

## Coherent radiation from thin films

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The spontaneous coherent radiation from thin crystal films of  $n$  layers is studied. By treating each layer as a unit and by employing the method of multiple-time-scale expansion, exact solutions for such systems, with no restriction on the value  $\lambda_0/d$ , are found. Here  $\lambda_0$  is the characteristic radiation wavelength and  $d$  is the average interparticle spacing. In the limit  $\lambda_0/d \gg 1$ , it is seen that there exist correlated states of the film which are exact analog of Dicke's superradiant states. The decay rate for the superradiant states is roughly enhanced by a factor of  $n(\lambda_0/d)^2$  compared to the single-atom decay rate  $\gamma$ .

### I. INTRODUCTION

As is well known, the correlation of atoms arising from their coupling to the common radiation field gives rise to a host of coherence phenomenon, a brief survey of which has been given in Ref. 1. In particular, the spontaneous-radiation process was first treated in the pioneering work of Dicke<sup>2</sup> for an atomic gas system whose linear dimension  $L$  is much smaller than the spontaneous radiation wavelength  $\lambda_0$ . Ernst and Stehle<sup>3</sup> extended the treatment to a large ( $L \gg \lambda_0$ ) system of randomly distributed atoms which are all excited initially by a generalized Wigner-Weisskopf method. The so-called photon trapping or the internal scattering of photons, the prime source of radiative correlation, has been taken into account by them in an approximate way, by averaging over the random positions of the atoms. Therefore, their analysis would not be exactly valid for several-atom systems where the radiative correlations depend significantly on the precise configuration of the atomic positions, nor for atoms in crystal lattice, where the crystal symmetry is of primary importance. More recently, Rehler and Eberly<sup>4</sup> also treated a large system of atoms, not necessarily all excited initially. In their work, which is not specifically for atoms with crystal symmetry, several physical assumptions, some more basic than others, have been made. These assumptions, as discussed carefully by the authors, would not be valid in every circumstance. However, their results appear very reasonable on physical ground and are qualitatively supported by the present work, although the initial state as well as the geometrical considerations involved in our investigation are quite different. On the other hand, in the usual theory of x-ray diffraction,<sup>5</sup> while the crystal symmetry is accurately considered the interaction of radiation with atoms is only calculated by first-order perturbation theory, thereby leaving out the photon-trapping mechanism completely.

Recently, the spontaneous radiation<sup>6</sup> as well as the resonance scattering<sup>7</sup> from systems of a few particles and from systems of atoms in lattice arrangements were analyzed. With the restriction to intermediate states of one quantum of excitation, within the resonance approximation, exact solutions to the coupled set of the resultant partial differential equations in the limit of large times have been found in these treatments by applying the multiple-time-scale perturbation theory<sup>1</sup> (MTSPT). The results are valid for arbitrary values of  $d/\lambda_0$ ,  $d$  being the average interparticle spacing. In the case of lattice, however, only the bulk effect on the radiation was obtained, owing to the use of the periodic boundary conditions, which leads to a considerable amount of trapping of photons in the system.<sup>8</sup> For the observation of coherent radiation that can leave the system, it is clearly important to study the boundary effect. A monolayer of atoms or a thin film of several layers constitutes the most suitable system for the investigation of the boundary effect, especially in view of the great experimental successes in making such ordered structures in recent years.<sup>9-11</sup> In this investigation, the spontaneous radiation from such thin films, in which the atoms are assumed tightly bound to the lattice sites, is calculated, again within the same resonance approximation and the same framework as used previously.<sup>4,6,7</sup> Both the multiple internal photon scattering mechanism and the crystal symmetry in two dimensions are accurately taken into account. The calculation of the boundary effect consists in treating the third dimension (the thickness) of the film exactly. Experimentally, the coherence effects on radiation,<sup>12</sup> such as the appearance of superradiant states, have been observed for gases<sup>13</sup> and crystal films.<sup>14</sup>

### II. FORMULATION

Consider a system of  $N$  two-level atoms, with  $\omega_0$  (we take  $\hbar = 1$ ) representing the spacing between

the two nondegenerate levels. It is assumed that initially, i.e., at  $t=0$ , only one unit of excitation energy  $\omega_0$  exists among the atoms. This initial condition can be justified if the number of excited atoms is small compared to  $N$  so that the correlations between excited atoms can be neglected.

