Quantitative criterion for cooperative or continuum response from an atomic assembly

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The purpose of this paper is to define the lowest pressure at which an atomic gaseous medium gives a cooperative or continuum response to an electromagnetic stimulus. This is done by examining the conditions for sufficient third-harmonic generation by the resonant atomic assembly. It is found that at such low pressures one would have 25 particles per $\lambda/10$-thick slice of the characteristic volume of the focused fundamental beam. The question of getting a cooperative response from an assembly of H atoms at such low pressures that strong interactions between two H atoms may be minimized is discussed. The criterion is also applied to get a cooperative response from the various nuclei in interaction with γ rays.

Recently a number of experimental and theoretical studies on multiphoton ionization of atomic gases have demonstrated the critical role played by the third-harmonic field generated inside the dense gas. These studies employ a pair of three-photon resonant states of the atomic gas which also have a one-photon allowed dipole moment. Careful experiments by Miller *et al.*[1] have established that the competition between multiphoton ionization and third-harmonic generation (THG) has a threshold nature. Below a critical pressure only the multiphoton ionization signal is observed and above the critical pressure, as the pressure is increased, the ionization signal at resonance starts to decrease, its excitation spectrum shifts towards the blue side of the resonance, and it broadens significantly. On the other hand, the third-harmonic signal starts to show up, its excitation spectrum also shifts towards the blue side of the resonance, and is broad. Payne, Garrett, and Baker[2] have shown that the critical pressure depends on the magnitude of the ac Stark shift, the laser band width, the diameter of the unfocused beam, and the focal length of the lens. It has been suggested[3] that the critical-pressure behavior of the competition between third-harmonic generation and multiphoton ionization separates the two regions called the single-atom-response and the continuum- or cooperative-response region. In the single atom response region, the number of atoms is so small that each atom interacts independently with the laser light, E_1, and the third-harmonic light, E_3, which is generated by the process of hyperresonance fluorescence. The TH light escapes from the sample without a significant effect on the atoms of the rest of the sample. On the other hand, in the continuum-response region the number of atoms in the sample is large. Each atom interacts with the third-harmonic field generated at its position by the rest of the atoms of the sample. The propagation of the third-harmonic field in the sample is then obtained by smoothing the fields over small volume elements. It is assumed in such a theoretical program that the number of atoms in the volume element is large enough to justify such a smoothing procedure. The number required is a question difficult to answer *a priori*. One could perhaps begin by allowing only two atoms within one wavelength, or fix another arbitrary number, say 10.

The experiment of Miller *et al.* gives us a basis to support such an argument. Note that the reduction in the resonant multiphoton ionization signal and its complete absence at high pressures in the experiments have been explained,[4] due to destructive interference between two paths of excitation of two-level atoms. One path of excitation is due to absorption of three photons of the fundamental beam and the second path is due to the absorption of one photon of the generated third harmonic. At the pressure where these two paths exactly cancel, the two-level atoms are found to stay effectively in their ground state. This leads to no multiphoton ionization signal. This also implies no fluorescence in the perpendicular direction—as was observed by Faisal *et al.*[4,5] Our numerical computations have shown[6] that soon after the approach of the pressure of exact cancellation, some phase-matched third-harmonic generation can take place in the forward direction at the blue side of the resonance. This has also been the observation of Miller *et al.* We made use of this fact and the prevailing reduced fluorescence to build an argument to develop a threshold pressure at which cooperative or continuum response can build up. We note that the efficiency of third-harmonic generation depends on the phase mismatch between the forced and the free waves. We then argue that, since the phase mismatch is determined in terms of the refractive index of the atomic gas, and since the refractive index is a bulk or continuum property of the gaseous sample, the threshold pressure at which the continuum response is manifested is the lowest pressure at which the phase-matching condition for a sufficient third-harmonic generation can be achieved.

