Resonantly two-photon pumped frequency converter*

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This letter describes a resonantly two-photon pumped frequency converter with application to the generation of tunable ultraviolet and vacuum ultraviolet radiation and also to infrared-to-visible up-conversion and imaging. Calculations show that up-conversion power efficiencies in excess of 100% should be obtainable with tunable dye lasers having peak powers in the hundred watt to kilowatt range.

In recent years, considerable success has been attained in utilizing metal vapors and mixtures of metal vapors and inert gases for frequency tripling of laser radiation into the ultraviolet and vacuum ultraviolet.[1,2] To date, this work has required picosecond time scale laser systems with peak powers in excess of 10^8 W. In this letter we describe a new technique which should allow generation of tunable ultraviolet (uv) and vacuum ultraviolet (VUV) radiation using any of a variety of tunable dye lasers and optical parametric oscillators having peak powers in excess of about 100 W. The device described here may also act as a broad-band ir-to-visible up-converter for coherent or incoherent ir radiation.

A schematic of the proposed device is shown in Fig. 1. To be specific, we consider the use of Mg to generate tunable uv and VUV radiation. A pump laser at frequency ω_p, in this case at 4597 Å, is tuned such that the sum of two photons is equal to the nonallowed $3s$-$4s$ transition of Mg. A second laser at frequency ω_t mixes with the pump laser to produce the sum or difference frequency to $\omega_p \pm \omega_t$. This process will be particularly efficient if the generated frequency lies within a certain range of any of the $np\,^1P^0$ levels. For example, if the tunable laser is tuned over a range of about ± 1000 cm^{-1} centered at 1.7 μ, then the theory to be derived below predicts approximately unity power conversion efficiency from tunable input to sum frequency centered at the $4p\,^1P^0$ level (2026 Å). A much broader range, about ± 10 000 cm^{-1}, centers at the $3p\,^1P^0$ level (2852 Å), though in this case the conversion efficiency will be several percent. For each higher np level, the conversion efficiency becomes higher, and the applicable range of this theory narrower.

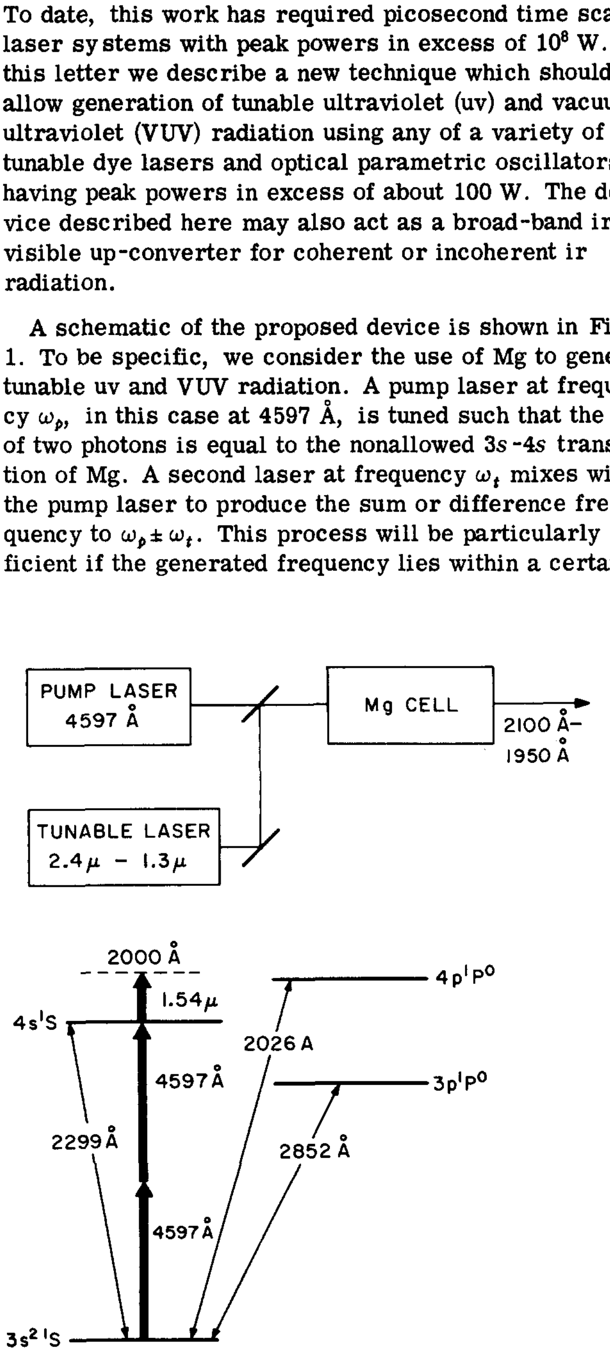

FIG. 1. Schematic of resonantly two-photon pumped frequency converter and pertinent energy levels of Mg.

