Optical parametric oscillators pumped by population-trapped atoms

S. E. Harris and Maneesh Jain

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

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We describe an optical parametric oscillator that is pumped by population-trapped atoms that are prepared with maximum coherence. The oscillator is based on the use of an effective nonlinear susceptibility that is of the same order as the linear susceptibility. Because the parametric gain is obtained in a single coherence length, the gain bandwidth can exceed the degenerate frequency. In Pb vapor the calculated gain is maximized at 1.88 μm and has a bandwidth of ~7500 cm^{-1}. © 1997 Optical Society of America

This Letter suggests a new type of gas-phase optical parametric oscillator with special properties: (1) a bandwidth that is on the order of the center (degenerate) frequency and (2) electronic tunability that is accomplished by small (gigahertz) tuning of the pumping laser. The essence of the device is the use of an effective nonlinear susceptibility that is of the same order as the linear susceptibility. The oscillator also differs from conventional optical parametric oscillators in that its operation depends on the temporal history of the pumping fields.

A schematic of the suggested technique is shown in Fig. 1. The signal and idler fields (dashed lines) are driven by population-trapped $|1\rangle - |2\rangle$ transition atoms. By use of electromagnetically induced transparency, strong-pumping fields are applied to the $|1\rangle - |3\rangle$ and $|2\rangle - |3\rangle$ transitions. The $|2\rangle - |3\rangle$ field is applied first, thereby setting up a quantum interference and allowing the $|1\rangle - |3\rangle$ field to propagate into what would otherwise be an opaque or refractively thick medium. The $|1\rangle - |3\rangle$ field is then increased slowly compared with the Rabi frequency of the $|2\rangle - |3\rangle$ field. This allows the ground-state atoms to evolve smoothly into a population-trapped superposition state with approximately equal and oppositely phased probability amplitudes in states $|1\rangle$ and $|2\rangle$ and a probability amplitude of 0 in state $|3\rangle$. In the ideal case, there is no dipole moment at either of the pumping fields.

With the above conditions, the gain coefficient of the signal and idler fields is determined by an effective nonlinear susceptibility that has a single nonresonant denominator and is therefore of the same order of magnitude as the linear susceptibility. Stated differently, the distance that, under more usual conditions, would cause $\Delta kL = \pi$, instead results in a gain coefficient of scale unity. This leads to gain bandwidths that are on the order of the degenerate frequency, i.e., $(\omega_2 - \omega_1)/2$.

Research on this oscillator is motivated by the recent experimental results of Jain *et al.*,[1] who, using strongly driven population-trapped atoms, have demonstrated 40% conversion efficiency from blue to ultraviolet light in a single coherence length of Pb atoms. We also note the research of Hemmer *et al.*,[2] in which population trapping is used to obtain unusually efficient phase conjugation. Other suggestions and experiments describing the use of electromagnetically induced transparency in nonlinear optics include those of Harris *et al.*[3] and Hakuta *et al.*[4] Even earlier, Tewari and Agarwal[5] showed that strong fields can modify **k**-vector matching. Adiabatic evolution into a population-trapped state in optically thin media was studied by Oreg *et al.*[6] and by Gaubatz *et al.*[7] Its use to eliminate optical self-focusing near resonance in an optically thick medium was demonstrated by Jain *et al.*[8]

In the following discussion we first calculate the parametric gain at the signal and idler fields as caused by the population-trapped atoms. These atoms are characterized by the density-matrix elements ρ_{ij} of the $|1\rangle - |2\rangle$ transition. Because the signal and idler fields are taken as small, and are also nonresonant, the ρ_{ij} are determined by the strong pumping fields at ω_p and ω_c and not by the fields at ω_s or ω_i.

