

Nonresonant third harmonic generation in thallium vapour

SHARADA ITAGI and ABDUL RAFE KHAN

Department of Physics, Marathwada University, Aurangabad 431 004, India

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Abstract. Feasibility of nonresonant third harmonic generation in thallium is investigated. The third order susceptibility, for driving frequencies in the visible region, is calculated and the phase matching with Ar as buffer gas is investigated. Other related quantities like coherence length, minimum pulse lengths required to get phase matching and power input requirements to achieve 50% conversion are also calculated.

Keywords. Nonlinear optics; third harmonic generation; nonlinear third order susceptibility; thallium coherence length; phase matching; pulse widths.

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1. Introduction

Third harmonic generation (THG) in metal vapours is an important method of producing coherent UV and soft x-ray radiation. THG was proposed and produced in gaseous media for the first time by Ward and New (1969) using inert gases. Harris and Miles (1971) showed that THG could be produced in alkali metal vapours if the driving frequency is chosen lower than and the third harmonic is higher than the frequency of the resonance line. This was feasible because of the anomalous dispersion in the vicinity of a resonance line. For some of the alkali metals nonresonant THG has been experimentally obtained (Bloom *et al* 1975a, b; Ohasi *et al* 1976; Puell *et al* 1976). Susceptibility values on which THG depends, are available for alkali metals (Miles and Harris 1973), and metal ions like Mg II and Ca II (Dinev and Ganev 1981). In this paper we report on the susceptibility for the third harmonic in thallium and also some related factors like phase matching requirements, coherence length and the minimum pulse widths required for THG.

2. Theory

The third harmonic power $P^{(3)}$ in terms of the incident power is given by (Miles and Harris 1973).

$$P^{(3)} = \frac{3\eta^4 \pi^2 c^2}{\lambda^4} N^2 [\chi^{(3)}(3\omega)]^2 |I|^2 [P^{(1)}]^3 \quad (1)$$

where $\eta = (\mu_0/\epsilon_0)^{1/2} = 377$; N —density of metal atoms/cc; I —an integral which accounts for the beam focussing and the mismatch between phase velocities of the fundamental and the third harmonic waves; $\chi^{(3)}(3\omega)$ —microscopic susceptibility for the third harmonic and is given by

$$\chi^{(3)}(3\omega) = \frac{1}{\hbar^3} \sum_g \sum_{abc} \mu_{ga} \mu_{ab} \mu_{bc} \mu_{cg} \rho_{gg} A_{abc} \quad (2)$$

where

μ_{ij} —matrix elements, ρ_{gg} —occupancy of the lowest state, A_{abc} —the frequency factors given by the expression

$$\begin{aligned} A_{abc} = & \frac{1}{(\Omega_{ag} - 3\omega)(\Omega_{bg} - 2\omega)(\Omega_{cg} - \omega)} \\ & + \frac{1}{(\Omega_{ag} + \omega)(\Omega_{bg} + 2\omega)(\Omega_{cg} + 3\omega)} \\ & + \frac{1}{(\Omega_{ag} + \omega)(\Omega_{bg} + 2\omega)(\Omega_{cg} - \omega)} \\ & + \frac{1}{(\Omega_{ag} + \omega)(\Omega_{bg} - 2\omega)(\Omega_{cg} - \omega)} \end{aligned} \quad (3)$$

Ω_{ij} are the atomic transition frequencies and ω is the driving frequency. The present calculations are carried out with the following assumptions.

(i) A single valence electron is involved in the interaction with the incident wave. (ii) The interaction is non-resonant. The fundamental and the harmonic frequencies do not coincide with the atomic transitions. (iii) The matrix elements are for the electric dipole interaction. While considering summation over different states in (2) the selection rules applicable to dipole transitions for one valence electron atoms are considered. (iv) Only bound states contribute to susceptibility. (v) The occupancy of the $^2P_{1/2}$ ground state is 1. Since the $^2P_{3/2}$ component is 7792 cm^{-1} higher, the assumption holds good.

