Model for laser action in vibronic systems

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The theory of laser action in solid-state systems is reconsidered with emphasis on the role of the interaction between lasing impurity states and lattice vibrations. The influence of a strong electron-phonon coupling on threshold conditions and field dynamics is presented, taking into account the vibronic-level structure, and compared with the usual four-level scheme.

I. INTRODUCTION

Laser action from impurities embedded in a host crystal is mainly understood as originating from stimulated emission between states which correspond to sharp electronic transitions in the fluorescence spectrum of the impurity. However, as was pointed out by McCumber in 1964,[1] the electron-phonon interaction may influence in a crucial way the lasing characteristics of a solid-state laser. This is the case when the fluorescence spectrum of the lasing impurity shows sidebands due to phonon-assisted photon emission. These sidebands have a rather broad fluorescence spectrum and their gain characteristic may be more favorable for laser action than that of the purely electronic zero-phonon line. This behavior, which is reminiscent of that found in dye lasers, indicates that these broad fluorescence bands may be exploited in order to obtain tunable laser emission. Recently, several examples of such vibronic laser devices have been considered using both different color centers and transition-metal ions as lasing impurities.[2–4] As a result, a number of tunable solid-state lasers in the near and middle infrared have been studied experimentally. Furthermore, evidence for the relevance of the electron-phonon interaction to the description of coherent emission processes from impurities in solids was given by experiments on many-color superfluorescent emission in $KCl:O_2^-$.[5]

These facts imply that it may be useful to reconsider the theory of laser action in vibronic systems from a more fundamental point of view. In fact, the influence of the lattice or molecular vibrations on lasing impurities in a solid-state laser is in general understood as being responsible for level broadening only. According to this interpretation a solid-state laser is well described by a four- or three-level scheme in which phonons are only responsible for the nonradiative transitions from the pumping to the excited level of the laser transition[6] or from the lower level to the ground state of the impurity. In these standard models the lasing transition happens between two electronic states. In the following we will call these models two-level models, concentrating on the relevant optical transition rather than on the underlying three- or four-level pump and depletion plus optical-transition scheme.

It is clear that the usual two-level laser models[7–9] cannot account for emission from vibronic states, because this emission is not described through transitions between electronic levels. Furthermore, as we will show in Sec. II, two electronic levels strongly coupled with optical phonons or vibrations give rise to a complicated level scheme which can hardly be reduced to two levels only. Therefore, systems which show laser emission in the phonon or vibration-assisted part of the fluorescence spectrum require an extension of the usual description which will account for the vibronic-level structure relevant to the emission. Eventually this new model will reduce to the known one when the direct influence of the lattice or molecular vibrations on the emission becomes irrelevant. We expect that such a theory will differ in several respects from the usual theories. The electron-phonon coupling should influence the relevant laser parameters such as the threshold inversion or the gain and saturation terms. Furthermore, different selection rules for transitions between vibronic levels will appear.

Some of these effects have already been discussed by the authors.[10] In this paper we generalize the results of Ref. 10 in several respects. In particular, we give gain curves which explicitly show the dependence of the threshold on the electron-phonon coupling and in which characteristic selection rules appear. The overall gain of the vibronic transition will be higher than that of an "isolated" vibronic line. This effect shows the importance of considering the influence of the overlap integrals between phonon wave functions on the emission properties. It also shows how tuning is possible in such a system as a consequence of the fact that several transitions have simultaneously a high gain and their behavior is mutually influenced. Furthermore, in lowest order we discuss dynamical field effects like saturation and mode-mode coupling, which again show a different behavior with respect to that known from the two-level model description. In particular, above threshold, different transitions are in resonance with different cavity modes and are mutually coupled. This effect is responsible for an intensity redistribution in the laser emission.

The theory is developed along the lines of the well-known theory of Lamb[11] for the pure electronic laser and

it offers a starting point for a better understanding of vibronic lasers. Although our analysis is quite general it should be possible to specialize it for realistic systems, without major efforts. A perturbative approach is used here for the sake of simplicity. However, a more complete analysis can be done, with some greater algebraic effort, starting from our equation, as we will point out in our discussion.

The paper is organized as follows. In Sec. II we briefly discuss the laser model and derive the field and matter equations in the mean-field approximation. The threshold condition and gain profiles are presented in Sec. III, whereas in Sec. IV the saturation effects at lowest order and the mode coupling are discussed.

II. VIBRONIC LASER MODEL

A. The model

Laser operation in a vibronic system is described through a generalization of the familiar two-level model which accounts for the effect of the interaction between impurity states and the host crystal lattice. Only two electronic levels of the impurity are considered in order to avoid effects such as absorption from other electronic states which would be fatal to laser action. Therefore, no other levels of the impurity except for those which originate the vibronic fluorescence in the emission spectrum have to be considered.[2-4] This assumption is common to most models which describe emission from vibronic states in crystals.

The electronic levels are coupled to the lattice and to the electromagnetic field in the laser cavity. In the usual dipole approximation the coupling to the electromagnetic field modes is given by

$$H_{e\text{-phot}}=\hbar\sum_k \omega_k a_k^\dagger a_k+\sum_j\sum_k g_{kj}(a_k\sigma_j^+ +a_k^\dagger\sigma_j^-)\ , \quad (2.1a)$$

where the index j refers to the positions of the impurities, k denotes the field modes in the cavity, and g_{kj} and ω_k are the dipole coupling constants and the mode frequencies, respectively. The phonon part of the interaction has the form

$$H_{e\text{-phon}}=\hbar\sum_j \Omega_j b_j^\dagger b_j+\hbar\varepsilon\sum_j \sigma_{zj}+\sum_j \lambda_j\sigma_{zj}(b_j^\dagger+b_j)\ , \quad (2.1b)$$

where ω_j are optical phonon frequencies, λ_j characterize the electron-phonon coupling, and ε is the electronic-level spacing. The electronic states are described by the usual pseudospin operators $\sigma_j^\pm,\sigma_{zj}$ with

$$[\sigma_j^+,\sigma_l^-]=\delta_{jl}\sigma_{zl},\quad [\sigma_j^\pm,\sigma_l^\pm]=0\ , \quad (2.2)$$

whereas a_k, $a_k^\dagger$, b_j, and $b_j^\dagger$ obey Bose commutation relations. We notice that the electronic states are coupled to localized optical phonons or vibrations, as is inferred from the index j which appears in the b operators as well as in the optical phonon frequencies Ω_j. This picture seems to be well suited to describing the vibronic structure of $KCl:O_2^-$.[5]

