Theoretical description of light amplification in $ZnTe:O: A$ vibronic-laser material

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Recent gain measurements in ZnTe:0 at 2 K are discussed in the framework of a vibronic-laser model. The behavior of the gain spectrum and of the saturation coefficient is understood within the model. Furthermore, the use of a one-dimensional amplifier equation when describing the dependence of the emitted intensity on the excitation length is justified.

I. INTRODUCTION

Gain measurements in ZnTe:0 at low temperature have been recently reported¹ as an example of a broadly tunable solid-state amplifier in the visible frequency range. Their interpretation raises some interesting questions, which can be analyzed in the framework of a vibronic-laser model.

First of all, let us briefly review the experimental results. As is well known, ZnTe is a direct-gap semiconductor with a band gap of about 2.38 eV. The impurity oxygen is isoelectronic with Te and gives rise to discrete levels in the forbidden band gap. The impurity is excited either indirectly via the valence-to conduction-band transition or directly by pumping into the discrete levels of the impurity. The absorption and luminescence spectra of ZnTe:0 at 2 K are shown in Fig. 1. They are well separated, having only the zero-phonon line in common, and show a peculiar structure that is associated with phonon-assisted processes. The properties of these spectra have been carefully discussed in Ref. 2, where further references are given. They are well understood in terms of a simple model for the luminescence from isoelectronic impurities, which has been used in discussing the optical properties of AgBr: $I³$ Here, however, one has to consider the coupling of both optical (longitudinal and transverse) and acoustic (longitudinal and transverse) phonons to the excitation in order to explain the rich structure of the spectra. The separation between absorption and emission spectra indicates that, with the exception of the zero-phonon line, the emitted light will not be reabsorbed within the material. This is different from what happens in atomic transitions as they are currently described in laser models. Furthermore, the emission takes place at wavelengths that are longer than that of the absorption. This indicates that the excitation decays to a state whose energy is that of the ground state of the electrons plus the energies of the phonons involved in the transition (see Fig. 2, in which we have given a schematic model of the transitions involved). We shall, nevertheless, stress that the scheme presented should not be taken too seriously because the localized electronic states should normally not be described in k space. There are indications that the lifetimes of the phonons in these states are very short. As a consequence, the virtual lower phonon states will not enter into the dynamics of the populations. Therefore, we have a situation that reminds one of a typical four-level laser model, where a rapidly decaying lower state favors the buildup of inversion and thus a large gain. This is also a feature of ZnTe:0 system, as shown in Ref. 1. Contrary to the four-level laser model, however, we have here the efFects of a number of phonon-

K.

FIG. 1. Absorption and luminescence spectra of ZnTe:O at 2 FIG. 2. Simple scheme of the transitions involved in the oxygen —bound-exciton recombination.

assisted transitions, or, roughly speaking, transition lines, which all share the same electronic excited state. The gain spectrum presented in Ref. ¹ reproduces well the luminescence spectrum of ZnTe:0. This result is interpreted¹ within a model for vibronic laser action.⁴⁻⁶ This model gives the general framework in which the experimental results on the amplifying properties of ZnTe:0 can be analyzed. As we shall see, it allows one to explain the gain spectrum as well as the saturation properties observed in ZnTe:O. The interplay of these two quantities in determining the emission intensity as a function of the excitation length is best illustrated in Fig. 3, where the intensity emitted at four different wavelengths is presented together with the solutions of (1), where the values of the gain and saturation are fitted from the experimental points and are, for λ =629.8 nm,

$$
g = 660 \text{ cm}^{-1}
$$
,
\n $\alpha = 10^{-4} \text{ arb. units}$,
\n $\kappa = 40 \text{ cm}^{-1}$,
\n $I_0 = 2 \times 10^6 \text{ arb. units cm}^{-1}$.