2.1 Dipole matrix elements

If the incident laser field is linearly polarized in the z direction, the matrix elements of the electric dipole operator which is a tensor of rank one, are given by (Eicher 1975):

$$\begin{aligned} & \langle n l s j m | P_z | n' l' s j' m' \rangle \\ & = -e \langle l s j m | T_0^{(1)} | l' s j' m' \rangle \langle n l \| r \| n' l' \rangle, \end{aligned} \quad (4)$$

where

$$\begin{aligned} & \langle l s j m | T_0^{(1)} | l' s j' m' \rangle \\ & = (-1)^{j-m} \begin{pmatrix} j & 1 & j' \\ -m & 0 & m' \end{pmatrix} \langle l s j \| T^{(1)} \| l' s j' \rangle, \end{aligned} \quad (5)$$

and

$$\begin{aligned} \langle l s j \| T^{(1)} \| l' s j' \rangle & = (-1)^{l+s+j'+1} (2j+1)^{1/2} (2j'+1)^{1/2} \\ & \times \begin{Bmatrix} j & 1 & j' \\ l' & s & l \end{Bmatrix} \langle l \| T^{(1)} \| l' \rangle, \end{aligned} \quad (6)$$

with

$$\langle l \| T^{(1)} \| l' \rangle = (-1)^l (2l+1)^{1/2} (2l'+1)^{1/2} \begin{pmatrix} l & 1 & l' \\ 0 & 0 & 0 \end{pmatrix}. \quad (7)$$

The symbols used here are the same as those used by Shore and Menzel (1968). The $3-j$ and $6-j$ symbols are given by Rotenberg *et al* (1960). The second factor in (4), is the radial matrix element. The calculation of the radial matrix elements is given by Bates and Demgaard (1949) and by Bebb and Gold (1966).

$$\langle n'l'|r|nl\rangle = N'N \sum_{p=0}^{p_0} A_p \left(\frac{1}{n^*} + \frac{1}{n^*} \right)^{-(v-p+1)} \Gamma(v-p+1) \quad (8)$$

where

$$N = (-1)^{n-l-1} (2/n^*)^{n^*} [\rho(n^*)n^{*2} \Gamma(n^*+l+1) \Gamma(n^*-l)]^{-1/2}$$

and N' refers to the upper state. n^* is the effective principal quantum number.

$$\rho(n^*) = 1 + \partial\mu/\partial n^*$$

$$A_p = \sum_{t=0}^p a_t a_{p-t}$$

and $a_t = a_{t-1} (n^*/2t) [l(l+1) - (n^*-t)(n^*-t-1)]$.

However, if the line strengths S or the oscillator strengths f are known, matrix elements can be evaluated using the following relations:

$$\langle n' l' s j' | P | n l s j \rangle = S^{1/2}. \quad (9)$$

$$f = \frac{4\pi mc}{3\hbar e^2 \lambda g} S, \quad (10)$$

where g is the statistical weight of the state from which the transition is taking place. Other symbols have the usual meaning.

We have used the f values calculated by Bardsley and Norcross (1980) for Tl. These authors have employed solutions of the Dirac equation with one-electron central field approximation and have chosen the parameters such that the observed energy levels are reproduced. They have also included core polarization effects in their calculations.

2.2 Phase matching

In the plane wave approximation or when the confocal parameter (Hanna *et al* 1979) is much larger than the linear dimensions of the gas cell the THG efficiency can be optimized by making the phase vector mismatch Δk equal to zero. Here

$$\Delta k = k_3 - k_1 = \frac{6\pi}{\lambda} (n_3 - n_1) \quad (11)$$

and n_3 and n_1 are refractive indices at the third harmonic and the fundamental wavelengths respectively. Δk can be made zero either by choosing the interacting frequency in the region of anomalous dispersion of the nonlinear medium around the resonance line or a normally dispersive buffer gas may be added to make the refractive indices at the fundamental and the third harmonic equal. In the absence of the buffer gas THG is possible over short distances where the first and third harmonics overlap. This distance is known as the coherence length L_c , and is given by

$$L_c = \pi/\Delta k. \quad (12)$$

If a buffer gas is mixed with the metal vapour the coherence can be achieved over longer distances and the conversion efficiency increases. The ratio of the buffer gas and the metal vapour is so adjusted that the refractive indices at the fundamental and the third harmonic are made equal. The matching ratio can be obtained by using Sellmeier equation:

$$n - 1 = \frac{Nr_e}{2\pi} \sum \frac{f_i}{[(1/\lambda_i)^2 - (1/\lambda)^2]}, \quad (13)$$

where $r_e = 2.818 \times 10^{-13}$ cm, f_i —oscillator strength of the line, λ_i —wavelength of the line which is absorbed from the ground state.

