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ABSTRACT

Competition between Raman and line-centre oscillation in multilevel lasers pumped with coherent radiation off resonance is studied by means of semi-classical laser theory. In a four level system consisting of a combination of a Λ and V system, it is shown that simultaneous Raman and line centre oscillation is expected on one of the transitions contrary to the case of a simple three level system where the Raman line completely suppresses the line centre oscillation. The crucial importance of a new highly nonlinear phase locking effect for the behaviour of the system is demonstrated.

Already for some time it has been known that off-resonantly pumped far infrared (FIR) lasers oscillate on the Raman line [1]. This feature has led to important applications such as the realisation of widely tunable CH_3F lasers (150 - 1200 μm) [2] and the use of notch-filters for Thomson scattering experiments in fusion plasma [3]. Only recently it has been shown that the preferred Raman oscillation over line centre oscillation in a three-level system can be explained by mode competition between the two lines: the line centre gain turns out be be suppressed by the combined action of simple population effects, population pulsations and off-diagonal coherence contributions [4]. The same considerations are valid in an equivalent problem of a single "refilling" transition shown in Fig. 1.

In certain experiments [5-8] both transition types depicted in Fig. 1 appear simultaneously. In [6] it has been shown that in the combined configuration of Fig. 2 also line centre oscillation is possible at one transition. The purpose of this letter is to study the combined system and explore the possible interference effects between Λ and V transitions and to predict the behaviour of the line centre and Raman modes in each transition. It will be shown that a line-centre emission develops asymptotically in one of the transitions and that nonlinear dispersion plays an important role in the evolution stage of the Raman modes.

The transition $1 \leftrightarrow 2$ is coupled to a single-mode (IR) pump field $\{1/2 E_p \exp[i(Kz-\Omega t)] + c.c.\}$. The signal fields (microwave or FIR)

$$\mathcal{E}_{ij} = \frac{1}{2} E_{ij,0} \exp\left[i(k_{ij,0} z - \nu_{ij,0} t)\right] + \frac{1}{2} E_{ij,1} \exp\left[i(k_{ij,1} z - \nu_{ij,1} t)\right] + c.c. \quad (1)$$

interact with the respective transitions, (ij) = (01) or (23) and may both be composed of two modes. In the following we assume for simplicity that the FIR modes indexed by 0 oscillate at the Raman resonances, i.e. $\nu_{01,0} = \Omega - \omega_{20}$ and $\nu_{23,0} = \Omega - \omega_{31}$ (ω_{ij} is the transition frequency), and the modes 1 at line centres, i.e. $\nu_{01,1} = \omega_{01}$ and $\nu_{23,1} = \omega_{23}$.

We have calculated the gain and dispersion of the above fields in a unidirectional amplifier configuration using semi-classical laser theory (the details are obtained by including the two-mode treatment according to [4]) in the four-level system of [12]. In the derivation of field equations and material polarization we have made the rotating wave and slowly varying envelope approximations, neglected Doppler shifts, assumed equal decay rates γ for all density matrix elements, and supposed thermal populations $n_0 = n_1 = n$ and $n_2 = n_3 = 0$ of the levels considered. For a detuned pump, such that $\Delta = \omega_{21} - \Omega \gg \gamma$ it is possible to account only for terms up to order Δ^{-2}. The resulting equations are then transparent enough to reveal several interesting aspects of the evolution of the FIR modes. A more accurate treatment can be done numerically. The validity restrictions of the present approximation are essentially the same as those given in [4].

It is convenient to introduce dimensionless complex field amplitudes (k = 0,1)

$$\mu_k = \frac{\mu_{01} E_{01,k}}{2\hbar\gamma} \quad , \quad \alpha = \frac{\mu_{12} E_{\uparrow}}{2\hbar\gamma} \quad , \quad \beta_k = \frac{\mu_{23} E_{23,k}}{2\hbar\gamma} \qquad (2)$$

where μ_{ij} is the dipole matrix element of the transition i→j. (α, β, and μ are Rabi flipping frequencies in units of γ). The field amplitudes are obtained, within the approximations mentioned above, from

