mathrm{cm}^{-1})$, so we approach three-photon resonance from the blue (red) side on the right (left) side of the graph.

as being axially symmetrical with a Gaussian intensity distribution, whereas in fact our laser beam is neither Gaussian nor symmetrical. Some of the discrepancy between predicted and observed values of $(R-1)/F$ may also be attributed to this source; the remainder may be due to residual two-photon absorption of the third harmonic, which would have the effect of increasing the suppression ratio above the predicted value.

In our analysis of the data, we have assumed $\Delta k=\Delta k_r$; that is, we have neglected two-photon absorption. To estimate α, we write for the excitation rate $\mathcal{R}$ of $|2\rangle$ by absorption of one photon at frequency $3\omega_1$ and one at ω_2

$$\mathcal{R}\simeq 2\frac{|\Omega_{02}^{(2)}|^2}{\Gamma_L}\simeq 2\frac{|\Omega_{01}^{(1)}|^2|\Omega_{12}^{(1)}|^2}{\Gamma_L\delta_1^2}\ ,$$

where $\Omega_{02}^{(2)}$ is the reduced two-photon Rabi frequency and Γ_L is the half width at half maximum (HWHM) of the transition. With N the number density, $F_{3\omega_1}$ the photon flux at $3\omega_1$, and β the two-photon-absorption coefficient for light at $3\omega_1$, we find

$$N\mathcal{R}=\beta F_{3\omega_1}$$

by equating the volume rate of excitation to the volume rate of absorption. Then

$$\alpha=\frac{\beta}{2}=\frac{N\mathcal{R}}{2F_{3\omega_1}}=\kappa_{01}\frac{|\Omega_{12}^{(1)}|^2}{\Gamma_L\delta_1^2}\ ,$$

where we have evaluated $|\Omega_{01}^{(1)}|^2$ in terms of $F_{3\omega_1}$ and κ_{01}. Following Ref. [19], we have $|\Omega_{12}^{(1)}|^2=1.7\times10^{16}I_2$, where I_2 is the intensity of laser 2 in W/cm^2. We find

$$\Delta k=\Delta k_r+i\alpha\simeq-\frac{\kappa_{01}}{\delta_1}\left[1-i\frac{|\Omega_{12}^{(1)}|^2}{\delta_1\Gamma_L}\right]$$
$$\simeq-\frac{\kappa_{01}}{\delta_1}\left[1-i\frac{\Delta_S}{\Gamma_L}\right]\ , \tag{4.1}$$

where Δ_S is the ac Stark shift in $|2\rangle$ due to laser 2. Note that two-photon absorption can increase the magnitude of Δk by at most a factor of 2, since if Δ_S becomes large compared to the other widths that make up Γ_L (γ_2 and Γ_I), one must use $\Gamma_L\simeq\Delta_S$.

From the measured energy and transverse spatial FWHM of the beam of laser 2, and assuming a Gaussian intensity distribution, we find $I_2=1.5\times10^7$ W/cm^2 for the peak intensity; from our scans of the four-photon resonance we estimate $\Gamma_L\simeq1$ cm^{-1}. For $\delta_1=28.6$ cm^{-1} and $P_{\mathrm{Xe}}=4$ Torr (corresponding to the largest value of $\Delta k_r b/2$ used), we have

$$\Delta k=-171(1-i0.25)\ \mathrm{cm}^{-1}\ .$$

For this value of Δkb, the series expansion in (2.9) is valid. To suggest the effect of an imaginary part of Δk we have, to first order in V,

$$M\propto\left|\frac{1}{(\Delta k)}\right|^2=\frac{1}{\Delta k_r^2+\alpha^2}\ ,$$

so absorption of the magnitude we predict here should not have a large effect on M or R, although observation of the total (far-field) third harmonic might show a dip at four-photon resonance, since the large IR beam will cause absorption over a distance many times as large as b.

In addition, we note from (4.1) that two-photon absorption comparable in magnitude to Δk_r should be accompanied by observable ac Stark broadening of the four-photon resonance. In our data we see, in fact, a small decrease in the width of the four-photon resonance as δ_1 decreases from 50 to 20 cm^{-1} on the blue side of three-photon resonance rather than any increase. For these reasons, we expect the effect of absorption to be relatively small, and the observed suppression to be due mainly to large $\Delta k_r b$.