In all curves in Fig. 3, all values of α remain the same and are independent of the wavelength, whereas the spectral dependence of the gain is the same as that of the luminescence. This behavior raises another question that we want to address. As shown in Ref. 1, the experimental points are well fitted by the solutions of the equation

$$
\frac{dI}{dx} = \frac{g(\omega)I}{1+\alpha I} - \kappa I + I_0(\omega) , \qquad (1)
$$

where $g(\omega)$ is the frequency-dependent gain, and α is the saturation parameter. The constant κ stands for the losses, and $I_0(\omega)$ is the spontaneous emission per unit length. This equation (1} is well established for a twolevel atomic system, whereas its use in the context of a vibronic system has to be justified. Our task will be to

FIG. 3. Emitted intensity vs excitation length for different wave lengths: (a) $\lambda = 629.8$ nm, (b) $\lambda = 638.1$ nm, (c) $\lambda = 646.6$ nm, and (d) $\lambda = 651.9$ nm. The solid lines correspond to the solution of (1) when the parameters g, α , κ , and I_0 are found with a fitting of the experimental points.

show that the pertinence of Eq. (1) to the description of amplification in ZnTe:0 is justified from a microscopical model. This means that we have to calculate the gain spectrum and the saturation properties of ZnTe:O considered as a vibronic amplifier. In order to do this we will use the model of Ref. 4, which will be adapted to the choice of this specific material.

The paper will then be organized as follows. In Sec. II the model of Ref. 4 is adapted to the characteristics of ZnTe:0. In Sec. III the theoretical and experimental results are compared and discussed.

II. THE MODEL

The simplest model that allows one to describe the emission properties of ZnTe:0 consists of two electronic levels coupled to optical and acoustic phonons and to the electromagnetic field. The main features of this model and the algebraic manipulation that allows one to write down equations for the vibronic transitions have already been discussed extensively elsewhere.^{4,5} Here we quote only the relevant difFerences, which are due to the specific choice of the material. The two main coupling terms in the Hamilton operator for this system are the dipole coupling to the electronic states and the electron-phonon coupling. The contribution of the first interaction is standard and can be found in Ref. 4. The electron-phonon contribution needs some comment: as already anticipated in the Introduction, the electronic states couple to both optical and acoustic phonons (longitudinal and transverse}. From the form of the phonon density of states of $ZnTe:O₁⁷$ one infers that a suitable model for the electron-phonon interaction is obtained by coupling the excitation to all four types of intervening phonons instead of considering optical phonons only in an Einstein model, as in Ref. 4. Therefore, we rewrite the electron interaction as

$$
H_{\text{electron-phonon}} = \sum_{i=1}^{4} \sum_{j} \left[\sigma_{zj} \sum_{q} \lambda_{q}^{i} (b_{q}^{i} + b_{q}^{i\dagger}) \right], \qquad (2)
$$

where b_q and b_q^{\dagger} are the phonon operators and σ_{zj} are electron operators at position j . The phonon frequencies will be Ω^i_{ρ} . The four constants λ^i_{ρ} describe the coupling to the four types of phonons. In order not to over complicate the notation, in the following we will consider only one type of phonon type and drop the index i and the sum over it. In the results the index i has to be restored. As usual, we have replaced the electronic variables by psedudospin operators, because we consider two electronic states only.

The quantity of interest is the global polarization of the emitting medium, which in the notation of Ref. 4 is given by

$$
P(x,t) = \sum_{n_q} \sum_{l_q} \left| f_{n_q, l_q}^+ \right| \frac{\lambda_q}{\Omega_q} \left| S_{n_q, l_q}^+ \right|
$$

+ $f_{n_q, l_q}^- \left| \frac{\lambda_q}{\Omega_q} \right| S_{n_q, l_q}^- \left| \left\langle r_{12} \right\rangle \right|$
= $P^+ + P^-$ (3)

Here the quantities $S^{\pm} = \sum_j S^{\pm}_j$ are mean values of the operators describing the transitions between the exited state and the lower vibronic states, and $\langle r_{12} \rangle$ is the matrix element of the dipole operator between the two electronic states. The vibronic states are labeled with the indices l_q and n_q . The quantities f_{ln} are the overlap integrals between the excited and nonexcited vibronic states.⁴ These states are the eigenstates of displaced harmonic oscillators and correspond to the eigenstates of the electron-phonon part of the interaction given by (2}. In order to calculate P, we need to know the explicit expressions for S^{\pm} and f_{in} . These are obtained from the equations governing the evolution of the different vibronic transitions. These equations, whose general form is given in Ref. 4, are greatly simplified owing to the characteristics of ZnTe:O. As one sees in Fig. 2, the luminescence spectrum indicates that the final state of all transitions, with the exception of the zero-phonon line, consists of the electronic ground state plus some phonons. In terms