$$\frac{d|\alpha|}{dz} = -\frac{c_\alpha \gamma^2}{\Delta^2}|\alpha|\left\{1+\frac{1}{D}\left[N\left(|\mu_0|^2+|\beta_0|^2\right)-4|\mu_0||\beta_0||\mu_1||\beta_1|\cos(\Phi)\right]\right\} \quad (3)$$

$$\frac{d|\mu_0|}{dz} = \frac{c_\mu \gamma^2}{\Delta^2}\frac{|\alpha|^2}{D}\left\{N|\mu_0| - 2|\mu_1||\beta_1|\cos(\Phi)|\beta_0|\right\} \quad (4)$$

$$\frac{d|\beta_0|}{dz} = \frac{c_\beta \gamma^2}{\Delta^2}\frac{|\alpha|^2}{D}\left\{N|\beta_0| - 2|\mu_1||\beta_1|\cos(\Phi)|\mu_0|\right\} \quad (5)$$

$$\frac{d|\mu_1|}{dz} = \frac{c_\mu \gamma^2}{\Delta^2}\frac{|\alpha|^2|\mu_1|}{1+4|\mu_1|^2}\left\{1-\frac{1}{D}\left(|\mu_0|^2-|\beta_0|^2\right)\left[3\left(1+|\mu_1|^2\right)+|\beta_1|^2\right]\right\} \quad (6)$$

$$\frac{d|\beta_1|}{dz} = \frac{c_\beta \gamma^2}{\Delta^2}\frac{|\alpha|^2|\beta_1|}{1+4|\beta_1|^2}\left\{1+\frac{1}{D}\left(|\mu_0|^2-|\beta_0|^2\right)\left[3\left(1+|\beta_1|^2\right)+|\mu_1|^2\right]\right\} \quad (7)$$

where $N = 1 + |\mu_1|^2 + |\beta_1|^2$ and $D = N^2 - 4|\mu_1|^2\,|\beta_1|^2 > 0$ and the small signal gain coefficients are denoted by

$$c_\mu = \frac{1}{2}\frac{\omega_{01}\mu_{01}^2 n}{\hbar\varepsilon_0 c\gamma},\; c_\alpha = \frac{1}{2}\frac{\Omega\,\mu_{12}^2 n}{\hbar\varepsilon_0 c\gamma},\; c_\beta = \frac{1}{2}\frac{\omega_{23}\mu_{23}^2 n}{\hbar\varepsilon_0 c\gamma} \quad (8)$$

The nonlinear dispersion enters via the equation

$$\frac{d\Phi}{dz} = \frac{\gamma^2}{\Delta^2}\left(\frac{c_\mu}{|\mu_0|^2}+\frac{c_\beta}{|\beta_0|^2}\right)\frac{|\alpha|^2}{D}|\mu_0||\beta_0||\mu_1||\beta_1|\sin(\Phi) \quad (9)$$

for the global phase

$$\Phi = \phi(\beta_0) - \phi(\beta_1) - \phi(\mu_0) + \phi(\mu_1) \quad (10)$$

Note that linear dispersion and absorption at FIR modes disappear because we have assumed $n_0 = n_1 = n$ and $n_2 = n_3 = 0$. Also, equations (3) - (7) and (10) are valid only if $|\mu_0|$, $|\beta_0| \neq 0$. However, if one wants to treat this case, or the case $\Phi(z = 0) = 0$, it is easy to return to the equivalent but less transparent complex equations.

Equations (3) - (7) and (9) determine the amplification of the fields. As expected, the equations are symmetric with respect to $\{\mu_0, \mu_1\}$ and $\{\beta_0, \beta_1\}$. When one of the pairs is neglected, we obtain the expressions given in [4]. Despite of the approximation of a detuned pump, the field gain factors still contain considerable saturation and mode coupling. A novel, unexpected feature is the high order dispersion represented by the global phase Φ. (In the calculations of [4] dispersion enters only to the order Δ^{-3} because the product $|\beta_1| \cdot |\mu_1|$ there is equal to zero). This nonlinear dispersion is due to a "five-photon interference" term whose real part is the second term of equations (4) and (5), and the imaginary part is the right hand side of the dispersion equation (9). Due to this phase, the Raman modes are coupled and behave as bichromatic modes [10, 11]. The line centre oscillations are strongly influenced by the Raman fields: each Raman mode decreases the gain of the line-centre oscillation coupled to the same transition, as predicted in [4], but enhances the line-centre gain of the other FIR transition. This is evidenced by the factor $(|\mu_0|^2 - |\beta_0|^2)$, appearing in (6) - (7). In (6), the first term of this factor is due to the competition effects discussed in [4], the second term is due to the three-photon resonance contribution and vice versa in (7).

