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Correlations in extended high-density superfluorescence: A self-organized distributed feedback laser

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We present a simple model that allows a description for the correlation of counterpropagating waves and the formation of spatial patterns in superfluorescent emission. No separation between left- and right-running waves is performed, and retardation effects are fully included. The results indicate qualitative agreement between our calculations and experiments in $KCl:O_2^-$.

I. INTRODUCTION

As it is well known in highly excited solid-state systems like $KCl:O_2^-$ (Refs. 1 and 2) and diphenyl:pyrene,[3-6] superfluorescent emission is observed (for a review see, e.g., Refs. 7–9). In particular, the experiments show that a spatial mode pattern develops in the emitted field, which seems to originate from a strong spatial correlation between counterpropagating pulses.[10,11] In this paper we present a simple model that accounts for this peculiar spatial pattern. For simplicity we will use a semiclassical description of the emission process, consisting of the Bloch-Maxwell equations, where, however, no slowly varying envelope approximation is done. The relevance of this last point will be discussed later.

We also will limit our calculation to the early stage of the emission, because already in this regime the spatial correlation pattern appears. Initial and boundary conditions as well as the other system parameters are appropriate for the experiments of superfluorescence (SF) in $KCl:O_2^-$. Although we do not calculate the whole time-dependent emission, our model reproduces the experimental situation in which superfluoresence is observed and thus gives an appropriate description of superfluorescent emission. Let us briefly recall the experimental situation which is the following.

A laser pulse excites the O_2^- centers in a well-defined volume of the crystal. The local centers themselves have a complicated level structure.[1] With a laser pulse a high-lying level is excited which will decay without radiation into the ground state of the upper potential sheet.[1] During this process the system loses all information about phase correlations. This features is exhibited in the experiment, because there are no memory effects of the polarization of the exciting field that shows up in the emitted field immediately after the excitation. Now an avalanche process starts because each local center emits light in all directions, influencing all other centers. A correlated area builds up in the sample from which superfluoresent pulses are emitted in special directions that depend on the geometry of the sample.[1-11] For a rod-shaped excitation volume the emission happens in the form of counterpropagating pulses along the axis of the rod. At high impurity density counterpropagating pulses show a mode structure which is reminiscent of the spatial standing-wave mode spectrum of a laser. In order to understand this peculiar behavior one has to assume that the emitted fields are correlated and that the mode pattern originates in a kind of interference effect between left- and right- emitted fields.

II. THE MODEL

It is not possible to explain this behavior in terms of the available theories, in which the counterpropagating fields are assumed to be noncorrelated,[12-15] and a slowly varying envelope approximation (SVEA) is calculated for the field. We can indicate a reason for this discrepancy, which is based on the space scale involved in our problem.

We notice that the average distance d between the centers is much smaller than the wavelengths λ of the emitted electromagnetic field in which we are interested. In fact, in the experiments the concentration of the centers is 10^{18} cm^{-3}, i.e., the average distance between neighboring centers is 10 nm, whereas the optical modes have λ values of the order of 600 nm. Therefore, we expect to observe field variations on a scale that is much smaller than λ and for which the SVEA does not hold anymore. On the contrary, most SF experiments deal with systems having much lower center densities such that $d \gg \lambda$, and therefore are well described within the SVEA.

The space scale implied by our physical system indi-

cates that it can be adequately described in terms of a polarization field $P(\mathbf{r},t)$. Furthermore, we simulate the initiation of the emission process by a $P(\mathbf{r},t)$, which is built up by randomly oriented dipoles in order to simulate the effect of an incoherent initial state. We start with the relation

$$\mathbf{E}(t,\mathbf{r})=\frac{1}{c^2}\int\frac{1}{|\mathbf{r}-\mathbf{r}'|}\ddot{\mathbf{P}}(t-\Delta t,\mathbf{r}')d^3\mathbf{r}' , \tag{1}$$

which is an exact consequence of the Maxwell equation, and where $\Delta t=|r-r'|/c=|\Delta r|/c$ is the retardation and P a dipole density. The positive frequency part of the polarization obeys the equation

$$[\dot{\mathbf{P}}(t,\mathbf{r})]^{(+)}=i\omega[\mathbf{P}(t,\mathbf{r})]^{(+)}+ign\mathbf{E}(t,\mathbf{r}) . \tag{2}$$

Here n is the inversion and depends on $(\mathbf{r},t)$ and $\mathbf{E}$. g is the dipole coupling and ω the transition frequency. All damping terms have been neglected. Equation (2) is a consequence of the Bloch equations for a two-level system whose use is justified in this context as follows. We know from experiments and previous theoretical models that transitions from the ground state of the upper potential sheet to several vibronic levels of the lower sheet contribute to these effects.[1] However, we will confine our study to the case when just one specific transition is observed so that the local system can then be described as a two-level system. There is, however, no loss of generality, because an extension of the model to the general case bears no difficulties.

