Interference Effects between Different Optical Harmonics

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(Received 17 June 1982)

It is shown that destructive interference between two coherent excitation pathways from different optical harmonics, a fundamental and a third harmonic, inhibits resonance-enhanced multiphoton ionization. Once the role of the third harmonic is realized, the interference follows naturally from simple considerations of Fermi's "golden rule" and Maxwell's equations.

PACS numbers: 32.90.+a, 41.10.Hv, 42.10.Jd, 42.10.Mg

In studying five-photon multiphoton ionization (MPI) of Xe I at vapor pressures >1 Torr, Aron and Johnson[1] focused attention on the unexpected and (previously) unexplained *absence* of a three-photon resonance enhancement by the $5p^5(^3P_{3/2}{}^{\circ})6s$ $J=1$ state (labeled 6s) at 68 045.67 cm^{-1}. In contrast, working at lower pressures (<1 Torr), Miller *et al.*[2] observed resonance-enhanced MPI near the 6s state. They noted the gradual weakening and disappearance of MPI with increasing pressure, while third-harmonic generation (THG), monitored concurrently, showed the opposite behavior. They initially interpreted their results in terms of a complex theoretical model, based on collective emission, in which enhanced THG competes with MPI. Subsequently, they developed a model based on the fact that the three-photon Rabi flopping frequency and the one-photon Rabi flopping frequency are equal and opposite near resonance.[3]

In this paper we demonstrate that the *apparent* competition is due to a *simple* interference between two different, *but coherent,* pathways to the 6s state: (1) a three-photon excitation driven by the electric field at the fundamental (laser) frequency ω_1, and (2) a one-photon excitation driven by the electric field at the third-harmonic frequency $\omega_3=3\omega_1$. The key experimental observation, which suggested that interference plays an important role, was reported by Glownia and Sander.[4] They discovered that *counterpropagating, circularly polarized* laser beams produced resonance-enhanced MPI *at* the 6s state at all pressures. Further, we obtained the same result, even with *linearly polarized* light (Fig. 1). These experiments demonstrate that the standing-wave excitation allows resonance-enhanced MPI. We will show that this result occurs because the magnitude of pathway (2) is diminished for a standing-wave excitation. Systematic studies characterizing pathway (2) and verifying its importance *away from resonance* will be presented in a subsequent publication.[5] The present paper shows that pathway (2) is important *on resonance.* This result is applicable to *any* one-photon, dipole-allowed transition, not just to the ground to 6s transition in Xe I.

Given pathway (2), explaining the anomalous on-resonance behavior reduces to a straightforward, but general, calculation of the MPI rate, $W_{fg}{}^{\mathrm{tot}}(5\omega_1)$, by Fermi's "golden rule":

$$W_{fg}{}^{\mathrm{tot}}(5\omega_1)=\frac{2\pi}{\hbar^2}\rho(5\omega_1)\left|\sum_k\frac{[ex_{fk}E(\omega_1)][ex_{kR}E(\omega_1)]}{\hbar(\omega_{kg}-4\omega_1)\hbar(\omega_{Rg}-3\omega_1)}\right|^2|\langle R|ex\mathfrak{F}^T(3\omega_1)|g\rangle|^2, \quad (1)$$

where $E(\omega_i)$ is the electric field at ω_i polarized in the x direction, $\rho(5\omega_1)$ is the density of final states, $x_{ij}=\langle i|x|j\rangle$, $\omega_{jg}=\omega_j-\omega_g-i\Gamma_{jg}/2$, $\Gamma_{jg}/2$ is the linewidth, and $\hbar(\omega_j-\omega_g)$ is the energy of the jth state relative to the ground state. In Eq. (1), $\mathfrak{F}^T(3\omega_1)$ represents the *net* transverse effective field at $3\omega_1$ driving the coherent polarization of the ground state to $|R\rangle$ transition by both pathways:

$$\langle R|ex\mathfrak{F}^T(3\omega_1)|g\rangle=\left(\sum_{mn}\frac{[ex_{Rm}E(\omega_1)][ex_{mn}E(\omega_1)][ex_{ng}E(\omega_1)]}{\hbar(\omega_{mg}-2\omega_1)\hbar(\omega_{ng}-\omega_1)}\right)+ex_{Rg}E(\omega_3). \quad (2)$$

(We remind the reader that $\omega_3=3\omega_1$.) Note that the correct treatment of coherence is to sum the pathways *before* squaring!

