

Interaction of light with an atom near the surface of a superlattice. II. Quasiperiodic case

Xiao-shen Li

*Center of Theoretical Physics, Chinese Center of Advanced Science and Technology (World Laboratory),
52 Sanlihe Road, P.O. Box 8730, Beijing 100 080, People's Republic of China
and Shanghai Institute of Metallurgy, Academia Sinica, 865 Chang Ning Road,
Shanghai 200 050, People's Republic of China**

Chang-de Gong

*Center of Theoretical Physics, Chinese Center of Advanced Science and Technology (World Laboratory),
52 Sanlihe Road, P.O. Box 8730, Beijing 100 080, People's Republic of China
and Department of Physics, Nanjing University, Nanjing, Jiangsu, People's Republic of China*

(Received 8 November 1988)

Spontaneous-emission properties and resonance fluorescence for a two-level adatom near the surface of a semi-infinite superlattice with quasiperiodic structure are studied by use of surface-dressed optical Bloch equations and Maxwell's equations. Effects of dielectric properties and the geometric structure of this superlattice on spontaneous-decay rate and frequency shift, resonance-fluorescence spectrum, time evolution of adatomic inversion, and fluctuations of the dipole moment for this adatom are discussed and some novel phenomena are discovered.

I. INTRODUCTION

In a recent paper¹ we obtained a set of new surface-dressed optical Bloch equations (SBE) by using Dekker's quantization procedure for dissipation systems^{2,3} and the reservoir theory⁴ and discussed resonance fluorescence of an adatom adsorbed at a surface of an ideal conducting metal or a dielectric with its transverse frequency coinciding with that of the adatomic transition. These SBE can be used to deal with the interactions of adatoms with light field. Very recently, we applied the above theory to luminescence and resonance fluorescence for an adatom adsorbed near a surface of a small metal particle⁵ or a multilayer thin film⁶ or a semi-infinite periodic superlattice.⁷ Recently, there has been also considerable interest in optical properties of adatoms and interactions of light with adatoms.⁸⁻¹¹ In a recent publication,¹⁰ Arnoldus and George discussed surface-enhanced correlations between polarized photons undergoing resonance fluorescence in the case of an atom near an ideal conducting metal surface. In another recent article¹¹ they studied spontaneous decay and adatomic fluorescence near a metal or an adsorbing dielectric.

In this paper we apply the theory described in Ref. 1 to the spontaneous-emission properties and resonance fluorescence of an adatom near a semi-infinite superlattice with quasiperiodic structure. The spectral properties of one-dimensional quasiperiodic Schrödinger operators¹² have attracted more interest^{13,14} since the discovery of quasicrystals.¹⁵ In Ref. 13 Lu *et al.* studied the properties of general one-dimensional quasilattices which can be generated from a finite set of basic cells by a generalized induction procedure and showed that quasiperiodicity is not necessarily related to irrationality of some physical parameters. Here we consider that the semi-infinite multilayer substrate having two constituents is uniform in the

X and *Y* directions but in the *Z* direction the layers are arranged according to the generalized induction procedure¹³ to form a semi-infinite quasiperiodic superlattice. The adatom is taken as an emitting dipole from which the electromagnetic field is emitted. This field is reflected by the surface and interfaces of the superlattice and coupled back with the dipole of which the dynamic behavior is thus influenced. We are interested in the effects of dielectric properties and quasiperiodic structures on the optical properties of the adatom. In what follows we use Maxwell's equations to calculate the reflected electric field for the quasiperiodic substrate. The reflected field at the dipole position is inserted into the SBE to find the luminescence and resonance fluorescence properties of this adatom.

II. BASIC EQUATIONS FOR THE ADATOM

In this section we calculate the surface-reflected field which can be inserted into the SBE (Ref. 1) to be solved to discuss optical properties of the adatom near a quasiperiodic superlattice surface. First, we may consider a general case, as shown in Fig. 1, there is an atom as an emitting dipole located at $\mathbf{r}=\mathbf{r}_0$ in the region $0 \leq z \leq -\infty$ containing a medium with dielectric constant ϵ near a surface of a solid composed of *N* layers.

To find the reflected electric field at the dipole position we use Maxwell's equations

$$\begin{aligned} \nabla \times \mathbf{E} &= -\frac{1}{c} \frac{\partial}{\partial t} \mathbf{B}, \\ \nabla \times \mathbf{B} &= \mathbf{0}, \\ \nabla \times \mathbf{H} &= \frac{1}{c} \frac{\partial}{\partial t} (\mathbf{D} + 4\pi \mathcal{P}), \\ \nabla \cdot (\mathbf{D} + 4\pi \mathcal{P}) &= 0, \end{aligned} \tag{2.1}$$

and the expression for the dipole moment

$$\mathcal{P}(\mathbf{r}, \omega) = \mathbf{p}(\omega) \delta(\mathbf{r} - \mathbf{r}_0), \tag{2.2}$$

and obtain the following equations for the field:^{1,7,16}

$$\nabla^2 \mathbf{E} + \epsilon k_0^2 \mathbf{E} = -4\pi [k_0^2 \mathcal{P} + \epsilon^{-1} \nabla(\nabla \cdot \mathcal{P})], \tag{2.3}$$

$$\mathbf{H} = \nabla \times \mathbf{E} / ik_0 \quad \text{for } -\infty < z < 0;$$

$$\nabla^2 \mathbf{E}_1 + \epsilon_1 k_0^2 \mathbf{E}_1 = 0, \quad \nabla \cdot \mathbf{E}_1 = 0, \tag{2.4}$$

$$\mathbf{H}_1 = \nabla \times \mathbf{E}_1 / ik_0 \quad \text{for } 0 \leq z \leq d_1;$$

$$\nabla^2 \mathbf{E}_2 + \epsilon_2 k_0^2 \mathbf{E}_2 = 0, \quad \nabla \cdot \mathbf{E}_2 = 0, \tag{2.5}$$

$$\mathbf{H}_2 = \nabla \times \mathbf{E}_2 / ik_0 \quad \text{for } d_1 \leq z \leq d_1 + d_2;$$