The usual way to handle the losses of a laser system consists in coupling its degrees of freedom to reservoirs. We do it in the standard way for the field losses.[7] However, a more natural way of describing the losses for the excited impurities and phonons which dissipate energy into the crystal consists of coupling the optical phonons or quasimolecular vibrations to the acoustical phonons. The latter are responsible for nonradiative energy dissipation into the surrounding crystal and therefore act as reservoirs for the optical modes. We describe the dissipation mechanism through

$$H_{\text{phon-phon}}=\hbar\sum_q \mu_q B_q^\dagger B_q +\sum_j\sum_q k_{jq}(B_q^\dagger+B_q)(b_j^\dagger+b_j)\ , \quad (2.3)$$

where $B_q,B_q^\dagger$ are the amplitudes of the acoustical phonon modes with wave vectors q and frequencies μ_q and k_{qj} are the coupling constants. As a result the laser Hamiltonian reads

$$H=H_{e\text{-phot}}+H_{e\text{-phon}}+H_{\text{phon-phon}}+H_{\text{phot-res}}\ . \quad (2.4)$$

The vibronic-level structure implied by (2.1b) is displayed in Fig. 1, where the different possible transitions between vibronic levels are also shown.

B. Transition operators

As we have already shown in an earlier paper,[12] the Hamiltonian (2.4) can be rewritten in a form which exhibits the transitions between vibronic states explicitly. It is obtained by the exponential transform

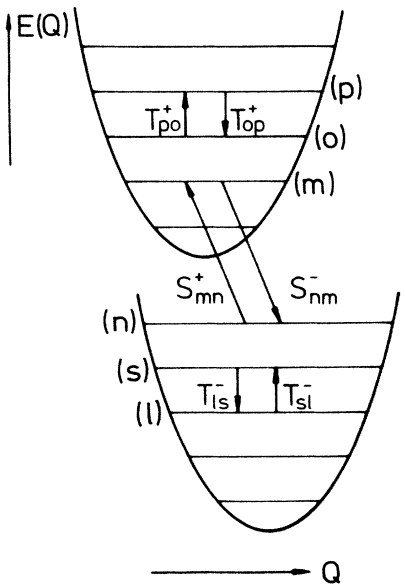

FIG. 1. Approximate vibronic-level structure. E is the energy and $Q=(1/\sqrt{2\Omega})(b+b^\dagger)$ is a phonon configuration coordinate.

$$U=\exp\left[-\sum_j \frac{\lambda_j}{\hbar\Omega_j}(b_j-b_j^\dagger)\sigma_{zj}\right], \tag{2.5}$$

which diagonalizes $H_{e\text{-phon}}$ and leads to the transformed Hamiltonian

$$H_{e\text{-phot}}=\hbar\sum_k \omega_k a_k^\dagger a_k + \sum_j\sum_k g_{kj}\left[a_k \exp\left[\sum_j \frac{\lambda_j}{\hbar\Omega_j}(b_j-b_j^\dagger)\right]\times\sigma_j^+ + \text{H.c.}\right], \tag{2.6}$$

which is best interpreted by projecting the interaction on the product spaces $|\uparrow\rangle\otimes|n\rangle$, $|\downarrow\otimes|m\rangle$ and obtaining

$$H_{e\text{-phot}}=\hbar\sum_k \omega_k a_k^\dagger a_k + \sum_k\sum_j\sum_{m,n} g_{kj}[f_{mn}(S_{mn}^+)_j a_k + f_{nm}(S_{nm}^-)_j a_k^\dagger]. \tag{2.7}$$

Here $|\uparrow\rangle, |\downarrow\rangle$ are the eigenstates of σ_z. m,n are phonon-state labels and

$$f_{mn}=\left\langle m\left|\exp\left[\sum_j \frac{\lambda_j}{\hbar\Omega_j}(b_j-b_j^\dagger)\right]\right|n\right\rangle = f_{nm}^\dagger \tag{2.8}$$

are the Frank-Condon overlap integrals.[13] We have already left out a term,

$$-\sum_{jl}\frac{\lambda_j\lambda_l}{\hbar\Omega_l}\sigma_{z_j}\sigma_{z_l},$$

which only redefines the frequency ε. Moreover, we have introduced the new transition operators

$$(S_{mn}^+)_j=|m\uparrow\rangle_{jj}\langle n\downarrow|, \quad (S_{nm}^-)_j=|n\downarrow\rangle_{jj}\langle\uparrow m|, \tag{2.9}$$

which are responsible for transitions between the different potential sheets in Fig. 1 and whose commutation rules are

$$[(S_{mn}^\pm)_j,(S_{kl}^\pm)_j]=0, \qquad [(S_{mn}^+)_j,(S_{kl}^-)_p]=\delta_{jp}[(T_{ml}^+)_j\delta_{kn}-(T_{kn}^-)_j\delta_{ml}], \tag{2.10}$$

where

$$(T_{mn}^+)_j=|m\uparrow\rangle_{jj}\langle\uparrow n|, \quad (T_{mn}^-)_j=|m\downarrow\rangle_{jj}\langle\downarrow n|. \tag{2.11}$$

The operators $T_{mn}^\pm$ describe transitions inside the upper- and lower-potential sheet, respectively.

C. Equations of motion

The equations of motion for the variables of the laser system have already been derived and briefly discussed elsewhere.[12] For later use we quote here their simplified expressions which entail the following approximations.