According to Eq. (9), the phase Φ tends to lock to a value $\Phi_\infty = \pi$ ($\Phi = 0$ is unstable). Therefore, in the long run $\cos(\Phi)$ turns negative and the two Raman modes assist each other according to (4) and (5). It is probable that the Raman mode having the higher linear gain will also grow faster even when saturation sets in. Which one of the Raman modes dominates will determine which one of the line-centre modes is ultimately suppressed - according to (6) and (7) it will be μ_1 in the case $|\mu_0|^2 > |\beta_0|^2$ and β_1 in the case $|\mu_0|^2 < |\beta_0|^2$ in the limit $|\mu_0|$, $|\beta_0| \gg 1$.

In conclusion, we expect that asymptotically the four-level system will oscillate on both the Raman and line-centre modes at the transition with lower small signal again and only at the Raman resonance at the higher gain transition. Whether this asymptotic solution is realized in an experiment will depend on the length of the amplifier as compared to the two gain lengths involved. In a highly asymmetric situation, e.g. $c_\beta \gg c_\mu$, the three-level model of [4] suffices. Equations (3) - (7) and (9) also predict that under some circumstances interesting transient behaviour may occur. For instance, one might anticipate regions where Φ has not yet reached its stationary value and there would appear competition between Raman modes if $\cos(\Phi) > 0$. These aspects are further illustrated by discussing some numerically integrated cases of (3) - (7) and (9).

The numerical examples are obtained by a Runge-Kutta-Merson integration method. We have neglected pump depletion represented by Eq. (3). In Fig. 3 we assume $c_\beta > c_\mu$ and for the entrance values we have chosen $|\mu_0| \gg |\beta_0|$ and $|\mu_1| \lessapprox |\beta_1|$. The field evolution follows the predictions above. The phase Φ relaxes gently towards π and even though initially $|\beta_0| < |\mu_0|$ for large propagation distances $|\beta_0|$ exceeds $|\mu_0|$ suggesting the survival of $|\mu_1|$ only. As a matter of fact, the line-center oscillation $|\beta_1|$ is very quickly suppressed and $|\mu_1|$ increased after $|\beta_0|$ has passed $|\mu_0|$, manifesting the dominance of the nonlinear part in the gain of the line centre-modes. Simultaneously a strong mutual enhancement of the Raman modes is observed due to the fact that $|\mu_1| \cdot |\beta_1|$ is maximum in eqs. (4) and (5).

In Figure 4 we have chosen such initial conditions that strong Raman competition might be expected: i.e. $\Phi(0) = 1.75 \times 10^{-4}$ and $|\mu_1| \gg |\beta_1|$ to confirm that the initial line-centre intensities do not influence qualitatively the asymptotic behaviour of the system. Consequently $|\beta_0|$ initially starts to decrease but this leads to a very rapid mode locking of Φ to π, because of the divergence $\sim 1/|\beta_0|$ in Eq. (9). After the 'sudden' locking, the system behaviour follows that of Fig. 3. Indeed, it may be shown analytically that, if μ_1, β_1 are relatively constant and the initial global phase Φ is zero, the phase locking is exactly step-like.

In Figures 5a) to c), we have studied possibilities to injection lock the system onto pure Raman emission on the transition having the <u>lower</u> small signal gain. Choosing a reasonably large initial value for $|\mu_0| \gg |\beta_0|$, we manage to suppress $|\mu_1|$ even though $c_\beta > c_\mu$. This situation prevails until $z \simeq 1200$ whereafter $|\mu_1|$ gets reamplified and all four modes oscillate. After a very long interaction length

($z \sim 7 \times 10^7$) the system attains its asymptotic form i.e. a state where $|\mu_1|$ only survives together with the Raman modes. This example clearly illustrates the possibility to select by injection locking the form of output modes in an amplifier of a fixed length.

