Control of Phase Matching and Nonlinear Generation in Dense Media by Resonant Fields

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We show that the atomic dispersion and therefore the efficiency of the nonlinear generation at a given frequency can be controlled by the addition of an extra resonant laser field. Using this change in dispersion, we explain some of the recent observations on the vacuum ultraviolet generation in Xe and Hg.

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Nonlinear generation of radiation in atomic vapors depends critically on the dispersion in the medium. For example, the efficient vacuum-ultraviolet (VUV) generation by four-wave mixing under tight focusing conditions takes place when the medium is negatively dispersive at the frequency of the generated VUV.[1] Consequently, such atomic vapors that have high $\chi^{(3)}(3\omega;\omega,\omega,\omega)$ for the generation of VUV but are positively dispersive cannot be used on their own. One well known method to overcome this difficulty is to use along with the original gas another gas which has a negative anomalous dispersion region at the generated VUV frequency.[2] In this Letter we propose another method of circumventing this difficulty by the addition of a strong saturating resonant laser field. This method of changing dispersion provides in general more than one frequency region in which efficient VUV generation can take place. We demonstrate the generation in the region where it was ordinarily forbidden. The proposed method can have practical advantage as both the intensity and the frequency of the saturating field can be used to control the dispersion and thus provides one with greater flexibility. Using the results on the change in the linear dispersion, we explain some of the recent observations of Compton and Miller[3] and that of Normand, Morellec, and Reif.[4] Note that in the recent experiments of Compton and Miller[3] on multiphoton ionization in Xe, the ionization signal was controlled by the addition of a resonant laser beam. We also present detailed results on the effect of absorption on the nonlinear generation.

In order to demonstrate the basic idea of use of the resonant field to control the dispersion and absorption at the frequency of the generated VUV we consider the atomic level structure shown in Fig. 1. In Xe, e.g., the $|3\rangle$, $|2\rangle$, $|1\rangle$ could correspond to $5p^6\,{}^1S_0$, $6s[\frac{3}{2}]_{J=1}$, and $7p[\frac{3}{2}]_{J=2}$ states whereas in Hg $|3\rangle$, $|2\rangle$, $|1\rangle$ could correspond to $6s^2 1S$, $6p\,{}^1P_1$, and $6d\,{}^1D_2$ states. Normally for third-harmonic (or VUV) generation the field E_2 is absent. In this case the power of the generated VUV is given by

$$I_{\rm VUV}=\frac{c}{4\pi}\int|E_{\rm VUV}|^2\,dx\,dy$$

$$=\frac{c}{4\pi}\left|\frac{K_{23}E_1^3}{d_{23}}\right|^2\frac{\pi\bar{\omega}_0^2}{6}\left|\frac{\Delta kb}{2}\right|^2|F(\Delta kb)|^2. \quad (1)$$

Here $\bar{\omega}_0$ is the beam waist at the focus of the fundamental beam and b is the confocal parameter. d_{mn} is the dipole matrix element between the states $|m\rangle$ and $|n\rangle$. $K_{23}E_1^3$ is the matrix element of the effective Hamiltonian for the absorption of three photons of field E_1 (frequency ω_1) between the states $|2\rangle$ and $|3\rangle$. The phase-matching integral F is given by

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

with $\beta=2(z-f)/b$ and ζ is β at $z=0$. z is the direction of propagation of the fundamental Gaussian-mode field E_1, and f is the focal distance from the face of

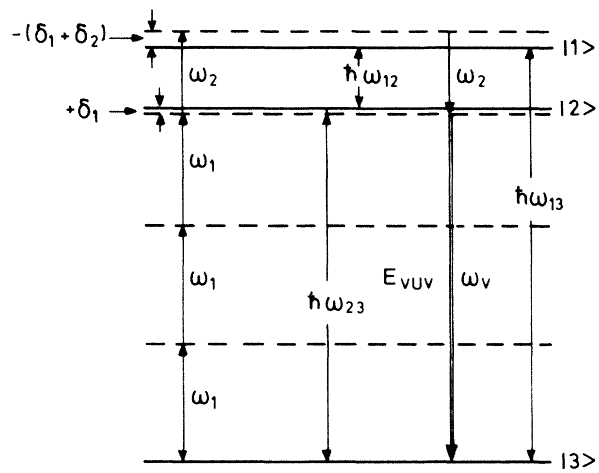

FIG. 1. The effective three-level structure of the atoms considered.

