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The Effect of Linewidth on the Efficiency of Two-Photon-Pumped Frequency Converters

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Abstract—**The efficiency of two-photon-pumped frequency converters as a function of the pump laser detuning and linewidth, and of the linewidth of the selected nonallowed atomic transition is studied. We show that pressure broadening of the atomic transition is a key parameter in the design of these converters. A technique is developed to measure this linewidth as a function of either self- or foreign-gas broadening using a broad-band pump laser. Measurements of the linewidth of the $3s^2S$-$3d^2D$ transition of Na, and its device implications, are presented.**

I. Introduction

TWO-PHOTON-PUMPED frequency converters are useful for the generation of tunable vacuum ultraviolet radiation, and provide an excellent technique for the conversion of infrared radiation into the visible and near ultraviolet [1]-[3]. Such a converter consists of a pumping laser whose frequency ω_0 equals approximately one-half the frequency of a nonallowed transition in an atomic vapor. The pump radiation sets up a strong excitation in the atom at a frequency $2\omega_0$ which cannot radiate because of the absence of a dipole moment. A third photon of frequency ω couples this excitation to an oscillation mode which does have a dipole moment, producing output radiation at frequencies $2\omega_0 \pm \omega$. The strength of the atomic excitation at $2\omega_0$, and thus the conversion efficiency, depends directly on the laser detuning, and on the atomic and laser linewidths.

In Section II below we calculate the functional dependence of the conversion efficiency of two-photon-pumped converters on the laser detuning and linewidth, and on the atomic linewidth, including both pressure and Doppler broadening. Our results show that the pressure broadening of the nonallowed transition is a key parameter in the design of UV generators and IR up-converters. Although a number of spectroscopic techniques currently exist to measure such broadening [4]-[7], they require rather sophisticated, narrow-bandwidth, tunable lasers. In Section III we describe a technique for measuring the pressure broadening of a nonallowed transition based on the results of Section II. The technique uses the broad-band laser source which would be used as the pump for the converter under evaluation. Section IV describes such measurements for an IR up-converter operating around 9 μ utilizing the $3s^2S$ to $3d^2D$ transition in Na [3].

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II. Calculation of Conversion Efficiency

The pumping radiation is described by a superposition of randomly phased discrete laser modes with center frequency ω_0, mode spacing $\Delta\omega$, amplitudes E_n, and phases φ_n. The second field is also assumed to be made up of a superposition of independent modes. Because of the randomness of the phases, each of these modes will be up-converted independently, and the power up-conversion efficiency is the same as that of a single mode. Therefore, in what follows, we treat this field as monochromatic with frequency ω and amplitude E. The dipole moment at the generated frequencies $2\omega_0 \pm \omega$ is given by [8]

$$P^{(3)}(t) = K_1 E\, e^{i\omega t} \sum_{k,n} E_n E_{k-n} \cdot \left\{ \frac{e^{-i(2\omega_0 + k\Delta\omega)t + i(\varphi_n + \varphi_{k-n})}}{(\omega_{02} - 2\omega_0 - k\Delta\omega) - i\gamma/2} \right\} + \text{c.c.} \quad (1)$$

where

$$K_1 = \sum_{\text{paths}} \frac{N \mu_{01}\mu_{12}\mu_{23}\mu_{30}}{8\hbar^3 (\omega_{01} - \omega_0)\,[\omega_{03} - (2\omega_0 \pm \omega)]}.$$

In this equation the ω_{0j} and μ_{ij} are atomic transition frequencies and matrix elements, γ is the pressure-broadened linewidth of the individual atoms, and N is the number density of the nonlinear species. A summation over all possible paths for the first and third steps in the perturbation sequence is also included [9]. To incorporate Doppler broadening in the theory we note that, for an observer traveling with the incident electric field, atoms moving at different velocities have different apparent resonance frequencies: $\omega_{02}(x) = \omega_{02} + x$. For a Maxwellian velocity distribution, the frequency deviation x has a Gaussian distribution function