The third-harmonic field $E(\omega_3)$, produced by a traveling plane wave $E(\omega_1)=\frac{1}{2}E_t(\omega_1)\exp[i(k_1z-\omega_1t)]$, is calculated from the wave equation with a nonlinear source term $P^{\mathrm{NLS}}(3\omega_1)=\frac{1}{2}P_t{}^{\mathrm{NLS}}(3\omega_1)\exp[i(3k_1z$

$-3\omega_1 t)]$:

$$\nabla^2 E(\omega_3) + \epsilon_3(\omega_3/c)^2 E(\omega_3) = -4\pi(\omega_3/c)^2[P_t^{\rm NLS}(3\omega_1)/2]\exp[i(3k_1 z - 3\omega_1 t)] \tag{3}$$

and has a solution of the form

$$E(\omega_3) = [E_t(\omega_3)/2]\exp[i(3k_1 z - 3\omega_1 t)] + (A_t/2)\exp[i(k_3 z - \omega_3 t)], \tag{4}$$

where $k_i = \epsilon_i^{1/2}\omega_i/c$, ϵ_i is the dielectric constant at ω_i, $E_t(\omega_3)$ is the amplitude of the particular solution (i.e., the driven response to the nonlinear polarization which propagates with the laser at a velocity $c/\epsilon_1^{1/2}$), and A_t is the amplitude of the homogeneous solution (i.e., the natural solution to the wave equation which propagates at $c/\epsilon_3^{1/2}$).[6] It is the interference of these two components that governs the net generation of $E(\omega_3)$ in the traditional treatments of THG.[7] When the nonlinear medium is assumed to be optically thin at ω_3, the homogeneous wave (A_t) propagates unattenuated. In contrast, in an optically thick medium (the situation that holds on resonance), the homogeneous wave is attenuated over a distance on the order of one absorption depth, leaving only the driven wave[6]

$$E_t(\omega_3) = 4\pi P_t^{\rm NLS}(3\omega_1)/(\epsilon_1 - \epsilon_3). \tag{5}$$

The appropriate expression for $P_t^{\rm NLS}(3\omega_1)$ when the major resonant contribution to the nonlinear response $[\chi^{(3)}(3\omega_1)]$ comes from the transition from the ground state to $|R\rangle$ is

$$P_t^{\rm NLS}(3\omega_1) = \frac{\chi^{(3)}(3\omega_1)}{4}E_t^3(\omega_1) = \frac{Ne^4}{4\hbar^3}\left(\sum_{mn}\frac{x_{gR}x_{Rm}x_{mn}x_{ng}}{(\omega_{Rg}-3\omega_1)(\omega_{mg}-2\omega_1)(\omega_{ng}-\omega_1)}\right)E_t^3(\omega_1), \tag{6}$$

where N is the atomic density. Furthermore, with the major resonant contribution to the linear response also coming from the ground-to-$|R\rangle$ transition, we have

$$\epsilon_1 - \epsilon_3 \cong -4\pi N e^2|x_{Rg}|^2/\hbar(\omega_{Rg}-\omega_3). \tag{7}$$

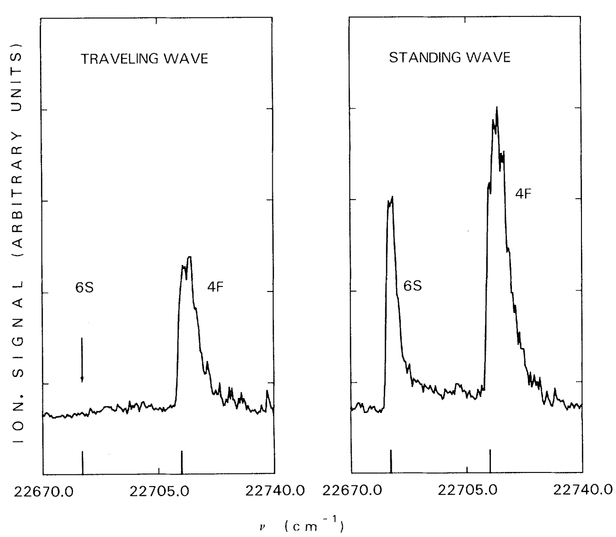

FIG. 1. Typical spectrum showing Xe I 6s and 4f ionization signals vs laser wave number when traveling-wave and standing-wave excitations are applied. The incoming power (77 kW in both scans) is retroreflected to produce the standing wave. The pressure is 1.5 Torr and the beam area at the focus is 1.8×10^{-7} cm^2. For reference, the unperturbed 6s and 4f resonance positions are indicated by tick marks below.