Following Refs. 1 and 6 closely, we can write, within the resonance approximation,<sup>2, 15</sup> the state of the system at any time  $t > 0$  as

$$|\Psi(t)\rangle = \sum_{j=1}^N b_j(t) |j; 0_{\vec{k}}\rangle + \sum_{\vec{k}} b_{\vec{k}}(t) |0; 1_{\vec{k}}\rangle, \quad (1)$$

where  $|j; 0_{\vec{k}}\rangle$  is the state in which the  $j$ th atom is excited with no photon present, and  $|0; 1_{\vec{k}}\rangle$  represents the state in which all atoms are unexcited with the  $\vec{k}$  photon present. The Schrödinger equation can then be reduced to the following equations for the amplitudes:

$$i \frac{d}{dt} b_j(t) = \sum_{\vec{k}} \mathcal{G}_{\vec{k}}^* e^{i\omega_0 k t} e^{i\vec{k} \cdot \vec{x}_j} b_{\vec{k}}(t), \quad (2)$$

$$i \frac{d}{dt} b_{\vec{k}}(t) = \mathcal{G}_{\vec{k}} e^{-i\omega_0 k t} \sum_{j=1}^N e^{-i\vec{k} \cdot \vec{x}_j} b_j(t), \quad (3)$$

where

$$\omega_{0k} \equiv \omega_0 - \omega_k \equiv \omega_0 - ck.$$

Here  $\mathcal{G}_{\vec{k}}$  is essentially the dipole matrix element for the transition between the two levels of the atom. The vector  $\vec{x}_j$  denotes the position of the  $j$ th atom.

By the use of the MTSPT, the oscillations of the amplitudes in the fast time scale of  $1/\omega_0$  can first be averaged out. The remaining time dependence of the amplitudes, which vary in a much slower time scale of  $1/\gamma_s$ ,  $\gamma_s$  being the natural linewidth for an isolated atom, can then be shown<sup>4, 6</sup> to be governed by

$$\frac{d}{dt} b_i(t) = - \sum_{j=1}^N f_{ij} b_j(t), \quad (4)$$

where

$$f_{ij} \equiv i \sum_{\vec{k}} |\mathcal{G}_{\vec{k}}|^2 e^{i\vec{k} \cdot \vec{x}_{ij}} \zeta(\omega_{0k}) \equiv i\omega_{ij} + \gamma_{ij}, \quad (5)$$

$$\vec{x}_{ij} \equiv \vec{x}_i - \vec{x}_j, \quad \zeta(\omega) = \mathcal{P}(1/\omega) - i\pi\delta(\omega).$$

The symbol  $\mathcal{P}$  means "taking the principal value."

It is observed from Eqs. (4) and (5) that, in the slow time scale of  $1/\gamma_s$ , the excitation energy of the system is transferred from one atom to another, with a probability amplitude proportional to  $\omega_{ij}$ , while part of the energy leaks away with a rate proportional to  $\gamma_{ij}$  during the transfer from atom  $i$  to atom  $j$ .

For convenience, yet without loss of generality, let us assume the basic lattice vectors to be parallel to the  $x$ ,  $y$ ,  $z$  axes, with their lengths denoted by  $a_1$ ,  $a_2$ ,  $a_3$ , respectively. The position of the  $j$ th atom of the  $\alpha$ th layer will be denoted by

$$\vec{x}_{\alpha j} = (\alpha - 1)a_3 \hat{z} + \vec{R}_j, \quad \alpha = 1, 2, \dots, \quad (6)$$

where  $\vec{R}_j$  denotes the position of the atom  $j$  as projected onto the  $x$ - $y$  plane, the surface of the crystalline film. Correspondingly, the probability amplitude of the atom  $\vec{x}_{\alpha j}$  being excited will be denoted by  $b_{\alpha j}$ . Equation (4) then becomes

$$\frac{d}{dt} b_{\alpha i}(t) = -i \sum_{\beta} \sum_j f_{\alpha i, \beta j} b_{\beta j}(t), \quad (7)$$

