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Intensity and Resonance Effects on the Three-Photon Resonant Third-Harmonic Generation in Hg

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Abstract. The possible mechanism for the generation of near three photon resonant third-harmonic in the low energy side of the 6^1P_1 state of Hg is described. It is shown that the existence of strong ac Stark effect, due to the presence of the additional resonance $6^1P_1 - 6^1D_2$ via the fundamental photon, results in a spatially dependent phase mismatch, which can lead to nonzero radiation in the $\Delta k > 0$ region. Additionally, the influence of the ac Stark effect as a saturation mechanism with increased incoming radiation power is described.

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Generation of coherent radiation in the VUV and XUV regions by frequency mixing in atomic media has been reported by several authors in recent years [1–10]. In most of the cases two- or three-photon resonances, focusing and phase matching are used in order to increase the efficiency of the process. However, together with the achieved enhancement of the efficiency, limits exist such as restriction in the spectral regions where radiation can be generated (Δk has to be negative), linear and nonlinear absorption of the incoming or the generated radiation, population transfer to excited states, ac Stark effect (shift or splitting of the states), nonlinearity of the refractive index, and parametric generation. Some of these restricting effects have been studied in detail by several authors [11–15], but there are still points that are not clear, the most important being the possibility of generation in spectral regions with $\Delta k > 0$, which, according to Bjorklund [11], is forbidden under tight focusing conditions.

It is the purpose of this work to present results and some theoretical calculations related to the third-harmonic (TH) generation near the 6^1P_1 level of Hg (both below and above the state), using a three-photon resonance enhancement. The existence of a spatially dependent wavevector mismatch, as a mechanism for the generation of radiation at spectral regions with positive dispersion, is described. Quantitative estimations concerning the ac Stark effect, the population transfer and the nonlinearities of the refractive index are presented, in order to support the given arguments. Finally, the operation of the ac Stark effect as a saturation mechanism with increased incoming radiation power is studied.

1 Experimental

The output of a KrF laser (Lambda Physics EMG 150) with 150 mJ, 15 ns pulses was used to pump a dye laser (Lambda Physics FL2002). The dye solution was Coumarin 153 dissolved in methanol which lases in the region 522–600 nm. The dye laser had a linewidth of 0.4 cm^{-1}, it was linearly polarized with a 10 ns FWHM pulse width and maximum energy 10 mJ. A 15 cm focal length lens was used to focus the dye laser radiation in the center of a heat pipe containing mercury, producing a maximum intensity of 50 GW/cm^2 (with a confocal parameter $b \sim 3$ mm). The heat pipe was properly shaped to allow return of the liquid mercury formed at the water cooled sides of the pipe [7, 8]. The active mercury column L was approximately 5 cm and argon was used to restrict the mercury vapor away from the windows and for phase matching. The generated VUV radiation at 184.9 nm was detected using a vacuum spectrometer equipped with a solar blind photomultiplier (EMR 541 G-08-17).

2 Results and Discussion

Figure 1 shows a simplified energy level diagram of the mercury atom relevant to this work. Figure 2 shows a typical spectrum of the TH when the dye laser is tuned between 555–552 nm. The dip in the generated VUV radiation at the 6^1P_1 level is due to the reabsorption by the mercury atoms and it becomes deeper as the interaction region moves backwards through the active region of the heat pipe. The FWHM of the spectrum is

approximately 0.5 nm and the TH signal does not drop to zero far from the resonance, although its intensity decreases by a factor of ten, approximately. The resonant line is shifted to shorter wavelengths by approximately 0.1 nm (30 cm^{-1}), taking into account the accuracy of the dye laser wavelength. This is in agreement with the ac Stark shift of 0.83 $cm^{-1}/GW/cm^2$ measured by Poirier et al. [16] for the 6^1P_1 level of mercury. For an input energy of 7 mJ (35 GW/cm^2) we expect a shift of approximately 29 cm^{-1}.

We calculated the wavevector mismatch for the case under study and we found that it is negative above the 6^1P_1 state and positive below it. Its value is in agreement with the one given by Hilbig et al. [17], who also generated nonresonant TH and 4-WSM radiation in the $\Delta k<0$ region, a result which agrees with the criteria developed by Bjorklung [11] for the tightly focused configuration. According to these criteria the third-harmonic generation is permitted only for those wavelengths at which the nonlinear medium is negatively dispersive, i.e., $\Delta k<0$, where Δk is the wavevector mismatch between the incident radiation ω_1 and the TH $3\omega_1$, $\Delta k=k(3\omega_1)-3k(\omega_1)$.