(a) The reservoirs have been eliminated by standard methods.[7–9] Their influence appears through the damping terms in the equations of motion for the laser variables. (b) We consider statistical mean values of the different variables. (c) We assume that the electric dipole matrix elements g_{jk} as well as the optical-phonon frequencies and coupling constants do not depend on the position index j. Therefore, we define collective transition operators $S_{mn}^\pm=\sum_j(S_{mn}^\pm)_j$, $T_{mn}^\pm=\sum_j(T_{mn}^\pm)_j$ and omit the index j in the following. (d) Expectation values of products of operators have been factorized according to the rule

$$\langle a_k^p(S_{mn}^+)^q(T_{ls}^-)^r\rangle=\langle a_k\rangle^p\langle S_{mn}^+\rangle^q\langle T_{ls}^-\rangle^r. \tag{2.12}$$

From (b) and (c) it follows that no diffusion effects will be described at this level of approximation. Therefore our description will be equivalent to the semiclassical description which is well established for the two-level models.[11] For the expectation values of the transition operators we get the following equations:

$$\frac{d}{dt}S_{mn}^+=-\gamma_{mn}S_{mn}^+ + 2\gamma_{m-1\,n-1}S_{m-1\,n-1}^+ + 2\gamma_{m+1\,n+1}S_{m+1\,n+1}^+ + i[\varepsilon-\Omega(n-m)]S_{mn}^+ - i\sum_{k,l} g_k a_k^\dagger(f_{ln}T_{ml}^+ - f_{ml}T_{ln}^-). \tag{2.13}$$

The equation for S_{nm}^- is obtained by taking the complex conjugate of (2.13) and interchanging the order of the indices in the summands. For the inversion $I_{mn}=T_{mm}^+-T_{nn}^-$ we obtain

$$\frac{d}{dt}I_{mn}=-\gamma_{mm}^T I_{mn} + 2\gamma_{m-1\,n-1}^T I_{m-1\,n-1} + \gamma_{mm}^T\Delta N_{mn} + 2\gamma_{m+1\,n+1}^T I_{m+1\,n+1} + i\sum_{k,l} g_{kl}[(f_{lm}S_{nl}^- + f_{nl}S_{lm}^-)a_k^\dagger - (f_{ln}S_{ml}^+ + f_{ml}S_{ln}^+)a_k], \tag{2.14}$$

where $\Delta N_{mn}=(T_{mm}^+-T_{nn}^-)^0$ is the pump-source term. The nondiagonal quantities $T_{mn}^\pm$ obey

$$\frac{d}{dt}T_{mn}^+=-[\gamma_{mn}^+ + i\Omega(n-m)]T_{mn}^+ + 2\gamma_{m-1\,n-1}^+ T_{m-1\,n-1}^+ + 2\gamma_{m+1\,n+1}^+ T_{m+1\,n+1}^+ - i\sum_{k,l} g_k(f_{nl}S_{ml}^+ a_k - f_{lm}S_{ln}^- a_k^\dagger),$$
$$\frac{d}{dt}T_{mn}^-=-[\gamma_{mn}^- + i\Omega(n-m)]T_{mn}^- + 2\gamma_{m-1\,n-1}^- T_{m-1\,n-1}^- + 2\gamma_{m+1\,n+1}^- T_{m+1\,n+1}^- + i\sum_{k,l} g_k(f_{lm}S_{ln}^+ a_k - f_{nl}S_{ml}^- a_k^\dagger). \tag{2.15}$$

Finally, the field is described by

$$\frac{d}{dt}a_k=-\Gamma a_k-i\omega_k a_k-i\sum_k\sum_{m,n}g_k f_{nm}S_{nm}^{-}\ ,$$
$$\frac{d}{dt}a_k^{\dagger}=-\Gamma a_k^{\dagger}+i\omega_k a_k+i\sum_k\sum_{m,n}g_k f_{mn}S_{mn}^{+}\ . \tag{2.16}$$

The quantities $\gamma_{mn}^{\pm}$, γ_{mn}, and γ_{mm}^{T} are damping constants for the transitions between levels belonging to the same potential sheet and different sheets, respectively. We give the explicit form for

$$\gamma_{mn}=\tfrac{1}{2}[(\gamma_1+\gamma_2)(m+n)+2\gamma_1]\ , \tag{2.17}$$

where γ_1 and γ_2 are the reservoir-induced transition rates corresponding to the processes $|m\uparrow\rangle\rightarrow|n\downarrow\rangle$, $|n\downarrow\rangle\rightarrow|m\uparrow\rangle$, respectively.

The quantities $\gamma_{mm}^{T},\gamma_{mn}^{\pm}$ have a similar form. The quantity Γ describes the field losses. Remember that all variables are c-number quantities. However, in order not to overcomplicate notations we use for them the same symbols that we have introduced for the operators. These equations are the starting point for our discussion of laser action in vibronic systems.

D. Perturbative solution

The system of equations, (2.13)–(2.16), as it stands is quite complicated, as it involves a large number of variables and indices. It is easy to realize that already in the stationary regime and considering only a small number of transitions an analytical solution of (2.13)–(2.16) becomes quite involved. Instead of doing cumbersome algebraic calculation or relying on numerical results only, we have chosen to discuss (2.13)–(2.16) in a perturbative approach which is of current use in the two-level laser theory.[11] We develop the relevant material variables around their stationary values near threshold assuming that the (unsaturated) inversion is at equilibrium with the pump and that the field amplitude and therefore also the expectation values of $S_{mn}^{\pm}$, $T_{mn}^{\pm}$, and a_k are zero. These assumptions are correct when the system is just below threshold and imply

$$(T_{mn}^{\pm})^{(0)}=0,\quad m\neq n$$
$$(I_{mn})^{(0)}=\Delta N_{mn}\ , \tag{2.18}$$
$$(S_{mn}^{\pm})^{(0)}=0\ .$$

The explicit calculations of the perturbation expansion of $S_{mn}^{\pm}$ are carried out in the Appendix up to third order in $g_k^2 a_k$. We will make use of them in Secs. IV and V when discussing threshold and mode-coupling effects.

III. THRESHOLD CONDITION

In order to discuss the threshold condition for a vibronic laser we need only to retain the first-order terms $(S_{mn}^{\pm})^{(1)}$ [cf. (A4)] in (2.16). Using (A1) as the ansatz and defining $A_k=|A_k|\exp i\psi_k$, we get in terms of amplitude and phase

$$\frac{d}{dt}|A_k|=-\Gamma|A_k|-g_k^2|A_k|\sum_{m,n}\frac{f_{mn}^2\Delta N_{mn}\gamma_{mn}}{\gamma_{mn}^2+[\varepsilon-(n-m)\Omega-(\omega_k+\varphi_k)]^2}\ , \tag{3.1a}$$

$$\frac{d}{dt}\psi_k=\omega_k+\varphi_k-g_k^2\sum_{m,n}f_{mn}^2\frac{\Delta N_{mn}[\varepsilon-(n-m)\Omega-(\omega_k+\varphi_k)}{\gamma_{mn}^2+[\varepsilon-(n-m)\Omega-(\omega_k+\varphi_k)]^2}\ . \tag{3.1b}$$

In writing (3.1) we have eliminated oscillating terms of the type $(\omega_k+\omega_{k'})t$. This happens because they oscillate at a frequency higher than ω_k and do not contribute around threshold. Their contributions become important when phase locking is present. This is the case well above threshold that is at field intensities for which the perturbative approach fails.