2.3 Group velocity matching

Since THG is achieved by using mode-locked pulses, it is necessary to get phase matching throughout the frequency spread of the pulse. This imposes a condition that a third harmonic pulse with group velocity $(d\omega/dk)_{3\omega}$, would not get out of step with the fundamental pulse with group velocity $(d\omega/dk)_\omega$, by more than the pulse duration Δt in passing through the length L of the medium. This is expressed mathematically as

$$\frac{n_1 n_3 \Delta v_g}{c^2} \leq \frac{\Delta t}{L}, \quad (14)$$

where

$$\Delta v_g = \left| \left(\frac{d\omega}{dk} \right)_{3\omega} - \left(\frac{d\omega}{dk} \right)_\omega \right|. \quad (15)$$

The minimum pulse duration will have to be

$$\Delta t = \frac{n_1 n_3 \Delta v_g L}{c^2}$$

2.4 THG power

In the plane-wave approximation, assuming a Gaussian beam and a confocal parameter equal to the cell length, the THG power is (Harris and Miles 1971)

$$\frac{P_3}{P_1} = 9\pi^2 \eta^4 c^2 \chi_T^2 P_1^2 \lambda^{-4}$$

where $\chi_T = \frac{\chi(\text{esu}) \times 10^{-17} N}{\text{degeneracy of the ground state}}$.

3. Results and discussion

The matrix elements are calculated using (9) and (10). Oscillator strengths used are those of Bardsley and Norcross 1980. The sign of the matrix elements is decided using equations (4)–(8). Since the ground state of Tl is $^2P_{1/2}$, the final state can be either $^2S_{1/2}$ or $^2D_{3/2}$. The intermediate states can be S and P , D and P or D and F . In the calculation of susceptibility from (2) and (3) the summation is taken over 50 combinations of states. The variation of susceptibility with wavelength, in the spectral range $0.4 \mu\text{--}1.2 \mu$ is shown in figure 1.

Phase matching is possible for Tl with Ar as buffer gas for driving frequencies in the visible region. The ratios of Ar atoms to those of Tl for phase matching at fundamental wavelengths 0.4μ , 0.5μ and 0.6μ have been calculated. In table 1, the third order susceptibility, the coherence length, the minimum pulse width required, the ratio of

