RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 30, NUMBER 5 NOVEMBER 1984

Competition between Raman and line-center oscillations in optically pumped far-infrared lasers

M. A. Dupertuis and M. R. Siegrist
Centre de Recherches en Physique des Plasmas, Assoiciation EURATOM–Confédération Suisse, Ecole Polytechnique Fédérale de Lausanne, CH-1007 Lausanne, Switzerland

R. R. E. Salomaa*
Max-Planck-Institut für Quantenoptik, D-8046 Garching, Federal Republic of Germany
(Received 20 July 1984)

Competition between Raman and line-center oscillations in off-resonantly pumped far-infrared lasers is studied by means of semiclassical laser theory. The observed preferential Raman line operation is shown to be caused by an inversion of the line-center gain when the Raman mode reaches intensities well below full saturation. This inversion is partly due to simple population effects. A further enhancement in the cross saturation at line center is caused by population pulsations. Numerical examples of mode competition are presented.

Semiclassical laser theory predicts an equal small-signal gain for both the laserlike line-center oscillation and the Raman-like transition in an off-resonantly pumped homogeneously broadened far-infrared (FIR) laser.[1,2] Contrary to expectations, however, only the Raman line is usually excited in experiments.[3] Mechanisms suggested as being responsible for the preferential Raman line oscillation include, e.g., ground-state absorption[4] and the different saturation behavior of the FIR and line-center modes.[5] For short-pulse excitation the build-up time for Raman oscillation may be too long in which case only the line-center transition is able to start lasing after the excitation pulse.[6] In this Rapid Communication we show that for long excitation pulses the preferred Raman oscillation is a result of mode competition between the two lines. To correctly describe the situation we must include mode-coupling terms into the gain equations of the two FIR modes. In the literature, mode competition in the present context has often improperly been discussed on the basis of results valid for a single pump and FIR mode. The effects discussed in this paper are also relevant for other kinds of optically pumped lasers as well as for some stimulated Raman scattering (SRS) experiments.

We consider a nondegenerate homogeneously broadened three-level system where the levels are indexed by 1 (ground level), 2 (upper FIR level), and 3 (lower FIR level). The transition $1\rightarrow 2$ is coupled by the optical pump field $\frac{1}{2}E_0\exp[i(Kz-\Omega t)]+\text{c.c.}$ The far-infrared field is assumed to contain two cotraveling modes

$$E_{\text{FIR}}=\tfrac{1}{2}\mathscr{E}_0\exp[i(k_0z-\nu_0t)]+\tfrac{1}{2}\mathscr{E}_1\exp[i(k_1z-\nu_1t)]+\text{c.c.} \quad (1)$$

Within the rotating-wave approximation the response of the three-level system to these three fields is exactly soluble with matrix continued-fraction techniques.[7] To simplify the analytical expressions below we assume that the pump laser is so much detuned that it suffices to include terms up to order (E_0^2) and that the FIR coupling is adequately described to lowest order. Also ac Stark shifts are ignored for brevity. These limitations are readily relaxed at the expense of longer expressions and we emphasize that the numerical results below are calculated with the full code described in Ref. 7.

Let us assume that the FIR mode 0 oscillates at the Raman resonance, i.e., $\Omega-\nu_0=\omega_{31}$ where ω_{31} is the atomic resonance frequency, and the mode 1 exactly at the line center $(\nu_1=\omega_{23})$. In this case the Raman gain is approximately

$$G_0(\beta_0,\beta_1)=(C\alpha^2/\Delta^2)(1+\beta_1^2/\gamma^2)^{-1} \quad (2)$$

and the line center gain

$$G_1(\beta_0,\beta_1)=(C\alpha^2/\Delta^2)(1+\beta_1^2/\gamma^2)^{-1}(1+4\beta_1^2/\gamma^2)^{-1}\times(1-3\beta_0^2/\gamma^2+\beta_1^2/\gamma^2)\ , \quad (3)$$