Substituting Eqs. (6) and (7) into Eq. (5), one sees that $E_t(\omega_3)$ reverses sign, putting it 180° out of phase with $E_t^3(\omega_1)$! With the substitution of $E(\omega_3)$ into Eq. (2), the matrix elements for the two pathways exactly cancel, exhibiting complete destructive interference. As a consequence, the resonance-enhanced MPI, calculated from Eq. (1), vanishes. Note that this result is independent of laser intensity $[E^2(\omega_1)]$ and gas pressure (N).

An alternative way of interpreting this formulation involves rewriting Eq. (5) as $E_t(\omega_3) \approx 2(\omega_3/c) \times [P_t^{\rm NLS}(3\omega_1)]l_{\rm coh}$, where the coherence length $l_{\rm coh}$ is the distance over which fields at ω_1 and ω_3 travel before dephasing. For the strongly absorbing case, $l_{\rm coh}$ is limited to the linear absorption depth. This depth, with its dependence on the matrix element x_{Rg} and the energy denominator $\omega_{Rg}-\omega_3$, "normalizes" the term $ex_{Rg}E(\omega_3)$ in Eq. (2) such that $\mathfrak{F}^T(3\omega_1)$ vanishes, independent of x_{Rg} or $\omega_{Rg}-\omega_3$. Physically, this means that *no work* is done by the fields to polarize the atom via the ground state to $|R\rangle$ transition.

In all of the above-referenced multiphoton-ionization experiments[1,2,4,5] the laser beam was focused into the gas. Nevertheless, the absorption depth, on resonance, was always shorter than the distance over which there were significant changes in the beam area and intensity. Therefore, propagation effects can be ignored; $E(\omega_3)$ has a local dependence on $E^3(\omega_1)$ so that the above plane-wave solution is applicable. This, then, explains the absence of resonance-enhanced

MPI in the traveling-wave experiments.[1,2,4,5]

For a standing-wave excitation, the startling appearance of resonance-enhanced MPI *via* the $6s$ state occurs because destructive interference between pathways (1) and (2) is *incomplete.* For a standing wave, we have $E(\omega_1) = E_s(\omega_1)(\cos k_1 z) \times \exp(-i\omega_1 t)$, and the nonlinear source term at $3\omega_1$ is

$$P^{\rm NLS}(3\omega_1) = P_s^{\rm NLS}(3\omega_1)(\cos k_1 z)^3 \exp(-i3\omega_1 t).$$

In the optically thick case, the solution of the driven-wave equation decomposes into two spatial Fourier components:

$$E(\omega_3) = \pi P_s^{\rm NLS}(3\omega_1)\left(\frac{\cos 3k_1 z}{\epsilon_1 - \epsilon_3} + \frac{3\cos k_1 z}{(\epsilon_1/9) - \epsilon_3}\right)\exp(-i3\omega_1 t). \tag{8}$$

The correct expression for $P_s^{\rm NLS}$ is

$$P_s^{\rm NLS}(3\omega_1) = \chi^{(3)}(3\omega_1)[E_s(\omega_1)]^3, \tag{9}$$

where $\chi^{(3)}$ is given in Eq. (6). [Note that this definition of $P_s^{\rm NLS}$ differs from that of Eq. (6) by a factor of 4 due to the way in which the amplitudes are defined for traveling and standing waves.] On substituting into Eq. (2), one again finds complete destructive interference for the spatial Fourier component at $3k_1$. On the other hand, the spatial Fourier component of $E(\omega_3)$ at k_1 contains a denominator, $(\epsilon_1/9) - \epsilon_3 \simeq \frac{8}{9}$, which no longer "normalizes" the term $ex_{Rg}E(\omega_3)$. Hence, the delicate balance between the two pathways, which is preserved for $3k_1$, is destroyed for k_1.

Interpreting Eq. (8) in terms of coherence lengths for each spatial Fourier component gives

$$E(\omega_3) = (\omega_3/2c)P_s^{\rm NLS}(3\omega_1)\{[(l_{\rm coh}^{(3k_1)})(\cos 3k_1 z) + 3l_{\rm coh}^{(k_1)}(\cos k_1 z)]\exp(-i3\omega_1 t)\}.$$

The component of $E(\omega_3)$ at $3k_1$ is still limited by the absorption depth. In contrast, the component at k_1 is proportional to a *much* shorter coherence length, on the order of half a wavelength. [For Xe I, knowing the oscillator strength for the ground-state-to-$6s$ transition ($f = 0.27$), one can calculate the relative amplitude of the contribution of the Fourier components of $E(\omega_3)$. At 1-Torr pressure, the ratio of the component at k_1 to that at $3k_1$ is $\sim 3\times10^{-2}$.] Thus the component of $E(\omega_3)$ at k_1 is negligible, leaving a sizable contribution from pathway (1), at k_1, to $\mathfrak{F}^T(3\omega_1)$. Physically, this means that work *is* done by the fields in polarizing the atom *via* the ground-state-to-$|R\rangle$ transition. The resulting net polarization is a stepping stone in the path to ionization. This explains the appearance of resonance-enhanced MPI, on resonance, for a standing-wave excitation.[4]