of the vibronic transitions, this means that the vibronic states corresponding to the lower oscillator have huge relaxation rates and do not contribute to the dynamics directly. This allows us to greatly reduce the number of transitions involved in the model of Ref. 4. We can then think of our system as consisting of an excited state coupled to an ensemble of nonexcited vibronic states which decay very rapidly. When considering one vibronic transition only and the explicit form of the pumping, one obtains the usual four-level model for a laser. This ensures that the gain of the system will be very large. However, in the case we are dealing with here, all transitions are coupled via the same excited state, and we have to consider their interplay, too. Furthermore, the global polarization is introduced as a variable. The equations are understood as describing the evolution of the mean values of the material quantities interacting with a classical field of amplitude $E(x,t) = \sum_k E_k(x) \exp(i\omega_k t)$. The material equations that are appropriate to our problem are then

$$
P^{+}(x,t) = -i \sum_{k} g_{k} E_{k}^{*}(x) \int_{0}^{t} \left[e^{-i(\omega_{k} + \varepsilon)(t - t')} \left[\sum_{q} \left(\frac{\lambda_{q}}{\Omega_{q}} \right)^{2} e^{-i\Omega_{q}(t - t')} \right]^{n} \frac{\exp \left[-\sum_{q} \left(\frac{\lambda_{q}}{\omega_{q} z} \right)^{2} \right]}{n!} T_{00}^{+}(t') \right] dt' \tag{4}
$$

and

$$
\frac{dT_{\omega}^+}{dt} = -\gamma_0 T_{\omega}^+ + \gamma_0 N - i \sum_k g_k E_k(x) e^{i\omega_k t} P^+
$$

+ $i \sum_k g_k E_k^*(x) e^{-i\omega_k t} P^-$. (5)

We have introduced here the explicit form of the overlap integrals for the relevant transitions, which are expressed as

$$
f_{0nq} = \prod_{q} \left[\frac{\lambda_q}{\Omega_q} \right]^{n_q} \frac{1}{n_q!} \exp \left[-\sum_{q} \left[\frac{\lambda_q}{\Omega_q} \right]^2 \right].
$$
 (6)

In (4) and (5) T_{00}^+ describes the population of the excited level, ε is the energy difference between the two electronic states, and g_k contains the dipole matrix element for the electronic transition and is the coupling constant to the field. Furthermore, we have introduced the excitation rate $\gamma_0 N$ for the excited state. There is also an equation for the population of the electronic ground state, which is not written here. It couples the two electronic states only and is not relevant to the gain, whereas its contribution to saturation will be automatically considered. For later use we have decomposed the time-dependent part of the field in plane waves of frequency ω_k . All of these equations contain relaxation rates that are related to spontaneous emission and nonradiative transitions or whatever process is supposed to contribute to the linewidth of optical transitions. In the material under study at 2 K

there are mainly two sources of linewidth: spontaneous emission, which determines the width of the zero-phonon line, and the width, which originates in the density of states of the phonons, whose relevance to the linewidth has been emphasized in Ref. 3. Apparently there are no relevant contributions of nonradiative transitions. Therefore, we have introduced into (5} the relaxation rate corresponding to the spontaneous linewidth γ_0 in the equations for the populations. The density of states contributes to the relaxation too. In the Appendix we show how this relaxation time, which afFects the global polarization P, can be made explicit in the equations. Using the results of the Appendix, the equation for the polarization is rewritten as

$$
P(x,t) = -i \sum_{k} g_k E_k^*
$$

$$
\times \sum_{n} \int_0^t \left[e^{-[i(\omega_k + \varepsilon - n\Omega) + \gamma n](t - t')} \times \frac{S^n}{n!} e^{-S^2} T_{00}^+(t') \right] dt', \qquad (7)
$$

where γ is the relaxation rate related to the phonon density of states, and S is defined in the Appendix.

As already mentioned, the electromagnetic field is treated as a classical variable $E(x,t)$ in one dimension. This approximation is justified in light of the experiments. In the usual slowly varying envelope approximation, and written with respect to a definite-frequency component in the stationary regime, the field intensity obeys the equation.

$$
\frac{dI_k}{dx} = g_k [E_k^*(x)P^- + E_k(x)P^+] , \qquad (8)
$$

where $P^{\pm}(x,t)$ have been defined in (3) and are calculated from (4) and from its complex-conjugate form for P^- . Our strategy will now be the following: we solve (4) and (5) by a perturbative expansion around the equilibrium value of the excitation in the stationary regime. 8 This is

the procedure already used in Ref. 4. Then we insert the result into Eq. (8) and compare it with the corresponding terms of the expansion of (1) for $\alpha I \ll 1$ This will allow us to identify the explicit expressions of $g(\omega)$ and α .