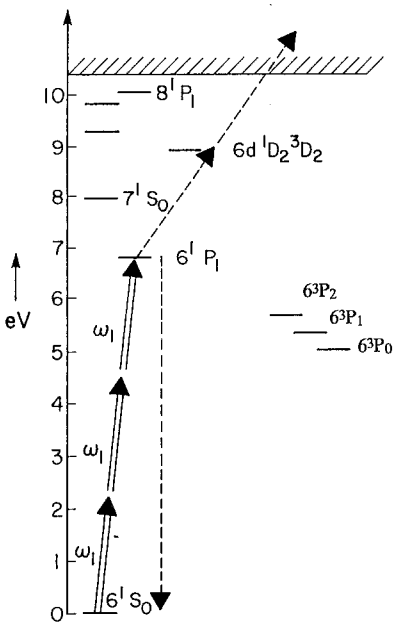

Fig. 1. A simplified energy level scheme of mercury

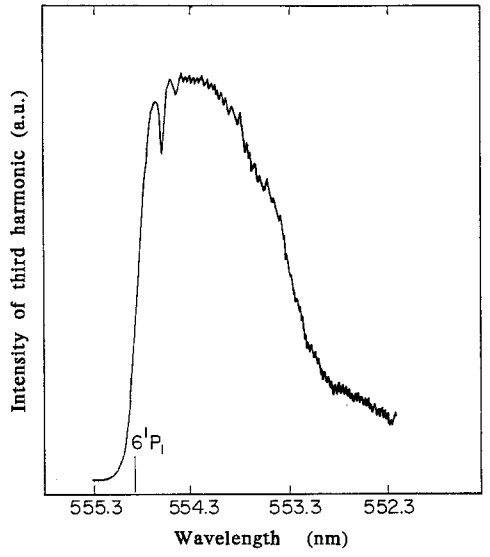

Fig. 2. A typical spectrum of the third-harmonic for $E_1=7$ mJ, $p_{Hg}=0.9$ mbar and $p_{Ar}=5$ mbar

The spectrum of Fig. 2 extends bellow and above the resonant level, a result which is in contrast with the criteria given by Bjorklund (in our case $b/L\sim 6\ 10^{-2}$), and the radiation has the same intensity in both spectral regions. Similar TH generation in the $\Delta k>0$ region of the 6^1P_1 level of Hg has been observed by Normand et al. [6], however, in their case, the radiation was an order of magnitude less intense than the one observed in the $\Delta k<0$ region. Observations concerning generation of radiation by TH and 4-WSM at spectral regions with $\Delta k>0$ have been also reported by Mahon and Tomkins [1], Hilbig and Wallenstein [3], Reintjes et al. [18], Smith [19], and Tsukiyama et al. [20].

For the explanation of the disagreement between experimental results and theoretical calculations, for the case of three-photon resonance enhancement, Blazewicz et al. [21] and Tewari and Agarwal [22] have proposed that the presence of a strong resonant field, between the final and some other state, can split the three-photon resonane state (ac Stark splitting) which alters the initially positive dispersion to a negative one. Consequently the generation of radiation is not forbidden any more. This mechanism can in principle be applied to the case under study, because the input field frequency is in near resonance with the transition $6^1P_1-6^1D_2$ ($\sim 800\ cm^{-1}$ detuning). However, we will show that the proposed mechanism of the ac Stark splitting can not explain our observations and it is rather the spatial dependence of the dispersion which is responsible for the generation of radiation in regions with $\Delta k>0$.

More specifically, the influence of the existing ac Stark effect on the dispersion of the system can be understood if we consider the wavevector mismatch of the TH generation, which is given by the equation:

$$\Delta k=12\pi^2\nu_1\,\mathrm{Re}[\chi^{(1)}(3\nu_1)], \tag{1}$$

where only the TH radiation contributes to the Δk, due to the presence of the resonance $\omega(6^1P_1)-\omega(6^1S_0)$, ν_1 is the incoming photon energy in cm^{-1}, and $\chi^{(1)}$ is the atomic linear susceptibility, given by

$$\chi^{(1)}(3\nu_1)=\frac{N|d_{30}|^2}{hc}\frac{1}{\delta_1-i\Gamma_{30}}. \tag{2}$$