In a previous paper we showed that in a three-level system coherently pumped off resonance, the gain on line-centre of the signal transition is efficiently suppressed in the presence of a moderately intense Raman oscillation. This desirable effect allows tunable single-mode operation of an FIR laser. The serial four-level configuration investigated in this paper is representative of commonly encountered optically-pumped FIR lasers with a refill transition [5-8]. We have derived a model which aids in predicting the mode of operation, i.e. which lines will appear. In addition to the prognosis of asymptotic behaviour, the model is also applicable to describe transients which alone might be observable in actual experiments. Our results indicate that suppression of line centre oscillation does not occur simultaneously on both signal transitions in this case. Under certain conditions, injection locking onto single mode Raman emission in either of the signal transitions can be achieved. We have also shown the important role played by nonlinear dispersion in the Raman-Raman mode interaction, resulting in mutual enhancement. Within the approximations used, the asymptotic states of the system are always multimode states. Therefore, in such a system, the use of a single mode theory is probably never justified.

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Figures Captions

Fig. 1: Two mathematically equivalent configurations:

(a) configuration Λ with $n_1 = n$, $n_2 = n_3 = 0$,

(b) configuration V (refilling) with $n_0 = n_1 = n$, $n_2 = 0$.

Index change to go from (a) to (b): $(1,2,3) \rightarrow (2,1,0)$.

Fig. 2: General configuration treated in this paper.

Fig. 3: Evolution of the system (amplifier) represented by equations (4) - (7) and (9). The pump absorption (eq. (3)) is neglected.

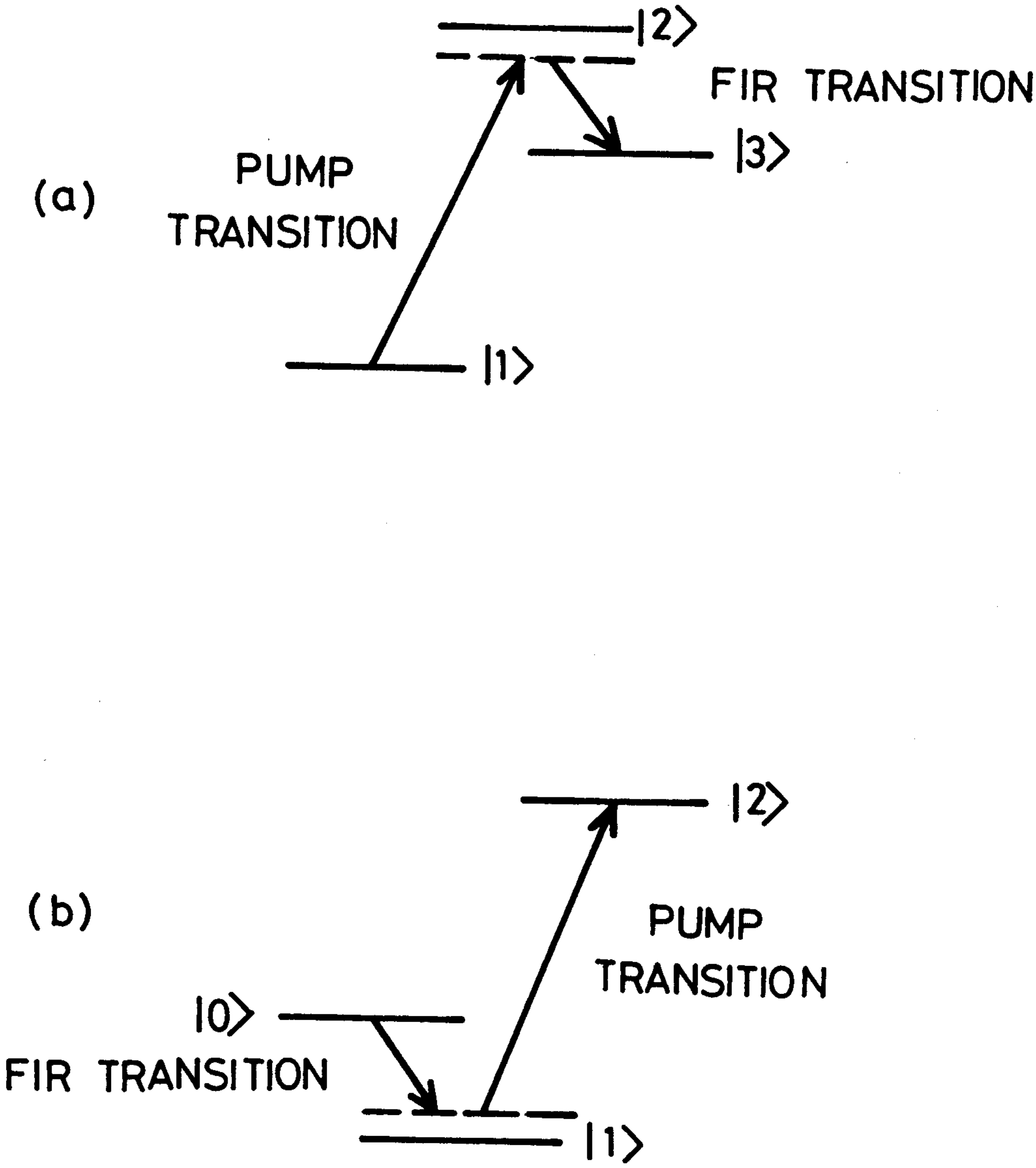
Full line : signal β coupled to the upper transition,