$$\nabla^2 \mathbf{E}_m + \epsilon_m k_0^2 \mathbf{E}_m = 0, \quad \nabla \cdot \mathbf{E}_m = 0,$$

$$\mathbf{H}_m = \nabla \times \mathbf{E}_m / ik_0 \quad \text{for } \sum_{i=1}^{m-1} d_i \leq z \leq \sum_{i=1}^m d_i \text{ and } 2 \leq m \leq N; \tag{2.6}$$

$$\nabla^2 \mathbf{E}_s + \epsilon_s k_0^2 \mathbf{E}_s = 0, \quad \nabla \cdot \mathbf{E}_s = 0,$$

$$\mathbf{H}_s = \nabla \times \mathbf{E}_s / ik_0 \quad \text{for } \sum_{i=1}^N d_i \leq z < \infty. \tag{2.7}$$

The solutions for the electric field of the above equations can be written as^{1,7,16}

$$\begin{aligned} \mathbf{E}(\mathbf{r}, \omega) &= \int \int \exp(i\mathbf{k}' \cdot \mathbf{r}) \mathcal{E}(u, v, \omega) du dv + \mathbf{E}_p(\mathbf{r}, \omega), \\ \mathbf{k}' \cdot \mathcal{E} &= 0, \quad \mathbf{k}' = (k_{\parallel}, -w), \quad w^2 = \epsilon k_0^2 - k_{\parallel}^2 \\ &\quad \text{for } \text{Im} w \geq 0 \text{ and } z < 0, \end{aligned} \tag{2.8}$$

where $k_{\parallel}^2 = u^2 + v^2$, the subscript \parallel means parallel to the interfaces, and

$$\begin{aligned} \mathbf{E}_p(\mathbf{r}, \omega) &= -(2\pi i)^{-1} \int \int \omega^{-1} [k_0^2 \mathbf{p} + \epsilon^{-1} \nabla(\nabla \cdot \mathbf{p})] \\ &\quad \times \exp[iu(x - x_0) + iv(y - y_0) \\ &\quad + iw|z - z_0|] du dv; \end{aligned} \tag{2.9}$$

$$\begin{aligned} \mathbf{E}_1(\mathbf{r}, \omega) &= \int \int [\mathcal{E}_1^{(+)}(u, v, \omega) \exp(i\mathbf{k}_1 \cdot \mathbf{r}) \\ &\quad + \mathcal{E}_1^{(-)}(u, v, \omega) \exp(i\mathbf{k}'_1 \cdot \mathbf{r})] du dv, \\ \mathbf{k}_1 \cdot \mathcal{E}_1^{(+)} &= 0, \quad \mathbf{k}'_1 \cdot \mathcal{E}_1^{(-)} = 0, \quad \mathbf{k}_1 = (k_{\parallel}, w_1), \\ \mathbf{k}'_1 &= (k_{\parallel}, -w_1), \quad w_1^2 = \epsilon_1 k_0^2 - k_{\parallel}^2, \quad \mathbf{k}_{\parallel} = (u, v, 0) \\ &\quad \text{for } 0 \leq z \leq d_1; \end{aligned} \tag{2.10}$$

$$\begin{aligned} \mathbf{E}_2(\mathbf{r}, \omega) &= \int \int [\mathcal{E}_2^{(+)}(u, v, \omega) \exp(i\mathbf{k}_2 \cdot \mathbf{r}) \\ &\quad + \mathcal{E}_2^{(-)}(u, v, \omega) \exp(i\mathbf{k}'_2 \cdot \mathbf{r})] du dv, \\ \mathbf{k}_2 \cdot \mathcal{E}_2^{(+)} &= 0, \quad \mathbf{k}'_2 \cdot \mathcal{E}_2^{(-)} = 0, \quad \mathbf{k}_2 = (k_{\parallel}, w_2), \\ \mathbf{k}'_2 &= (k_{\parallel}, -w_2), \quad w_2^2 = \epsilon_2 k_0^2 - k_{\parallel}^2 \\ &\quad \text{for } d_1 \leq z < d_1 + d_2; \end{aligned} \tag{2.11}$$

$$\begin{aligned} \mathbf{E}_m(\mathbf{r}, \omega) &= \int \int [\mathcal{E}_m^{(+)}(u, v, \omega) \exp(i\mathbf{k}_m \cdot \mathbf{r}) \\ &\quad + \mathcal{E}_m^{(-)}(u, v, \omega) \exp(i\mathbf{k}'_m \cdot \mathbf{r})] du dv, \\ \mathbf{k}_m \cdot \mathcal{E}_m^{(+)} &= 0, \quad \mathbf{k}'_m \cdot \mathcal{E}_m^{(-)} = 0, \quad \mathbf{k}_m = (k_{\parallel}, w_m), \\ \mathbf{k}'_m &= (k_{\parallel}, -w_m), \quad w_m^2 = \epsilon_m k_0^2 - k_{\parallel}^2 \\ &\quad \text{for } \sum_{i=1}^{m-1} d_i \leq z \leq \sum_{i=1}^m d_i; \end{aligned} \tag{2.12}$$

$$\begin{aligned} \mathbf{E}_s(\mathbf{r}, \omega) &= \int \int \mathcal{E}_s(u, v, \omega) \exp(i\mathbf{k} \cdot \mathbf{r}) du dv, \\ \mathbf{k} \cdot \mathcal{E}_s &= 0, \quad \mathbf{k} = (k_{\parallel}, w) \quad \text{for } \sum_{i=1}^N d_i \leq z < \infty. \end{aligned} \tag{2.13}$$