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the medium at which the field E_1 enters. Note that Δkb is complex and is given by

$$\Delta kb = 2\pi\omega_v b\chi^{(1)}(3\omega_1)/c = A/(\delta_1 - i\Gamma_{23}), \quad (3)$$

with

$$A = 6\pi\omega_1 n|d_{23}|^2 b/\hbar c.$$

Here $\chi^{(1)}(3\omega_1)$ is the linear susceptibility at the frequency ($\omega_v = 3\omega_1$) of the generated VUV, and n, Γ_{23}, and δ_1 are respectively the density of atoms, the relaxation rate of the density matrix element ρ_{23}, and the detuning $\omega_{23} - 3\omega_1$. Because of the three-photon resonance, $3\omega_1 \simeq \omega_{23}$, the susceptibility $\chi^{(3)}(3\omega_1;\omega_1,\omega_1,\omega_1)$ has the structure $\simeq nd_{32}K_{23}/\hbar(\delta_1 - i\Gamma_{23})$. Note that the matrix element d_{23} in the denominator of Eq. (1) cancels with the one in the Δkb. Notice further that the VUV for the resonant medium is governed by $|\Delta kbF(\Delta kb)|^2$ rather than by $|F(\Delta kb)|^2$ which is the case for the VUV generation in a nonresonant system. The properties of $F(\Delta kb,\beta)$ are well known for the case of no absorption in the medium. The efficient generation is determined by the dispersion of $\chi^{(1)}(3\omega_1)$. We now investigate how the nonlinear generation changes by an additional laser field.

Using density-matrix equations for the system shown in Fig. 1 we have shown that in the presence of the extra resonant plane-wave field E_2 (frequency ω_2) the linear and nonlinear susceptibilities become intensity dependent and are found to have the explicit forms

$$\bar{\chi}^{(1)}(3\omega_1) = \frac{n|d_{23}|^2}{\hbar}\left[\delta_1 - i\Gamma_{23} - \left(\frac{|g|^2}{(\delta_1+\delta_2) - i\Gamma_{13}}\right)\right]^{-1}, \quad (4)$$

$$\bar{\chi}^{(3)}(3\omega_1;\omega_1,\omega_1,\omega_1) = \frac{nd_{32}K_{23}}{\hbar}\left[\delta_1 - i\Gamma_{23} - \left(\frac{|g|^2}{(\delta_1+\delta_2) - i\Gamma_{13}}\right)\right]^{-1} = \frac{K_{23}}{d_{23}}\bar{\chi}^{(1)}(3\omega_1), \quad (5)$$

where $g = d_{12}E_2/\hbar$, $\delta_2 = \omega_{12} - \omega_2$, and Γ_{13} is the relaxation rate of ρ_{13}. Using (5) and (4) we have proved that the VUV generation is still given by Eqs. (1)–(3), but with $\chi^{(1)}(3\omega_1) \to \bar{\chi}^{(1)}(3\omega_1)$. We next discuss the important properties of $\bar{\chi}^{(1)}(3\omega_1)$. The dispersion $[\mathrm{Re}\bar{\chi}^{(1)}(3\omega_1)]$ and absorption $[\mathrm{Im}\bar{\chi}^{(1)}(3\omega_1)]$ can be varied at will by use of the intensity and the frequency (detuning δ_2) of the field E_2. For example, for positive detuning δ_1, one can induce a net negative $\mathrm{Re}\bar{\chi}^{(1)}(3\omega_1)$ by choosing appropriate values of E_2 and δ_2, whereas it is easily seen that for positive δ_1 and $E_2 = 0$, $\mathrm{Re}\chi^{(1)}(3\omega_1)$ is positive. In Figs. 2(a) and 2(b) we show the changes in $\mathrm{Re}\Delta kb$ and $\mathrm{Im}\Delta kb$ for various values of detunings δ_1, δ_2 and the parameters A, g. The absorption peak in the absence of the field E_2 splits into the well known Autler-Townes doublet,[5] shown in Fig. 2(a) for a range of g and δ_2 values. The medium in the presence of the field E_2 shows two separated anomalous dispersion regions. Note the change in the sign of $\mathrm{Re}\Delta kb$ for $|\delta_1| < |g|$. It is this change in sign coupled with sufficiently large pressure which is useful to generate VUV in the frequency range where it is normally forbidden. Note further that the splitting is symmetric about $\delta_1 = 0$ if the second laser is exactly on resonance with the transition $|1\rangle \leftrightarrow |2\rangle$. For off-resonant field E_2 ($\delta_2 \neq 0$) the splitting becomes asymmetric with respect to δ_1. Consequently the medium can be made negatively dispersive even at exact resonance, i.e., $\delta_1 = 0$. Thus for spectroscopic studies one has the possibility of generating VUV at desired frequencies.