where $C=\nu\mu_{23}^2n_1^0/(\hbar\epsilon_0c\gamma)$. In (2) and (3) we have denoted the flipping frequencies by $\alpha=\mu_{21}E_0/2\hbar$ (μ_{mn} is the dipole matrix element between states n and m) and $\beta_k=\mu_{23}\mathscr{E}_k/2\hbar$ $(k=0,1)$, the pump detuning by $\Delta=\omega_{21}-\Omega$ (note that the assumed resonance implies that $\omega_{23}-\nu_0=\nu_1-\nu_0=\Delta$). Relaxation is contained in the rate parameter γ (taken equal for all density matrix elements) and only ground-level population n_1^0 is assumed to be present in the absence of fields. We do not include in this discussion the effect of the small nonzero equilibrium populations n_2^0 and n_3^0, although we have taken it into account in some calculations. As soon as the pump is strong enough, i.e., $\alpha^2n_1^0>\Delta^2(n_3^0-n_2^0)$ this effect is negligible. The gain functions (2) and (3) are valid to order $\alpha^2/\Delta^2,\beta_k/\Delta$. For notational simplicity we have also neglected the pump-induced ac Stark shifts (of the order α^2/Δ). In the limit $\beta_1\rightarrow 0$ Eq. (2) reduces to the familiar single-mode Raman-gain expression, and Eq. (3) in the limit $\beta_0\rightarrow 0$ to the corresponding line-center gain. Despite the intensity limitations Eqs. (2) and (3) still contain considerable saturation and mode coupling.

The stability analysis of an assumed single-mode operation is readily performed with the aid of (2) and (3). Stable single-mode Raman-line oscillation $(\beta_0\neq 0,\ \beta_1=0)$ is expected for $\beta_0^2>\gamma^2/3$ because then the small signal line-center gain $G_1(\beta_0,0)$ is negative according to (3) and small fluctuations around $\nu_1=\omega_{23}$ are damped. In the case of an oscillator the Raman intensity is solved from $G_0(\beta_0, 0) = l$ where l represents cavity losses. Stability requires that $G_1(\beta_0, 0) < l$. Note that for very high values of $\beta_0(\beta_0^2/\gamma\Delta \gtrsim 0.5)$ Eqs. (2) and (3) are not valid and, therefore, they should be applied only to study the initial mode competition period, i.e., to study whether β_0 can grow large enough to suppress β_1. According to (2) line-center oscillation is not able to suppress Raman oscillation: $G_0(0, \beta_1)$ remains positive throughout the validity range of (2).

Part of the mode competition is easily understood. The two-photon Raman transition transfers ground-level population to level 3. This reduces (for the line-center oscillation required) population inversion between levels 2 and 3 created by off-resonant single-photon pumping $1 \rightarrow 2$. On the other hand, line-center oscillation tends to equalize only populations between the FIR levels and thus has a small influence on the Raman line which mainly depends on ground-level population. The single-photon pumping rate $1 \rightarrow 2$ is roughly $\gamma\alpha^2/\Delta^2$ and the two-photon pumping rate $1 \rightarrow 3$ is $\alpha^2\beta_0^2/\Delta^2\gamma$ which would suggest that the population inversion between levels 2 and 3 is destroyed for $\beta_0^2 > \gamma^2$. As expected, simple population arguments are insufficient and coherence effects have to be included to explain the smaller observed critical value $\beta_0^2 = \gamma^2/3$. (Already in the single-mode case the coherence part gives an important contribution; e.g., at line center the population part interferes destructively with the coherence part, reducing it to one-half). The line-center oscillation in the presence of three waves is affected, in addition to the "stepwise" processes $[1 \rightarrow (\Omega) \rightarrow 2$ and $1 \rightarrow (\Omega + \nu_1) \rightarrow 3]$, also by a three-photon process $1 \rightarrow 2$ where a pump photon Ω and a line-center photon ν_1 get absorbed and simultaneously a Raman photon ν_0 is emitted (parametric absorption). Owing to the fact that the intermediate level 3 is resonant, it is not easy to distinguish this process from a stepwise one without considering clearly unequal relaxation rates or pump laser incoherence.[8]