Here N is the Hg number density, δ_1 is the detuning $\nu(6^1P_1)-3\nu_1$, d_{30} is the electric dipole matrix element for the transition $6^1P_1\rightarrow 6^1S_0$, and Γ_{30} is the linewidth of this transition. Using the formalism of Tewari and Agarwal [22], the presence of the additional resonance results in a change of the linear susceptibility, which is now given by the equation

$$\chi^{(1)}(3\nu_1)=\frac{N|d_{30}|^2}{hc}\left[\delta_1-i\Gamma_{30}-\left(\frac{|g|^2}{\delta_1+\delta_2-i\Gamma_{40}}\right)\right]^{-1}, \tag{3}$$

where $\delta_1+\delta_2$ is the detuning $\nu(6^1D_2)-4\nu_1$, Γ_{40} is the linewidth of the transition between 6^1D_2 and 6^1S_0, and the extra term is an effective Rabi frequency of the additional resonance, with $|g|$ given by

$$|g|=2\pi|d_{43}|E_1/h, \tag{4}$$

where d_{43} is the electric dipole matrix element for the transition $6^1P_1\rightarrow 6^1D_2$, and E_1 is the electric field intensity of the incoming Gaussian laser beam. Taking into account that δ_1, $\delta_1+\delta_2 \gg \Gamma_{30}$, Γ_{40} (Γ_{30}, Γ_{40} are the atomic linewidths which, for the conditions of the experiment, are expected to be less than $1\,\mathrm{cm}^{-1}$, as the Doppler broadening is $0.05\,\mathrm{cm}^{-1}$ and some collisional broadening is expected, while the detunings δ_1, $\delta_1+\delta_2$ are much larger), the wavevector mismatch is given by

$$\Delta k=12\pi^2\nu_1\frac{N|d_{30}|^2}{hc}\frac{1}{\delta_1-\dfrac{|g|^2}{\delta_1+\delta_2}}. \tag{5}$$

Using the Hg energy spacings [23] and the definitions of δ_1, δ_2, the $\delta_1+\delta_2$ value is calculated to be

$$\delta_1+\delta_2=(4/3)\delta_1-759\,\mathrm{cm}^{-1}. \tag{6}$$

The $\delta_1+\delta_2$ value remains negative down to a detuning δ_1 of $569\,\mathrm{cm}^{-1}$, which means that on the low energy side of the shifted 6^1P_1 state in the spectrum of Figs. 2 ($\delta_1=\delta_{\mathrm{eff}}-30\sim-30$ to $30\,\mathrm{cm}^{-1}$); the wavevector mismatch (5) remains always positive, whatever the input intensity is, which prohibits the emission of radiation. Consequently, in the case of the spectrum under study, the proposed splitting, because of the additional strong resonance, does not alter the sign of the wavevector mismatch, and the proposed mechanism can not explain the observed emission of radiation.

An alternative explanation for the emission of radiation at the spectral region with $\Delta k>0$ can be given, if we consider the fact that the wavevector mismatch depends on the spatial coordinates. According to the work of Reintjes et al. [18], Ganeev et al. [24, 25], and Drabovich et al. [26] in the case that the wavevector mismatch is influenced by the spatially dependent nonlinearity of the refractive index, the destructive interference of the harmonic generated in the regions before and after the focal plane is incomplete and the generation is possible for $\Delta k>0$. Furthermore, Drabovich et al. [26] calculated the phase matching integral corresponding to the spatially dependent wavevector mismatch, due to the nonlinearity of the refractive index, which was found to have non zero value in the spectral regions with positive dispersion, in contrast to the criteria developed by Bjorklund [11]. Consequently, it seems that when the wavevector mismatch is a function of the spatial coordinates, caused by nonlinearities of the refractive index or some other mechanism, the value of the phase matching integral is nonzero for $\Delta k>0$ and the emission of radiation is possible. This explanation seems also consistent with the results of Mahon and Tomkins [1], Hilbig and Wallenstein [3] and Tsukiyama et al. [20], because in all cases, large intensity dependent population transfer is expected, which introduces spatial dependence of the wavevector mismatch Δk.

For the case under study, where the uniformity of the vapor number density and the symmetry of the focus of the beams were tested carefully, the ac Stark effect, the population transfer, and the nonlinearity of the refractive index are the possible mechanisms that can introduce spatial dependence to the wavevector mismatch. In the following sections, these mechanisms will be discussed in detail.