We now proceed to define quantitatively what we mean by sufficient THG. Using standard procedure one finds that the envelope of the generated third harmonic at a point z in the gaseous sample is given by

$$E_3(xyz)=-\frac{i\Delta kb}{2}\left[\frac{K_{Rg}\epsilon^3}{d_{Rg}}e^{-3k(x^2+y^2)/1+i\beta}\right]\times F(\Delta kb,\zeta,\beta)\ , \quad (1)$$

where F is the phase-matching integral given by

$$F(\Delta kb,\zeta,\beta)=\int_{\zeta}^{\beta}\frac{e^{[-(i\Delta kb/2)(\beta'-\beta)]}}{(1+i\beta')^2}d\beta'\ . \quad (2)$$

The incident light of frequency ω has been assumed to be in the fundamental Gaussian mode with confocal parameter $b=k\bar{\omega}_0^2$ where k is the one-dimensional wave vector of the incident beam in the direction of propagation z and $\bar{\omega}_0$ is the diameter of the beam at its focus in the middle of the cylindrical sample. $\beta=[2(z-f)/b]$ with $\zeta=\beta$ at $z=0$. $K_{Rg}\epsilon^3$ is the slowly varying part of the three-photon effective Hamiltonian in the rotating wave approximation between the ground state g and the excited state R. d_{Rg} is the one-photon allowed dipole moment between the same states. The phase matching parameter $\mathrm{Re}\Delta kb$ and the absorptive parameter $\mathrm{Im}\Delta kb$ are dependent on the atomic density n, and the two-level line shape, Γ, through the equation

$$\Delta kb=\frac{6\pi n\omega\,|d_{Rg}|^2 b}{\hbar c(\delta-i\Gamma)},\quad \delta=\frac{E_R-E_g}{\hbar}-3\omega\ . \quad (3)$$

Under tight and symmetric focusing, the conditions for efficient THG can be determined[7] analytically from Eq. (1) and are given by

$$\mathrm{Im}\Delta kb\approx 0\ ,\qquad \mathrm{Re}\Delta kb=-4\ . \quad (4)$$

These are well known from the theory of third-harmonic generation. In the situation of three-photon resonant levels with allowed dipole transition ($\Delta l=\pm 1$) one notes that these conditions are satisfied at large pressures on the far blue side $\delta=-|\delta|$ of the resonance, i.e., at $|\delta|\gg-|\Gamma|$. At lower pressures the absorption of the generated third harmonic becomes significant at the detuning at which $\mathrm{Re}\Delta kb=-4$ is satisfied. One must then examine the conditions of efficient THG in the presence of absorption. We have analyzed[6] the function

$$G(\Delta kb)=|\Delta kbF(\Delta kb)|^2 \quad (5)$$

numerically for a range of values of the absorption parameter $\mathrm{Im}\Delta kb$. It has been found that the peak height which is $\simeq 12$ at $\mathrm{Im}\Delta kb=0$ reduces sharply to $\simeq 1$ when $\mathrm{Im}\Delta kb\simeq 0.3$. At values of $\mathrm{Im}\Delta kb>0.3$ the nature of the curves shows marked changes because the absorption length becomes of the order of the confocal parameter and Maker fringes start to determine the coherence length of the medium.

One notes that at some very low pressure, $\mathrm{Re}\Delta kb=-4$ can be satisfied at $\delta=-\Gamma$. This detuning corresponds to the minimum of the dispersion curve and the lowest pressure at which this happens is given by

$$n=\frac{12\pi}{\left[\frac{\lambda\pi\bar{\omega}_0^2}{4}\right]}\frac{\Gamma}{\gamma};\quad \gamma=\frac{2}{3}\frac{|d|^2\omega_v^3}{hc^3}\ . \quad (6)$$

The value of $\mathrm{Im}\Delta kb$ is -4 at the conditions of Eq. (6). Thus $|G|$ remains much less than unity. Hence, conditions of Eqs. (6) are not used to define the region of sufficient third-harmonic generation. Nevertheless, it is worth noting that the number density (n) in (6) is scaled by the characteristic volume element ($\lambda\pi\bar{\omega}_0^2/4$) which may be viewed as the volume of a cylinder of length λ along the direction of propagation and an area determined by the beam waist $\bar{\omega}_0$. Concerning the ratio Γ/γ one notes that if it is unity Eq. (6) determines a unique number of 12π particles in the characteristic volume element of the tightly focused beam. This number may sound satisfactorily large to ensure small fluctuations, of the order of $1/N\sim 1/12\pi$, determined by the system size. It may be of interest at this stage to recall that while attempting a smoothing procedure in a medium with pencil-shaped geometry, one divides the sample into thin vertical slices of width, in the direction of propagation, less than the wavelength λ, and assumes sufficiently large number of particles in each slice.[2,8] This is done to ensure dipole approximation for each atom interacting with the field, and to avoid any fluctuation effects that may arise due to a small number of particles in the slice. For the situation of Eq. (6) one observes that each $\lambda/10$ slice of the characteristic volume has approximately four particles. In such a situation only the forced component of the polarization wave survives—the free wave component is virtually absent. This is the pressure range at which the destructive interference in forward direction starts to exist.