With (2.8)–(2.13), applying Maxwell boundary conditions at $N + 1$ interfaces and using the same procedure as Refs. 1, 7, and 16, we obtain the component of the reflected electric field along the dipole direction at \mathbf{r}_0 ,

$$E_R^{\parallel} = (i/2) \sqrt{\epsilon} k_0^3 \rho \int_0^{\infty} k \mu_0^{-1} (R_0^{\parallel} + \mu_0^2 R_0^{\perp}) dk, \tag{2.14}$$

$$E_R^{\perp} = -i \sqrt{\epsilon} k_0^3 \rho \int_0^{\infty} k^3 \mu_0^{-1} R_0^{\perp} dk, \tag{2.15}$$

where the superscript \parallel (\perp) of E_R stands for parallel (perpendicular) to the interfaces, and we have defined

$$R_0^{\perp, \parallel} = \frac{R_{0, \parallel}^{\perp, \parallel} + R_1^{\perp, \parallel}}{1 + R_{0, \parallel}^{\perp, \parallel} R_1^{\perp, \parallel}} \exp(i2\mu_0 \hat{d}), \tag{2.16}$$

$$R_1^{\perp, \parallel} = \frac{R_{1, 2}^{\perp, \parallel} + R_2^{\perp, \parallel}}{1 + R_{1, 2}^{\perp, \parallel} R_2^{\perp, \parallel}} \exp(i2\mu_1 \hat{d}_1), \tag{2.17}$$

$$\begin{aligned} \dots, \\ R_{m-1}^{\perp, \parallel} &= \frac{R_{m-1, m}^{\perp, \parallel} + R_m^{\perp, \parallel}}{1 + R_{m-1, m}^{\perp, \parallel} R_m^{\perp, \parallel}} \exp(i2\mu_m \hat{d}_m), \quad m = 2, 3, \dots, N \end{aligned} \tag{2.18}$$

$$R_N^{\perp, \parallel} = R_{N, N+1}^{\perp, \parallel} \exp(i2\mu_N \hat{d}_N), \tag{2.19}$$

where

$$R_{m-1, m}^{\perp} = \frac{\epsilon_{m-1} \mu_m - \epsilon_m \mu_{m-1}}{\epsilon_{m-1} \mu_m + \epsilon_m \mu_{m-1}}, \tag{2.20a}$$

$$R_{N, N+1}^{\perp} = \frac{\epsilon_N \mu_{N+1} - \epsilon_s \mu_N}{\epsilon_N \mu_{N+1} + \epsilon_s \mu_N}, \tag{2.20b}$$

$$R_{m-1, m}^{\parallel} = \frac{\mu_{m-1} - \mu_m}{\mu_{m-1} + \mu_m}, \tag{2.21a}$$

$$R_{N, N+1}^{\parallel} = \frac{\mu_N - \mu_{N+1}}{\mu_N + \mu_{N+1}}, \tag{2.21b}$$

$$\mu_0 = (1 - k^2)^{1/2}, \tag{2.22}$$

$$\mu_m = (\epsilon_m / \epsilon - k^2)^{1/2}, \quad \mu_{N+1} = (\epsilon_s / \epsilon - k^2)^{1/2}, \tag{2.23}$$

$$\hat{d}_m = \epsilon^{1/2} \omega d_m / c. \tag{2.24}$$

For semi-infinite substrate occupying the half-space $z \geq 0$, we can just take $N \rightarrow \infty$. In general, we found through numerical analysis that when N is large enough, the reflected field E_R is nearly independent of N , especial-

ly when the constructing layers contain absorbing materials.

In Ref. 1 we have derived a set of new SBE and in Refs. 5-7 we have used them to study the interaction of light with an atom near a small metallic particle or a multilayer thin film or a semi-infinite periodic superlattice surface. The SBE in the rotating frame are

$$\frac{d}{dt} \begin{pmatrix} \langle S^+ \rangle \\ \langle S^z \rangle \\ \langle S^- \rangle \end{pmatrix} = \begin{pmatrix} i(\Delta + \Omega^s) - \gamma & i\Omega & 0 \\ i\Omega^*/2 & -2\gamma & -i\Omega/2 \\ 0 & -i\Omega^* & -i(\Delta + \Omega^s) - \gamma \end{pmatrix} \begin{pmatrix} \langle S^+ \rangle \\ \langle S^z \rangle \\ \langle S^- \rangle \end{pmatrix} - \begin{pmatrix} 0 \\ \gamma \\ 0 \end{pmatrix}, \quad (2.25a)$$

where the total decay rate $\gamma = \gamma^0 + \gamma^s$, γ^0 is the decay rate without the presence of the solid surface,^{6,7,17} detuning $\Delta = \omega_{21} - \omega_L$, and Rabi frequency $\Omega = |p|E_L$;

$$\gamma^s = |p|^2 \text{Im}f(d) \quad (2.25b)$$

and

$$\Omega^s = |p|^2 \text{Re}f(d) \quad (2.25c)$$

are the surface-induced spontaneous decay rate and frequency shift, respectively; ω_{21} is the adatomic transition frequency, $|p|$ is the matrix element of electric dipole operator, ω_L and E_L are the frequency and amplitude of the external laser field, respectively; $f(d)$ is a function of the distance d and determined by¹

$$E_R = |p|f(d)S^- = pf(d), \quad (2.26)$$

where E_R is the component of \mathbf{E}_R in the direction of \mathbf{p} .

With the SBE (2.25) and the expression for the reflected field E_R we can study the interaction between radiation fields and the adatom.

III. LUMINESCENCE PROPERTIES

As shown in Fig. 1, we consider the fact that the solid substrate is uniform in the X and Y directions, but in the Z direction its structure is something like a one-dimensional lattice. According to Ref. 13, a one-dimensional quasiperiodic lattice structure can be obtained by using the (concurrent) substitution rule

$$\mathbf{a} \rightarrow \underline{M}\mathbf{a}, \quad (3.1)$$

where \mathbf{a} is a column vector $\mathbf{a} = (a_1, a_2, \dots, a_g)^T$ and \underline{M} is a $g \times g$ transform matrix with non-negative integer entries. As in Refs. 6 and 7, we still consider the typical case that the semi-infinite quasiperiodic superlattice has two constituents of which one is a metal called medium A with thickness d_A and dielectric function⁷

$$\epsilon_A = 1 - \omega_p^2 / (\omega^2 + i\omega\Gamma_p), \quad \Gamma_p = 0.01\omega_p \quad (3.2)$$

where ω_p is the plasmon frequency, while the other is a insulator called medium B with d_B and $\epsilon_B = 3.0$. Therefore, we have¹³ $g=2$, $a_1=A$, $a_2=B$. We may consider three typical cases with the transform matrices:

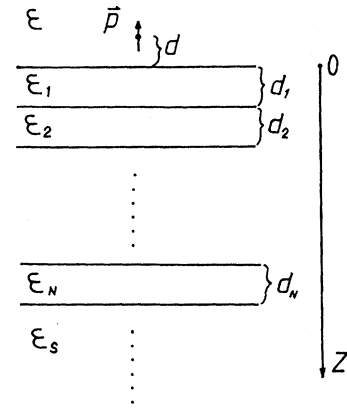


FIG. 1. An adatom with electric moment \mathbf{p} is located at a distance d from a surface of a semi-infinite superlattice occupying region $z > 0$. This adatom is in the region $z < 0$ occupied by a medium with dielectric constant ϵ .

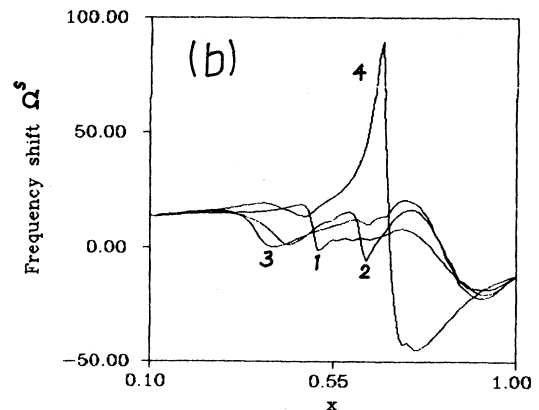
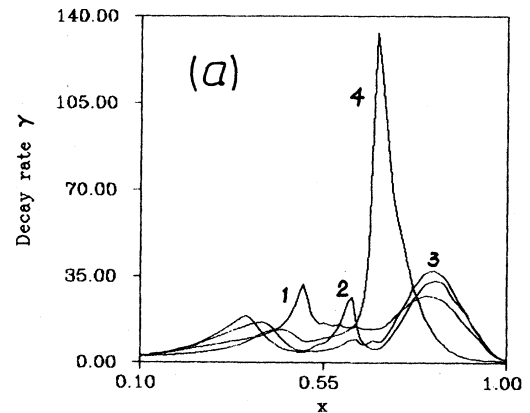


FIG. 2. (a) Spontaneous-decay rate γ and (b) frequency shift Ω^s vs x for various choices of structures: curve 1, structure 1; curve 2, structure 2; curve 3, structure 3; curve 4, structure 4.

$$\underline{M}_1 = \begin{bmatrix} 1 & 1 \\ 1 & 0 \end{bmatrix}, \quad \underline{M}_2 = \begin{bmatrix} 1 & 2 \\ 1 & 0 \end{bmatrix}, \quad \underline{M}_3 = \begin{bmatrix} 1 & 2 \\ 1 & 1 \end{bmatrix}^{\#}, \quad (3.3)$$

respectively, and the corresponding quasiperiodic structures

$$\begin{aligned} \underline{M}_1 &\Rightarrow AB (\epsilon_1 = \epsilon_A, \epsilon_2 = \epsilon_B) \rightarrow ABA (\epsilon_{1,3} = \epsilon_A, \epsilon_2 = \epsilon_B) \\ &\rightarrow ABAAB \rightarrow ABAABABA \rightarrow \dots \text{ (structure 1) }, \end{aligned} \quad (3.4)$$

$$\underline{M}_2 \Rightarrow AB \rightarrow ABBA \rightarrow ABBAABBA \rightarrow \dots \text{ (structure 2) }, \quad (3.5)$$

$$\underline{M}_3 \Rightarrow AB \rightarrow ABBAAB \rightarrow ABBAABABAABBAAB \rightarrow \dots \text{ (structure 3) }. \quad (3.6)$$

To compare the results for the present cases with those for periodic case as in Ref. 7, we define the periodic structure as "structure 4."

For simplicity, in the present paper we only consider the vertical orientation of the dipole. With Eqs. (2.15), (2.25), and (2.26) we obtain the spontaneous-decay rate and frequency shift in the unit^{6,7} of γ^0 for the adatom near this superlattice,

$$\gamma = 1 - \frac{3}{2} \text{Re} \int_0^\infty dk \mu_0^{-1} k^3 R_0^{\frac{1}{2}} \Big|_{N \rightarrow \infty} \quad (3.7)$$

and

$$\Omega^s = \frac{3}{2} \text{Im} \int_0^\infty dk \mu_0^{-1} k^3 R_0^{\frac{1}{2}} \Big|_{N \rightarrow \infty}, \quad (3.8)$$

respectively. Through numerical evaluation of Eqs. (3.7) and (3.8) we have found that up to 200 layers are enough for the convergence when the order of magnitude of \hat{d}_i is 10^{-1} , due to the dissipation in medium *A*. In addition, we numerically calculated Eqs. (3.7) and (3.8) with $N=200$ for the same semi-infinite periodic structure as in Ref. 7 and compared the results with those obtained in Ref. 7. We found that they are in good agreement with each other, although they are obtained from different formulas. Therefore the present calculations are reliable.