First we will deal with the influence of the ac Stark effect on the wavevector mismatch for the $\Delta k>0$ region. Under the influence of the ac Stark effect, due to the additional resonance, the dynamical detuning in the denominator of the Hg contribution to the wavevector mismatch (5) is given by

$$\Delta=\delta_1-(|g|^2/\delta_1+\delta_2)=\delta_1+(|g|^2/|\delta_1+\delta_2|) \tag{7}$$

for the region of interest and where the parameters have been defined above. For values of the second term comparable to the δ_1 value, the detuning will depend on the incoming radiation electric field intensity (4) and consequently on the spatial coordinates. Figure 3 shows the ratio $(|g|^2/\delta_1+\delta_2)/\delta_1$ as a function of the detuning from the shifted state of the spectrum of Fig. 2 ($\delta_{\mathrm{eff}}=\delta_1+30\,\mathrm{cm}^{-1}$) and for the spectral region with $\Delta k>0$. The calculation was carried out for an incoming radiation intensity of $35\,\mathrm{GW/cm}^2$ ($g=182\,\mathrm{cm}^{-1}$) and it was assumed that the main contribution on the ac Stark effect comes from the near resonance $6^1P_1-6^1D_2$ because other states such as 7^1S_0, 8^1S_0, 7^1D_2 contribute approximately 4% to the total process (where the values of the corresponding electric dipole matrix elements were taken from the work of Mukherjee et al. [27]). The intensity dependent term is greater than the δ_1 value for all wavelengths corresponding to $\Delta k>0$, thus the detuning, and the wavevector mismatch are always a function of the field which is spatially dependent due to the focusing.

Next, we will consider the influence of the population transfer on the phase matching integral. Under the influence of the possible population transfer to the 6^1P_1

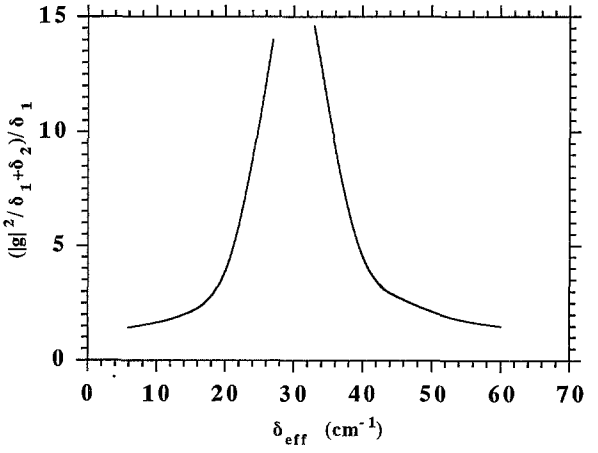

Fig. 3. The ratio $(|g|^2/\delta_1+\delta_2)/\delta_1$ as a function of the effective detuning ($\delta_{\mathrm{eff}}=\delta_1+30\,\mathrm{cm}^{-1}$) in the Δk positive region of the spectrum in Fig. 2

state, following either the three-photon absorption of the incoming radiation or the one-photon absorption of the TH, the wavevector mismatch can be spatially dependent, because in both cases it is a function of the electric field intensity through the equation

$$\Delta k_{\mathrm{Hg}} = (1-a)\, N\, C_{\mathrm{Hg}} - aN\, C_{\mathrm{Hg}}. \tag{8}$$

Here, it was assumed that the 6^1S_0 and 6^1P_1 states have opposite contribution to the wavevector mismatch (as they both depend on the resonance $6^1P_1 - 6^1S_0$), N is the total Hg number density, and a is the percentage of the excited atoms, which is spatially dependent through the electric field intensity.