The intensity of the generated VUV, as given by Eq. (1), depends on the phase-matching integral which in turn depends on Δkb. In our study Δkb is complex because we are dealing with resonant situations and because we also have another resonant field which changes the absorption characteristics. The complex Δkb makes the evaluation of $F(\Delta kb)$ quite complex. Even in the tight-focusing limit no analytic result for $F(\Delta kb)$ can be obtained.

The behavior of $F(\Delta kb)$ has been analyzed by the fast Fourier transform method. In Fig. 3 the function $G(\Delta kb) = |\Delta kbF(\Delta kb)/2|^2$ is plotted against $\mathrm{Re}\Delta kb$ for various values of $\mathrm{Im}\Delta kb$. For these figures we have taken $\beta = 10$ at the output face. The field E_1 is assumed to be focused in the middle of the sample. Note that there is no VUV generation in the positive dispersion region. The peak of $G(\Delta kb)$ for $\mathrm{Im}\Delta kb = 0$ occurs at $\mathrm{Re}\Delta kb = -4$. This peak starts to fall in magnitude sharply when $\mathrm{Im}\Delta kb$ changes from 0.005 to 0.3; however, its position remains roughly unchanged. For larger $\mathrm{Im}\Delta kb > 0.3$, the function G shown in the inset of Fig. 3 exhibits oscillations on the negative dispersive side. Thus Maker fringes are obtained for $\mathrm{Im}\Delta kb > 0.3$, i.e., when the absorption length of the medium becomes comparable to the confocal parameter b. These and the related effects shall be discussed elsewhere. Figure 2(a) shows two regions of the detuning δ_1 for which the value of $\mathrm{Re}\Delta kb$ is equal to -4 at sufficiently large pressures. These regions correspond to $\delta_1 < -d_{12}E_2/\hbar$ and positive $\delta_1 < |d_{12}E_2/\hbar|$. Asymmetric regions are obtained when δ_2 is nonzero. Note that the region of positive δ_1 is the one which is normally forbidden for VUV generation if the extra resonant field E_2 were absent. To have efficient VUV generation in this region one needs $\mathrm{Im}\Delta kb < 0.005$. This can be achieved by ad-