We suggest that the conditions of noticeable and sufficient third-harmonic generation may be taken to be

$$\mathrm{Im}\Delta kb=0.3\ ,\qquad \mathrm{Re}\Delta kb=-4\ . \quad (7)$$

These equations determine a value of detuning $\delta=-13.3\Gamma$ and a pressure

$$n\simeq\frac{80\pi}{\left[\frac{\pi}{4}\lambda\bar{\omega}_0^2\right]}\frac{\Gamma}{\gamma}\ . \quad (8)$$

For the condition of the experiments of Miller *et al.*,[1] one has $\lambda=440.8$ nm, $\bar{\omega}_0=20\ \mu m$, $\delta\simeq 68$ GHz, and $\gamma=0.22$ GHz. From (8) one finds $n=5.6\pm 10^{14}/\mathrm{cm}^3$ which is in fair agreement with their observed pressure lying between 10^{-3} and 0.05 Torr at which they observe the starting of the shift to the blue side of the third harmonic as well as the ionization signal. At these conditions there are as many as 750 particles per ($\lambda/10$) slice of the characteristic volume of the tightly focused beam. This number is fairly large to ensure that the assumption of smoothing is valid. It may be noted, however, that this large number is a result of the large Γ/γ ratio in the experiments. It is due to multimode nature of laser light, Stark broadening under conditions of tight focusing of intense beams, and collisional broadenings. Some of these limitations could be lifted, for example, by choosing single mode laser and reducing the intensity of focussed light to reduce Stark broadening. A reduction in the other limitation may then follow, as then the study can be done at still lower pressures.

A suggestion of the present study therefore is to do experiments to achieve the limit of observing sufficient THG at a particle number of 25 in one $\lambda/10$ slice of the characteristic volume of a focused beam.

We may attempt to make use of the same criterion to determine the existence of cooperative or continuum conditions for other regions of wavelength. Consider, for example, the problem of generating an intense beam of Lyα (1216 Å) radiation. The neighborhood of this wavelength has potential applications in plasma diagnostics[9] and H_2-molecule spectroscopy. Various methods of generating this range of wavelength by four-wave mixing have been demonstrated and interested readers may consult a recent review by Vidal.[10] Here we are interested in the possibility of generating this range of wavelength by making use of three-photon resonant atomic H. One major problem[11] in using H-atomic gas as a nonlinear medium is that at fairly low atomic densities two H atoms interact to form an H_2 molecule. The half-life of a H atom decreases with increasing pressure of the atomic gas. At about 0.2 Torr pressure the lifetime of the H atom is about 1 s.[12] This time is long enough to allow experiments to be performed with a laser pulse of a few nanoseconds duration. A relevant question for the success of these experiments is whether the system will give a cooperative response at the range of pressure at which H atoms have long lifetimes. On the basis of Eq. (8) and assuming condition of tight focusing similar to that of experiments on Xe, we get $n \simeq 7.54\times10^{14}$ particles per cm^3. In a pressure range of 0.02–0.2 Torr one could expect a continuum-response region for sufficient and efficient THG at a required wavelength. It may be recalled that requirement of negative phase matching for single component H-atom gas can be met by controlling phase matching by an additional resonant beam as suggested recently.[6] The second color also gives a tunability control over the wavelength for efficient third-harmonic generation.

Another example where Eq. (8) can be used to test for a cooperative response is in the attempt to investigate the cooperative response in the range of γ rays. Taking 0.01 nm to be the typical γ ray wavelength and assuming focusing geometry such that the confocal parameter $b \simeq 1$ cm which would in turn imply $\bar{\omega}_0 \simeq 1$ μm, one would then require a concentration of 10^{34} nuclei per cm^3 to take part in a cooperative response. Achieving such a density of atomic nuclei appears to be a formidable task but this density might occur in some neutron stars. It may then be of interest to attempt to look for coherent γ rays from such objects in the universe. Problems concerning γ ray lasers have recently been noted by Lipkin.[13]

In conclusion, we have given a quantitative criterion to determine if a particular low-pressure assembly of atoms will give a cooperative or continuum response to an electromagnetic stimulus. The criterion is based on THG in media which are in resonance with generated third harmonic. It is shown that this criterion can be used to furnish relevant informations in two typical situation of interest.

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