METHOD TO LOCK AN OPTICAL PARAMETRIC OSCILLATOR TO AN ATOMIC TRANSITION*

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A method to lock the output frequency of an optical parametric oscillator to an absorbing atomic transition is proposed. The effective linewidth of the oscillator becomes approximately that of the absorbing transition.

In recent years optical parametric oscillators which are tunable over most of the visible and near infrared spectra have been constructed.[1-6] The linewidth of these oscillators is determined by the length and type of nonlinear crystal, and by how close to degeneracy they are operated. Observed linewidths have been as small as 3 cm^{-1} and have more typically been between 30 and 100 cm^{-1}.

In this letter, we propose a technique which allows the output frequency of an optical parametric oscillator to be locked to the frequency of an absorbing atomic transition. The effective linewidth of the oscillator becomes approximately that of the absorbing transition, which for a gas may be 0.03 cm^{-1} or less.

The basic configuration for the proposed technique is shown in Fig. 1. The usual single nonlinear crystal is replaced by two nonlinear crystals which have the direction of their positive z axes reversed. For example, in $LiNbO_3$ the positive end of the z axis is that end which becomes negative upon compression in the z direction. Between the reversed nonlinear crystals is placed a cell containing the gas to which it is desired to lock the output frequency of the oscillator. We term the frequency of the absorbing transition as the signal frequency, and assume the mirrors of the oscillator to have high reflectivity at only the idler frequency. Oscillators having only their signal or idler frequencies resonant have recently been successfully demonstrated.[7-9]

To understand the operation of the oscillator, first consider the case where the two nonlinear crystals are of equal length, and the absorbing gas is removed. At the center of the parametric linewidth where $\Delta k = 0$, the parametric gain of the first crystal is exactly cancelled by the second crystal. That is, as a result of the reversal of the $+z$ axes of the two crystals, the relative phases of the signal, idler, and pump on entering the second crystal

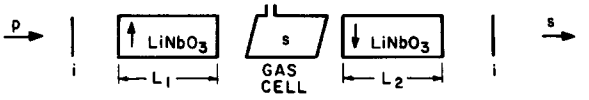

Fig. 1. Schematic of frequency locking technique. The letters s, i, and p denote the signal, idler, and pump, respectively. The gas transition is assumed to absorb at only the signal frequency; and the mirrors are assumed to have high reflectivity at only the idler frequency. The vertical arrows in the $LiNbO_3$ crystals denote the direction of their positive z axes.

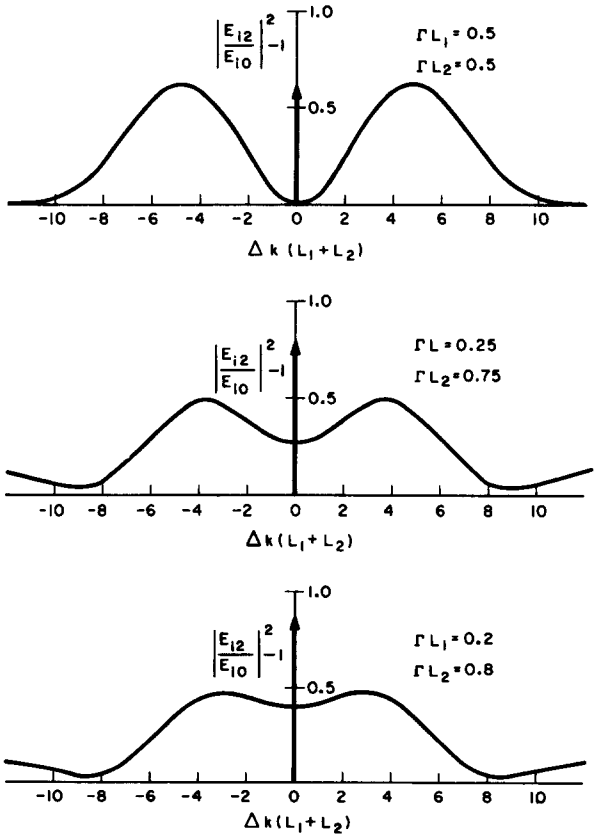

Fig. 2. Incremental power gain versus $\Delta k(L_1 + L_2)$.