where

$$f_{\alpha i, \beta j} \equiv i \sum_{k_z} \sum_{\vec{k}} |\mathcal{G}_{\vec{k}}|^2 e^{i\vec{k} \cdot \vec{R}_{ij}} e^{ik_z(\alpha - \beta)} \zeta(\omega_0 - \omega_k), \quad (8)$$

$$\vec{k} = \hat{z}k_z + \vec{k}, \quad \vec{R}_{ij} = \vec{R}_i - \vec{R}_j.$$

To solve Eq. (7), let us try a solution of the form

$$b_{\alpha i}(t) \sim B_{\alpha}(\vec{p}, t) e^{i\vec{p} \cdot \vec{R}_i}, \quad (9)$$

where the two-dimensional vectors  $\vec{p}$  are determined by the periodic boundary conditions in the  $x$  and  $y$  directions,

$$\vec{p} = 2\pi \frac{n_1}{N_1} \frac{1}{a_1} \hat{x} + 2\pi \frac{n_2}{N_2} \frac{1}{a_2} \hat{y}, \quad (10)$$

where

$$n_1 = 0, \pm 1, \pm 2, \dots, \pm(\frac{1}{2}N_1 - 1), +\frac{1}{2}N_1,$$

$$n_2 = 0, \pm 1, \pm 2, \dots, \pm(\frac{1}{2}N_2 - 1), +\frac{1}{2}N_2,$$

and  $N_1$ ,  $N_2$  are the number of unit cells in the  $x$ ,  $y$  directions. Substituting Eq. (9) into (7), we obtain

$$\frac{d}{dt} \hat{B}_{\alpha}(\vec{p}, t) = - \sum_{\beta} F_{\alpha\beta}(\vec{p}) B_{\beta}(\vec{p}, t), \quad (11)$$

where

$$\begin{aligned} F_{\alpha\beta}(\vec{p}) &= i \sum_{k_z} \left( \sum_{\vec{k}} \sum_j |\mathcal{G}_{\vec{k}}|^2 e^{i(\vec{k} - \vec{p}) \cdot \vec{R}_{ij}} \zeta(\omega_0 - \omega_k) \right) \\ &\quad \times e^{ik_z(\alpha - \beta)a_3} \\ &= i\Omega_{\alpha\beta}(\vec{p}) + \Gamma_{\alpha\beta}(\vec{p}), \end{aligned} \quad (12)$$

$$\begin{aligned} \Gamma_{\alpha\beta}(\vec{p}) &= \pi \sum_{k_z} \left( \sum_{\vec{k}} \sum_j |\mathcal{G}_{\vec{k}}|^2 e^{i(\vec{k} - \vec{p}) \cdot \vec{R}_{ij}} \delta(\omega_0 - \omega_k) \right) \\ &\quad \times e^{ik_z(\alpha - \beta)a_3} \\ &= \Gamma_{\beta\alpha}(\vec{p}). \end{aligned} \quad (13)$$

Note that  $F_{\alpha\beta}(\vec{p})$  is independent of  $\vec{R}_i$ , owing to the crystal symmetry. The interpretation of Eq. (11) is clear. It describes the transfer of excitation energy of the system from the layer  $\alpha$  to the other

layers  $\beta$ , with a leakage rate of the energy proportional to  $\Gamma_{\alpha\beta}$  during the process of the transfer. When it interacts with other layers, each layer of atoms essentially acts as a unit, represented effectively by the terms in large parentheses in Eqs. (12) and (13).

The explicit evaluation of  $\Gamma_{\alpha\beta}(\vec{p})$  of Eq. (13) is clumsy but straightforward. The result can be expressed in terms of  $\gamma_s$  as

$$\Gamma_{\alpha\beta}(\vec{p}) = \frac{3\pi}{2k_0^3} \frac{N}{A} \gamma_s \times \sum_{\vec{g}} \left( \frac{[k_0^2 - (\vec{p} + \vec{g})^2] |\hat{e} \times \hat{z}|^2 + |\hat{e} \times (\vec{p} + \vec{g})|^2}{[k_0^2 - (\vec{p} + \vec{g})^2]^{1/2}} \right) \times \cos \{ [k_0^2 - (\vec{p} + \vec{g})^2]^{1/2} (\alpha - \beta) a_3 \}, \quad (14)$$

where  $N/A$  is the number of atoms per unit area of the crystalline film,  $\hat{e}$  is a unit vector in the direction of the electric dipole responsible for the transition between the two levels of the atom separated in energy by  $\omega_0 = ck_0$ . The sum in Eq. (14) is restricted to those reciprocal lattice vectors  $\vec{g}$  for which  $k_0^2 - (\vec{p} + \vec{g})^2 \geq 0$ , as shown in Fig. 1.