$$p(x) = \frac{1}{\sqrt{2\pi\gamma_D^2}}\, e^{-x^2/2\gamma_D^2} \tag{2}$$

where

$$\gamma_D = \sqrt{\frac{kT\omega_{02}^2}{Mc^2}}.$$

Combining (1) and (2), and substituting into Maxwell's equations, we find that the power conversion efficiency for the second field is given by

$$\mathcal{E}(\gamma, \gamma_D, \delta, \Delta\omega) = K_2 \frac{P_{\text{av}}^2}{\gamma_D^2} \sum_k G(k)\,[F_R^2(k) + F_I^2(k)] \tag{3}$$

where

$$P_{\text{av}} = \frac{1}{2}\sum_n E_n^2$$

$$G(k) = \sum_n \frac{E_n^2 E_{k-n}^2}{P_{\text{av}}^2}$$

$$F_R(k) = \int_{-\infty}^{\infty} \frac{(\omega_{02} - 2\omega_0 + x - k\Delta\omega)\, e^{-x^2/2\gamma_D^2}\, dx}{(\omega_{02} - 2\omega_0 + x - k\Delta\omega)^2 + (\gamma/2)^2}$$

$$F_I(k) = \int_{-\infty}^{\infty} \frac{(\gamma/2)\, e^{-x^2/2\gamma_D^2}\, dx}{(\omega_{02} - 2\omega_0 + x - k\Delta\omega)^2 + (\gamma/2)^2}$$

$$K_2 = \frac{2\eta_0^2\omega_s^2 L_c^2 K_1^2}{\pi^3} \sin^2\left(\frac{\pi}{2}\frac{L}{L_c}\right).$$

In these expressions, P_{av} is the average power in the pump field, $G(k)$ is the normalized autoconvolution of the power spectrum of the pump field, $F_R(k)$ and $F_I(k)$ are the real and imaginary parts of the plasma dispersion function [10], η_0 is the impedance of vacuum, ω_s is the generated frequency, L is the length of the medium, and L_c is the coherence length for the process [9]. If the coherence length is set by the nonlinear species itself (no phase matching), then its value is inversely proportional to N and the factor K_2 is independent of pressure.

Given the values of the parameters δ, $\Delta\omega$, γ_D for a particular system it is quite easy to evaluate the efficiency given by (3) numerically as a function of the atomic linewidth γ. In most cases of practical interest, however, (3) can be reduced to a simple closed-form relationship. Generally the laser spectrum consists of a large number of modes with a spacing much smaller than both the atomic and Doppler linewidths, in which case the summations over n and k in (3) can be approximated by integrals. In addition, the spectrum of the laser can usually be represented by a Gaussian distribution:

$$E_n^2 = E_0^2\, e^{-4n^2\Delta\omega^2/\delta^2} \tag{4}$$

where δ is the laser linewidth, defined as $(\ln 2)^{-1/2}$ times the full width at half-maximum (FWHM).

Consider first the case when the laser is detuned several linewidths from one-half the two-photon transition frequency. For this case (3) reduces to

$$\mathcal{E} = \frac{8\pi K_2 P_{\text{av}}^2}{(\omega_{02} - 2\omega_0)^2}. \tag{5}$$

This is the well-known quadratic reduction of efficiency with detuning, independent of the laser, atomic, and Doppler linewidths.

When the laser is tuned on-resonance and the pressure is low enough so that Doppler broadening dominates pressure broadening, i.e., $\gamma_D > \gamma$, then

$$\mathcal{E} = 4(\pi^2\sqrt{2} + 8\sqrt{\pi})\, K_2 \frac{P_{\text{av}}^2}{\gamma_D \delta}. \tag{6}$$

In the high-pressure region where pressure broadening dominates

$$\mathcal{E} = 16\,\pi^2 \sqrt{\frac{2}{\pi}}\, K_2 \frac{P_{\text{av}}^2}{\gamma\delta}. \tag{7}$$

In view of (5) one might not initially expect the *linear* reduction of efficiency with laser linewidth of (6) and (7). Physically, however, it is clear that a resonant atomic response is produced not only by the pump mode exactly at half the transition frequency, but also by all combinations of two modes whose sum is within the atomic linewidth [11]. Thus efforts to significantly narrow the pump linewidth, usually at the expense of simplicity and peak power, are often not effective in improving efficiency. A similar analysis of two-photon absorption can be used to show that, on resonance, the absorption cross section is also a linear, rather than quadratic, function of laser linewidth.