III. GAIN AND SATURATION COEFFICIENTS

The calculation of the gain spectrum is performed as outlined in the previous section. The first-order contribution to the global polarization is

$$
P^{+(1)}(x,t) = -i \sum_{k} g_k E_k^*(x) \int_0^t dt' e^{-i(\omega_k + \varepsilon)(t-t')} N \sum_n \left\{ \left[\sum_q \left(\frac{\lambda_q}{\Omega_q} \right)^2 e^{-i\Omega_q(t-t')} \right]^n \exp \left[-\sum_q \left(\frac{\lambda_q}{\Omega_q} \right)^2 \right] \right\}.
$$
 (9)

For a given observation frequency ω_0 the gain is then determined by

$$
g(\omega_0) = g_k^2 N \sum_n \left[\sum_q \left(\frac{\lambda_q}{\Omega_q} \right)^2 \delta(\Omega_q - \varepsilon - \omega_0) \right]^n
$$

$$
\times \exp \frac{\left[-\sum_q \left(\frac{\lambda_q}{\Omega_q} \right)^2 \right]}{n!} . \tag{10}
$$

This quantity shows the same behavior as the luminescence spectrum⁴ when ω_0 is varied. Therefore, we see explicitly that the gain spectrum and the luminescence spectrum have the same dependence on energy, which corresponds well to the experimental results reported in Ref. [1]. As an illustration of the inffuence of the interplay of the different vibronic transitions in the gain spectrum, we evaluate the gain spectrum $g(\omega)$ using the simplified expressions in the Appendix, with a phonon density of states approximated by Lorentzian curves centered on the experimental phonon energies. The values of

luminescence spectrum, Fig. 2. The result is shown in Fig. 4, taking into account the contributions of all phonon types, and can be compared to the measured luminescence spectrum. To each different transition corresponds a sharp line in the spectrum. However, due to the finite width γ of the lines, they superimpose in the sum and also lead to substantial gain in those spectral regions in which the single line would not show any. A more quantitative calculation can be done within the model presented in Ref. 3 for AgBr:I, which corresponds well with the one used here, using an experimental phonon density of states, and leads to the gain spectrum shown in Fig. 5, where it is compared to the experimental luminescence curve. The agreement between both curves is satisfactory. Values for the gain, however, are more difficult to determine because the density of O centers is not known with sufficient accuracy. From (10) one also sees that the gain has as a lower bound the gain of the

the parameter S_i , Ω_i , and γ_i are obtained from the

FIG. 4. Approximate theoretical gain spectrum from (A5). The dashed line corresponds to the gain spectrum for the longitudinal-phonon contribution alone.

FIG. 5. Gain spectrum calculated with the model of Testa et al. (Ref. 3) (a) compared to the experimental luminescence curve (b) for liquid-helium temperature.

pure electronic transition. This is useful in order to evaluate the order of magnitude of the gain to be obtained in this material.

The calculation of the saturation coefficient is more involved and requires the use of third-order terms in the field amplitude. The corresponding expressions are cumbersome and are found in the Appendix of Ref. 4 [see Eqs. (A7) and (AS)]. These expressions are inserted into (3) and give the third-order contribution $P^{(3)}$ to the polarization. As is well known, $⁸$ this quantity is expressed as</sup> the product of the third power of the field amplitude times the gain and the saturation coefficient α . Because we want to give an expression for α , we need to discuss $P^{(3)}$ in some detail. In our system with only one excited state, $P^{(3)}$ reduces in the sum of two terms, one of which is the complex conjugate of the other. Therefore we need to discuss only this single term, which we call M in the following. Its explicit expression is

$$
M = 2 \int_0^{\infty} dt \int \int d\omega_{k_1} d\omega_{k_2} d\omega_{k_3} \left\{ e^{i(\omega_0 - \omega_{k_1} + \omega_{k_2} - \omega_{k_3})t} N g_{k_1} g_{k_2} g_{k_3} \right\}
$$