Dashed line: signal μ coupled to the lower transition.

Numerical values of the parameters are $c_\beta = 21$, $c_\mu = 20$, $|\alpha| = 1$, $|\Delta| = 10\gamma$.

Fig. 4: As fig. 3 with different initial values.

Fig. 5: As fig. 3 with different initial values.



(a)
|2⟩
FIR TRANSITION
|3⟩
PUMP
TRANSITION
|1⟩
(b)
|2⟩
PUMP
TRANSITION
|0⟩
FIR TRANSITION
|1⟩

Fig. 1

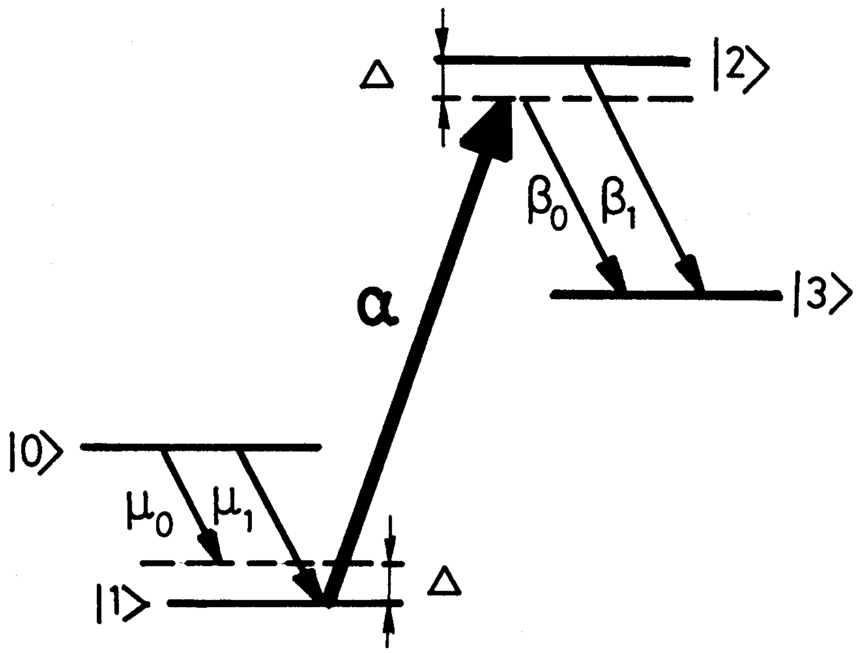
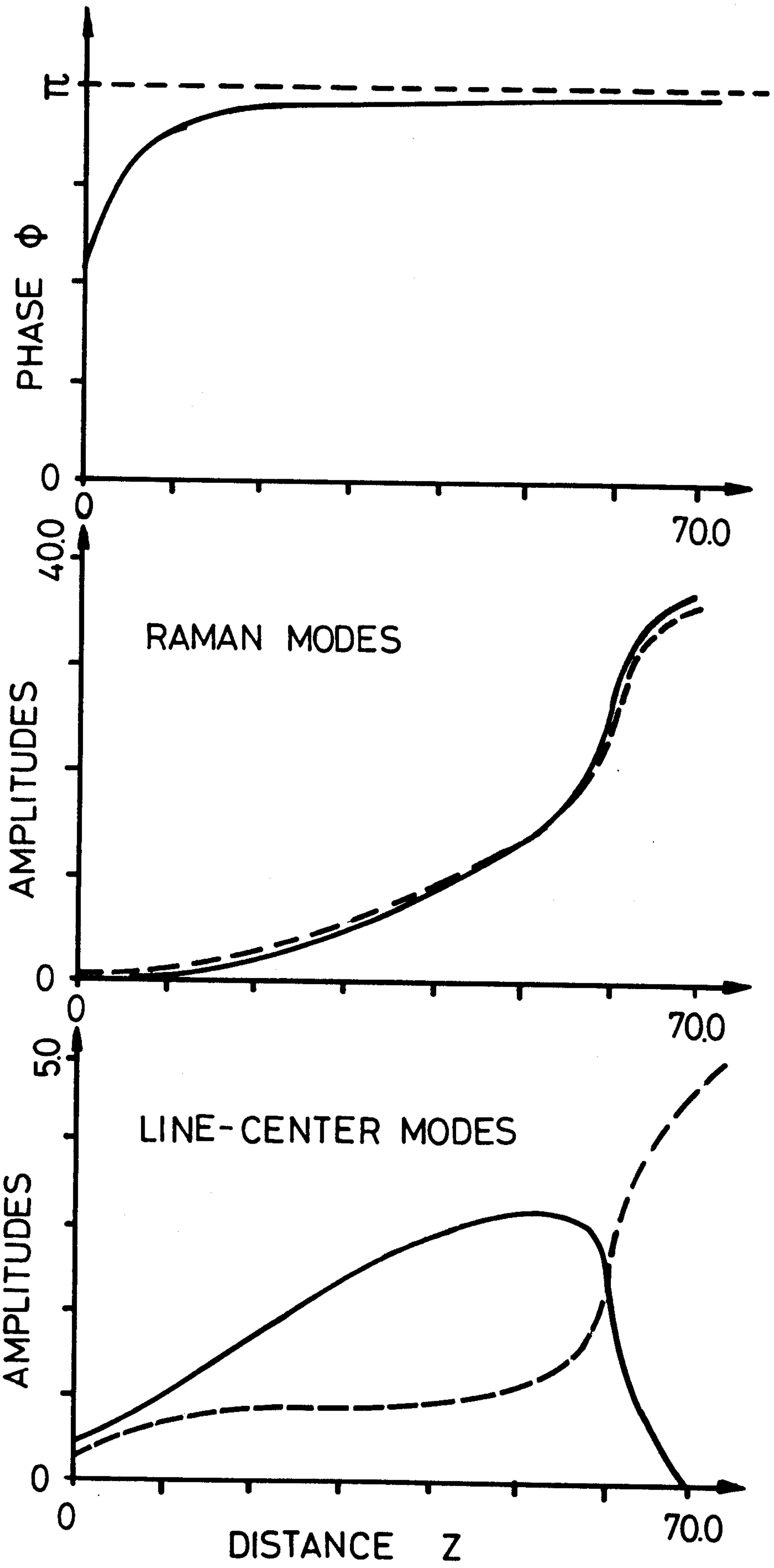