As has long been known,[3] the advantages of using a nonallowed transition to resonantly enhance the nonlinear optical susceptibility are the absence of both loss and dispersion at the input and generated frequencies. However, as a result of the resonantly enhanced two-photon absorption, the power density of the pumping laser is restricted to a value which is several orders of magnitude lower than would be the case if this resonance had not been employed.

In the following, we write formulae for conversion efficiency from tunable laser power to sum (or difference) frequency power, with the assumption that the pump power density is limited by two-photon absorption of an intermediate nonallowed transition (for example, the $3s^2\,^1S - 4s\,^1S$ transition in Mg). The additional assumptions, that the conversion occurs in a single coherence length and that this length is determined strictly by the upper level (the $4p\,^1P^0$ level in Mg), lead to a strikingly simple formula for predicted conversion efficiency. Before proceeding, we note that resonant nonallowed transitions have recently been used in a somewhat different manner for the generation of tunable infrared radiation.[4]

From Maxwell's equations, the power density generated in a single coherence length at the sum frequency ω_s is given by $P/A(\omega_s) = (1/2\pi^2)\eta\omega_s^2 |\mathcal{P}(\omega_s)|^2 L_c^2$, where $\mathcal{P}(\omega_s)$ is the generated dipole moment, L_c is the coherence le length, and $\eta = (\mu/\epsilon_0)^{1/2}$. For an energy level system such as that shown in Fig. 1, the dipole moment and coherence length are closely approximated by

$$\mathcal{P}(\omega_s) = N\mu_{01}\mu_{12}\mu_{23}\mu_{30}E_p^2E_t/4\hbar^3(\Delta\omega_1)(\Delta\omega_2 + \tfrac{1}{2}j\delta\omega_2)\Delta\omega_3 \qquad (1)$$

and

$$L_c = \frac{2\pi\hbar}{N\eta}\frac{1}{\omega_s}\frac{\Delta\omega_3}{\mu_{03}^2},$$

where N is the atom density, μ_{ij} are the various dipole matrix elements, $\Delta\omega_1 = \omega_p - \omega_{01}$, $\Delta\omega_2 = 2\omega_p - \omega_{02}$, $\Delta\omega_3$

TABLE I. Systems for ultraviolet and vacuum ultraviolet generation. The (–) signs denote the difference frequency process $\omega_s = \omega_p - \omega_t$.

Element	Pump wavelength (Å)	Tunable wavelength (μ)	Generated wavelength (Å)
Mg	$4s$: 4597	2.4 → 1.3	2100 → 1950
	$3d$: 4310	(–) 0.467 → 11.0	4000 → 2200
Cd	$6s$: 3752	1.8 → 1.4	1700 → 1650
	$5d$: 3377	(–) 0.395 → 1.9	2950 → 1850
Zn	$5s$: 3585	1.5 → 1.1	1600 → 1550
	$4d$: 3202	(–) 0.393 → 1.9	2700 → 1750
Hg	$7s$: 3129	1.6 → 1.1	1425 → 1375
	$6d$: 2804	(–) 0.372 → 1.5	2250 → 1550

$= 2\omega_p \pm \omega_t - \omega_{03}$; ω_p is the pump frequency, ω_t is the tunable frequency, and ω_{01}, ω_{02}, and ω_{03} denote the pertinent atomic transition frequencies from ground. (For the example of Fig. 1, these transitions occur to the $3p\,^1P^0$, $4s\,^1S$, and $4p\,^1P^0$ levels, respectively). $\delta\omega_2$ is the half-power linewidth of the ω_{02} transition, and E_p and E_t are the electric field strengths produced by the pump and tunable lasers. Combination of the above formulae leads to a predicted last coherence length conversion efficiency of

$$\mathcal{E} = \frac{\eta^2 \mu_{01}^2 \mu_{12}^2 \mu_{23}^2}{\hbar^4 \Delta\omega_1^2 (\Delta\omega_2^2 + \frac{1}{4}\delta\omega_2^2) \mu_{03}^2} \left(\frac{P}{A}\right)_p^2, \tag{2}$$

where $(P/A)_p$ is the power density of the pump laser.