\n
$$
\times E_{k_1}(x) E_{k_2}^*(x) E_{k_3}(x) \exp \left[-2 \sum_q \left(\frac{\lambda_q}{\Omega_q} \right)^2 \right] \frac{1}{\gamma_0 + i(\omega_{k_2} - \omega_{k_3})}
$$

\n
$$
\times \sum_n \left[\sum_q \left(\frac{\lambda_q}{\Omega_q} \right)^2 \delta(\omega_q - \varepsilon - \omega_{k_1} + \omega_{k_2} - \omega_{k_3}) \right] \frac{1}{n!}
$$

\n
$$
\times \sum_n \left[\sum_q \left(\frac{\lambda_q}{\Omega_q} \right)^2 \delta(\Omega_q - \varepsilon + \omega_{k_3}) \right] \frac{1}{m!} \right],
$$

\n(11)

where $\omega_0 = k_0/c$. For the component of the field with frequency ω_0 , we find that (11) reduces to

 ϵ

$$
M = 2g_{k_0}^3 E_{k_0}(x) N \sum_{n} \left[\sum_{q} \delta(\Omega_q - \omega_0 - \epsilon) \left[\frac{\lambda_q}{\Omega_q} \right]^2 \right]_{n}^{\alpha} \frac{1}{n!} \exp \left[- \sum_{q} \left[\frac{\lambda_q}{\Omega_q} \right]^2 \right]
$$

$$
\times \int \int d\omega_{k_1} d\omega_{k_2} \left\{ \frac{E_{k_2}^*(x) E_{k_1}(x)}{\gamma_0 + i(\omega_0 - \omega_{k_1})} \right\}
$$

$$
\times \sum_{m} \left[\sum_{q} \left[\frac{\lambda_q}{\Omega_q} \right]^2 \delta(\Omega_q - \epsilon + \omega_{k_2} - \omega_0 - \omega_{k_1}) \exp \left[- \sum_{q} \left[\frac{\lambda_q}{\Omega_q} \right]^2 \right] \right].
$$
 (12)

The first term in the product is the gain spectrum. The rest of the expression under the integral contains once more the gain spectrum. In a good approximation we can then write the last two terms in the product.

$$
M = 2g(\omega_0)E_{k_0}|E_{k_0}|^2 \int \int d\omega_{k_1} d\omega_{k_2} \left[\sum_n \left\{ \sum_q \left[\delta(\epsilon - \Omega_q + \omega_{k_1} - \omega_{k_2} + \omega_0) \left(\frac{\lambda_q}{\Omega_q} \right)^2 \right] \right\}^n \frac{\exp\left[-\sum_q \left[\frac{\lambda_q}{\Omega_q} \right]^2 \right]}{n! \gamma_0 + i(\omega_0 - \omega_{k_1})} \right], \quad (13)
$$

where $g(\omega_0)$ is the gain given in (10). This approximation relies on the following properties of the functions under the integral in (11): the first term has a sharp maximum around ω_0 , and therefore the function $E_{k_1}(x)$ can be approximate by $E_{k0}(x)$. The other term has the form of the gain curve on which ω_0 lies. Therefore it may be approximated by a δ function with the argument $\omega_{k_1} - \omega_{k_2}$, thus giving the desired result. Taking now the sum of M and its complex conjugate, and remembering that $P^{(3)}$ is expressed as the product $g(\omega)|E_{k_0}|^2|E_{k_0}|\alpha$, we find from (13) that the saturatio coefficient is given by

$$
\alpha = \int \int d\omega_{k_1} d\omega_{k_2} \left\{ \frac{\exp \left[-\sum_q \left(\frac{\lambda_q}{\Omega_q} \right)^2 \right]}{\gamma_0 + i(\omega_0 - \omega_{k_1})} \sum_n \frac{1}{n!} \left[\sum_q \delta(\epsilon - \Omega_q + \omega_{k_2} - \omega_{k_1} + \omega_0) \left(\frac{\lambda_q}{\Omega_q} \right)^2 \right]^n \right\} + \text{c.c.} \quad (14)
$$

which is independent of ω_0 because the integrals extend over the full ω range. This gives theoretical support to the experimental result on the independence of the saturation coefficient of the frequency stated in Ref. 1.