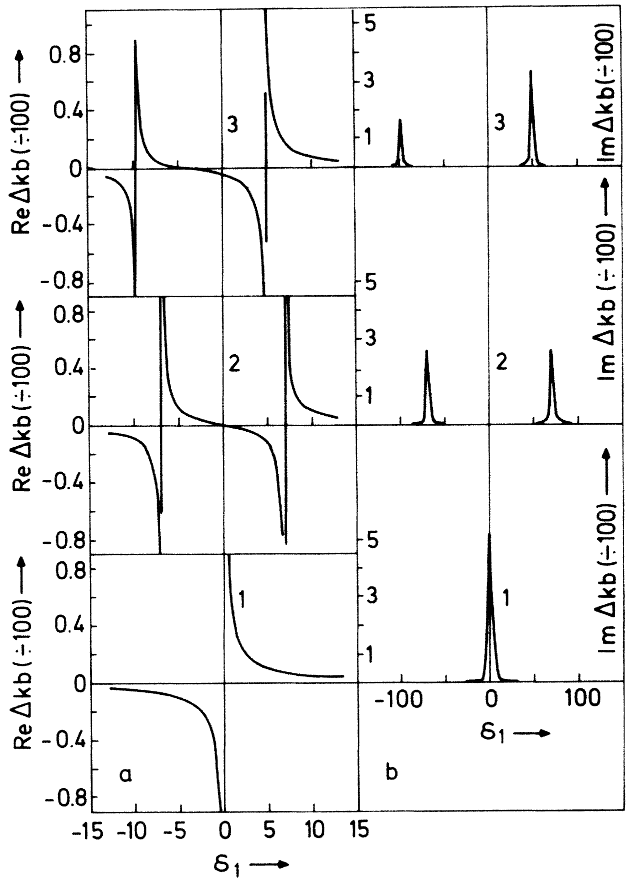

FIG. 2. (a) The real and (b) the imaginary parts of the complex Δkb vs δ_1, in units of Γ_{23}, are plotted for three values of g and δ_2, viz., (1) $g=\delta_2=0.0$; (2) $g=155$ cm^{-1} ($E_2 \simeq 1$ GW/cm^2, $\delta_2=0.0$; and (3) $g=155$ cm^{-1}, $\delta_2=113$ cm^{-1}. These curves correspond closely to the experiments of Compton and Miller at 0.32 Torr with $A=116$ cm^{-1}, $\Gamma_{23}=\Gamma_{13}\simeq 68$ GHz as observed experimentally at low pressures. The curves marked (3) correspond to the situation when the field E_2 is detuned by $50\Gamma_{23}$.

justing the control parameter δ_2, i.e., the frequency of the second laser.

Therefore the above analysis shows that by having an additional resonant field one can (1) change the absorption properties of the medium considerably, (2) develop two regions of negative dispersion, and (3) generate VUV efficiently in the region in which it is forbidden.

In the case of Hg vapor Normand, Morellec, and Reif[4] have reported the observation of third-harmonic generation on both sides of the resonance $\delta_1=0$ with a dip in third-harmonic generation at exact resonance. For their experiment the role of the extra laser field E_2 is played by the fundamental field itself. Their observation is consistent with the two regions of negative dispersion as obtained in our analysis.

We now use the results displayed in Figs. 2 and 3 to explain experiments of Compton and Miller.[3] They have observed two color multiphoton ionization signals much in the manner of this Letter. They, however, did not report the simultaneous observation of a VUV signal. Nevertheless, it is interesting to analyze their two color multiphoton ionization experiments in view of the possible competition effects due to VUV generation. It is well understood now that the ionization signal depends crucially on the VUV generation which can be very efficient as a result of the three-photon resonance.[6,7] Depending on the absorption in the medium at different detuning δ_1 and pressure (n) we distinguish three different regions of operations for $E_2=0$: (1) $\mathrm{Im}\Delta kb \gg 1$.—This condition occurs at large pressures in the region of resonance. There is no resonant ionization signal although a weak VUV signal may exist. This happens through the cancellation of the contribution from the two coherent paths of excitation to the resonant state. (2) $\mathrm{Im}\Delta kb \simeq 0$ with $\mathrm{Re}\Delta kb = -4$.—This condition is satisfied in the far blue region of the three-photon resonance, i.e., when the pressure is large enough to make $\mathrm{Re}\Delta kb = -4$ at large detuning $\delta_1 < 0$. The excitation wavelength in this case moves so much out of the resonance profile of the three-photon resonant level that one has $\mathrm{Im}\Delta kb \simeq 0$. There is no ionization signal in this region but an intense VUV signal exists. (3) $0.01 < \mathrm{Im}\Delta kb < 0.2$ with $\mathrm{Re}\Delta kb = -4$.—This condition is satisfied at those pressures at which the $\mathrm{Re}\Delta kb = -4$ occurs in the region nearer to the resonance. In this region the generation of VUV is not as efficient as for $\mathrm{Im}(\Delta kb) < 0.001$. However, the contribution of the third-harmonic path of excitation to level $|2\rangle$ as compared to the contribution of the path of excitation due