For the calculation of the population transfer to the 6^1P_1 state, the linewidth of the transition $6^1P_1 - 6^1S_0$ is needed. The Doppler broadening of this transition is calculated to be $0.05\,\mathrm{cm}^{-1}$. However, a considerable influence on this value is expected through the existence of collisions and the ac Stark effect, which can not be calculated directly. However, we can estimate the population transfer if we consider the effect of the Ar buffer gas on the phase matching. Figure 4 shows the TH signal as a function of the Ar number density, for TH photon energy $54123\,\mathrm{cm}^{-1}$ (where Δk is negative) and other parameters the same as in the spectrum of Fig. 2. According to the criteria given by Bjorklund [11] for the $\Delta k<0$ spectral region, the maximum TH signal is expected at $\Delta k = -2/b$ (a criterion which has been probably changed slightly because of the existence of a spatially dependent wavevector mismatch). Using data reported by Chashchina et al. [28], the Ar contribution to the phase mismatch is estimated to be $5\times10^{-18}\,\mathrm{cm}^2$. Using this value, the above mentioned criterion, the Hg number density in use ($1.8\times10^{16}\,\mathrm{cm}^{-3}$), the Ar density at the TH maximum ($1.2\times10^{17}\,\mathrm{cm}^{-3}$), and the electric dipole matrix element reported by Mukherjee et al. [27], we estimated the total Hg contribution to the wavevector mismatch to be $-4.4\times10^{-16}\,\mathrm{cm}^2$, a value smaller than the calculated one of $-1.2\times10^{-14}\,\mathrm{cm}^2$ (where the influence of the ac Stark effect has been included through (5)). The existing difference between the experimental and the calculated value of the wavevector mismatch can be justified if we consider the contribution of the excited states population and/or the contribution of the non-

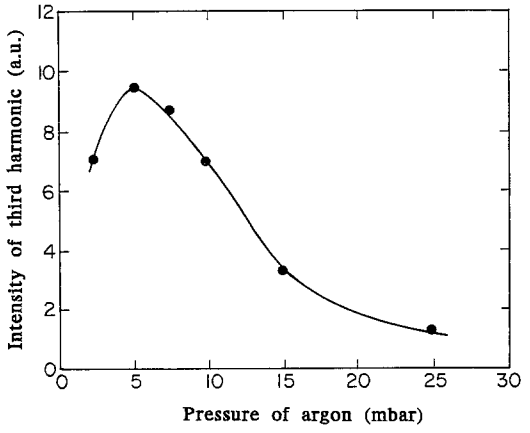

Fig. 4. The third harmonic signal as a function of the buffer gas pressure for fixed Hg pressure of $p_{\mathrm{Hg}}=0.9$ mbar

linearity of the refractive index, as it has been presented in a previous work [29]. An estimation of the contribution to the wavevector mismatch, coming from the nonlinearity of the refractive index, was carried out, using the formalism presented by Reintjes [30a] for the three-photon resonance case and electric dipole matrix element values reported by Mukherjee et al. [27]. The maximum contribution (very near the 6^1P_1 state) is approximately $10^{-18}\,\mathrm{cm}^2$, thus, the nonlinearities of the refractive index can be neglected. Consequently, it is the population of the 6^1P_1 state that alters the Hg contribution to the wavevector mismatch. Using the assumptions and the values presented above, the population transfer is estimated to be 48%. This population transfer, corresponding to an input intensity of $35\,\mathrm{GW/cm}^2$, a Hg number density of $1.8\times10^{16}\,\mathrm{cm}^{-3}$ and a TH photon energy of $54123\,\mathrm{cm}^{-1}$, can be due either to the one-photon absorption of the generated radiation or the three-photon absorption of the fundamental, as it was mentioned above. In the following sections, these mechanism will be studied in more detail.

First we shall estimate the population transfer via three-photon absorption of the incoming radiation, for the case of Fig. 4, using the formalism of Reintjes [30b] and the electric dipole matrix elements reported by Mukherjee et al. [27]. Even at high values of the $6^1P_1 - 6^1S_0$ transition linewidth, i.e., $1\,\mathrm{cm}^{-1}$, the population transfer is estimated to be 3%, thus it can be neglected, compared to the total of 48%. Consequently, the main population transfer mechanism is the one-photon absorption of the TH radiation.

Next we will estimate the ratio of the excited state population as a function of the detuning from the shifted 6^1P_1 state and for the spectral region with $\Delta k>0$. Using the measured value of the generated TH photons ($\sim10^{12}$ photons/pulse, corresponding to $2\,10^7\,\mathrm{W/cm}^2$), and the calculated value of the absorption cross section in linewidth units (σ/Γ) for photon energy $54123\,\mathrm{cm}^{-1}$ and including the influence of the ac stark effect [imaginary part of (3)], the linewidth of the $6^1P_1 - 6^1S_0$ transition was found to be $0.2\,\mathrm{cm}^{-1}$. This value, corresponding to an input radiation intensity of $35\,\mathrm{GW/cm}^2$ and a Hg number density of $1.8\times10^{16}\,\mathrm{cm}^{-3}$, is the same for all the wavelengths in the spectrum of Fig. 2. Then the calculation of the ratio N_{exc}/N_0 was carried out, using this linewidth value and the TH radiation intensity from Fig. 2. The results of the calculation are shown in Fig. 5. It can be seen that at the spectral region with $\Delta k>0$, the population transfer is considerable only near the new position of the state. Consequently, the population transfer can introduce spatial dependence to the wavevector mismatch only for wavelengths very near resonance.