We now determine the maximum allowed value of $(P/A)_p$ by the condition that $W^{(2)}\tau = \frac{1}{2}$, where $W^{(2)}$ is the two-photon transition probability and τ is the incident laser pulse length Δt or the decay time T_1 of the $4s\,^1S$ level, whichever is shorter. The maximum allowed power density of the pump laser is then[5]

$$\left(\frac{P}{A}\right)_{\max} = \frac{\sqrt{2}\,\hbar^2 \Delta\omega_1}{\mu_{01}\mu_{12}\eta} \frac{1}{(\tau\delta\omega_2)^{1/2}} \left[(\Delta\omega_2)^2 + \left(\frac{\delta\omega_2}{2}\right)^2\right]^{1/2}. \tag{3}$$

We substitute this value of $(P/A)_{\max}$ into Eq. (2) to obtain a conversion efficiency of

$$\mathcal{E} = \frac{T_2}{\Delta t} \frac{\mu_{23}^2}{\mu_{03}^2} = \frac{T_2}{T_1} \frac{\mu_{23}^2}{\mu_{03}^2}, \tag{4}$$

where $T_2 = 2/\delta\omega_2$ is the dephasing time of the $4s\,^1S$ level. The first and second equalities of Eq. (4) apply for laser pulse lengths less than T_1 and greater than T_1, respectively (T_1 must include radiative trapping). For lasers with a pulse length longer than T_1, a molecular quencher may be used to substantially reduce T_1.[6] For the $4p\,^1P^0$ level of Mg, $\mu_{23}^2/\mu_{03}^2 = 48$.[7]

Note that the detuning from the $4s\,^1S$ level, and thus implicitly the linewidth of the pump laser, does not enter into the conversion efficiency formula. Larger detunings require larger power densities [Eq. (3)], but yield the same conversion efficiency. Before using Eq. (4), it must be ascertained that it is indeed the p level nearest the generated signal that determines the coherence length. An approximate condition for the validity of this assumption is that $\mu_{03}^2/\Delta\omega_3 > \mu_{01}^2/\Delta\omega_1$. We should note that if phase-matching techniques[1] are used to further increase the predicted conversion efficiency, the increase will only be linear with the number of coherence lengths which are matched. This results because increasing the number of coherence lengths reduces the allowable value of $W^{(2)}\tau$ and thus of $(P/A)_{\max}$.

As a first example, consider a Mg cell at a vapor pressure of 10 Torr. Assuming a laser pulse length of 4 nsec and a $4s\,^1S$ linewidth of 0.1 cm^{-1}, then for conversion to the vicinity of the $4p\,^1P^0$ level, Eq. (4) yields a power efficiency of 126%. (Note that only photon conversion efficiencies are limited to 100%.) Assuming $\Delta\omega_2 = 0.1$ cm^{-1}, we require a pump power density of 9.7×10^7 W/cm^2. For this case, the range of validity of Eq. (4) is about ± 1000 cm^{-1}. At the ends of this band, at a pressure of 10 Torr, the coherence length will be 0.5 cm; and thus for confocal focusing, a pumping laser power of 557 W is required.

As noted earlier, other applicable ranges of this theory center at each of the $np\,^1P^0$ levels. Table I gives a number of other examples of metal vapor systems and applicable ranges for the generation of tunable uv and VUV radiation.

As another and somewhat different example, we consider the use of resonantly two-photon pumped Na for conversion of an ir signal at 10.6 μ into the near ultraviolet. For a pump laser at 6856 Å (i.e., two-photon pumping of the $3s$-$3d$ transition), a 10.6-μ signal will be converted to 3320 Å. Since in this case the generated frequency is somewhat outside the allowed range of this theory, a more exact computer calculation was employed. Assuming $\Delta\omega_2 = \delta\omega_2 = 0.1$ cm^{-1} and Δt or $T_1 = 10$ nsec, we find a conversion efficiency of 8.8% at an allowed incident power density of 5.25×10^6 W/cm^2. The conversion will be very broad band, allowing up-conversion and imaging of thermal radiation.

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