In the present paper we are interested in the structure effects and dielectric effects of the metal layers when the adatom is located very near the surface. So we take $\hat{d}=0.3$, $\epsilon=1.2$, $\hat{d}_A=0.1$, and $\hat{d}_B=0.15$. By integrating numerically Eqs. (3.7) and (3.8), Figs. 2(a) and 2(b) show the dependence of γ and Ω^s on the ratio of frequency ω/ω_p for the above four different structures. With these figures one can easily compare with one another the results for these four structures. Evidently, different structure of the substrate will lead to different number, posi-

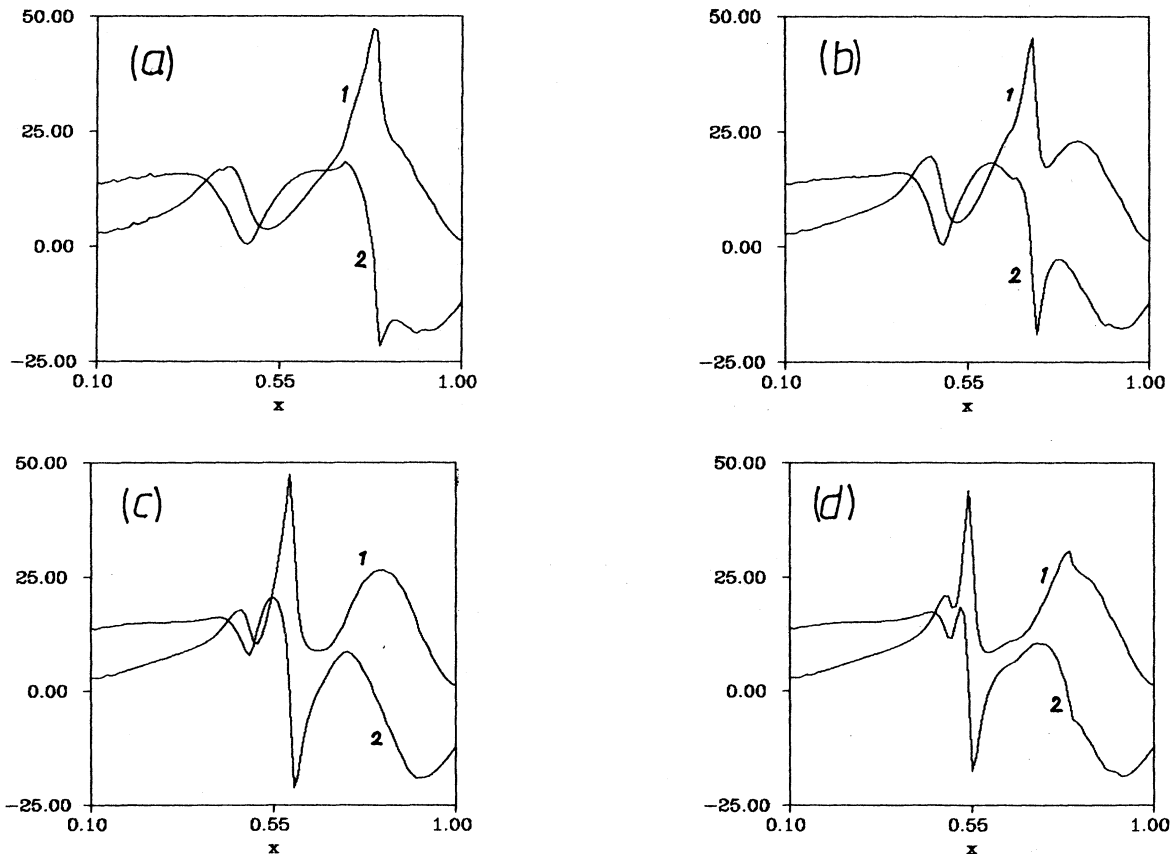


FIG. 3. Decay rate γ (curves 1) and frequency shift Ω^s (curves 2) vs $x (= \omega/\omega_p)$ for structure 1, $\epsilon_A = 1 - \omega_p^2 / (\omega^2 + i\omega\Gamma_p)$, $\Gamma_p = 0.01\omega_p$, $\epsilon_B = 3.0$, (a) $N = 5$, (b) $N = 8$, (c) $N = 13$, (d) $N = 21$, (e) $N = 34$, (f) $N = 55$, (g) $N = 89$, and (h) $N = \infty$.