### III. LIFETIMES, RADIATION INTENSITIES, AND SUPERRADIANT STATES

#### A. Monolayered film

For a monolayered crystal,  $\alpha = \beta = 1$  and Eq. (14) reduces to

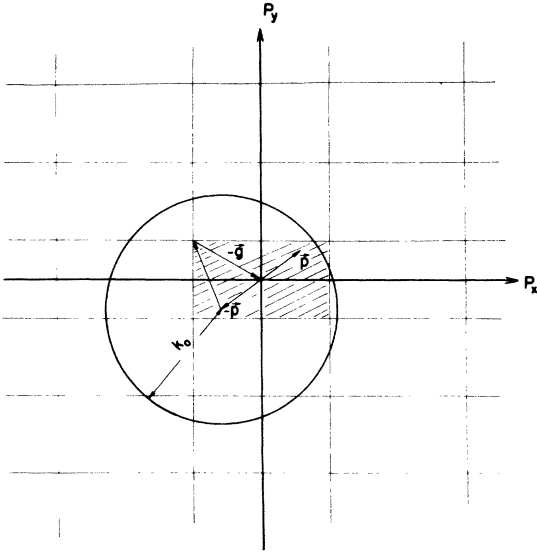


FIG. 1. Only those  $\vec{g}$  vectors or the grid points within the circle centered at  $-\vec{p}$  and with a radius  $k_0$  will contribute to the sum in Eq. (14). The shaded area represents the first Brillouin zone of Eq. (10).

$$\Gamma_s(\vec{p}) \equiv \Gamma_{11}(\vec{p})$$

$$= \frac{3\pi}{2k_0^3} \frac{N}{A} \gamma_s \sum_{\vec{g}} \frac{[k_0^2 - (\vec{p} + \vec{g})^2] |\hat{e} \times \hat{z}|^2 + |\hat{e} \times (\vec{p} + \vec{g})|^2}{[k_0^2 - (\vec{p} + \vec{g})^2]^{1/2}}. \quad (15)$$

To further examine  $\Gamma_s(\vec{p})$  we look into two cases: (a) when  $\lambda_0 = 2\pi/k_0 \ll d$ , and (b) when  $L \gg \lambda_0 \gg d$ . In case (a), it follows more directly from Eq. (13) that

$$\Gamma_s(\vec{p}) = \pi \sum_{\vec{k}} |\mathcal{G}_{\vec{k}}|^2 \delta(\omega_0 - \omega_{\vec{k}}) \equiv \gamma_s$$

by noting that only the  $j=i$  term in the sum over  $j$  will contribute, the other terms being averaged to zero upon summing over  $\vec{k}$ . In case (b), we can easily show that  $\Gamma_s(\vec{p})$  is either roughly equal to  $\gamma_s(\lambda_0/d)^2$  when  $\vec{p}$  is such that there is one  $\vec{g}$  vector in the restricted sum over  $\vec{g}$ , or equal to zero when no  $\vec{g}$  vector satisfies the restriction imposed on that sum. The  $\vec{p}$  vectors for which  $\Gamma_s(\vec{p}) \neq 0$  are just those within the  $k_0$  neighborhood of the four corners of the first Brillouin zone (see Fig. 2). The number of those  $\vec{p}$ 's is  $\pi k_0^2 / (2\pi/L) \sim k_0^2 L^2$ , which is much greater than one, but much smaller than the number of atoms in the layer.