We notice that Eqs. (3.1) coincide with the equations already presented by the authors in a previous paper[10] which are derived by neglecting the "phonon-term" contributions in (2.13). The equivalence is not surprising because the phonon terms do not contribute to the first-order equations. The equation for ψ_k [Eq. (3.1b)] is a quite complicated equation which is solved by assuming that the corrections due to the presence of the medium are small, which is a fairly good approximation around threshold.[11] As a result we get in the stationary case

$$\varphi_k+\omega_k=g_k^2\sum_{m,n}f_{mn}^2\frac{\Delta N_{mn}[\varepsilon-(n-m)\Omega-(\omega_k+\varphi_k)]}{\gamma_{mn}^2+[\varepsilon-(n-m)\Omega-(\omega_k+\varphi_k)]^2}\ , \tag{3.2}$$

which shows the characteristic pulling term, and we can assume $\varphi_k=0$ in (3.1).

The laser threshold follows from (3.1a) in the stationary regime by imposing the condition that field losses and gain be equal. This means that

$$\Gamma=g_k^2\sum_{m,n}f_{mn}^2\frac{\Delta N_{mn}\gamma_{mn}}{\gamma_{mn}^2+[\varepsilon-(n-m)\Omega-\omega_k]^2}\ . \tag{3.3}$$

As already pointed out by the authors,[10] the differences from the two-level model are evident. In fact, neglecting all nonresonant terms in (3.3) for a given mode frequency $\omega_k=\varepsilon-(n-m)\Omega$, one obtains

$$\Gamma=\frac{f_{mn}^2 g_k^2}{\gamma_{mn}}\Delta N_{mn}\ , \tag{3.4}$$

which has to be compared with the one-mode two-level model result $\Gamma=(g_k^2\Delta N)/\gamma$. The difference comes from the Frank-Condon integral f_{mn} which is smaller than 1 and may vanish as function of $m,n,(\lambda/\hbar\Omega)$. The physical implications of (3.3), however, can only be appreciat-

ed when discussing the contribution of all terms which appear in the sum. For simplicity we assume that only one vibronic upper state is excited, i.e., $m=0$. Moreover, in order to remain as general as possible we do not specify the different parameters according to one of the known lasing materials.

Expression (3.3) then gives the gain profile of the laser in the whole emission region. We choose for the damping constants the value $\gamma_{mn}/\Omega=0.02$ which ensures that the lines corresponding to each of the possible transitions from the excited state will be resolved, provided that we normalize all quantities with the frequency difference Ω between vibronic levels. The normalized-gain spectra for the initial upper states $m=0$ and 1 are presented in Figs. 2 and 3, where the spectra are normalized with the factor $g_k^2 N$. The height of each peak is proportional to the gain of the corresponding line. Notice that in Figs. 2 and 3 the 0-phonon line corresponding to the transition between the two pure electronic states is on the extreme right. From these spectra some features of the vibronic laser are deduced.

Both spectra consist of a sequence of lines of different height, which is determined by the Frank-Condon factors f_{mn}. For $m=1$ (Fig. 3) the distribution of the maxima is different with respect to that found for $m=0$ and one line is missing. This shows how selection rules are introduced by the overlap integrals.

Furthermore, depending on the field damping Γ, several lines may be above threshold at the same time. This is clearly shown in Fig. 2 where the two adjacent lines corresponding to the three- and four-phonon lines, respectively, have the same height and therefore lead to the same laser threshold. When several upper states are occupied and equilibrium with the pump is established, which we may simulate by a thermal-occupation distribution starting from the state $m=0$, the gain curve has the form shown in Fig. 4. From these curves and from the threshold condition (3.3), we infer that the simultaneous laser emission from several transitions which is the condition for having a tunable laser can be achieved in the vibronic systems. When higher values of γ_{mn}/Ω are chosen, the overlapping between the different lines in Figs. 2–4 becomes important. As a result a higher gain is obtained. This is shown in Fig. 5 for several values of γ_{mn}/Ω. The curves are normalized to the peak values of Fig. 2 where the overlapping is negligible and the one-transition threshold condition (3.4) holds. As is shown in Fig. 5 for values of γ_{mn}/Ω, an appreciable growth in the overall-gain profile is obtained. This result indicates that a vibronic system may have higher gain than that which would be found when concentrating on a transition between a pair of vibronic levels only, i.e., with an effective two-level model. This indicates how the complicated level structure originating from the strong electron-phonon interaction influences the laser action in a solid-state laser. In order to appreciate this point, we recall that an electronic transition which is located not far from the vibronic band may have a smaller gain and therefore not be laser active, although its gain is comparable with that of a vibronic transition when the latter is considered independently from the other transitions in the vibronic spectrum.[2]

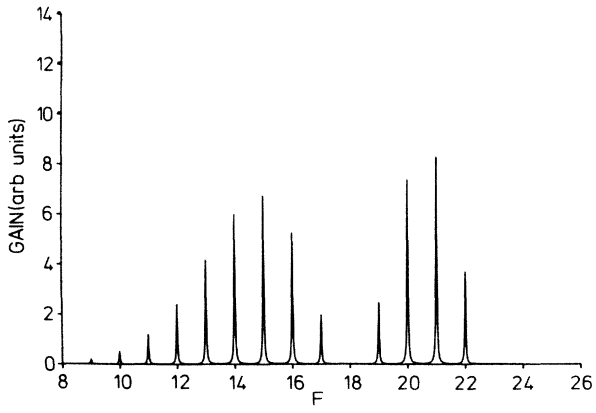

FIG. 3. Same as Fig. 2 with $m=1$.