Fig. 2


π
PHASE Φ
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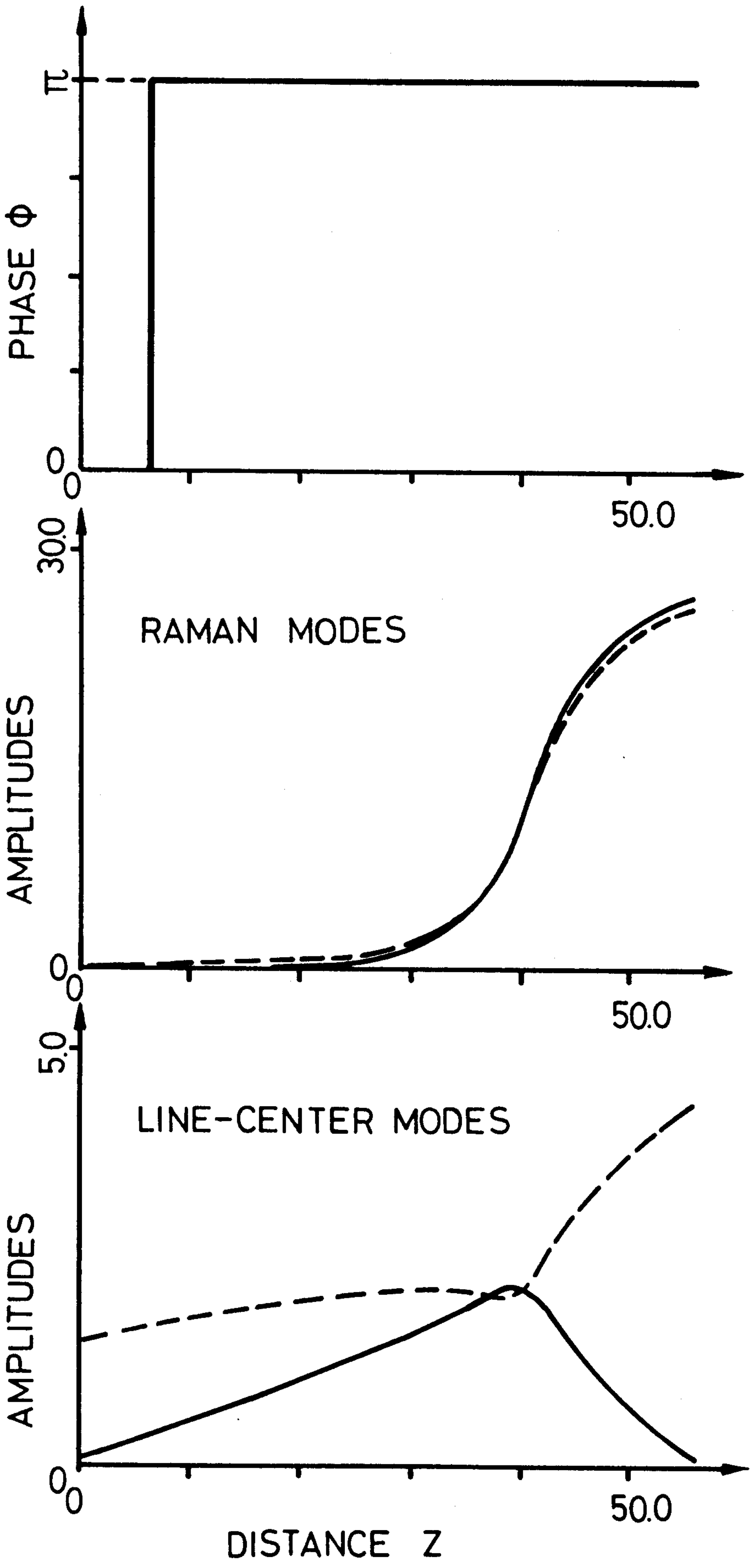
0
70.0
40.0
RAMAN MODES
AMPLITUDES
0
0
70.0
5.0
LINE-CENTER MODES
AMPLITUDES
0
0
70.0
DISTANCE Z