We have studied the questions above by plotting the various contributions to the density matrix elements $\rho_{23}(1)$ and $\rho_{23}(0)$ which give the gain at line center and at the Raman transition, respectively. In the equation of motion for $\rho_{23}(1)$,

$$\dot{\rho}_{23}(1) = -(\gamma + i\Delta)\rho_{23}(1) + i\beta_1[\rho_{22}(0) - \rho_{33}(0)] + i\beta_0[\rho_{22}(1) - \rho_{33}(1)] - i\alpha\rho_{31}^*(-1) \ ; \quad (4)$$

there appears a dc population term $[\rho_{22}(0) - \rho_{33}(0)]$, a term $[\rho_{22}(1) - \rho_{33}(1)]$ arising from population pulsations at the beat frequency $\nu_1 - \nu_0$, and an off-diagonal term $\rho_{31}^*(-1)$ describing coherence between levels 1 and 3. These give contributions $2 - 2\beta_0^2/\gamma^2$, $-\beta_0^2/\gamma^2$, and -1, respectively, to the factor $1 - 3\beta_0^2/\gamma^2$ occurring in Eq. (3) in the limit $\beta_1 << \gamma$. The dc population difference indeed changes its sign at $\beta_0^2/\gamma^2 = 1$ as was qualitatively argued above. The two other source terms representing pure coherence effects reduce the small signal gain [factor -1 due to $\rho_{31}^*(-1)$] and increase the cross saturation (population pulsations). The line-center gain, therefore, changes sign already at $\beta_0^2 = \gamma^2/3$.

The discussion above elucidates part of the physics behind the asymmetric mode coupling appearing in (2) and (3). We will now discuss if a FIR laser or amplifier can reach a single-mode operating point after an initial transient period which would verify that mode competition is the main reason for the preferred Raman-line oscillation in FIR lasers. Assuming a steady-state amplifier (the oscillator case is analogous in the case of long pulse excitation when pulse propagation and transient effects are negligible), we obtain the normalized mode intensities from

$$\frac{d\beta_k^2}{dz} = G_k(\beta_0^2, \beta_1^2)\beta_k^2 \quad (k = 0, 1) \ . \quad (5)$$

Figure 1 gives an example of a numerical integration of Eq. (5) where G_k are calculated with the full code instead of the approximate equations (2) and (3). At the entrance plane the modes are assumed to have the same boundary value $\beta_0^2(0) = \beta_1^2(0) = \mathscr{E}^2(0) << \gamma^2$. Initially both modes grow exponentially with the same small signal gain. Once β_0^2 and β_1^2 become of the order γ^2 the Raman line suppresses the growth of the line-center oscillation. On the other hand, the increase of the Raman intensity is only moderately slowed down by the presence of line-center oscillation. The maximum of β_1^2 appears when $\beta_0^2 \approx 0.4\gamma^2$ which is in good agreement with the small signal ($\beta_1^2 = 0$) prediction $\beta_0^2 = \gamma^2/3$. After this point β_1^2 starts to decrease, but β_0^2 continues to increase. Notice that higher-order terms which are for simplicity neglected in Eq. (2) are responsible for the saturation behavior of the Raman mode. For comparison we have also shown in Fig. 1 the line-center evolution in the case when the Raman mode is neglected. Single-mode

