Two-color interference effect involving three-photon excitation and four-wave-mixing processes

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We have extended observations of total cancellation of three-photon atomic excitation by internally generated fields due to four-wave mixing. The process was found to occur as predicted in an excitation mode involving two photons of frequency ω_1 and one of frequency ω_2, plus the generated four-wave-mixing field at frequency $2\omega_1+\omega_2$. Experiments were conducted on the $6s$ state of Xe.

I. INTRODUCTION

Experimental[1-5] and theoretical[6-10] studies of three-photon excitation of atomic states which are also accessible by one-photon transitions have shown that the accumulated nonlinear response of individual atoms, through the internally generated third-harmonic field, can destructively interfere with the normal three-photon excitation process to completely cancel (to first order) any resonant excitation. Both theoretical and experimental studies have shown that the cancellation effect between three-photon and third-harmonic excitation persists under very general circumstances including excitation with pulsed broad-bandwidth lasers at gas pressures from a few mTorr to hundreds of Torr, and under pressure broadening through self-induced and buffer-gas interactions. It has also been shown that the cancellation effect is spoiled in three-photon excitation involving absorption of photons from two counterpropagating laser beams[4] of the same frequency, and the result has been qualitatively and quantitatively described theoretically.[10]

This rather interesting aspect of cooperative atomic response should be evident in any odd-photon resonant excitation which is also allowed by a one-photon process. More specifically, in the three-photon case the photons need not be of the same frequency. If laser beams of frequencies ω_1 and ω_2 are copropagated and tuned to three-photon resonance at $2\omega_1+\omega_2$, where the two frequencies are otherwise essentially arbitrary, then at elevated pressures, the four-wave mixing process at $2\omega_1+\omega_2$ should again interfere with three-photon excitation at resonance, and no resonant signal should be evident. If the two beams are counterpropagated the three-photon resonance should reappear (in ionization or fluorescence measurements, for example).

If we assume that light at ω_1 and at ω_2 is very far from any one-photon resonance such that ω_1 and ω_2 propagate with similar phase velocities, then the formalism of Refs. 7 and 10 can be easily extended to the more general circumstance involving excitation by two different frequencies. The phase mismatch between the laser beams and the generated field at $2\omega_1+\omega_2$ becomes

$$\Delta k = 2k_{\omega_1}+k_{\omega_2}-k_{2\omega_1+\omega_2}-i\beta$$

where k_{ω_i} is the wave vector for a field at frequency ω_i. The complex term is responsible for absorption of the generated photons, where 2β is the absorption coefficient. The formulas in Ref. 10 can be trivially extended to show that, at elevated concentrations,[10] the polarization $Y(z,t)$ at frequency $2\omega_1+\omega_2$ tends to zero at three-photon resonance unless the two frequencies are produced by beams which overlap at nonzero angle.

The present study is an addendum to the experimental findings reported in Ref. 5. Here we show that in resonantly enhanced multiphoton measurements at number densities above 10^{16} cm^{-3}, the cancellation effect involving two colors is essentially complete in copropagating lasers at three-photon resonance, and strongly evident in otherwise identical counterpropagating beams.

II. METHOD AND RESULTS

Resonance ionization measurements were made at energies corresponding to $\hbar(2\omega_1+\omega_2)=E_{6s}$ in Xe, where E_{6s} is the energy of the $5p^5 6s[\frac{3}{2}]_1^0$ level. The basic experimental apparatus has been described earlier.[5] In the present instance a Lumonics 861 excimer laser, with 4 ns, 80 mJ pulses was used to pump two dye lasers, as shown in two configurations in Fig. 1. The first dye laser, a modified Molectron DL14, was set at fixed frequency (200 μJ/pulse with QUI dye) while the second dye laser, a Lumonics EPD 330, operated with rhodamine B at 1 mJ/pulse, was scanned through three-photon resonance at $2\omega_1+\omega_2$. Multiphoton ionization measurements were made in a shielded proportional counter. This and the data acquisition system have been described earlier.[5]

In Fig. 2 are shown ion signal traces from three different experiments in which laser one was set at 387.90 nm while laser two was scanned from $\sim$606.1 to 607.7 nm. Under this circumstance three-photon excitation of the $6s$ state can occur through $2\omega_1+\omega_2$ absorption at $\lambda_2=606.576$ nm. The xenon pressure was fixed at 250 Torr, where earlier studies involving single-color excitation have shown that strong pressure broadened ionization signals can be obtained.[5]

The top trace in Fig. 2 represents the resonant ion yield when laser two is tuned through the three-photon Xe $6s[\frac{3}{2}]_1^0$ resonance (at $\lambda_2=606.6$ nm) in the *counterpro-*

pagating geometry shown in Fig. 1. In this configuration there is no traveling wave nonlinear polarization at $2\omega_1+\omega_2$, and normal three-photon resonant excitation of the state is expected, as observed.