Fig. 3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PHASE Φ
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0
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30.0
RAMAN MODES
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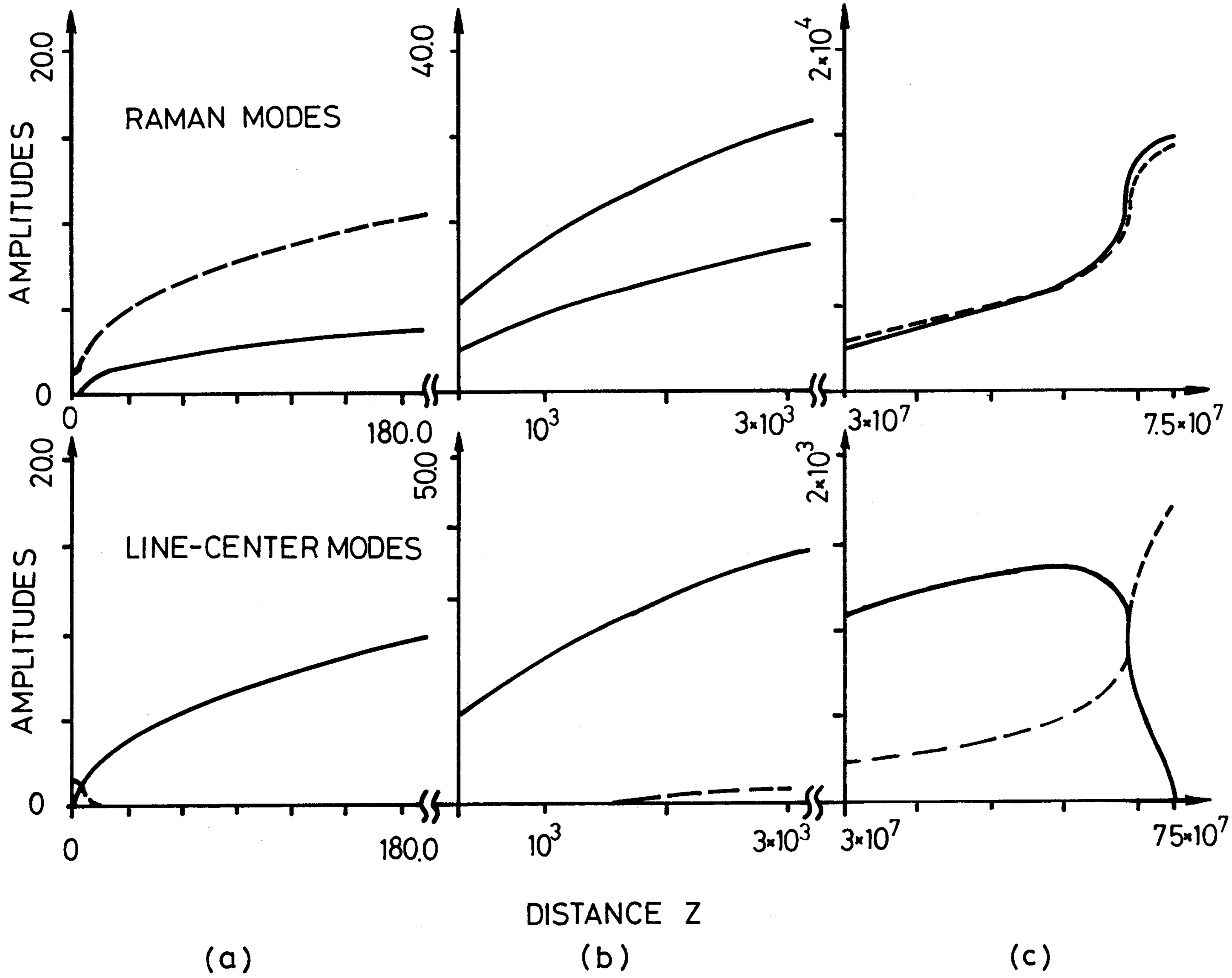
0
0
50.0
5.0
LINE-CENTER MODES
AMPLITUDES
0
0
50.0
DISTANCE Z

Fig. 4



RAMAN MODES
LINE-CENTER MODES
AMPLITUDES
AMPLITUDES
20.0
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40.0
2×10^4
20.0
50.0
2×10^3
0
180.0
10^3
3×10^3
3×10^7
7.5×10^7
75×10^7
DISTANCE Z
(a)
(b)
(c)

Fig. 5