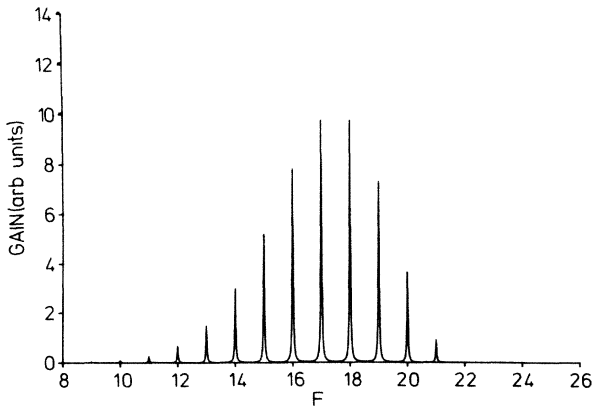

FIG. 2. Gain profile for the excited level $m=0$. Here the parameters are fixed as follows: $E/\Omega=20$, $\gamma_{mn}/\Omega=0.02$. The curve is normalized with $g_k^2 N$ and $F=\omega_k/\Omega$.

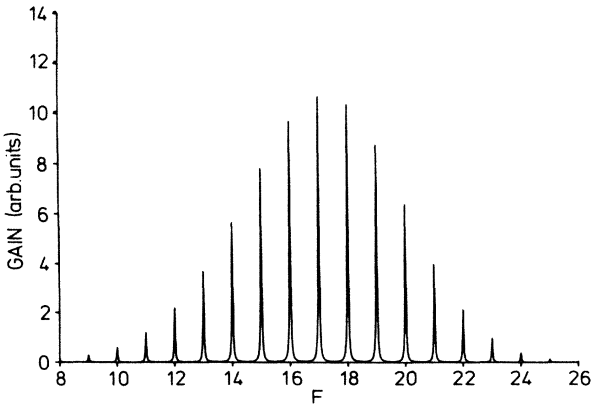

FIG. 4. Same as Fig. 2 where the initial inversion results from a thermal admixture of upper states. Here $\hbar\Omega/kT=1.1$.

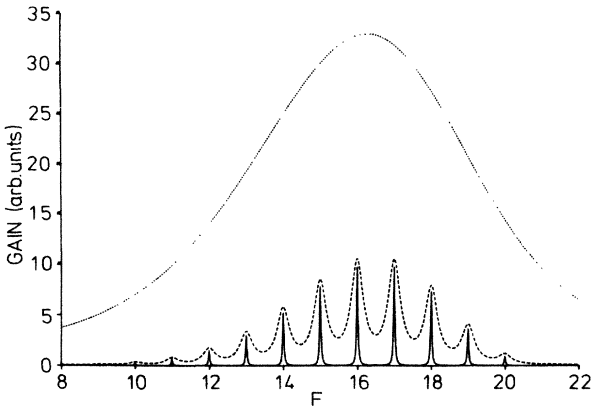

FIG. 5. Gain profiles for $E/\Omega=20$, and $m=0$ as a function of $F=\omega_k/\Omega$, $\gamma_{mn}/\Omega=0.02$ (solid curve), $\gamma_{mn}/\Omega=0.2$ (dashed curve), $\gamma_{mn}/\Omega=2$ (dotted curve).

Finally, we notice that the gain curves corresponding to large values of gain have a form which is qualitatively similar to that seen in the experiments. The tuning at this level is understood as follows: different vibronic transitions have a high gain. This gain is enhanced by the cooperation of the different summands in (3.3). Therefore, a threshold condition is found for a set of transitions leading to a situation similar to that which is found in dye lasers. Since several cavity modes are contained in the broad emission line, resonance is achieved for different transitions simultaneously and tunable laser action is possible.

IV. NONLINEAR EFFECTS

In order to account for dynamical effects in the light field above threshold, the third-order terms in the field expansion have to be considered. In our model these terms have the property of showing explicitly the influence of the transitions between levels inside the same potential sheet in Fig. 1, i.e., of the phonon terms on laser action. Since these terms are absent in the two-level models we expect them to be responsible for effects which are peculiar to vibronic lasers. Notice that the name phonon terms has its origin in the expression of the phonon creation (or annihilation) operator $b,b^\dagger$ in terms of $|m\uparrow\downarrow\rangle\langle\downarrow\uparrow n|$ projectors as

$$b=\sum_{m,n}\sqrt{m}\,(T^+_{mn}+T^-_{mn})\delta_{n\,m-1}\,. \tag{4.1}$$

Therefore, $T^\pm_{mn}$ which are introduced via the commutation rules (2.10) can be directly related to the creation or annihilation of phonons.

The complicated form of these third-order terms [(A7) and (A8)] which contain a lot of summation indices requires some simplification. We will consider only resonant terms, i.e., terms in which the frequency-dependent denominators reduce to $\gamma_{mn},\gamma^\pm_{mn}$. However, we will discuss also the contributions coming from the nonresonant terms, although we shall not write them out explicitly.

Furthermore, we will distinguish between the contributions to the third order coming from the expansion of the expression for the population inversion $T^+_{mm}-T^-_{nn}$ (population terms) and that coming from the phonon terms. As in the case of the threshold calculations we will start with the situation analogous to that which is currently considered in the two-level model, i.e., the single-mode approximation.

A. The one-mode case

We assume that only one cavity mode is resonant with a vibronic transition and neglect nonresonant terms. The result is

$$\begin{aligned}(S^+_{mn})^{(3)}=&\frac{4ig_k^3}{\gamma^2_{mn}\gamma^T_{mm}}\,|A_k|^2A_k^*f^3_{mn}\Delta N_{mn}\\&+I^+_{mn}\left[\frac{f^2_{nn}}{\omega^+_{mn}}+\frac{f^2_{mm}}{\omega^-_{mn}}\right]\\&+I^-_{mn}f_{mm}f_{nn}\left[\frac{1}{\omega^+_{mn}}+\frac{1}{\omega^-_{mn}}\right],\\(S^-_{nm})^{(3)}=&\frac{4ig_k^3}{\gamma^2_{mn}\gamma^T_{mm}}\,|A_k|^2A_kf^3_{mn}\Delta N_{mn}\\&+I^+_{mn}f_{nn}f_{mm}\left[\frac{1}{\omega^+_{mn}}+\frac{1}{\omega^-_{mn}}\right]\\&+I^-_{mn}\left[\frac{f^2_{nn}}{\omega^+_{mn}}+\frac{f^2_{mm}}{\omega^-_{mn}}\right].\end{aligned} \tag{4.2}$$

In the following we will neglect the terms in (4.2) which contain frequency differences between the vibronic states and originate from the phonon terms. They merely redefine the coefficients and do not influence the dynamics in a specific way.