The solution to Eq. (11) is then given by  $B_\alpha(\vec{p}, t) \sim e^{-i\Omega_s(\vec{p})t} e^{-\Gamma_s(\vec{p})t}$ , with  $\Omega_s(\vec{p}) \equiv \Omega_{11}(\vec{p})$ , and the solution to Eq. (7) is given by

$$b_{\alpha i}(t) = \sum_{\vec{p}'} B_\alpha(\vec{p}') e^{-i\Omega_s(\vec{p}')t} e^{-\Gamma_s(\vec{p}')t} e^{i\vec{p}' \cdot \vec{R}_i}, \quad (16)$$

where the coefficients  $B_\alpha(\vec{p}')$  are determined from

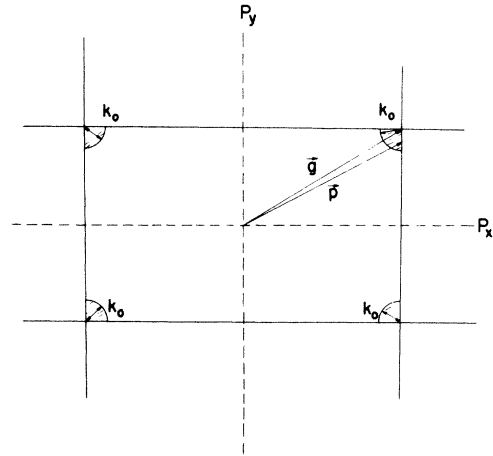


FIG. 2.  $\vec{p}$  vectors in the first Brillouin zone for which  $\Gamma_s(\vec{p}) \neq 0$  are those within the  $k_0$  neighborhood of the four corners of the first zone. The shaded areas indicate the  $k_0$  neighborhoods.

the initial conditions. If we assume that  $B_\alpha(\vec{p}') = (1/\sqrt{N})\delta_{\vec{p}, \vec{p}'}$ , i.e., only a definite  $\vec{p}$  mode is excited initially, then it follows from Eq. (16) that in subsequent times, only the same  $\vec{p}$  mode and no other  $\vec{p}' \neq \vec{p}$  modes will be involved. It is also seen that this mode will, in case (b), either decay exponentially with a coherently enhanced rate  $\sim \gamma_s(\lambda_0/d)^2$ , or not at all (i.e., the excitation is trapped indefinitely in the system), depending on what  $\vec{p}$  is. This result agrees qualitatively with that of Rehler and Eberly,<sup>4</sup> showing that the radiative decay half-life of a large many-atom system is much shorter than the natural life time of a single atom, but not so short as the half-life of a small atomic system with the same number of atoms. Those  $\vec{p}$  modes with the enhanced decay rates are the exact analogue of Dicke's superradiant states<sup>2</sup> in a crystallized thin film. This result also confirms a previous qualitative assertion<sup>16</sup> that a large system may be considered as a collection of small subsystems, each of which consisting of particles within a half-wavelength or so of each other, participates coherently in the radiation process.

The amplitude  $b_{\vec{k}}(t)$  can be obtained by substituting Eq. (16) into (3) and integrating with respect to  $t$ . Thus, with  $\alpha=1$  for the monolayer and  $\vec{k} = \hat{z}k_z + \vec{k}$ , we get

$$b_{\vec{k}}(t) = \mathcal{G}_{\vec{k}} \sum_{\vec{p}'} B_\alpha(\vec{p}') \times \left( \frac{\exp\{-i[\omega_{0\vec{k}} + \Omega_s(\vec{p}')]t - \Gamma_s(\vec{p}')t\} - 1}{\omega_{0\vec{k}} + \Omega_s(\vec{p}') - i\Gamma_s(\vec{p}')} \times \sum_j e^{-i(\vec{k} - \vec{p}') \cdot \vec{R}_j} \right). \quad (17)$$

Making use of

$$\sum_j e^{-i(\vec{k} - \vec{p}') \cdot \vec{R}_j} = N_1 N_2 \delta_{\vec{k} + \vec{g}, \vec{p}'}, \quad (18)$$

and denoting the vector  $\vec{k}$  as reduced to the first Brillouin zone by  $\vec{k}_r$ , we obtain

$$b_{\vec{k}}(t) = N_1 N_2 \mathcal{G}_{\vec{k}} B_1(\vec{k}_r) \times \frac{\exp\{-i[\omega_{0\vec{k}} + \Omega_s(\vec{k}) - i\Gamma_s(\vec{k})]t\} - 1}{\omega_{0\vec{k}} + \Omega_s(\vec{k}) - i\Gamma_s(\vec{k})}, \quad (19)$$