For image up-converters of this type it is easy to show that the conversion efficiency for a given resolution and pump power increases quadratically with the atomic number density [12]. As the density is increased, however, pressure broadening will dominate the atomic linewidth, and (7) indicates that the efficiency obtainable in a single coherence length will be reduced. In some cases there may also be a significant contribution to the broadening from a foreign buffer gas. Thus the pressure broadening of the selected nonallowed transition is a key parameter in the design of two-photon-pumped frequency converters.

III. Measurement of Pressure Broadening

Conceivably one could determine the limiting efficiency of a given frequency converter by merely measuring conversion efficiency while raising the atomic and/or buffer gas density. However, other effects such as atomic and molecular absorption, change of coherence length caused by the foreign gas, thermal defocusing, and the Kerr effect also influence the efficiency. Thus an independent measurement of pressure broadening (both self- and foreign-gas) is required to optimize the performance of the device.

Several methods are currently available to measure the linewidth of nonallowed transitions [4]-[7] but the required laser sources are not often available. Using the results of Section II, it is possible to quickly and conveniently measure these parameters in cases of practical interest using just the unnarrowed pump laser.

Equation (5) shows that when the laser is sufficiently detuned, the efficiency is independent of the linewidth of both the laser and the atomic transition, but it will still be influenced by the other processes mentioned above. Thus these effects may be normalized out by making efficiency measurements both on-resonance and off-resonance as a function of pressure, and taking a ratio. The theoretical result, in the high-pressure region where $\gamma > \gamma_D$, is the ratio of (7) to (5)

$$R = 2\sqrt{2\pi} \times \frac{(\omega_{02} - 2\omega_0)^2}{\gamma \times \delta}. \tag{8}$$

The detuning and laser linewidth can be eliminated by normalizing to the low-pressure asymptote of R, R_0 found from (6) and (5):

$$\frac{R}{R_0} = \frac{4\pi}{\pi\sqrt{\pi} + 4\sqrt{2}} \frac{\gamma_D}{\gamma}. \tag{9}$$

Thus one can determine the pressure-broadened linewidth γ from the measured values of R and R_0:

$$\gamma = 1.12 \times \gamma_D \times \frac{R_0}{R}. \tag{10}$$

The procedure does not require detailed knowledge of the laser linewidth and spectral composition, but is valid only in the higher pressure regime where $\gamma > \gamma_D$. Practically, this is usually the only region where values of γ are of interest, and extrapolation to lower pressures can usually be made with confidence.

A more rigorous determination of γ, valid at all pressures, can be made using an exact evaluation of (3) for the efficiency. This is useful if detailed knowledge of γ at intermediate pressures is required, and/or the assumptions used to reduce (3) to closed form are not valid. The experimental procedure is identical to that explained above. Two sets of efficiency measurements, on- and off-resonance, are made and the normalized ratio R_m/R_{m0} is plotted versus the pressure of the broadening agent (the subscript m refers to measured values, c to calculated values). Using the fixed parameters γ_D, δ, and $\Delta\omega$, the ratio R_c/R_{c0} is computed as a function of γ from (3) and also plotted. Note that since the efficiency off-resonance is a constant independent of γ, only one numerical evaluation of (3) per point is required to calculate R_c/R_{c0}. The normalized curves of measured and calculated efficiency are used to determine γ from the relation $[R_m(P)/R_{m0}] = [R_c(\gamma)/R_{c0}]$. Thus for any given pressure P, the linewidth γ may be read from the point on the calculated curve associated with the value $R_m(P)/R_{m0}$. Although this technique requires more detailed knowledge of the system, and also some computation, it is completely general and can determine pressure-broadened linewidths considerably smaller than the Doppler linewidth. The two procedures are illustrated below in Section IV.

