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Saturation Effects in a Tunable Coherent Near-Infrared Source

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also be used for far-ir generation, where instead of the sum frequency one is interested in the difference frequency. The limitation on the tunability because of the finite spectral range of the active medium can be overcome by using different dyes in regions A′ and B′ of Fig. 1.

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Saturation effects in a tunable coherent near-infrared source*

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A saturation effect in a tunable infrared source utilizing four-wave parametric conversion in potassium vapor is reported and is shown to be the result of parasitic oscillations. A hundredfold increase over previously attained power levels has been effected via elimination of these oscillations.

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We have repeated the experiments of Sorokin, Wynne, and Lankard[1-3] on the generation of coherent tunable near-ir radiation utilizing a resonantly enhanced four-wave parametric process in potassium vapor. The energy level diagram for the process is shown in Fig. 1. Our experimental arrangement is identical to that described in Refs. 1 and 2. The orthogonally polarized beams of two individually tunable dye lasers at frequencies ν_L and ν_P pumped by the same nitrogen laser are collinearly combined in a Glan prism and then focused into a heat-pipe oven containing potassium vapor. The third beam at ν_S necessary for the four-wave process results from stimulated electronic Raman scattering (SRS) in the potassium vapor and thus automatically fulfills the condition for resonant enhancement. The ir power output as a function of wavelength is given in Table I. The following observations have been made:

(i) The ir power output reaches a maximum value of 9.5 W at 2.2 μ. This improvement of a factor of 95 over the power quoted in Refs. 1–3 is obtained with only a factor-of-2 increase in input power.[3]

(ii) The ir power is not maximized by apportioning two-thirds of the available input power to the dye laser at ν_L as would follow from the third-order susceptibility given in Eq. (2) of Ref. 2 and repeated in Eq. (1) below. Instead, we find the optimum power level to be 2.6 kW at all ir wavelengths.[4] After this value is reached, the ir power depends linearly on the input power at ν_p and is unaffected by further increases in the power at ν_L.

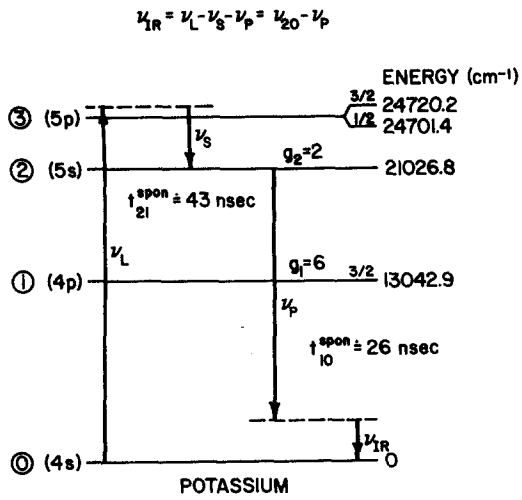

FIG. 1. Energy level scheme for four-wave parametric mixing in potassium vapor. The input beams at ν_L and ν_p are provided by two independently tunable dye lasers (Molectron model D200) pumped by the same nitrogen laser (Molectron model UV1000). The beam at ν_S is produced by stimulated electronic Raman emission induced by tuning ν_L to energies slightly above the 5p resonance. The power at ν_S is linearly proportional to the power of the beam at ν_L. The spontaneous lifetimes were obtained from Ref. 5.

TABLE I. Infrared output power as a function of wavelength. The power was measured with a cooled Ge:Au photoconductive detector which was calibrated using the SRS emission at 2.66 μ. We compared the output of the Ge:Au detector with that of a calibrated joulemeter. This 2.66-μ calibration was extrapolated to other wavelengths by using standard Ge:Au sensitivity vs wavelength curves. We estimate the accuracy of our calibration to be $\pm 10\%$. The input power at ν_L was a constant 2.6 kW. At the ir wavelength of 2.2μ the input power at ν_p was 41 kW. The heat-pipe pressure was 15 Torr.

ir wavelength (μ)	ir power (W)
2.0	4.4
2.1	7.6
2.2	9.5
2.3	2.9
2.4	1.1
2.5	1.1
2.6	0.65
2.7	0.61
2.9	0.48
3.0	0.38
3.3	0.06
3.9	0.01
4.0	0.01

(iii) The SRS power at ν_s is a linear function of the input power at ν_L from threshold (~100 W) to 10.8 kW (the maximum power available from our system).