We assume that the distribution of the O_2^- centers is at random and that their concentration is low enough so that there is no direct interaction between the centers. We assume now that the system is highly excited so that n is positive (total inversion of the centers) and that it does not vary during the time in which the special pattern develops. This means that we limit our discussion to the early stage of the emission when the equation for the inversion density n in (2) can be linearized around its initial value n_0. As we will see in the simulation spatial patterns already build up in a time which is a multiple of $\lambda/c<L/c$ (photon lifetime in the sample). Equation (2) for the medium polarization then reads

$$\dot{\mathbf{P}}(t,\mathbf{r})=i\omega\mathbf{P}(t,\mathbf{r})+ign_0\mathbf{E}(t,\mathbf{r})=i\omega\mathbf{P}(t,\mathbf{r})+\beta\mathbf{E}(t,\mathbf{r}) , \tag{3}$$

where $\beta=ign_0$.

We justify this approximation, which is mainly consistent with the experiments, as follows. The left-right correlation is already present in the early stage of the emission process when the inversion does not yet strongly deviate from its initial value. Already weak modulations of the inversion in the $\lambda/2$ period are expected to enhance the behavior found in our treatment. We will discuss this point in more detail after our calculation method has been presented.

Finally, in the following, the polarization of the host crystal will be neglected because it is built up through scattering processes, the contribution of which are several orders of magnitude smaller than the resonant processes of the centers ($P_{\text{res}}/P_{\text{host}}\approx 10^{-8}$).

III. CALCULATION METHOD

We now briefly sketch the calculation method. From Eq. (3) we get the inhomogeneous solution

$$\mathbf{P}(t,\mathbf{r})=e^{i\omega t}\hat{\mathbf{P}}(t,\mathbf{r}) , \tag{4}$$

with

$$\hat{\mathbf{P}}=\int_0^t\beta\mathbf{E}e^{-i\omega t}dt=\int_0^t\dot{\hat{\mathbf{P}}}\,dt . \tag{5}$$

From (4) and (5) it is obvious to choose

$$\mathbf{E}=\hat{\mathbf{E}}e^{i\omega t} , \tag{6}$$

where $\hat{\mathbf{E}}$ and $\hat{\mathbf{P}}$ are functions slowly varying in time. We want to point out that in our problem this is not an approximation but a requirement caused by the structure of the differential equation (3). For the space dependence no assumption is made. For our numerical treatments we write

$$\hat{\mathbf{P}}(t+dt)=\hat{\mathbf{P}}(t)+\dot{\hat{\mathbf{P}}}\,dt=\hat{\mathbf{P}}(t)+\beta\hat{\mathbf{E}}\,dt+\cdots . \tag{7}$$

When choosing

$$dt\ll P(t)/\beta E , \tag{8}$$

we can neglect higher-order terms in dt. Combining Eqs. (1), (4), and (7) we get ($|k|=\omega/c$)

$$\hat{\mathbf{P}}(t+dt)+\hat{\mathbf{P}}(t)-\frac{\beta}{c^2}dt\int\frac{\omega^2\hat{\mathbf{P}}-2i\omega\dot{\hat{\mathbf{P}}}-\ddot{\hat{\mathbf{P}}}}{|\Delta r|}e^{ik|\Delta r|}d^2r' . \tag{9}$$