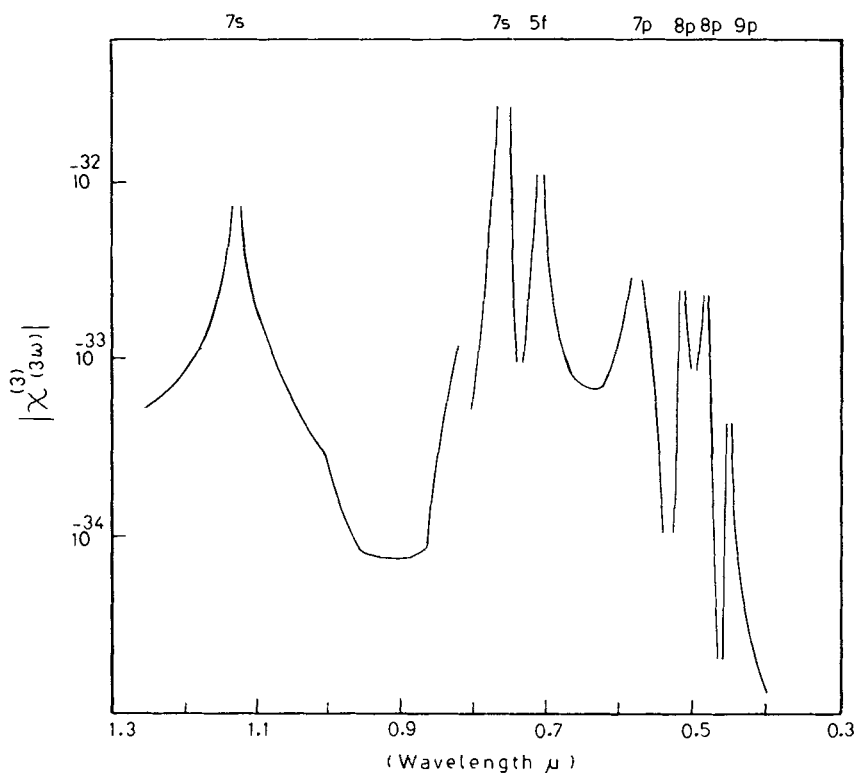


Figure 1. Nonlinear susceptibility of Tl vs wavelength.

Table 1. Calculated parameters for THG in thallium.

Wavelength of fundamental (μ)	Metal vapour	Buffer gas	$\chi^3(3\omega)$ (esu)	L_c (cm)	N_{Ar}/N_{Tl}	Δt (psec)	P_F (50%) (Watts)
0.4	Tl	Ar	1.33×10^{-35}	0.06	36	0.182	3×10^8
0.5	Tl	Ar	6.69×10^{-34}	0.17	39	0.082	9.3×10^6
0.6	Tl	Ar	1.1×10^{-33}	0.19	74	0.089	8.1×10^6
0.6	Na	Xe	-6.36×10^{-33}	0.11	476	7.6	7.61×10^7

buffer gas atoms to Tl atoms and incident power required to produce 50% conversion at wavelengths 0.4 μ , 0.5 μ and 0.6 μ , are tabulated. In the last column similar values for sodium are given for 0.6 μ , for comparison. The relevant data for sodium are from the work of Harris and Miles (1971).

In figure 2, the refractive indices of Tl and Ar are plotted in the spectral range of interest. In the spectral range beyond 0.22 μ because of absorption by

- $6^2P_{1/2}$ ————— $7d \quad 2D_{3/2}$ (0.238 μ),
- $6^2P_{1/2}$ ————— $8d \quad 2D_{3/2}$ (0.224 μ),
- $6^2P_{1/2}$ ————— $9s \quad 2S_{1/2}$ (0.232 μ),
- $6^2P_{1/2}$ ————— $10s \quad 2S_{1/2}$ (0.221 μ),

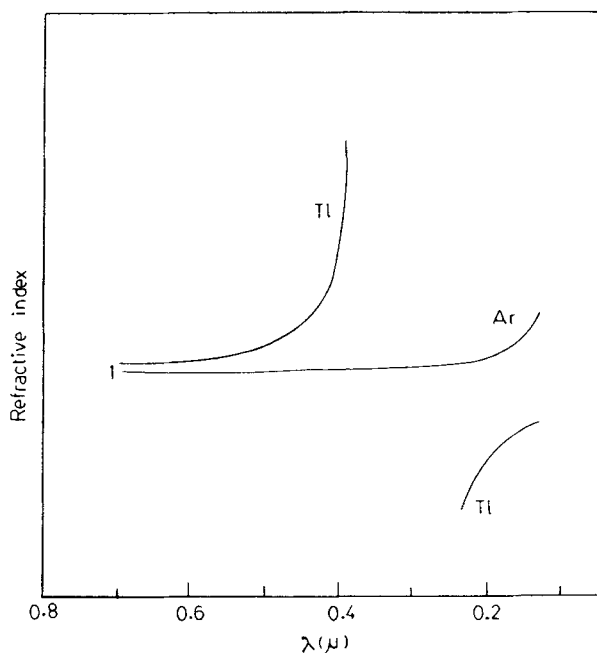


Figure 2. Refractive indices of Tl and Ar vs wavelength.

the refractive index variation with wavelength is not smooth and the theory of nonresonant THG does not hold good.

There is no experimental data on nonresonant THG in Tl. However, the two-photon resonant THG has been experimentally achieved by Wang and Davis (1975). The data in table 1 is for Tl atom concentration of 10^{18} atoms/cc. Temperatures in the range of 1100–1200°C are required to get the necessary atomic concentration.

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