Another physical interpretation of this problem may be obtained by viewing it in terms of the widely used vector model[8] for a two-level system. In this model, the traveling-wave case corresponds to having the pseudovector always pointing straight down, a position corresponding to zero transverse polarization and all of the atoms in the lower level. This position is stable because the two opposing transverse forces applied to the pseudovector are self-adjusting, maintaining an exact balance [i.e., $\mathfrak{F}^T(3\omega_1) = 0$]. For the standing-wave case, the self-adjusting balance is destroyed [i.e., $\mathfrak{F}^T(3\omega_1) \neq 0$], allowing the pseudovector to tip, producing polarization and, hence, ionization.

In conclusion, we have shown the existence of a destructive interference mechanism between two coherently superimposed pathways from fields at two different harmonics, one at the fundamental (ω_1) and one at the third-harmonic (ω_3) frequency. This interference is completely destructive for a traveling-wave excitation. This suggests that the nonlinear medium tries to minimize its polarization energy. For the optically thick medium, once a steady-state condition is reached, there is no net polarization due to the transition from the ground state to $|R\rangle$, even though an electric field at the third harmonic exists.

We thank Dr. D. S. Bethune and Dr. H. Zacharias for helpful discussions.

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Effect of a Static Electric Field on the Trapping of Beam Electrons in a Slow Wave Structure

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(Received 3 May 1982)

The effect of an applied static electric field on trapped beam electrons in a traveling wave tube has been observed. In particular, it was found that the wave power can be increased. It was found that beam space charge can play an important role in limiting the wave-power enhancement, and that the wave enhancement is strongly dependent on the rf input drive level. By launching large-amplitude waves over 10 dB of enhanced wave growth has been observed.

PACS numbers: 41.70.+t, 42.60.-v, 52.40.Mj. 85.10.Hy

In a previous Letter[1] it was predicted that if an external force is applied to particles executing nonlinear trapping oscillations in a plasma wave, the wave power can be increased. We have observed this effect in a traveling wave tube[2] (TWT). The equations[3] that describe the wave-particle interaction in a TWT are identical to those[4,5] that describe the beam-plasma instability in the small-cold-beam limit. The effect of a static electric field on trapped beam particles has been studied theoretically both in a small-cold-beam–plasma system and in a TWT. In the beam-plasma system the theory was first done by Morales.[1] In the TWT, it was done by Hess.[6]

Although the physics of this effect in this highly nonlinear system is interesting in and of itself, there are more practical reasons for studying it as well. One of the reasons that motivates the study of this effect in the beam-plasma case is that it provides a possible model for the interaction of runaway electrons with cavity modes in a tokamak.[1] The study of this effect in the TWT case is motivated by the possibility of enhancing[7] the wave growth past saturation in TWT's. Indeed, a closely related enhancement technique, velocity tapering, has been well studied[7] theoretically and experimentally. In addition, because of the analogy between free-electron lasers and TWT's, recent ideas[8] concerning power enhancement in free-electron lasers may also stimulate interest in the study of this effect.

The main qualitative features of the effect can be understood by considering the following simplified physical picture. If a weak force is applied to particles trapped in the potential well of a wave of essentially fixed phase velocity, the response of the particles cannot be a uniform acceleration because they are constrained to move on the average at the wave phase velocity. Since the particles cannot change their momentum in response to the applied force, the wave responds by changing its momentum. And because the wave power is proportional to the wave momentum, the wave power can be increased in this way. If the applied force is strong enough to detrap the particles, the particles accelerate, and the wave-power enhancement is destroyed.

We have observed these effects in a TWT. The effects are also predicted by our computer simulations and the computer solutions agree well with the experiment. The apparatus, which has been described elsewhere,[9] differs from most conventional TWT's in that it is 3–4 times longer when measured in scaled units. A cold electron beam is directed down the axis of a wire helix slow wave structure which is held together by a support structure and is enclosed by a glass vacuum tube. Outside of the glass tube are electrostatic probes which are used to transmit and receive radio-frequency waves. This assembly is enclosed by a grounded cylindrical conductor which is slotted so that the probes can be moved axially. The grounded cylinder acts as a waveguide beyond cutoff and insures that waves