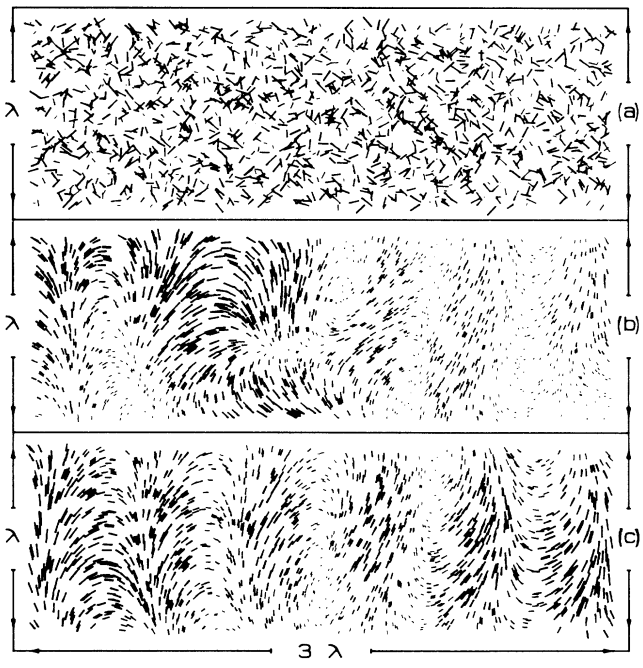

FIG. 1. The polarization field $\hat{\mathbf{P}}$ in an area of $\lambda\times 3\lambda$, where 1500 grid points are distributed randomly. The length and thickness of the lines correspond to the amplitudes, the direction to the phase of $\hat{\mathbf{P}}$. (a) Starting situation, (b) after two iteration steps with $\beta\,dt=1$, and (c) after 16 iteration steps.

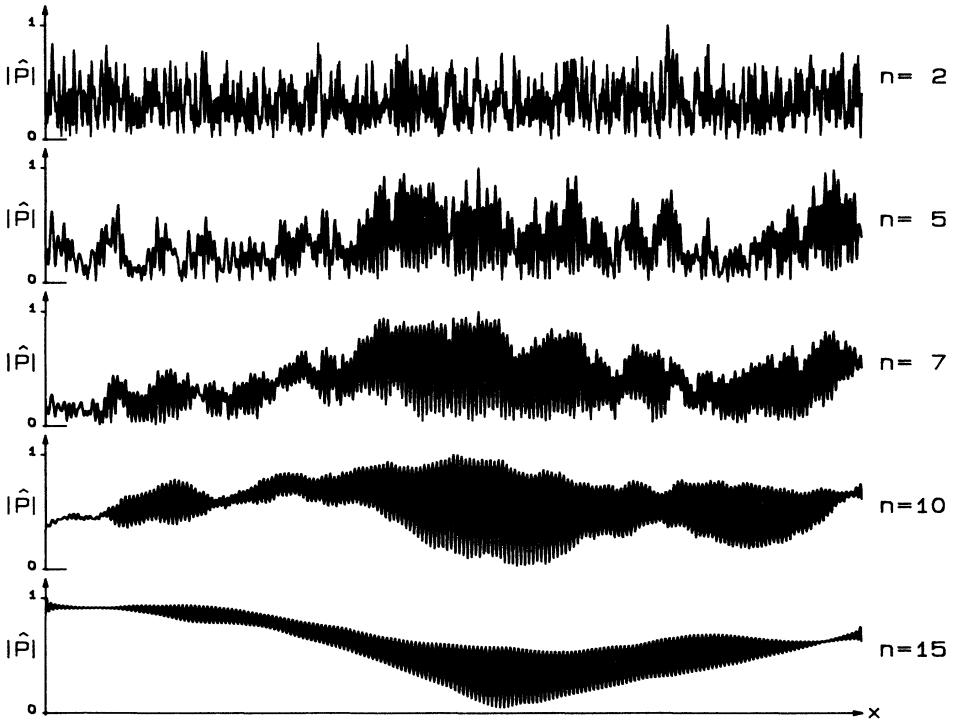

FIG. 2. The absolute of $\hat{\mathbf{P}}$ is drawn as a function of x for an array of the dimension of 100λ for a selected number n of iterations. In each case the absolute is normalized to $|\hat{\mathbf{P}}_{\max}| = 1$.

This equation can be solved by an iteration procedure. During the time of each calculation cycle we assume $\hat{\mathbf{P}}$ to be constant, which means we can put $\ddot{\hat{\mathbf{P}}}$ and $\dot{\hat{\mathbf{P}}}$ equal to zero. The value of the time step of each iteration has to obey relation (8). The electromagnetic field which results from each iteration step induces a polarization field which again acts as a source for the polarization buildup calculated in the next iteration cycle.