In Fig. 4 we see that as we approach the three-photon resonance from either the blue side (the right side of Fig. 4) or the red, the suppression of the four-photon resonance by the interference occurring at the three-photon level reaches very large values. In contrast, as we approach the phase-matched value of $-\Delta k_r b/2=2$, the suppression becomes negligible. As we tune closer to the three-photon resonance, the phase mismatch becomes larger, so destructive interference among components of the third-harmonic light generated at smaller values of z results in less net third-harmonic intensity. Expressed another way, the correlation length for third-harmonic generation $I_c=\pi/\Delta k_r$ becomes smaller, and an atom at any given position z sees a third harmonic produced over a shorter region. The first term in the expansion of the diffraction integral in Eq. (2.9) does not depend on the phase mismatch; it is the adiabatic polarization that follows (and cancels) the three-photon laser field exactly. So for large $\Delta k_r b$, ionization signals are strongly suppressed, since we have (1) little excitation by one third-harmonic photon plus one photon at ω_2, because the bad phase mismatch kills the propagating third harmonic, and (2) little excitation by laser photons, since the adiabatic part of the polarization at $3\omega_1$ cancels the contribution from three laser photons.

For $\Delta k_r b$ near the phase-matching point, the third-harmonic intensity can grow large enough to provide substantial pumping of the four-photon level by the absorption of one photon at $3\omega_1$ and one at ω_2; note that for equal intensities the two-photon process is vastly more likely than the four-photon $(\omega_1+\omega_1+\omega_1+\omega_2)$ process [20]. Thus near $-\Delta k_r b/2=2$ we see little evidence of suppression. In fact, in this region the ionization is significantly enhanced by the presence of the third-harmonic field.

V. HYBRID RESONANCE EXPERIMENT

In this section we describe an experiment closely related to the four-photon suppression experiment just described. In the absence of three-photon interference, a bright laser tuned close to a three-photon resonance will create some population in the three-photon level by pressure-induced wing absorption. If a second laser were tuned resonant-resonant between the three-photon level and a higher-lying dipole-coupled level, an enhancement in the MPI would be observed as a result of pumping of

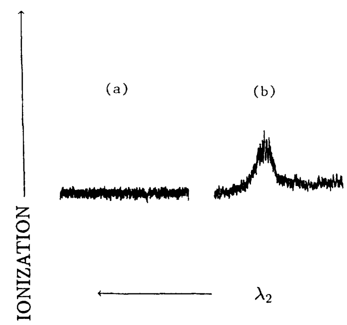

FIG. 5. Suppression of wing absorption into the three-photon resonance by the three-photon-interference effect. We scan λ_2 across the $6s \rightarrow 6p$ transition with the beam time delayed. λ_1 is tuned 19 cm^{-1} to the blue side of the $6s$ and $P_{Xe}=25$ Torr. In (a), λ_1 is not counterpropagated and the interference prevents absorption. In (b), counterpropagation overcomes the interference and MPI due to population in the three-photon state is seen.

population from the three-photon state to the higher-lying state and subsequent ionization. This is a classic "hybrid" resonance.

We tune the first laser 19 cm^{-1} to the blue of the $6s[\frac{3}{2}](J=1)$ three-photon transition. The second, infrared laser is now time delayed by 12 ns so that it no longer overlaps temporally with the first laser. Tuning the IR laser across the $6s[\frac{3}{2}](J=1)\rightarrow 6p[\frac{1}{2}](J=0)$ resonance should produce a peak in the MPI signal proportional to the population excited into the $6s$ during the blue-laser pulse; note that any population in the $6s$ is effectively trapped on the time scale of the delay between laser pulses due to the opacity of the medium. In Fig. 5 we show two scans of this sort taken with and without counterpropagation of the nearly three-photon-resonant laser. Here the xenon density is 25 Torr; I_1 is the same as before, while I_2 is about four times smaller. In Fig. 5(a), taken without counterpropagation, there is no ionization signal peak, while in Fig. 5(b), taken with the blue beam counterpropagated and the sensitivity reduced by a factor of 10, there is a prominent peak. This demonstrates clearly that the three-photon interference effect suppresses wing absorption into the three-photon level.

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