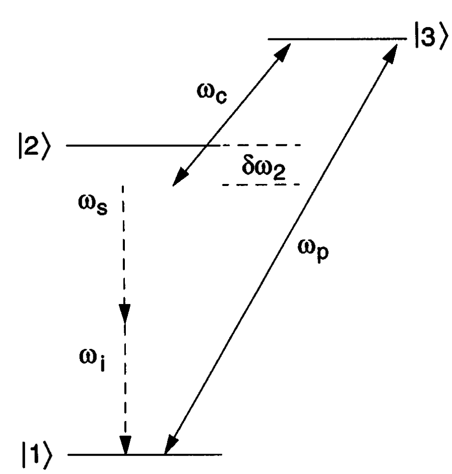

Fig. 1. Energy schematic for an optical parametric oscillator pumped by population-trapped atoms. Electromagnetic fields at frequencies ω_p and ω_c drive population-trapped $|1\rangle - |2\rangle$ transition atoms. The phase variation of these atoms as a function of z is determined by the detuning from two-photon resonance $\delta\omega_2$. A small detuning of $\delta\omega_2$ (typically a few hundred megahertz) causes phase matching and parametric gain at signal and idler frequencies $(\omega_s + \omega_i) = (\omega_p - \omega_c)$.

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We assume that the signal and idler fields are monochromatic with slowly varying envelopes. We also assume that the detuning of these fields from the virtual states that determine the effective susceptibility is large compared with the linewidths of these states. The behavior of the system is then characterized by the constants

$$a_q = \frac{1}{2\hbar^2}\sum_j\left[\frac{|\mu_{1j}|^2}{(\omega_j - \omega_1) - \omega_q} + \frac{|\mu_{1j}|^2}{(\omega_j - \omega_1) + \omega_q}\right],$$

$$d_q = \frac{1}{2\hbar^2}\sum_j\left[\frac{|\mu_{2j}|^2}{(\omega_j - \omega_2) - \omega_q} + \frac{|\mu_{2j}|^2}{(\omega_j - \omega_2) + \omega_q}\right],$$

$$b_q = \frac{1}{2\hbar^2}\sum_j\left[\frac{\mu_{1j}\mu_{2j}}{(\omega_j - \omega_1) - \omega_q} + \frac{\mu_{1j}\mu_{2j}}{(\omega_j - \omega_2) + \omega_q}\right],$$

$$c_q = b_q^*. \tag{1}$$

Here, ω_j are the energies of the virtual states and μ_{ij} are the matrix elements from states $|1\rangle$ and $|2\rangle$ to state j.

The equations for the envelopes of the signal and idler fields as a function of distance are

$$\frac{\partial E_s}{\partial z} = -j\eta_s\hbar\omega_s N[(a_s\rho_{11} + d_s\rho_{22})E_s + b_s\rho_{12}E_i^*],$$

$$\frac{\partial E_i}{\partial z} = -j\eta_i\hbar\omega_i N[(a_i\rho_{11} + d_i\rho_{22})E_i + c_i\rho_{12}E_s^*]. \tag{2}$$

The quantity N is the atom density, $\eta = (\mu/\epsilon_0)^{1/2}$, and ρ_{ij} are the density-matrix elements of the $|1\rangle$–$|2\rangle$ transition. The signal and idler **k** vectors, relative to vacuum, and the coupling constants κ_s and κ_i are

$$k_s = \eta_s\hbar\omega_s N(a_s\rho_{11} + d_s\rho_{22}),$$

$$k_i = \eta_i\hbar\omega_i N(a_i\rho_{11} + d_i\rho_{22}),$$

$$\kappa_s = \eta_s\hbar\omega_s N b_s|\rho_{12}|,$$

$$\kappa_i = \eta_i\hbar\omega_i N c_i|\rho_{12}|. \tag{3}$$

Because of the two strong fields that are used to drive the ρ_{ij}, the coupling constants have only a single nonresonant denominator and at maximum coherence ($|\rho_{12}| = 0.5$) are of the same magnitude as the signal and idler **k** vectors. When we write the coherence ρ_{12} and the **k**-vector mismatch as

$$\rho_{12} = |\rho_{12}|\exp(-j\delta kz),$$

$$\Delta k = \delta k - (k_s + k_i), \tag{4}$$

Eqs. (2) become

$$\frac{\partial E_s}{\partial z} = -jk_sE_s - j\kappa_sE_i^*\exp(-j\delta kz),$$

$$\frac{\partial E_i}{\partial z} = -jk_iE_i - j\kappa_iE_s^*\exp(-j\delta kz). \tag{5}$$