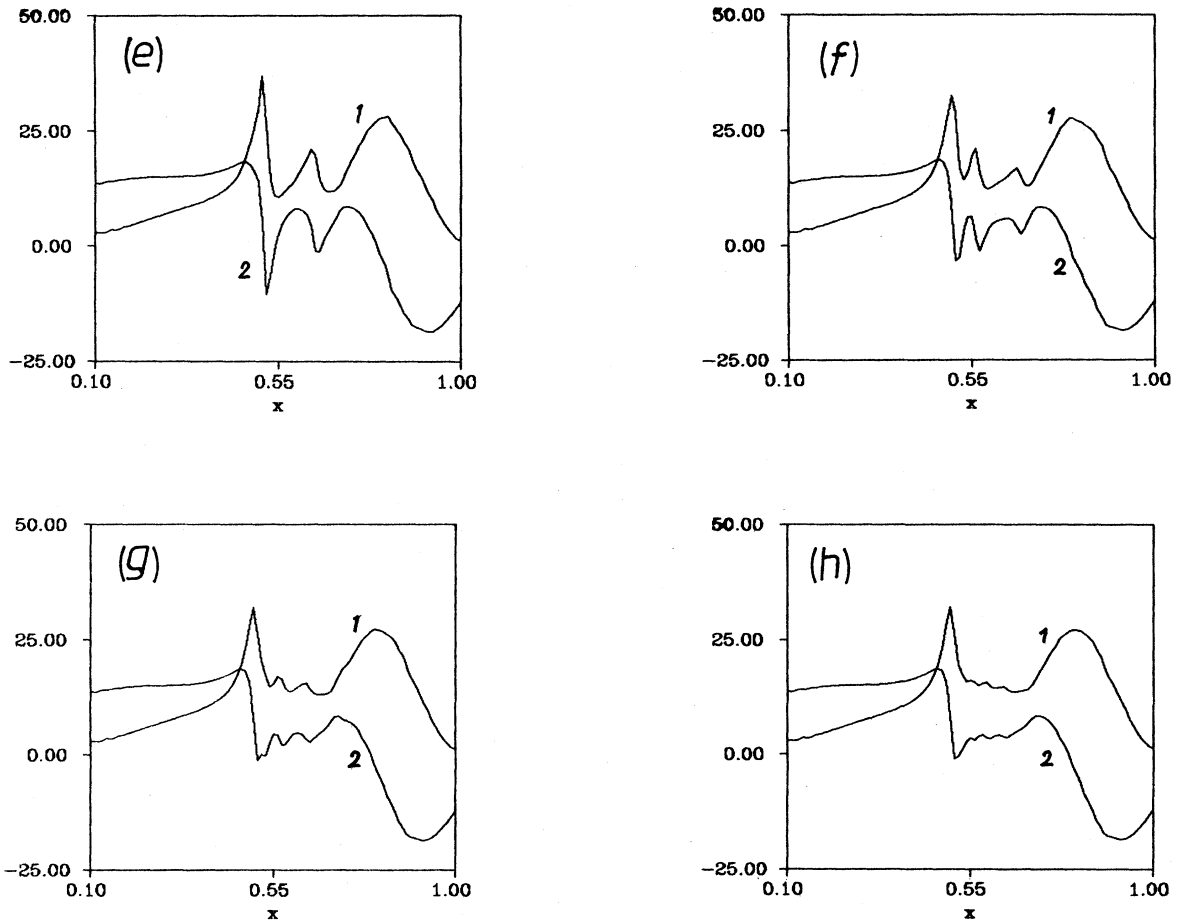


FIG. 3. (Continued).

tion, and height of the peaks. In Fig. 2(a), for each structure the right peak may be resulted from multibeam interference of the nonplane electromagnetic waves refracted at and reflected from each interface of the superlattice. In the periodic case the interference is much stronger, so the right peak is much higher. The other peaks resulted from the energy transfer of the adatom to the surface plasmons. The processes of the interference and the energy transfer strongly depend on the structures and the dielectric properties of the superlattice substrate. Comparing Fig. 2(b) with 2(a), we can easily see that each peak in γ -versus- x ($=\omega/\omega_p$) curves is accompanied by negative $d\Omega^s/dx$ in Ω^s -versus- x curves. Therefore the present situation is something like that of a beam of light transmitting through a medium, with the energy transfer of the adatom corresponding to the light absorption process and the frequency shift of the adatomic transition to the light dispersion.

Figures 3(a)–3(h) show the transition effect of the substrate from a finite thin film to a semi-infinite superlattice for the dependence of γ and Ω^s on x . Evidently, for smaller layer number N , the effects of increasing layers become stronger. However, when $N \geq 34$ (i.e., successively using \underline{M}_1 for more than six times), the left and the

right peaks in the γ -versus- x curves are rarely changed by increasing the layer number N . When N becomes very large ($N \geq 134$), the smaller peaks between these two peaks tend to disappear. We also have found that this phenomenon appears when we take other thicknesses for medium A and B .

IV. EXCITATION BY AN EXTERNAL LASER FIELD

In this section we consider the behavior of the adatom driven by an incident laser field with frequency ω_L and constant amplitude E_L . As in Ref. 8, when the intensity of the laser is weak, it is more probable for the adatom to stay in the lower state than in the upper state. In this case we approximately have

$$\langle S^z(t) \rangle = -\frac{1}{2}. \quad (4.1)$$

So, we can linearize the SBE (2.25) to obtain the equations for $\langle S^-(t) \rangle$ and $\langle S^+(t) \rangle$, e.g.,

$$\frac{d}{dt} \langle S^-(t) \rangle = -[i(\Delta + \Omega^s) + \gamma] \langle S^-(t) \rangle \frac{i}{2} \Omega^*. \quad (4.2)$$

Inserting (4.2) and its conjugation into the equation of $\langle S^z(t) \rangle$ in the SBE and considering the adatom initially

in the lower state, i.e., $\langle S^z(0) \rangle = -\frac{1}{2}$, one can easily obtain

$$\langle S^z(t) \rangle = |\Omega|^2 [1 + e^{-2\gamma t} - 2e^{-\gamma t} \cos(\Delta + \Omega^s)t] / 4|z|^2 - \frac{1}{2}. \quad (4.3)$$

With Eq. (4.3) we numerically analyzed time evolution of the mean value of the inversion operator, and some results are shown in Fig. 4 for different structures, where time t is in the unit of $1/\gamma^0$. From this figure one can see how the occupation probabilities of the adatomic levels are affected by the dielectric properties and the structure of the semi-infinite superlattice. Evidently, different geometric structure leads to different time behavior of the mean atomic inversion. Meanwhile, the frequency and amplitude of the oscillations strongly depend on both frequency ratio x and geometry of the substrate. E.g., in Fig. 4(d), $\langle S^z(t) \rangle$ around $x=0.67$ rarely changes as t increases but constantly take the value of $-\frac{1}{2}$ due to the strong energy-transfer process. In this case the intensity of the exciting field is not strong enough to cause the adatom to make upward transition.

By means of the SBE (2.25), one can easily obtain the

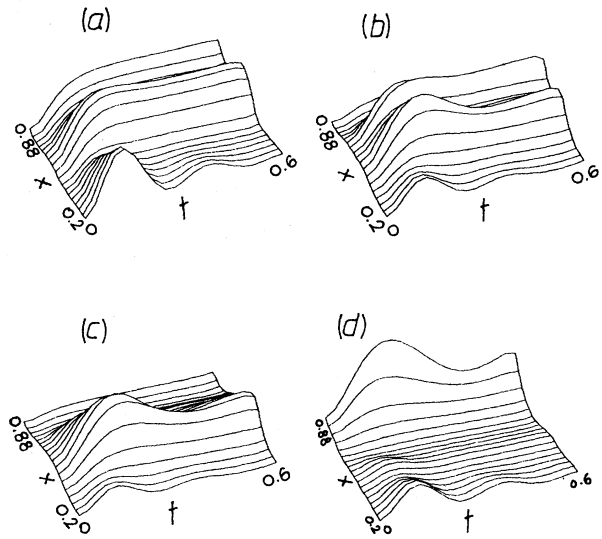


FIG. 4. $[\langle S^z(t) \rangle + \frac{1}{2}] / |\Omega|^2$ vs time t for various choices of x , $\Delta=10.0$. (a) Structure 1, (b) structure 2, (c) structure 3, and (d) structure 4.