The contributions to the one-mode resonant terms come from the population inversion only. In this sense the one-mode theory is only a slight modification of the usual two-level result where the dipole-matrix element is replaced by its product times the Frank-Condon integrals.

For the field mode we then obtain the equations

$$\begin{aligned}\frac{d}{dt}|A_k|=&-\Gamma|A_k|-\frac{g_k^2}{\gamma_{mn}}|A_k|\Delta N_{mn}f^2_{mn}\\&-\frac{8g_k^4|A_k|^3}{\gamma^2_{mn}\gamma^T_{mm}}\Delta N_{mn}f^4_{mn}\,,\end{aligned} \tag{4.3a}$$

$$\frac{d}{dt}\psi_k=-(\omega_k+\varphi_k)-\frac{g_k^2f^2_{mn}\varphi_k}{\gamma^2_{mn}+\varphi_k^2}+\frac{4g_k^4|A_k|^2f^4_{mn}\varphi_k}{\gamma^T_{mm}\gamma_{mn}(\gamma^2_{mn}+\varphi_k^2)}\,. \tag{4.3b}$$

The amplitude equation (4.3a) contains a saturation term whose magnitude is determined by the quantities f_{mn}. The equation for the phase shows a field-dependent resonant shift. This shift depends on f^4_{mn} and will, in general, be small.

This term introduces a slight modification with respect to the two-level results where the field-dependent shift is

nonresonant. Apart from this modification the dynamics of the one-mode case will be the same as that of the two-level model. This result is not astonishing because by considering only one resonant mode we have restricted ourselves to an effective two-level system.

B. Many-mode case

As we have already pointed out in Sec. III, many transitions can coexist above threshold. This means that the one-mode case just discussed corresponds to a strongly idealized situation. In order to investigate the behavior of a realistic vibronic-laser system we need to consider the case in which more than one transition is excited and is resonant with its own cavity mode. For simplicity let us consider the case in which only two transitions are above threshold and each of them is resonant with a cavity mode. This is the case for a system having a gain profile such as that shown in Fig. 3, where two adjacent lines have the same gain. Each transition will give rise to saturation terms such as that found in the one-mode case. Furthermore, there will be a nonresonant mode coupling which is due to the presence of the nonresonant mode-coupling terms in the inversion part of the third-order expansion. Besides these terms which are also expected from the two-level results, new contributions arise from the phonon terms. Besides nonresonant terms which we do not write here explicitly and which are of the same form of that originating from the expansion of $T^{+}_{mm}-T^{-}_{nn}$, we find the resonant terms

$$
\begin{aligned}
(S^{+}_{mn})^{(3)}_{\text{phon}} &= -ig_{k_1}g^{2}_{k_2}\mid A_{k_2}\mid^{2}A^{*}_{k_1}\Delta N_{ml} \\
&\quad\times\frac{f_{ml}f^{2}_{mn}}{\gamma_{mn}\gamma_{ml}\gamma^{T}_{mm}}\exp(-i\alpha_{k_1}t)\ , \\
(S^{+}_{ml})^{(3)}_{\text{phon}} &= -ig^{2}_{k_1}g_{k_2}\mid A_{k_1}\mid^{2}A_{k_2}\Delta N_{m} \\
&\quad\times\frac{f^{2}_{lm}f_{mn}}{\gamma_{mn}\gamma_{ml}\gamma^{T}_{mm}}\exp(-i\alpha_{k_2}t)\ ,
\end{aligned}
\tag{4.4}
$$

and the analogous terms for $(S^{-}_{mn})^{(3)}_{\text{phon}}$.

These terms are completely new and couple the two modes "resonantly." This means that they are more effective than their nonresonant counterparts. We stress the fact that this resonant mode-mode coupling is a feature of the vibronic model, because the corresponding terms originate in the phonon terms, i.e., in the transitions between vibronic states belonging to the same potential sheet. Furthermore, their contribution cannot be neglected in a "zeroth-order approximation," because both transitions and both modes occur above threshold simultaneously. From this viewpoint the vibronic laser appears to be intrinsically a many-mode device. The dynamics of the two coupled modes when all nonresonant terms are neglected is described by

$$
\begin{aligned}
\frac{d}{dt}\mid A_{k_1}\mid &= -\Gamma\mid A_{k_1}\mid -\frac{g^{2}_{k_1}f^{2}_{mn}}{\gamma_{mn}}\Delta N_{mn}\mid A_{k_1}\mid \\
&\quad -\frac{8g^{4}_{k_1}f^{4}_{mn}}{\gamma^{2}_{mn}\gamma^{T}_{mm}}\Delta N_{mn}\mid A_{k_1}\mid^{3} \\
&\quad -\frac{2g^{2}_{k_1}g^{2}_{k_2}f^{3}_{mn}}{\gamma_{mn}\gamma_{ml}\gamma^{T}_{mm}}f_{ml}\Delta N_{ml}\mid A_{k_2}\mid^{2}\mid A_{k_1}\mid\ , \\
\frac{d}{dt}\mid A_{k_2}\mid &= -\Gamma\mid A_{k_2}\mid -\frac{g^{2}_{k_2}f^{2}_{ml}}{\gamma_{mn}}\Delta N_{ml}\mid A_{k_2}\mid \\
&\quad -\frac{8g^{4}_{k_2}f^{4}_{ml}}{\gamma^{2}_{mn}\gamma^{T}_{mm}}\Delta N_{ml}\mid A_{k_2}\mid^{3} \\
&\quad -2\frac{g^{2}_{k_1}g^{2}_{k_2}f^{3}_{ml}}{\gamma_{mn}\gamma_{ml}\gamma^{T}_{mm}}f_{mn}\Delta N_{mn}\mid A_{k_1}\mid^{2}\mid A_{k_2}\mid\ .
\end{aligned}
\tag{4.5}
$$

These equations show explicitly that the saturation terms belong to one transition only (only f^{4}_{mn} quantities appear in the coefficients), whereas the mode-coupling terms imply a transition to different vibronic states because they contain the $f_{ml}f_{mn}$ products. This means that the mode-coupling terms describe a redistribution of the emitted intensity between the two modes which is demonstrated by (4.5). We want to stress the fact that the mode coupling which thus appears is rather a "transitions coupling" in the sense that different modes are coupled by different transitions. This point makes clear that the dynamics of a vibronic laser is quite complicated and should be discussed on the basis of a numerical solution of (4.5). However, already in the framework of our very simplified analysis some of its relevant features can be extracted.