From (14) it follows that the saturation coefficient calculated from the data for the pure electronic transition gives an upper bound for the realistic values of α . In fact, the short lifetime of the lower vibronic states implies that the saturation will not be very different from that found for the pure electronic transition. This gives a qualitative explanation of the result expressed by (12). This point deserves some additional comments. In order to understand the saturation process, we use the following picture: one photon of frequency ω_0 is emitted. Because of the rapid decay of the lower vibronic states, reabsorption, if it happens at all, cannot be influenced by transitions between lower vibronic states. This is expressed in (11) by the presence in the integrals of a frequency-dependent function which does not contain phonon frequencies. In the next step there is once more emission from the same excited state at the same frequency, and so on. Saturation builds up from the repetition of these processes to an arbitrary order, and therefore, it is expected to give the same frequency-independent value for all emission frequencies that share the same excited state. This is evident when using the approximate form (7) of the equation for $P(x, t)$. Having carried out the calculations that lead to (14), one sees by inspection that α is approximately given by the expression for the saturation of the pure electronic transition. A different situation would arise if an appreciable population of the lower vibronic levels as well as transitions between these levels would be present. In this case we expect α to be frequency dependent. An illustration of this different behavior is discussed in Ref. 9. When the terms in the expansion of the solution for material variables in the vibronic model are substituted into (8), we obtain an equation that is equivalent of the expansion of (1) in powers of αI . This means that (1) gives a satisfactory description of the amplification properties of ZnTe:O.

In conclusion, we have shown that the one-dimensional amplifier equation (1) gives a good description of the emission properties of ZnTe:0, and that the vibronic model of Ref. 4 allows the determination of the gain and saturation parameters in (1).

APPENDIX

The expressions (9) and (14) require numerical calculations in order to give quantitative results. An insight into their structure is obtained from approximate analytical results which rely on an assumption about the form of the phonon density of states. We need to discuss the sum over the phonon wave vector q which appears in (4) in some detail. The sum over q is rewritten as an integral using the phonon density of states $\rho(\Omega_a)$,

$$
S(t) = \sum_{q} \left[\frac{\lambda_{q}}{\Omega_{q}} \right]^{2} e^{-i\Omega_{q}(t-t')}
$$

= $\int d\Omega_{q} \left[\left(\frac{\lambda_{q}}{\Omega_{q}} \right) \right]^{2} e^{-i\Omega_{q}(t-t')} \rho(\Omega_{q}) \right]$. (A1)

For an Einstein model, we would find the expression used in Ref. 4. When the density of states is approximated by a sum of a Lorentzians with centers at frequencies Ω corresponding to the maximum of the density of states and widths corresponding to the experimental ones, we interpret (Al) as a Fourier transform and rewrite

$$
S(t) = \int_0^\infty e^{-(\gamma + i\Omega)(t-\tau)} \varphi(\tau) d\tau , \qquad (A2)
$$

where $\varphi(\tau)$ ($\tau = t - t'$) is the time Fourier transform of the normalized coupling and the explicit form of the Lorentzian has been introduced. It is evident that this expression is equivalent to

$$
S(t) = e^{-(\gamma + i\Omega)t} S \tag{A3}
$$

with

$$
S = \int d\Omega_q \rho(\Omega_q) \left[\frac{\lambda_q}{\Omega_q} \right]^2.
$$
 (A4)

When inserting (A3) into (7) we obtain the same expression that can be derived from the equations in Ref. 4, with a decay rate that is determined by the density of states. For general forms of the density of states, one can only quote an implicit expression. In this spirit, the material equations become

$$
\frac{dP^+}{dt} = -i \sum_k g_k E_k^*(x) \sum_n \frac{S^n}{n!} e^{-S^2}
$$

$$
\times \int_0^t e^{-[i(t+\omega_k - n\Omega) + \gamma n]t'} \times T_{00}^+(t-t')dt', \quad (A5)
$$

and the gain is given as in Ref. 4 by

$$
g(\omega_0) = g_k^2 \sum_{i=1}^4 \left[\sum_n \left[\frac{(n+1)\gamma_i S_i^n e^{-S_i^2} N}{\gamma_i^2 + (\omega_0 + \varepsilon - n \Omega_i)^2} \frac{1}{n!} \right] \right], \quad (A6)
$$

where a sum over all involved phonon types is carried out.

This expression was used to calculate the approximated gain spectrum in Fig. 4. The values of linewidths and peak frequencies are taken from the experimental data.

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