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are such that instead of further growth, the signal and idler decay to the values which they had on entering the first crystal. Since relative phases are involved, the physical spacing between the two crystals is not of consequence. Suppose now that an absorbing gas is inserted into the gas cell and the pressure adjusted such that the gas is nearly opaque at the pertinent transition. The signal generated in the first crystal is now absorbed by the gas, and therefore cancellation of gain no longer occurs in the second crystal. The result is a sharply peaked gain function centered at the frequency of the atomic transition. It will be seen below that the peak height of this gain function is between 50 and 100% of the gain which would exist if the $+z$ axes of the two crystals were aligned instead of opposed. It will also be seen that it will be of advantage to make the second crystal a number of times longer than the first.

We will take the gas to have a loss $\alpha(\omega)$ and phase shift $\phi(\omega)$ such that the signal frequency $\bar{E}$ field at its output is related to that at its input by $\exp -[\alpha(\omega) + j\phi(\omega)]$; and assume the gas to be completely transparent at the idler frequency. We take the first nonlinear crystal to have length L_1 and the second to have length L_2. The magnitude of the parametric gain at the idler frequency for a single pass through the two reversed crystals separated by the gas cell is given by

$$\left|\frac{E_{i2}}{E_{i0}}^2\right| = \left| \left(\cosh sL_1 + \frac{j\Delta k}{2s}\sinh sL_1\right) \times \left(\cosh sL_2 + \frac{j\Delta k}{2s}\sinh sL_2\right) - \exp\left[-\alpha(\omega)\right] \times \exp j\phi(\omega)\,\frac{\Gamma^2}{s^2}\sinh sL_1 \sinh sL_2\right|^2, \quad (1)$$

where

$$s = (\Gamma^2 - \tfrac{1}{4}\Delta k^2)^{1/2}.$$

The wave vector mismatch $\Delta k = k_p - k_s - k_i$ is zero at the center of the parametric gain curve and may be written $\Delta k = b\Delta\omega_s$, where $\Delta\omega_s$ is the excursion from line center of the signal frequency. For $LiNbO_3$ at 6328 Å, the constant $b = 6.2 \times 10^{-10}$ sec/m.[10] Γ^2 is the parametric gain constant and is dependent on the strength of the pump. To obtain Eq. (1), pump depletion was neglected and thus Γ was taken to be the same in both crystals. For 90° phase matching in $LiNbO_3$ with $\lambda_s \cong \lambda_i \cong 1\,\mu$, $\Gamma^2 \cong 0.1\,P_p/A$, where A is the area of the pumping beam and P_p/A has units of MW/cm^2.

We first examine the shape of the parametric gain curve which is predicted by Eq. (1) in the absence of the absorbing gas, i.e., with $\alpha(\omega) = \phi(\omega) = 0$. Figure 2 shows the incremental power gain $|E_{i2}/E_{i0}|^2 - 1$ plotted versus $\Delta k(L_1 + L_2)$, for three cases where $\Gamma L_1 = 0.5$, $\Gamma L_2 = 0.5$; $\Gamma L_1 = 0.25$, $\Gamma L_2 = 0.75$; and $\Gamma L_1 = 0.2$, $\Gamma L_2 = 0.8$, respectively. For the case of two crystals of equal length the gain is zero at line center and rises off line center. Crystals of differing length (note that the sum of the lengths is held constant) increase the gain at line center and reduce the ripple.

Next, consider the gain in the presence of the absorbing gas. We will assume that the linewidth of the absorbing transition is many times smaller than that of the parametric gain, and that the temperature of the nonlinear crystals have been adjusted such that their parametric linewidths center at approximately the atomic transition. For such cases Δk may be taken equal to zero in the vicinity of the atomic transition, and Eq. (1) gives

$$\left|\frac{E_{i2}}{E_{i0}}\right|^2 = \left|\cosh \Gamma L_1 \cosh \Gamma L_2 - \exp\left[-\alpha(\omega)\right]\exp j\phi(\omega)\sinh \Gamma L_1 \sinh \Gamma L_2\right|^2. \quad (2)$$

For large $\alpha(\omega)$, i.e., for a nearly opaque gas, $|E_{i2}/E_{i0}|^2 = \cosh^2\Gamma L_1 \cosh^2\Gamma L_2$. This gain is shown plotted as a delta function at $\Delta k = 0$ for each of the three cases in Fig. 2. For the case of a 4 to 1 ratio of crystal lengths, i.e., $\Gamma L_1 = 0.2$, $\Gamma L_2 = 0.8$, the ratio of gain at the atomic transition to the maximum parametric gain which occurs elsewhere on the line is about 1.8. Choosing the second crystal longer than the first has the additional advantage that the signal power entering the gas from the first crystal varies as L_1^2, and thus the possibility of saturating the gas is reduced.