We conclude this section with one more remark. The coupling between transitions which is represented by (4.5) indicates how the many color superfluorescence experiments[5] can be interpreted in the framework of our model. In fact, different resonant transitions are always coupled together with coupling strengths which depend on the Frank-Condon overlap integrals. Transitions whose transition frequencies are not quite different and with not too different strengths will interact. Therefore, a redistribution of the emission between these transitions will follow which can lead to superfluorescent emission between different states at the same time. For a simplified model consisting of an upper- and two lower-vibronic levels the model used in Ref. 14 is found.

V. CONCLUSIONS

We summarize our results as follows. We have presented an analysis of the vibronic laser action which is based on the whole vibronic level scheme as it is determined by the electron-phonon interaction. The resulting description, which may have some formal analogy with that of a many-level laser, allows us to give a detailed picture of laser action in these systems. We have presented gain profiles which are characteristic for vibronic systems because the interplay between the different possible tran-

sitions results in a higher-gain profile than what is expected for a single transition. Furthermore, we are able to give selection rules and line strengths for the different transitions involved as a function of the electron-phonon coupling as expressed through the Frank-Condon integrals. Finally, an insight into the dynamics of laser action in a vibronic system showing interesting effects of transition-mode coupling and intensity redistribution between modes has been given. Furthermore, we have given a general scheme in which optical effects in vibronic systems and their dynamics can be studied. Application of these ideas to realistic systems is the goal of future work.

APPENDIX

We collect here all algebraic details concerning the perturbation calculation which leads from (2.13)–(2.15) to the first- and third-order terms [(A4), (A7), and (A8)] needed in Secs. III and IV. Before going over calculations we make the following ansatz for the field amplitude:

$$a_k = A_k \exp[i(\omega_k + \varphi_k)t] , \tag{A1}$$

where φ_k is an unknown phase which describes the frequency shift induced in the field modes by the lasing material. In general, φ_k will depend on the field intensity as is determined from the theory.[7–9]

First of all rewrite (2.13) and (2.15) in integral form

$$\begin{aligned} T^{+}_{mn} = -i \sum_k \sum_l \int_0^t \exp\{ -[\gamma^{+}_{mn} + i(n-m)\Omega](t-t') \} \\ \times [A_k f_{nl} S^{+}_{ml} \exp(i\alpha_k t') \\ - A^{*}_k f_{lm} S^{-}_{ln} \exp(-i\alpha_k t') \\ + \delta_{mn} \gamma_{mn} \delta_{nl} \Delta N_{ml}] dt' , \end{aligned} \tag{A2}$$

$$\begin{aligned} S^{+}_{mn} = -i \sum_k \sum_l \int_0^t \exp\{ -[\gamma_{mn} + i\varepsilon \\ - i(n-m)\Omega](t-t') \} \\ \times [A^{*}_k f_{ln} T^{+}_{ml} \exp(-i\alpha_k t') \\ - A_k f_{ml} T^{-}_{ln} \exp(i\alpha_k t')] dt . \end{aligned} \tag{A3}$$

Then introduce the zeroth-order approximations (2.18) and perform the integrals over t in the limit $t \gg 1/\gamma_{mn}$ obtaining

$$\begin{aligned} &(T^{\pm}_{mn})^{(1)} = 0, \quad m \neq n \\ &(I_{mn})^{(1)} = 0 , \\ &(S^{+}_{mn})^{(1)} = -i \sum_k g_k A^{*}_k \frac{I^{+}_{mn}}{\varepsilon_{nm} - i\alpha_k} \exp(-i\alpha_k t) , \\ &(S^{-}_{nm})^{(1)} = i \sum_k g_k A_k \frac{I^{-}_{mn}}{\varepsilon^{*}_{nm} - i\alpha_k} \exp(i\alpha_k t) , \end{aligned} \tag{A4}$$

where we have introduced, in order to simplify the notation,

$$\begin{aligned} &\varepsilon_{nm} = \gamma_{mn} + i[\varepsilon - \Omega(n-m)] , \\ &I^{+}_{mn} = f_{mn} \Delta N_{mn} , \\ &I^{-}_{mn} = f_{nm} \Delta N_{mn} . \end{aligned}$$

The terms $\gamma_{m\pm1\,n\pm1}\, S^{+}_{m\pm1\,n\pm1}$ have not been considered. Their effect consists merely in a redefinition of the coefficient of the field amplitude in (A3) and (A4) by a factor $\gamma_{m\pm1\,n\pm1}/(\varepsilon - (n-m)\Omega - \alpha_k)$ which is always much smaller than one on the time scale of the field variation, which is the relevant time scale in this context. In fact, the denominators which appear in it are always nonresonant and contain a frequency difference which grows with n,m. These terms could play a role in describing transient effects which we do not consider in this paper.

The second-order contributions are obtained by following the same procedure and give for the population inversion terms

$$\begin{aligned} (T^{+}_{mm})^{(2)} = -\sum_{k_1 k_2} \sum_l g_{k_1} g_{k_2} \Bigg[A_{k_2} A^{*}_{k_1} f_{ml} I^{+}_{ml} \exp[i(\alpha_{k_2} - \alpha_{k_1})t] \frac{1}{(\varepsilon_{lm} + i\alpha_{k_1})[\gamma^{T}_{mm} - i(\alpha_{k_2} - \alpha_{k_1})]} \\ + A_{k_1} A^{*}_{k_2} f_{lm} I^{-}_{lm} \frac{\exp[i(\alpha_{k_1} - \alpha_{k_2})t]}{(\varepsilon^{*}_{lm} - i\alpha_{k_1})[\gamma^{T}_{mm} - i(\alpha_{k_1} - \alpha_{k_2})]} \Bigg] , \end{aligned} \tag{A5}$$