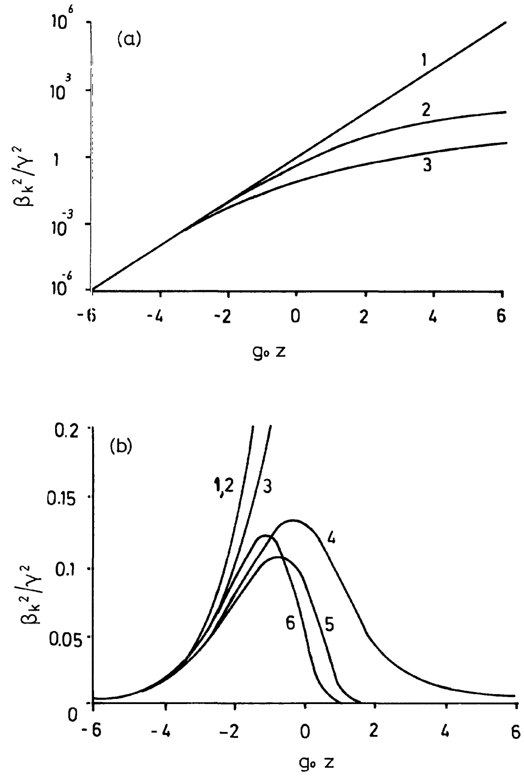

FIG. 1. Line-center (LC) and Raman line (R) intensity vs distance of propagation in an amplifier. ($\alpha = 1, \Delta = 4, \gamma = 1$.) Curve 1: $\exp(g_0 z)$; curve 2: single R mode; curve 3: single LC mode; curve 4: LC mode in presence of R mode; curve 5: as 4, but $\alpha = 0.1$, $\Delta = 30$; curve 6: Eq. (7).

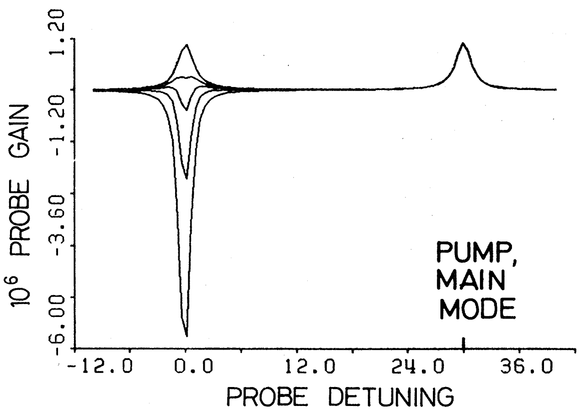

FIG. 2. Probe gain $[\rho_{23}(1)]$ profile vs detuning for $\alpha=0.1$, $\beta_1=0.1$, $\Delta=30$, $\gamma=1$, and $\beta_0=0.1, 0.5, 0.7, 1., 1.5$, respectively (in order of decreasing peak value).

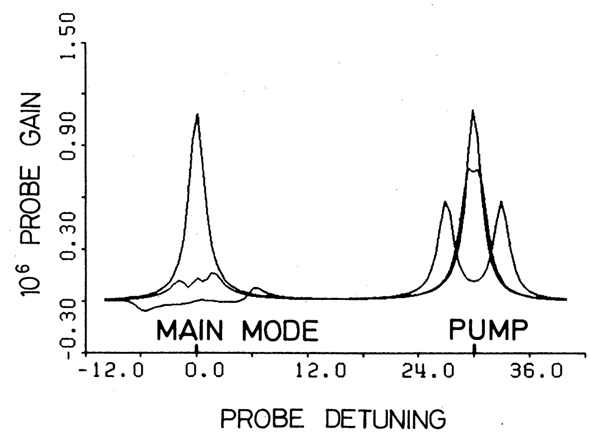

FIG. 3. As Fig. 3 but the main mode on line center. $\beta_0=0.1$, $\beta_1=0.01, 0.07, 3.0$ (in order of decreasing peak value).

line-center oscillation saturates more strongly than Raman oscillation, but this effect does not suppress the line-center oscillation and cannot explain the observed preferential Raman operation. Mode competition effects instead lead to true extinction of the line-center oscillation.