(iv) As the optimal power of 2.6 kW at ν_L is exceeded, a highly directional output signal at 1.25 μ commences. Wynne *et al.*[2] have suggested that the ir power output should be proportional to the cube of the power of the nitrogen laser which pumps the dye lasers unless saturation effects limit the conversion. Our findings in (ii) above indicate that saturation effects *do* occur very quickly and that afterwards ir power is only linearly proportional to the additional nitrogen pump laser power. One likely explanation of this phenomenon is that the SRS process has saturated. This possibility is excluded, however, by our observations in (iii). We propose an alternative mechanism:

Figure 1 indicates that stimulated emission at 1.25 μ, as observed in (iv) above, is possible between levels 2 and 1 because the condition for inversion, $g_1 t_2^{\text{spont}} > g_2 t_1^{\text{spont}}$, is satisfied. Once the SRS mechanism has populated level 2 to the critical inversion density, no increase in pump power density (corresponding to 2.6 kW total power for our geometry) will increase the density of atoms in this state. The consequences for the third-order susceptibility[2]

$$\chi^{(3)} = \frac{iN_0 e^4}{h^3} \frac{x_{01}x_{12}x_{23}x_{30}}{(\nu_{10}-\nu_p)(\nu_{30}-\nu_L)\Gamma} \tag{1}$$

can be seen by writing $\chi^{(3)}$ in terms of the relevant density matrix element ρ_{20}. Then

$$\chi^{(3)} = \{E_L E_S^* \exp[-2\pi i(\nu_L-\nu_S)t]\}^{-1}[e^2 N_0 x_{10}x_{12}/h(\nu_{10}-\nu_p)] \times \rho_{20}^{(2)}(\nu_L-\nu_S). \tag{2}$$

By definition,

$$\rho_{20}(\nu_L-\nu_S) = \overline{a_2 a_0^*}\exp(-2\pi i\nu_{20}t),$$

where the a_i's are the standard coefficients of time-dependent perturbation theory and only those terms are included for which the frequency dependence of the product is $(\nu_L-\nu_S)$. Because virtually all of the atoms are in the ground state before the perturbation fields are applied, it is permissible to remove the ensemble average and set $a_0^*(t)=1$. The ir power is proportional to $|\chi^{(3)}|^2$ and thus to $a_2 a_2^*$. Furthermore, $\rho_{20}(\nu_L-\nu_S)$ is the only important term in the perturbation expansion of ρ_{20}, and thus

$$a_2(\nu_L-\nu_S)a_2^*(\nu_L-\nu_S) = N_2 N_0^{-1} \times \text{const}.$$

Therefore, once stimulated emission at 1.25 μ begins, $\rho_{20}(\nu_L-\nu_S)$ is proportional to $(N_2^{\max}N_0^{-1})^{1/2}$, a constant. With this value substituted for $\rho_{20}(\nu_L-\nu_S)$ in Eq. (2), it is apparent that the resultant ir power, $|E_L(t)E_S^*(t)E_p^*(t)\chi^{(3)}|^2$, is independent of E_L and E_S and thus exhibits the observed functional behavior. In the unsaturated region, Eq. (1) is still correct.

Even though this effect limits the realizable ir power possibilities to less than those predicted in Ref. 2, the power levels we have already achieved indicate that ir sources based on four-wave parametric conversion in the alkali metal vapors should become practical spectroscopic tools. With the incorporation of a concentric heat pipe into our system, as discussed in Ref. 2, we are confident that ir beam powers on the order of 1 mW can be generated out to 30 μ. Because we have determined that the optimal power density at ν_L corresponds to the total power of 2.6 kW for *all* ir wavelengths, the next step for increasing the ir power will be to improve the efficiency of the dyes used to generate the beam at ν_p. This is particularly important for the wavelength region $\lambda_{ir} > 2.8$ μ where the rhodamine dyes can no longer be used to generate the radiation beam at ν_p.

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