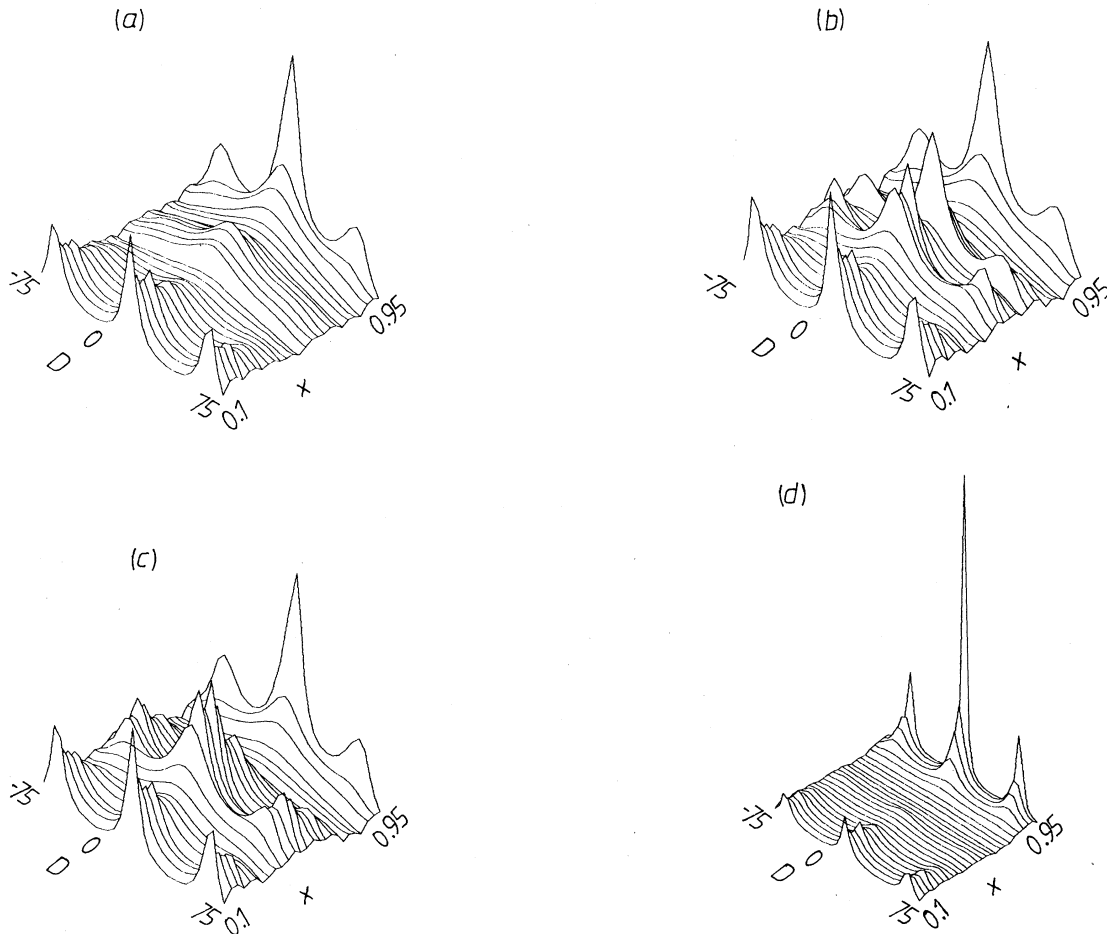


FIG. 5. Resonance fluorescence spectrum for various choices of x , $|\Omega|=45.0$, $\Delta=10.0$. (a) Structure 1, (b) structure 2, (c) structure 3, and (d) structure 4.

incoherent part of the resonance fluorescence spectrum^{1,5-8}

$$\bar{g}(\nu) = \frac{\frac{1}{2}|\Omega|^4\gamma(D^2 + \frac{1}{2}|\Omega|^2 + 4\gamma^2)}{(X^2 + Y^2)(\frac{1}{2}|\Omega|^2 + |z|^2)}, \quad (4.4)$$

where

$$D = \nu - \omega_L, \quad z = \gamma + i(\Delta + \Omega^s),$$

$$X = 2\gamma(\frac{1}{2}|\Omega|^2 + |z|^2 - 2D^2),$$

$$Y = D(|\Omega|^2 + |z|^2 + 4\gamma^2 - D^2).$$

Since both γ and Ω^s are determined by the dielectric properties and geometry of the solid substrate, the incoherent part of the resonance fluorescence spectrum for an adatom near a surface of a superlattice is also determined by the dielectric properties and structure of the layers composing this superlattice via both γ and Ω^s . We have numerically calculated the incoherent spectrum given by Eq. (4.4) and some results are shown in Fig. 5, in which each figure part demonstrates how the spectrum depends on the frequency ratio x ($=\omega/\omega_p$) of the adatomic transition to the plasmon in the metal layers. This dependence is nearly oscillatory. Comparing one part with the others in Fig. 5, one can see different results from different structures of the substrate. We know that for certain values of x the energy-transfer processes becomes much stronger so that two sidebands or even all the three peaks are suppressed.⁷ However, these values of x strongly depend on the geometric structure of the substrate. On the other hand, the frequency ratio depen-

dence of the spectrum for periodic structure remarkably differs from those for quasiperiodic structures and the shapes of the spectrum for different kind of quasiperiodic structures are different.