IV. Measurement of the Na $3s^2S$-$3d^2D$ Transition

We have measured the foreign-gas broadening by He of the $3s^2S$-$3d^2D$ transition in Na using the above techniques. A pumping wavelength of 6856 Å was obtained from an optical parametric oscillator, and the idler frequency at 2.37 μ was used as the input frequency ω to generate the observed lower sideband signal at $2\omega_0 - \omega$ (4006 Å). The pumping radiation had a linewidth of 2 cm^{-1} and a mode spacing of 0.028 cm^{-1}. Both incident beams had a peak power of about 1 kW and were focused into a small Na heat pipe at a temperature of about 700 K, yielding a Doppler linewidth γ_D = 0.049 cm^{-1}, and an Na pressure of 0.75 torr (1×10^{16} cm^{-3}).

Efficiency measurements were made both on- and off-resonance while varying the He pressure from a few torr to 2 atm. Fig. 1 shows the ratio $R_m(P)/R_{m0}$ as a function of the He pressure, and Fig. 2 is a plot of the computed ratio $R_c(\gamma)/R_{c0}$ for this case. Fig. 1 also shows the atomic linewidth γ as a function of pressure, both using the exact method and, for $P > 800$ torr, using the approximate method. Note that the two techniques are consistent, and that the numerical technique permits one to measure pressure broadenings sev-

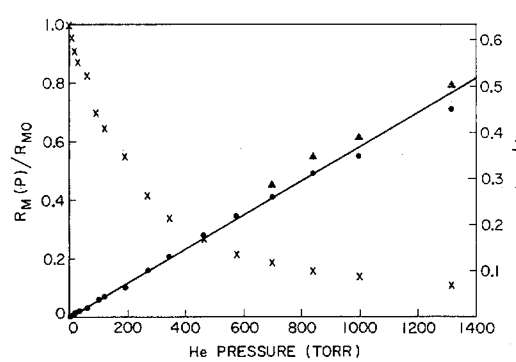

Fig. 1. Measured radio $R_m/R_{m0}(x)$ and corresponding atomic linewidth γ of the Na $3s^2S$-$3d^2D$ transition versus He pressure. Linewidths indicated by (▲) are from the simplified, high-pressure procedure; those indicated by (•) are found from the numerical technique.

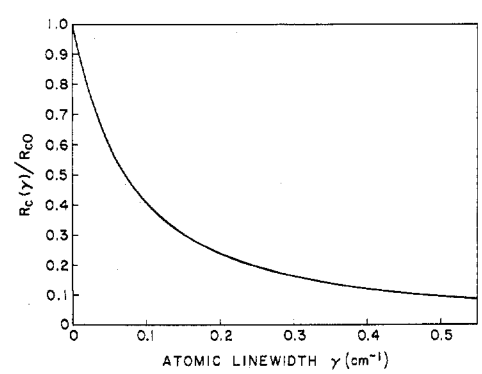

Fig. 2. Calculated ratio R_c/R_{c0} versus atomic linewidth.

eral times smaller than the Doppler broadening. The measured broadening coefficient is 0.3 cm^{-1}/atm.

A similar procedure was used to determine the self-broadening of this transition by varying the Na density from 0.75 torr to 150 torr. Unfortunately, at these high Na densities, it was necessary to introduce 300 torr of He buffer gas to protect the cell windows, and the Na self-broadening was not observable above the 0.1-cm^{-1} He broadening. Thus we infer a self-broadening of less than 0.1 cm^{-1} at a density of 10^{18} cm^{-3}. From a device point of view this result is important in two respects. First, it indicates that the converter may be operated at Na pressures of at least 150 torr without a significant reduction of efficiency, and second, it indicates that it is very important to keep the buffer gas pressure as low as possible. Fig. 1 shows, for example, that at an He buffer gas pressure of about 300 torr, the efficiency is about a factor of 3 less than the value with no He at all.

Using the results of these measurements, we predict that a Na image up-converter operating on this transition at an Na pressure of 150 torr will require 2.5 MW of pump power at 6856 Å to up-convert 10^5 resolvable spots from the 9.0-9.5-μ band to the 3302-3308-Å band at a photon conversion efficiency of 10 percent. Development of devices of this type is currently underway in our laboratory.

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