IV. RESULTS

When starting the iteration procedure we assume the polarization to consist of a random distribution of amplitudes and phases. The three-dimensional space integral is transcribed into a sum taken at up to 1500 grid points randomly distributed in a plane. The result of the iteration is seen in Fig. 1 for three different times. The iteration time step is of the order λ/c. The time scale on which the pattern formation occurs is therefore much shorter than all characteristic times of the emission processes. It is implied that outside the active volume the polarization is zero and the refraction index is constant. For this calculation, it is not necessary to separate left- and right-running waves, as is usually done. On the contrary, our calculations show that in general left- and right-running waves should be treated separately since the components are mixed through the retardation. After two iteration steps a near-field organization in phases and amplitudes shows up and results after 16 iterations in an organization in space with a period of $\lambda/2$. This clearly shows that the slowly varying envelope approximation in space is not allowed, because the polarization is modulated in the range of $\lambda/2$.

In a sequence of simulations it turned out that no

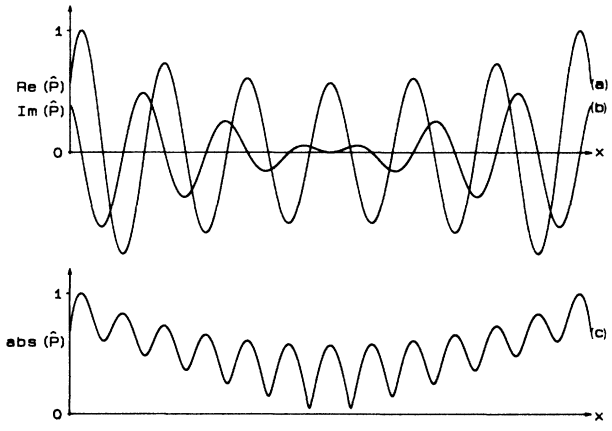

FIG. 3. The imaginary, real, and absolute of $\hat{\mathbf{P}}$ is drawn for a linear array of length 6.5λ. The figure describes self-stabilized polarization which appears in the limit $n \to \infty$. Numerically, this condition is satisfied for $n > 1000$.

spurious effects appear when going from a random grid to an equidistant grid. Therefore, in the following we will use equidistant grid points with a separation of $\lambda/10$ which ensures us that all information about the direction of the waves is held. We want to consider the starting process of the self organization in more detail. As an example which corresponds to one possible experimental setup we choose a pencil-shaped (quasi-one-dimensional) excitation volume. We still have to work with the three-dimensional Maxwell equations in order to account for field losses in transverse directions.

From Fig. 2 one already sees that after two iteration steps local interference structures for the polarization in small domains are evident. These domains grow increasingly with time. This is seen for the iteration $n=5$. For $n=10$ and 17 small interference structures appear at the left and right end of the one-dimensional area. This is due to the fact that at the end of the inversion area small reflections caused by the gain variation are induced, which, however, are not significant in the further time evolution of the system.

In Fig. 3 the real and imaginary parts of the polarization amplitude are shown separately for an organized linear array of length 6.5λ. It is clearly seen that in the central region of the array there is no phase shift between both parts, which implies that this is a standing-wave area. On the other hand, at both ends of the array a phase shift of $\pm 90°$ appears implying areas of either left- or right-running waves. These self-organized structures remind us of the distributed feedback (DFB) laser, where the experimental conditions force periodic inverted structures with strongly coupled waves.[16] There it could be shown that even an inversion modulation as small as 10^{-6} is enough to induce DFB lasers. The main effect will be Bragg reflection on the internal polarization modulation.

V. CONCLUSION

In conclusion, we want to emphasize the dominant role of the retardation term $\exp(-i\omega\Delta r/c)$ in the iteration equation (9). As one can verify by a simple calculation, the retardation part mixes the left- and right-running waves together in such a way that their contribution to the field at the point r cannot be separated. This indicates again that in a more thorough calculation the whole spatial dependence of the field must be taken into account and that the currently used slowly varying envelope approximation (SVEA) for the space dependence of E in the longitudinal direction is not adequate to describe the experimental results. To our knowledge this point has been already considered only in Ref. 16. A more comprehensive description of the time-dependent emission, including pulse duration and time delays, is also in progress. Calculations for two-dimensional arrays show preliminary results in agreement with two-dimensional experiments[10,11] and will be published elsewhere.

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