With the change of variables $\tilde{E}_s = E_s\exp(jk_sz)$ and $\tilde{E}_i = E_i\exp(jk_iz)$ and the boundary condition $\tilde{E}_i(0) = 0$, the solution of Eqs. (5) is

$$\frac{\tilde{E}_s(L)}{\tilde{E}_s(0)} = \left[\exp\left(-j\frac{\Delta kL}{2}\right)\right]\left(\cosh sL + j\frac{\Delta k}{2s}\sinh sL\right),$$

$$\frac{\tilde{E}_i(L)}{\tilde{E}_s^*(0)} = -j\frac{\kappa_i}{s}\left[\exp\left(-j\frac{\Delta kL}{2}\right)\right]\sinh sL,$$

$$s = \left(\kappa_s\kappa_i - \frac{\Delta k^2}{4}\right)^{1/2}. \tag{6}$$

If ρ_{12} contains no spatial dependence over and above vacuum, i.e., $\delta k = 0$, then using the constants of Eqs. (1), we find that the parameter s is imaginary, and therefore there is no parametric gain. We obtain gain by adjusting the two-photon detuning $\delta\omega_2$ of the pump and coupling lasers (Fig. 1) to set $\Delta k = 0$ at band center. Because of the large dispersion of near-resonance electromagnetically induced transparency, this can typically be accomplished with detunings of $\delta\omega_2$ of less than 1 GHz.

In the usual four-frequency parametric oscillator, in which all fields are nonresonant, the dipole moments at the signal and idler frequencies depend on the instantaneous values of all of the fields. In our case, the pumping fields must be applied so as to establish the population-trapped state. One way to do this is to apply pulses that have nearly identical envelopes (matched pulses) on the $|1\rangle$–$|2\rangle$ and $|2\rangle$–$|3\rangle$ transitions. The front edge of these pulses prepares the population-trapped state, and in the ideal case there is no absorption or group delay thereafter. To accomplish this, it is essential that the number of photons in the coupling laser pulse be large compared with the oscillator-strength-weighted number of atoms in the laser path.[9] In practice, the pulses should be short compared with the dephasing time of the $|1\rangle$–$|2\rangle$ transition and also must have sufficient intensity that $d_c|E_c|^2$ is large compared with the transition linewidth.

We limit the discussion to the special case of maximum coherence.[1] With E_p and E_c as the field amplitudes at ω_p and ω_c, respectively, maximum coherence is obtained when $a_p|E_p|^2 = d_c|E_c|^2$. One can then show that for two-photon detunings $\delta\omega_2$, which are small compared with $d_c|E_c|^2$, $|\rho_{11}| = |\rho_{22}| = -\rho_{12} = 0.5$ and that the spatially varying phase δkz of ρ_{12} [Eqs. (4)] is

$$\delta k = -\frac{[k_p^{(0)} + k_c^{(0)}]}{2}\frac{\delta\omega_2}{d_c|E_c|^2}. \tag{7}$$

Here, the quantities $k_p^{(0)} = \eta_p\hbar\omega_pNa_p$ and $k_c^{(0)} = \eta_c\hbar\omega_cNd_c$ are the **k** vectors (relative to vacuum) of the fields at frequencies ω_p and ω_c, if all the atoms are in state $|1\rangle$ or state $|2\rangle$, respectively. Because these quantities are very large compared with the nonresonant a_q and d_q evaluated at the signal and idler frequencies, only small detunings of $\delta\omega_2$ are necessary for phase matching.