In conclusion, we have shown that for the case under study, it is rather the spatial dependence of the wavevector mismatch Δk, due to the ac Stark effect (and the population transfer for the very near resonance case) which give a non zero value to the phase matching integral for the positive dispersion region of the 6^1P_1 state.

Figure 6 shows the third harmonic signal as a function of the input dye laser energy, for several vapor pressures

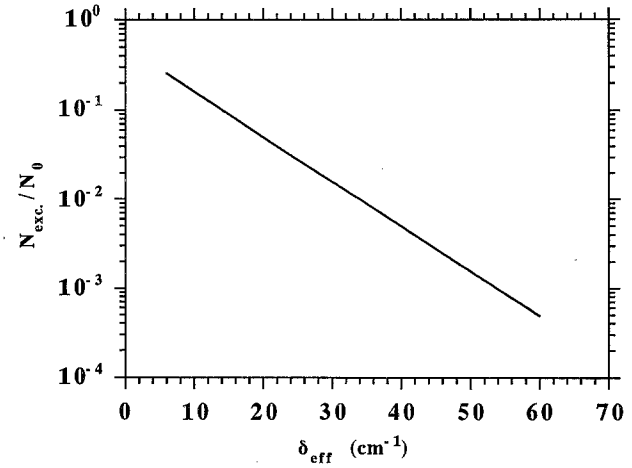

Fig. 5. The ratio N_{exc}/N_0 as a function of the effective detuning ($\delta_{eff}=\delta_1+30\,cm^{-1}$) in the Δk positive region of the spectrum in Fig. 2

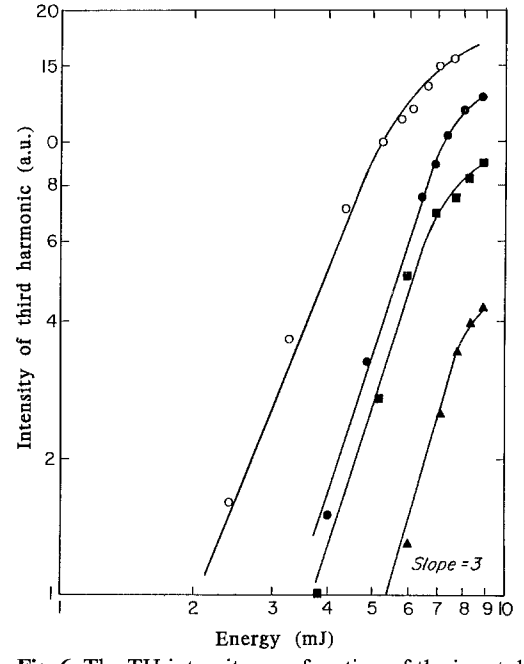

Fig. 6. The TH intensity as a function of the input dye laser energy for several vapor pressures (mbar) of Hg (▲ 0.23; ■ 0.58; ● 0.90; ○ 1.73)

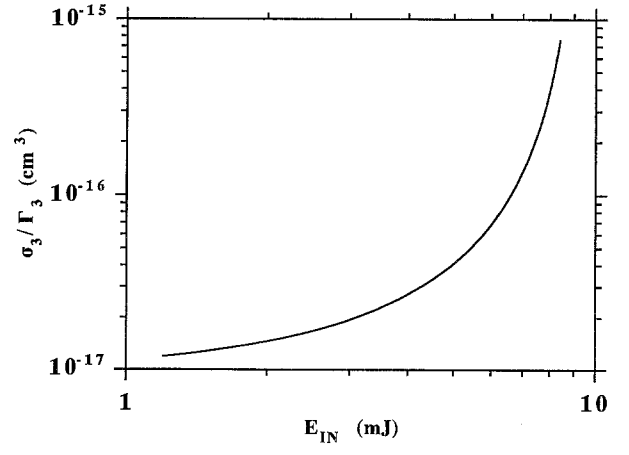

Fig. 7. The absorption cross section at the harmonic wavelength, divided by the transition linewidth as a function of the incoming laser energy