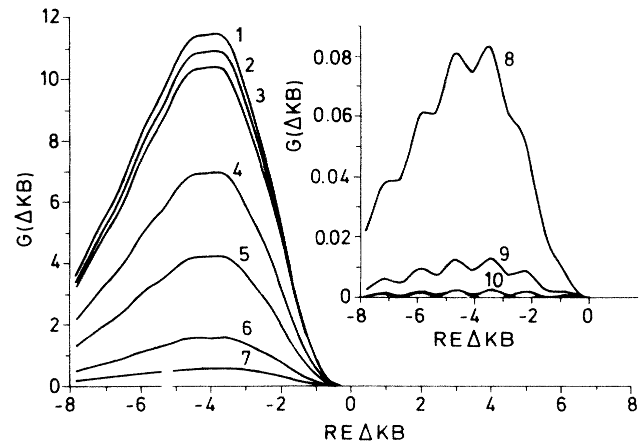

FIG. 3. The behavior of the function $G(\Delta kb)$, which is proportional to the power of the generated VUV, is shown as a function of $\mathrm{Re}\Delta kb$ for various values of $\mathrm{Im}\Delta kb$: (1) 1×10^{-4}, (2) 5×10^{-3}, (3) 1×10^{-2}, (4) 5×10^{-2}, (5) 0.1, (6) 0.2, (7) 0.3. Inset: The region of Maker fringes for $\mathrm{Im}\Delta kb$ equal to (8) 0.5, (9) 0.7, (10) 0.9.

to the three photons of the fundamental is large. Consequently when the pressure of the gas is varied the ionization signal shows shift and broadening similar to the shift and broadening of the generated VUV.

We now examine the situation when the *extra resonant field is switched on.* Consider first the one-color experiment at pressure 0.32 Torr at which no ionization signal is observed because of the conditions (1) and (2) prevalent in the regions described above. At this pressure one finds $\mathrm{Im}\Delta kb \simeq 513$ for $\delta_1 = 0$, and the detuning δ_1 at which $\mathrm{Re}\Delta kb = -4$ is satisfied is found to be 8.72×10^{12} Hz. For this detuning δ_1, $\mathrm{Im}\Delta kb$ is small, $\mathrm{Im}\Delta kb \simeq 2.806\times10^{-2}$. On switching on the field E_2 with intensity of the order of 10^9 W/cm^2, one finds that for $\delta_1=\delta_2=0$, $\mathrm{Im}(\Delta kb)$ drops from 513 to 0.1 thus disallowing the possibility of exact cancellation of the two coherent paths of excitations and hence there is *recovery* of *ionization signal* at this position due to condition (3). On the other hand, for $\delta_1 = 8.72\times10^{12}$ Hz and $\delta_2=0$, one finds that $\mathrm{Im}(\Delta kb)$ increases to 0.08 and $\mathrm{Re}(\Delta kb)$ becomes -5.6. This means that the far blue region which corresponded to condition (2) in one-color situation gets transformed into the condition (3) when the second field is switched on. *Hence ionization signal on the far blue region is also recovered.* Note further the development of the region corresponding to condition (3) on the red side of the position $\delta_1=0$, $\delta_2=0$, i.e., on the $\delta_1>0$ side. Because of this region there is recovery of ionization signal on the red side also. This is consistent with the experiment of Compton and Miller which shows a broad peak covering both the blue and the red sides of the three-photon resonance. It should be borne in mind that in the two-color experiment another channel of ionization opens up via the resonant state $|1\rangle$. This fact coupled with the above analysis explains the broad peak for the ionization signal when field E_2 is switched on.

On increasing the pressure from 0.32 to 14.4 Torr in the *two-color* experiment, Compton and Miller observed the vanishing of the broad peak of ionization. From Eq. (4) one sees that with increase of the pressure from 0.32 to 14.4 Torr the value of $\mathrm{Im}(\Delta kb)$ increases approximately by a factor of 50. This leads to a broad region where the exact cancellation of the two coherent paths of excitation exists.

The analysis given above shows how the generation of VUV is crucial in determining the ionization signal via three-photon resonance in the two-color multiphoton ionization experiments of Compton and Miller.

In conclusion, we have shown how the control of the dispersive and absorptive properties of a medium by the addition of an extra resonant laser field can be used to generate VUV at some preselected range of frequencies around the third harmonic of the fundamental. Needless to say, the third-harmonic generation has been selected here to illustrate the basic point though the method should be equally useful for other types of nonlinear generation.

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