We illustrate these ideas with parameters for the Pb vapor system that was used in Ref. 1 to demonstrate frequency upconversion at maximum coherence.

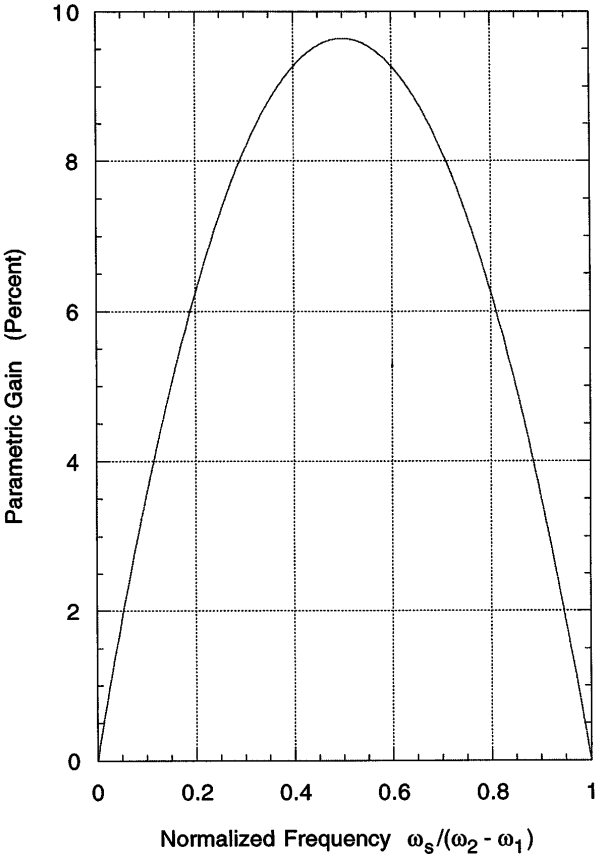

Fig. 2. Parametric gain versus normalized signal frequency. For Pb vapor, the gain maximizes at 1.88 μm and has a bandwidth of ~7500 cm^{-1}.

Here, the energies of states $|2\rangle$ and $|3\rangle$ relative to the ground state are 10 650 and 35 287 cm^{-1}, respectively. To make a pure three-state system, we use right circularly polarized light at ω_p and left circularly polarized light at ω_c. The $|1\rangle$–$|3\rangle$ and $|2\rangle$–$|3\rangle$ transition-matrix elements are then 0.79 and 1.37 atomic units, respectively. (The small contribution of higher transitions to the gain is neglected.)

As in Ref. 1, we assume a cell length of 25 cm. To have reasonable gain, we assume an atom density of 5×10^{16} atoms/cm^3. The power densities at ω_p and ω_c that were chosen to equalize the Rabi frequencies of these fields are 100 and 32.7 MW/cm^2, respectively.

Figure 2 shows the parametric (power) gain as a function of the signal frequency. Here, $\delta\omega_2$ is chosen to set $\Delta k = 0$ at line center and is not varied as the signal frequency varies. The gain peaks at the degenerate frequency of $(\omega_2 - \omega_1)/2 = 5325\ cm^{-1}$ and remains within 50% of its maximum over a bandwidth of 7560 cm^{-1}. To attain phase matching at line center ($\omega_2/2$) requires a two-photon detuning of −110 MHz. (For detunings that are within the rotating-wave approximation, this value is independent of the common detuning from state $|3\rangle$.)

It should be noted that the gain of Fig. 2 can, instead, be interpreted [see Eqs. (6)] as the photon-conversion efficiency from the signal to the idler. Therefore, if one used a conventional optical parametric oscillator to generate radiation in the 0.94–1.88-μm spectral region, the Pb cell, acting as a frequency downconverter, and with fixed $\delta\omega_2$, would generate radiation that is tunable from 1.88 μm to nearly dc.

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