In the bottom trace of Fig. 1 we show a similar scan of laser two but in this case the laser beams at ω_1 and ω_2 are copropagated through the use of a beam combining dichroic mirror as shown in Fig. 1. In this beam configuration there is no hint of a resonant enhancement of the ionization signal at the three-photon resonant wavelength. Indeed, the cancellation of three-photon excitation by the four-wave mixing process at $2\omega_1+\omega_2$ is complete within the error limits of the experiment. This result is in complete agreement with our theoretical prediction.

The total absence of a resonantly enhanced signal, as in the bottom trace, could be considered less than convincing that the predicted cancellation effect is responsible for the observed result. As a check on our experimental arrangement (beam overlap, etc.) we inserted a mirror in the exit region of the copropagating beams to retroreflect *both* frequencies. This procedure produced the result shown in the middle trace of Fig. 2. That is, the resonant enhancement reappeared at 606.576 nm as expected, indicating that the absence of resonantly enhanced ionization in the single-pass copropagating experiment (bottom trace) is not an artifact but is associated with the predicted cancellation effect. (With counterpropagating beams, as in the middle trace, the cancellation effect is not operative in three-photon excitation through all combinations of $2\omega_1+\omega_2$ where two photons are taken from one direction of propagation and one from the opposite direction.)

Finally, in order to show explicitly that the process under observation is indeed three-photon excitation of the $6s$ state, as described, we repeated the experiments after setting the fixed frequency laser at a new wavelength. The examples shown in Fig. 3 correspond to a setting of $\lambda_1=387.743$ nm for laser one, in which case three-photon excitation occurs at $\lambda_2=607.346$ nm. The results under the conditions indicated in the figure caption conform exactly to prediction.

III. CONCLUSIONS

We have extended previous results to show that the nature of the nonlinear atomic response which produces complete cancellation between three-photon and one-photon excitation of atomic states, where the one-photon process arises from internally generated third harmonic fields, is also applicable to the more general case involving two-color excitation. If two different frequencies are used

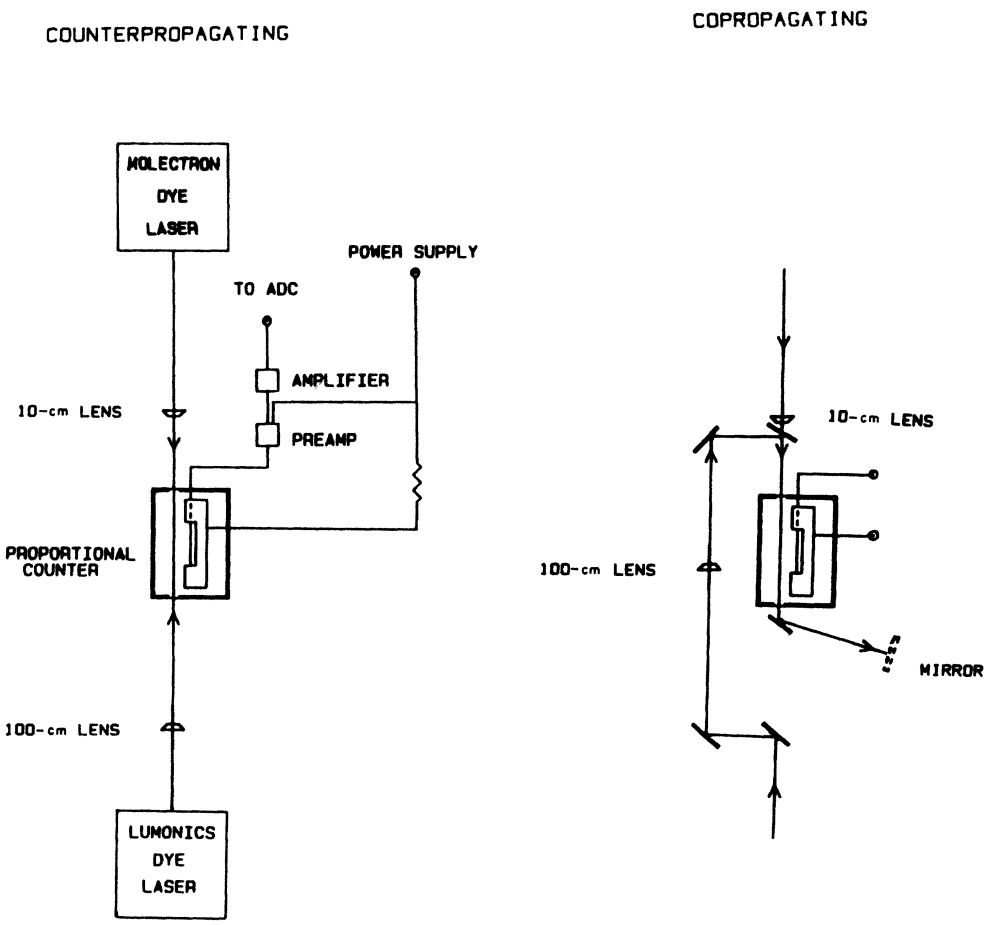

FIG. 1. Experimental arrangements for counterpropagating and copropagating geometries. In the copropagating arrangement a retroreflecting mirror, M, shown as a dashed line in the figure, was added for one set of experiments.