where  $B_1(\vec{k}_r)$  depends on the initial condition. The radiation intensity distribution is then given by  $\lim_{t \rightarrow \infty} |b_{\vec{k}}(t)|^2$ . If the system starts in a certain superradiant  $\vec{p}$  state, the radiation will then only consist of photons with  $\vec{k}_r = \vec{p}$ , and

$$\lim_{t \rightarrow \infty} |b_{\vec{k}}(t)|^2 = \frac{N_1 N_2 |\mathcal{G}_{\vec{k}}|^2 \delta_{\vec{k}_r, \vec{p}}}{[\omega_{0\vec{k}} + \Omega_s(\vec{p})]^2 + \Gamma_s^2(\vec{p})}. \quad (20)$$

We can check the normalization that

$$\lim_{t \rightarrow \infty} \sum_{\vec{k}} |b_{\vec{k}}(t)|^2 = \sum_{\vec{k}} \left( \frac{|\mathcal{G}_{\vec{k}}|^2}{[\omega_{0\vec{k}} + \Omega_s(\vec{p})]^2 + \Gamma_s^2(\vec{p})} \sum_j e^{i(\vec{k} - \vec{p}) \cdot \vec{R}_j} \right) = \frac{\pi}{\Gamma_s(\vec{p})} \sum_j \sum_{\vec{k}} |\mathcal{G}_{\vec{k}}|^2 \delta(\omega_{0\vec{k}}) e^{i(\vec{k} - \vec{p}) \cdot \vec{R}_j} = 1,$$

where we have used Eq. (18) and the  $\delta$ -function representation  $\delta(x) = (1/\pi) \lim_{\epsilon \rightarrow 0} [\epsilon/(x^2 + \epsilon^2)]$ , as well as Eq. (13).

### B. Films of two layers

Here  $\alpha=1, 2$ ;  $\beta=1, 2$ . The solution to Eq. (11) is of the form

$$B_\alpha(\vec{p}, t) \sim e^{F_\pm(\vec{p})t}, \quad (21)$$

where

$$F_\pm(\vec{p}) = -i[\Omega_s(\vec{p}) \pm \Omega_{12}(\vec{p})] - [\Gamma_s(\vec{p}) \pm \Gamma_{12}(\vec{p})].$$

The solution to Eq. (7) is

$$b_{\alpha j}(t) = \sum_{\vec{p}} [B_{\alpha+}(\vec{p}) e^{F_+(\vec{p})t} + B_{\alpha-}(\vec{p}) e^{F_-(\vec{p})t}] e^{i\vec{p} \cdot \vec{R}_j}, \quad (22)$$

where the coefficients  $B_{\alpha\pm}$  are to be determined from the initial conditions. We see that for each  $\vec{p}$ , there are now two modes with complex frequencies  $F_+(\vec{p})$  and  $F_-(\vec{p})$ , corresponding to the existence of two layers of atoms.

In the limit of  $k_0 a_3 \ll 1$  we see from Eqs. (14) and (15) that  $\Gamma_{12}(\vec{p}) \approx \Gamma_s(\vec{p})$ . In this limit,  $\Gamma_-(\vec{p}) \approx 0$  and

the excitation energy in this mode will be trapped.<sup>8</sup> On the other hand,  $\Gamma_+(\vec{p}) \approx 2\Gamma_s(\vec{p})$ , implying that the two layers in the  $F_+(\vec{p})$  mode will radiate coherently, leading to a doubly enhanced decay rate for this superradiant mode, provided  $\Gamma_s(\vec{p}) \neq 0$ .

### C. Films of $n$ layers

Equation (11) can again be solved by trial solutions of the form  $B_\alpha(\vec{p}, t) \sim e^{F(\vec{p})t}$ . The complex frequencies  $F(\vec{p})$  are just the roots of an  $n \times n$  determinantal equation  $|F_{\alpha\beta}(\vec{p}) - F(\vec{p})\delta_{\alpha\beta}| = 0$ . In general, there will be  $n$  complex-frequency modes corresponding to each  $\vec{p}$ . In the interesting limiting case that the thickness of the film is much smaller than  $\lambda_0$ , there will again be, for each  $\vec{p}$  with  $\Gamma_s(\vec{p}) \neq 0$ , a superradiant mode which decays with a rate approximately equal to  $n \cdot \Gamma_s(\vec{p})$ , while the other modes with the same  $\vec{p}$  lead to energy trapping. In the case of  $\lambda_0 \sim a_3$ , the values of the complex frequencies and the associated radiation

spectrum will depend on the details of the crystal structure of the film as well as on the two levels of the atom, as is already evident from Eq. (15) for a monolayered film. It is also conceivable that one might learn about the crystal structure by observing the coherent radiation.