Simple analytical results are obtained by neglecting β_1^2 in (2) and (3). In this case we get

$$\beta_0^2(z)=\mathscr{E}^2(0)\exp(g_0z) \ , \qquad (6)$$

$$\beta_1^2(z)=\mathscr{E}^2(0)\exp(g_0z)\exp\{-[3\mathscr{E}^2(0)/\gamma^2](e^{g_0z}-1)\} \ , \qquad (7)$$

where $g_0=c\alpha^2/\Delta^2$ is the small-signal gain.

According to (7) the maximum value of β_1^2, $\max\beta_1^2 =\gamma^2/3\exp[3\mathscr{E}^2(0)/\gamma^2-1]$, appears at $g_0z_{\max}=+\ln[\gamma^2/(3\mathscr{E}^2(0))]$. The decay of the line-center mode back to an insignificant level takes place over a very short distance. According to (6) and (7) an intensity ratio $R=\beta_0^2/\beta_1^2$ of the two modes is obtained at a distance $g_0z=\ln[\ln(R)]$ from the peak of the line-center mode (e.g., $R=100$ at $g_0z=1.53$). After this transient period single-mode operation is established. Figure 1 reveals that Eq. (7) is a reasonably good approximate solution of the line-center evolution.

According to the results above single-mode Raman-line oscillation in an optically pumped FIR laser is well explained by mode competition effects. So far we have assumed that the two modes are exactly on line center and on the Raman line. In Fig. 2 we show the small-signal gain spectrum for a weak pump at various values of Raman intensity β_0^2. The gain peak around the line center becomes negative for $\beta_0^2\simeq\gamma^2/3$ and oscillations in this region are suppressed.

Figure 3 shows the small-signal gain spectrum for the case when the strong FIR mode oscillates at line center, i.e., $\beta_1\neq 0$. Apart from Stark splitting at higher intensities, the gain near the Raman peak is only weakly affected. In contrast the behavior near the line center shows very strong saturation due to the resonance of the oscillating mode β_1. The gain spectra in Figs. 2 and 3 differ drastically from the single-mode results which are reproduced only when $\beta_0^2\rightarrow 0$ in Fig. 2 and when $\beta_1^2\rightarrow 0$ in Fig. 3. This demonstrates the importance of a proper mode interaction treatment.

---

*Permanent address: Department of Technical Physics, Helsinki University of Technology, SF-02150 Espoo 15, Finland.

[1]S. J. Petuchowski, A. T. Rosenberger, and T. A. De Temple, IEEE J. Quantum Electron. **QE-13**, 476 (1977).

[2]R. L. Panock, and R. J. Temkin, IEEE J. Quantum Electron. **QE-13**, 425 (1977).

[3]P. Woskoboinikow, H. C. Praddaude, W. J. Mulligan, and B. Lax, J. Appl. Phys. **50**, 1125 (1979); R. Frey, F. Pradère, and J. Ducuing, Opt. Commun. **23**, 65 (1977); R. Marx *et al.*, IEEE J. Quantum Electron. **QE-17**, 1123 (1981).

[4]T. A. De Temple, in *Infrared and Millimeter Waves*, edited by K. J. Button (Academic, New York, 1979), Vol. I, pp. 129–184.

[5]Z. Drozdowicz, R. J. Temkin, and B. Lax, IEEE J. Quantum Electron. **QE-15**, 170 (1979); **QE-15**, 865 (1979).

[6]J. D. Wiggins, Z. Drozdowicz, and R. J. Temkin, IEEE J. Quantum Electron. **QE-14**, 23 (1978).

[7]M. A. Dupertuis, R. R. E. Salomaa, and M. R. Siegrist, IEEE J. Quantum Electron. **QE-20**, 440 (1984).

[8]R. R. E. Salomaa, J. Phys. B **11**, 3745 (1978); J. Mostowski and S. Stenholm, Phys. Scr. **26**, 211 (1982).