and for the phonon terms

$$\begin{aligned} (T^{+}_{ml_1})^{(2)} = -\sum_{k_1 k_2} \sum_{l_2} g_{k_1} g_{k_2} \Bigg[A_{k_2} A^{*}_{k_1} f_{l_1 l_2} I^{+}_{ml_2} \frac{\exp[i(\alpha_{k_2} - \alpha_{k_1})t]}{(\varepsilon_{l_2 m} + i\alpha_{k_1})[\omega^{+}_{l_1 m} - i(\alpha_{k_2} - \alpha_{k_1})]} \\ + A_{k_1} A^{*}_{k_2} f_{l_2 m} I^{-}_{l_1 l_2} \frac{\exp[i(\alpha_{k_1} - \alpha_{k_2})t]}{(\varepsilon^{*}_{l_2 l_1} - i\alpha_{k_1})[\omega^{+}_{l_1 m} - i(\alpha_{k_1} - \alpha_{k_2})]} \Bigg] . \end{aligned} \tag{A6}$$

The $\gamma^{\pm}_{m\pm1\,n\pm1}$ terms in (A5) and (A6) have been left out using the same argument which has been introduced for the $\gamma_{m\pm1\,n\pm1}$ in the first-order terms. An analogous argument allows us to disregard the contributions of the $\gamma^{T}_{m\pm1\,n\pm1}$ in the expression for the population inversion. Thus, also in this case the influence of these terms may be disregarded on the time scale which is relevant for our consideration. As has been shown in Ref. 14, their contribution becomes essential when transient effects like superfluorescence are discussed.

$$
\begin{aligned}
(S^{+}_{mn})^{(3)} = i \sum_{k_1,k_2,k_3} g_{k_1} g_{k_2} g_{k_3} \frac{1}{\varepsilon_{nm} + i(\alpha_{k_1} - \alpha_{k_2} + \alpha_{k_3})} f_{mn}
\\
\times \Bigg\{ \sum_{l_1} \Bigg[A^{*}_{k_3} \exp(-i\alpha_{k_3} t) f_{l_1 n} \sum_{l_2} \Bigg[A_{k_2} A^{*}_{k_1} \exp[i(\alpha_{k_2} - \alpha_{k_1})t] f_{l_1 l_2} I^{+}_{m l_2} \frac{1}{(\varepsilon_{l_2 m} + i\alpha_{k_1})[\omega^{+}_{l_1 n} - i(\alpha_{k_2} - \alpha_{k_1})]}
\\
+ A_{k} A^{*}_{k_2} \exp[i(\alpha_{k_1} - \alpha_{k_2})t] f_{l_2 m} I^{-}_{l_2 l_1}
\\
\times \frac{1}{(\varepsilon^{*}_{l_2 l_1} - i\alpha_{k_1})[\omega^{+}_{l_1 m} - i(\alpha_{k_1} - \alpha_{k_2})]} \Bigg] \Bigg]
\\
+ \Bigg[A^{*}_{k_3} \exp(-i\alpha_{k_3} t) f_{m l_1} \sum_{l_2} \Bigg[A^{*}_{k_1} A_{k_2} \exp[i(\alpha_{k_2} - \alpha_{k_1})t] f_{l_1 l_2} I^{+}_{l_1 n}
\\
\times \frac{1}{(\varepsilon_{n l_2} + i\alpha_{k_1})[\omega^{-}_{n l_1} + i(\alpha_{k_2} - \alpha_{k_1})]}
\\
+ A_{k_1} A^{*}_{k_2} \exp[i(\alpha_{k_1} - \alpha_{k_2})t] f_{n l_2} I^{-}_{l_1 l_2}
\\
\times \frac{1}{(\varepsilon^{*}_{l_1 l_2} - i\alpha_{k_3})[\omega^{-}_{n l_1} - i(\alpha_{k_1} - \alpha_{k_2})]} \Bigg] \Bigg] \Bigg\}, \qquad \text{(A7)}
\end{aligned}
$$

$$
\begin{aligned}
(S^{-}_{mn})^{(3)} = i \sum_{k_1,k_2,k_3} g_{k_1} g_{k_2} g_{k_3} \frac{1}{\varepsilon^{*}_{nm} - i(\alpha_{k_1} - \alpha_{k_2} + \alpha_{k_3})} f_{nm}
\\
\times \Bigg\{ \sum_{l_1} \Bigg[A^{*}_{k_3} \exp(-i\alpha_{k_3} t) f_{n l_1} \sum_{l_2} \Bigg[A_{k_2} A^{*}_{k_1} \exp[i(\alpha_{k_2} - \alpha_{k_1})t] f_{m l_2} I^{+}_{l_1 l_2} \frac{1}{(\varepsilon_{l_1 l_2} + i\alpha_{k_1})[\omega^{+}_{m l_1} - i(\alpha_{k_2} - \alpha_{k_1})]}
\\
+ A_{k_1} A^{*}_{k_2} \exp[i(\alpha_{k_1} - \alpha_{k_2})t] f_{l_2 l_1} I^{-}_{l_2 m}
\\
\times \frac{1}{(\varepsilon^{*}_{l_2 m} - i\alpha_{k_1})[\omega^{+}_{m l_1} - i(\alpha_{k_1} - \alpha_{k_2})]} \Bigg] \Bigg]
\\
+ \Bigg[A^{*}_{k_3} \exp[-i\alpha_{k_3} t] f_{l_1 m} \sum_{l_2} \Bigg[A^{*}_{k_1} A_{k_2} \exp[i(\alpha_{k_2} - \alpha_{k_1})t] f_{l_2 n} I^{+}_{l_2 l_1}
\\
\times \frac{1}{(\varepsilon_{l_1 l_2} + i\alpha_{k_1})(\omega^{-}_{l_1 n} + i[\alpha_{k_2} - \alpha_{k_1}])}
\\
+ A_{k_1} A^{*}_{k_2} \exp[i(\alpha_{k_1} - \alpha_{k_2})t] f_{l_1 l_2} I^{-}_{n l_2}
\\
\times \frac{1}{(\varepsilon^{*}_{n l_2} - i\alpha_{k_3})[\omega^{-}_{l_1 n} - i(\alpha_{k_1} - \alpha_{k_2})]} \Bigg] \Bigg] \Bigg\}. \qquad \text{(A8)}
\end{aligned}
$$

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