#### IV. DISCUSSIONS AND CONCLUSIONS

In the calculations presented in Secs. II and III, the atoms are assumed to be stationary. It would not be difficult to include the effect<sup>17</sup> of phonons. More significantly we have assumed that the atomic levels remain intact when the atoms form a lattice. This is just the tight-binding approximation, valid only for the low-lying levels of the atomic

core.

It was pointed out previously<sup>6</sup> that a relatively large fraction of excitation energy will generally be trapped<sup>8</sup> and only a small fraction can escape as spontaneous radiation from the bulk of a crystal. The consideration of the boundary effect,<sup>18</sup> which is prominent in a thin film geometry, changes greatly the ratio of the trapped energy to the radiated energy, which is of course important for the observation of such radiation. The superradiant state may actually be produced by passing a plane wave pulse through the system.<sup>2,4</sup> The effect of an incident photon beam with frequencies centered around  $\omega_0$  is currently under investigation.

<sup>1</sup>P. S. Lee, Y. C. Lee, and C. T. Chang, *Phys. Rev. A* **8**, 1722 (1973).

<sup>2</sup>R. H. Dicke, *Phys. Rev.* **93**, 99 (1954).

<sup>3</sup>V. Ernst, and P. Stehle, *Phys. Rev.* **176**, 1456 (1968); V. Ernst, *Z. Phys.* **229**, 432 (1969).

<sup>4</sup>N. Rehler and J. Eberly, *Phys. Rev. A* **3**, 1735 (1971).

<sup>5</sup>See, for example, R. E. Peierls, *Quantum Theory of Solids* (Clarendon, Oxford, 1955).

<sup>6</sup>P. S. Lee and Y. C. Lee, *Phys. Rev. A* **8**, 1727 (1973).

<sup>7</sup>C. T. Chang, Ph. D. dissertation (State University of New York at Buffalo, 1973) (unpublished).

<sup>8</sup>See also, C. Stroud, J. Eberly, W. Lama, and L. Mandel, *Phys. Rev. A* **5**, 1094 (1972). In this work, it is also found that a small system of atoms cannot radiate all its energy coherently; some of it remains trapped.

<sup>9</sup>J. J. Lander and J. A. Morrison, *Surf. Sci. (Neth.)* **4**, 103 (1966).

<sup>10</sup>J. J. Lander and J. A. Morrison, *Surf. Sci. (Neth.)* **5**, 163 (1966).

<sup>11</sup>J. J. Lander and J. A. Morrison, *Surf. Sci. (Neth.)* **6**, 1 (1967).

<sup>12</sup>H. G. Kuhn and J. M. Vaughan, *Proc. R. Soc. A* **277**, 297 (1964); H. G. Kuhn, E. L. Lewis, and J. M. Vaughan, *Phys. Rev. Lett.* **15**, 687 (1965).

<sup>13</sup>N. Skribanowitz, I. P. Herman, J. C. MacGillivray, and M. S. Feld, *Phys. Rev. Lett.* **30**, 309 (1973), and references contained therein.

<sup>14</sup>F. Varsanyi, *Appl. Phys. Lett.* **19**, 169 (1971).

<sup>15</sup>Y. C. Lee and D. L. Lin, *Phys. Rev. A* **6**, 388 (1972).

<sup>16</sup>Y. C. Lee and D. L. Lin, *Phys. Rev.* **183**, 147 (1969).

<sup>17</sup>For the effect of random motion, see Y. C. Lee, and D. L. Lin, *Phys. Rev.* **183**, 150 (1969); also P. S. Lee and Y. C. Lee (unpublished).

<sup>18</sup>The effect of the geometry of the system on the pattern of the radiation has been considered in Refs. 3 and 4, which show a strongly directional characteristic of the radiation.