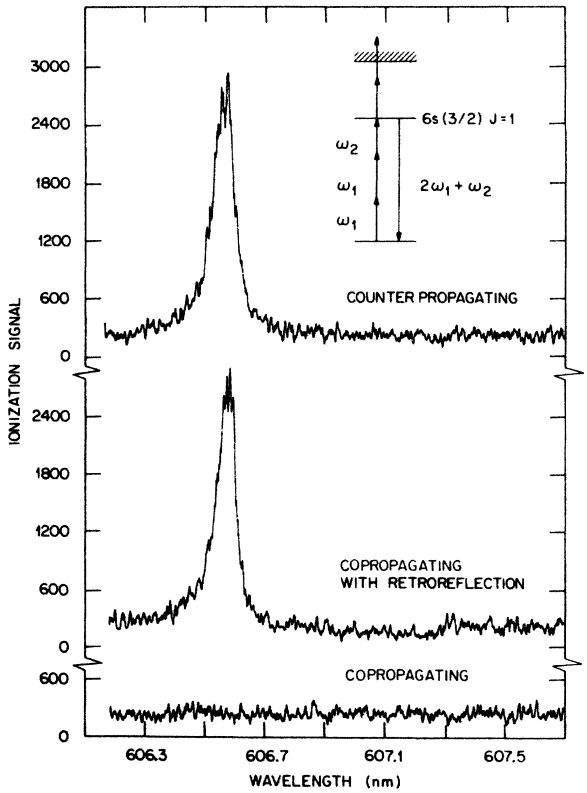

FIG. 2. Resonance ionization signals obtained through three-photon excitation of Xe $6s[\frac{3}{2}]^0_{J=1}$ in three different geometries. Top trace, counterpropagating beams. Bottom trace, copropagating single pass geometry. Middle trace, copropagating beams with retroreflecting mirror to create counterpropagating beams at both frequencies. Insert shows excitation scheme and subsequent two-photon ionization out of the resonance level. Laser one set at 387.900 nm, laser two tuned through wavelength shown on abscissa.

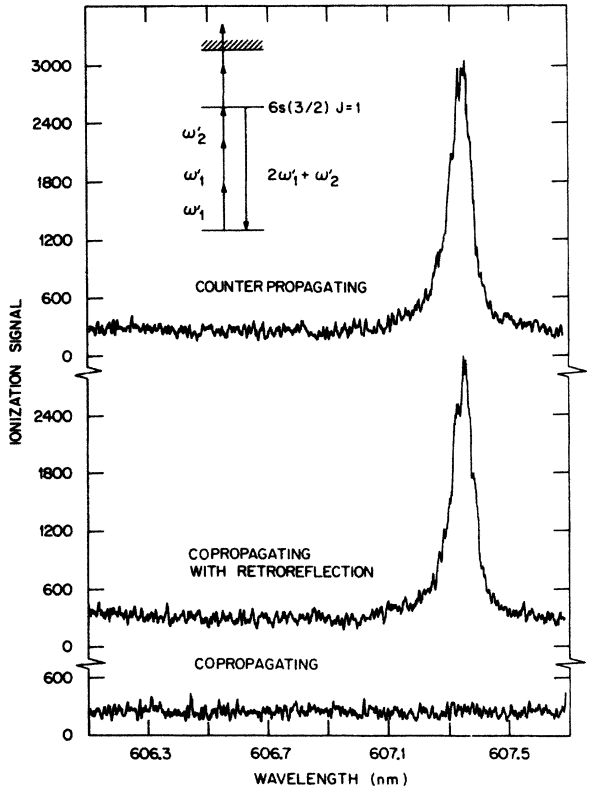

FIG. 3. Same as Fig. 1, but for laser one set at 387.743 nm.

in the three-photon excitation, then the internally generated four-wave mixing field at $2\omega_1+\omega_2$ again destructively interferes with the direct normal three-photon excitation process if the geometry is such that a traveling wave polarization is generated at the sum frequency.

We have demonstrated the rather novel appearing feature of two broadband laser beams at frequencies ω_1 and ω_2 which can excite a three-photon resonance at $2\omega_1+\omega_2$ when traveling in opposite directions, but show no measurable excitation signal when copropagating under otherwise identical conditions. The present results are all in agreement with earlier theoretical predictions,[7,10] and they represent an addendum to earlier experiments[5] which were designed to test several features of the detailed theoretical description of this subject.

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