The detailed shape of the parametric gain in the vicinity of the transition is determined by the relation of $\alpha(\omega)$ to $\phi(\omega)$. For an ideal Lorentzian line, $\phi(\omega)/\alpha(\omega)$ is proportional to the detuning from the atomic line center. For small $\alpha(\omega)$, the peak parametric gain occurs at the center of the atomic line. However, for larger $\alpha(\omega)$, e.g., higher pressure or longer cell length, higher gain results toward the wings of the atomic line where the ratio of $\phi(\omega)$ to $\alpha(\omega)$ is larger. [Note that the most favorable case for Eq. (2) is $\alpha(\omega) = 0$, $\phi(\omega) = \pi$.] In this case, the gain becomes double peaked, dipping at the center frequency of the atomic transition. As a result of this phase contribution of the atomic line, the peak gain may exceed the height of the delta functions shown in Fig. 2, and the width of the gain function may be a few times wider than that of the atomic transition.

In constructing an oscillator of the type described here, it may be desirable to make the $c/2L$ frequency spacing of the idler modes less than the width of the atomic transition. Even though the pump frequency is randomly fluctuating, there would then always be at least one idler mode such that the difference between the pump frequency and its frequency falls within the width of the atomic transition.

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A THERMALLY PUMPED CO_2 LASER*

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Power of 20 mW and gain of 11% have been obtained by using thermally excited nitrogen to pump a flowing-gas carbon dioxide laser. One immediate use of this new pumping technique is the study of molecular laser physics in a simplified environment; the thermal laser is essentially free of chemical and charged-particle effects.

In an earlier paper[1] we reported the observation of 10.6-μ optical gain in a thermally pumped CO_2–N_2 system, a development originally proposed by Basov, Oraevskii, and Shcheglov.[2] With a few changes in apparatus we have increased the gain sufficiently to make a laser, whose peak output is estimated to be 20 mW, and whose peak small-signal gain is 11%. The apparatus, which appears in Fig. 1, may be described as follows:

(1) Nitrogen at a pressure of about 400 Torr is heated in an oven to 1200°C, as measured by a thermocouple in the gas stream. The estimated flow rate is about 25 cm/sec as the gas enters the oven at 27°C and about 125 cm/sec when it reaches 1200°C; these flow rates should be low enough to permit the vibrational temperature T_v to come to equilibrium with the translational temperature T_t.[3] The experimental evidence that T_v does have time to reach equilibrium with T_t is that the laser performance is essentially unchanged by doubling the length of the oven.

(2) The nitrogen expands through a converging-diverging nozzle of 1-mm throat diameter to a condition of low pressure and supersonic velocity,[4] and passes out of the oven into a section of tubing whose walls are near room temperature. Standing shock waves[4] in this cooling tube decelerate the gas to a subsonic velocity near 3.4 cm/sec, while the pressure reaches a value of about 6 Torr. Neither the supersonic velocity nor the shocks were directly measured in the laser apparatus; we infer their presence from pitot tube measurements we have made on very similar nozzle-duct apparatuses. The subsonic velocity was calculated from measurements of pressure and mass flow rate.

The purpose of the cooling tube is to rapidly lower the nitrogen's translational temperature. The low pressure, the high flow velocity, and the low vibrational relaxation rate that accompanies a low translational temperature[3] result in the nitrogen's entering the laser tube with $T_v > T_t$. Note that the present apparatus does not utilize the rapid gas cooling which accompanies acceleration to supersonic velocity—the nitrogen reheats as it decelerates in the standing shocks, and must ultimately lose its translational energy by conduction to the walls of the cooling tube. Eventually it should be more efficient to utilize the supersonic cooling directly, particularly in large-bore tubes, where conduction cooling would become very slow.

(3) After the excited N_2 has entered the laser tube, it is mixed with 1–1.5 Torr of room-temperature CO_2. Because of the near coincidence between the first excited vibrational state of N_2 and the $00^\circ1$ state of CO_2, excitation is transferred to the CO_2 in a time on the order of 50 μsec.[5] This leads to inversion on the $00^\circ1$–$10^\circ0$ transition of CO_2, and allows us to construct a 10.6-μ laser. (At certain cavity lengths we have also detected weak oscillation on the 9.6-μ transition, $00^\circ1$–$02^\circ0$.) The fact that we observe both R and P transitions of the $00^\circ1$–$10^\circ0$ band implies the existence of complete

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