of mercury and a wavelength at the blue side of the 6^1P_1 level ($3\nu_1=54123\,cm^{-1}$, with $\Delta k<0$). The slope of the plots is approximately 3. At high input energies saturation appears and the saturation point shifts slightly to lower energies for higher Hg pressures. The presented behavior can be attributed to the presence of the strong ac Stark effect due to the additional resonance, because for δ_1 negative and with increased incoming laser energy, the dynamical detuning Δ (7) decreases. Consequently, the absorption cross section at the harmonic wavelength, the Hg contribution to the wavevector mismatch and the nonlinear susceptibility increase, because they depend on this dynamical detuning. The large absorption cross sections together with the high TH radiation intensities, following the increase of the fundamental energy, result in an enhanced population transfer to the 6^1P_1 state. The increased Hg contribution to the wavevector mismatch leads Δk to values higher than the optimum one. These processes, together with the possible saturation of the nonlinear susceptibility, are responsible for the saturation of the generated TH radiation as a function of the input energy.

Figures 7 and 8 show the variation of the absorption cross section at the harmonic wavelength, divided by the linewidth of the transition $6^1P_1-6^1S_0$, and the contribution of Hg to the wavevector mismatch, respectively, as a function of the input energy and for a TH photon energy of $54123\,cm^{-1}$ (the nonlinear susceptibility is expected to have similar behavior as in Fig. 8). It is clear that for energies higher than 4 mJ all quantities increase rapidly, thus the third-harmonic generation should be strongly affected. However, the exact influence of each process on the saturation is rather complicated, because the increased incoming energy causes broadening of the $6^1P_1-6^1S_0$ transition linewidth (which increases the absorption cross section) and the final value of the wavevector mismatch depends on the population transfer to the 6^1P_1 state (which depend on the exact value of the absorption cross section). This becomes more complex if we include the effect of the Hg number density. The increased Hg number densities result in an increase of the $6^1P_1-6^1S_0$ transition

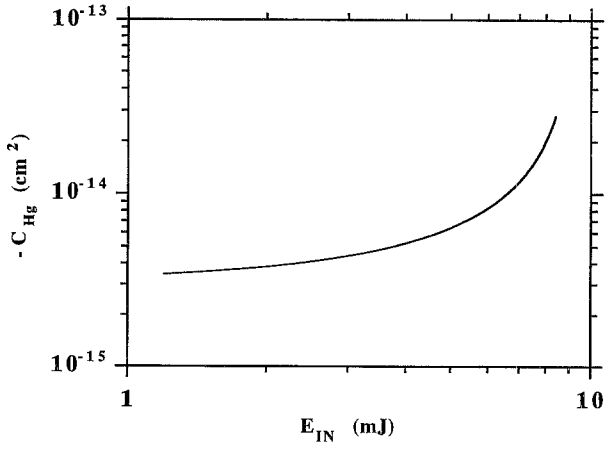

Fig. 8. The Hg contribution to the phase mismatch as a function of the incoming laser energy

linewidth (due to collisional broadening) and in a decrease of the phase matching function F value, which depends on the quantity σNl, as it has been discussed by Tewari and Agarwal [22]. A comparison between the curves in Fig. 6 shows that for this specific wavelength, the large Hg number densities are preferable to the high input energies, for intense TH generation. However, because the exact influence of the additional resonance on the saturation mechanisms depends on the generated wavelength, special care has to be taken for the choise of the optimum experimental conditions in every case.

3 Conclusions

In this work we have studied the three-photon resonant third-harmonic generation near the 6^1P_1 state of Hg with high incoming laser energy and under the influence of the additional strong resonance $6^1P_1-6^1D_2$, via the ω_1 photon. We have demonstrated that the third-harmonic generation is possible in the region with $\Delta k>0$ and its intensity is of the same magnitude as for the $\Delta k<0$ region. The presence of the additional resonance does not seem to change the sign of the dispersion in this particular case, as it has been proposed, and it is rather the dependence of wavevector mismatch on the spatial coordinates which is responsible for the observed radiation. This spatial dependence is introduced mainly by the existing strong ac Stark effect, because for all the wavelengths corresponding to $\Delta k>0$, the intensity dependent detuning term is larger than the initial detuning δ_1. Additionally, for wavelengths very near the shifted 6^1P_1 state, spatial dependence can also be introduced by the existing population transfer.

It was also shown that the existence of the additional strong resonance introduces a number of saturation mechanisms with increased incoming laser energy. Under the influence of the ac Stark effect, the final detuning changes, resulting in a change of the absorption cross section at the harmonic wavelength, the Hg contribution to the wavevector mismatch and the nonlinear susceptibility. Consequently, optimum conditions depending on the generated wavelength have to be chosen in order to increase the conversion efficiency of the process.

It should be of interest to study the TH generation with the introduction of an additional resonance using an independent laser of frequency ν_2, both for the 6^1P_1 state and the continuum, where laser induced continuum structure (LICS) is expected to occur.

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References

1. R. Mahon, F.S. Tomkins: IEEE J. QE-**18**, 913–920 (1982)
2. J.C. Miller, R.N. Compton: Phys. Rev. A **25**, 2056–2063 (1982)
3. R. Hilbig and R. Wallenstein: IEEE J. QE-**19**, 1759–1770 (1983)
4. T. Srinivasan, H. Egger, H. Pummer, C.K. Rhodes: IEEE J. QE-**19**, 1270–1275 (1983)
5. R. Hilbig, R. Wallenstein: IEEE J. QE-**19**, 194–201 (1983)
6. D. Normand, J. Morellec, J. Reif: J. Phys. B, Atom. Mol. Phys. **16**, L227–L232 (1983)
7. P.R. Herman, P.E. Larocque, R.H. Lipson, W. Jamroz, B.P. Stoicheff: Can. J. Phys. **63**, 1581–1588 (1985)
8. P.R. Herman, B.P. Stoicheff: Opt. Lett. **10**, 502–504 (1985)
9. K. Miyazaki, H. Sakai, T. Sato: IEEE J. QE-**22**, 2266–2271 (1986)
10. G. Hilber, A. Lago, R. Wallenstein: J. Opt. Soc. Am. B **4**, 1753–1764 (1987)
11. G.C. Bjorklund: IEEE J. QE-**11**, 287–296 (1975)
12. E.A. Stappaerts: IEEE J. QE-**15**, 110–118 (1979)
13. H. Scheingraber, C.R. Vidal: IEEE J. QE-**19**, 1747–1757 (1983)
14. M.G. Paynne, W.R. Garett: Phys. Rev. A **28**, 3409–3429 (1983)
15. A.V. Smith, W.J. Alford, G.R. Hadley: J. Opt. Soc. Am. B **5** 1503–1519 (1988)
16. M. Poirier, J. Reif, D. Normand, J. Morellec: J. Phys. B, Atom. Mol. Phys. **17**, 4135–4149 (1984)
17. R. Hilbig, G. Hilber, R. Wallenstein: Appl. Phys. B **41**, 225–230 (1986)
18. J. Reintjes, M. Dlabal, L.L. Tankersley: AIP Conf. Proc. **90**, 491 (Boulder 1982)
19. A.V. Smith: Opt. Let. **7**, 341–343 (1985)
20. K. Tsukiyama, M. Tsukakoshi, T. Kasuya: Jpn. J. Appl. Phys. **27**, 1552–1553 (1988)
21. P.R. Blazewicz, M.G. Payne, W.R. Garrett, J. Miller: Phys. Rev. A **34**, 5171–5174 (1986)
22. S.P. Tewari, G.S. Agarwal: Phys. Rev. Let. **56**, 1811–1814 (1986)
23. C.E. Moore: *Atomic Energy Levels,* ed by USA Dept. of Com., Nat. Bur. Stand., **3**, 191–195 (1971)
24. R.A. Ganeev, V.V.Gorbushin, I.A. Kulagin, T. Usmanov: Appl. Phys. B **41**, 69–71 (1980)
25. R.A. Ganeev, V.V. Gorbushin, I.A. Kulagin, T. Usmanov: Optics Spect. **61**, 807–810 (1986)
26. K.N. Drabovich, I.A. Kulagin, T. Usmanov: Sov. J. Quantum Electron **15**, 402–404 (1985)
27. N. Mukherjee, A. Mukherjee, J.C. Diels: Phys. Rev. A **38**, 1990–2004 (1988)
28. G.I. Chashchina, V.I. Gladushchak, E.Y. Shreider: Opt. Spect. **24**, 542–543 (1968)
29. E. Koudoumas, T. Efthimiopoulos: Submitted to J. Opt. Soz. Am. B (1991)
30. J.F. Reintjes: In *Nonlinear Optical Parametric Processes in Liquids and Gases* (Academic, New York, 1984) a) pp. 251, b) pp. 273