Now we proceed to discuss the squeezing phenomenon of the adatomic dipole moment in this interaction. As in Ref. 7, the dispersive and the absorptive components of the adatomic dipole moment are

$$\sigma_1 = (S^+ + S^-)/2, \quad (4.5)$$

$$\sigma_2 = (S^+ - S^-)/2i, \quad (4.6)$$

respectively. Letting $\sigma_3 = S^z$, we have the commutation relation

$$[\sigma_1, \sigma_2] = i\sigma_3 \quad (4.7)$$

and the corresponding uncertainty relation

$$(\Delta\sigma_1)^2(\Delta\sigma_2)^2 \geq \langle\sigma_3\rangle/4. \quad (4.8)$$

Hence, the adatomic state is said to be squeezed when one of the operators σ_1 and σ_2 satisfies the relation

$$(\Delta\sigma_i)^2 < |\langle\sigma_3\rangle|/2, \quad i=1,2 \quad (4.9)$$

and the adatom is said to be in the coherent state when both σ_1 and σ_2 satisfy

$$(\Delta\sigma_i)^2 = |\langle\sigma_3\rangle|/2. \quad (4.10)$$

With Eqs. (2.25), (4.5), and (4.6), we have the steady-state results

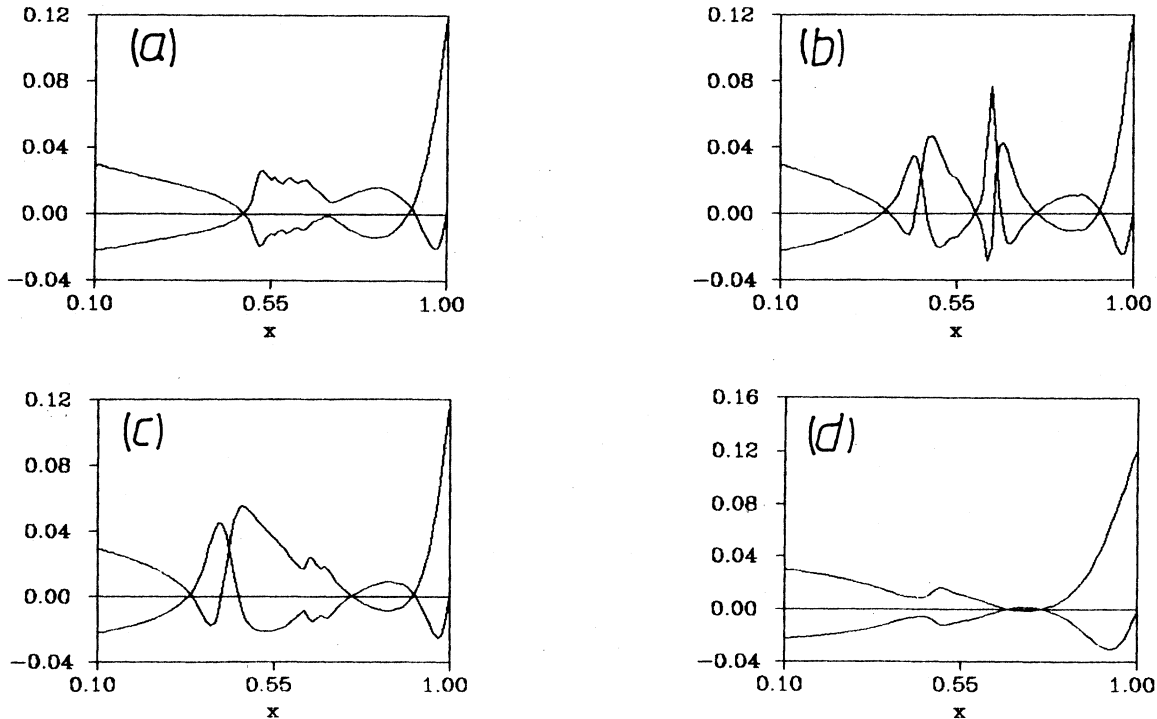


FIG. 6. Q_1 (curves 1) and Q_2 (curves 2) vs x , $\Delta=5.0$, $|\Omega|=10.0$, (a) structure 1, (b) structure 2, (c) structure 3, and (d) structure 4.

$$(\Delta\sigma_1)^2 = [1 - (\Delta + \Omega^s)^2 |\Omega|^2 / (|\Omega|^2/2 + |z|^2)] / 4, \quad (4.11)$$

$$(\Delta\sigma_2)^2 = [1 - \gamma^2 |\Omega|^2 / (|\Omega|^2/2 + |z|^2)] / 4, \quad (4.12)$$

$$\langle \sigma_3 \rangle = -|z|^2 / 2(|\Omega|^2 + |z|^2). \quad (4.13)$$

Defining

$$Q_i = (\Delta\sigma_i)^2 - |\langle \sigma_3 \rangle| / 2, \quad i = 1, 2 \quad (4.14)$$

we may rewrite condition (4.9) as

$$Q_i < 0 \quad (4.15)$$

for squeezing in the dispersive component or in the absorptive component.

We know that fluctuations of the adatomic dipole moment strongly depend on the optical properties and the geometric structure of the substrate.⁷ Here we study these fluctuations under strong excitation of the external laser field. Figure 6 shows some numerical results. We found that the two components of the dipole moment exhibit squeezing nearly in the whole ratio region ($x = 0.1$ to $x = 1.0$) and they are squeezed alternatively. This alternatively squeezing effect is stronger for structures 2 and 3. We also found that for some special values of x the adatom is in the coherent state, i.e., Eq. (4.10) is satisfied. These values are not the same for different

structures. The above phenomena are due to the nonlinear interaction of the adatom with the surface-reflected field and the exciting laser field. Through this interaction the dielectric effect and the structure effect are revealed.

V. SUMMARY

So far we have studied fluorescence and light near-resonance scattering of an adatom near a quasiperiodic superlattice surface, and compared the results for quasiperiodic structures with those for periodic one, and compared with one another the results for different kinds of quasiperiodic structure. The fluorescence decay rate and frequency shift as well as the scattered spectrum carry information of the properties and structures of the superlattice. A lot of novel phenomena, such as absorptionlike and dispersionlike behavior and transition behavior when the number of the quasiperiod gradually approaches infinity for the decay rate and frequency shift, are revealed and they are absent in Ref. 6, in which we considered a substrate of a thin film with limited alternatively arranged layers and frequency-independent real dielectric constants. In addition, the present results are also different from those obtained in Ref. 7 and reflects the effects of the structure.

*Mailing address.

¹X.-s. Li and C.-d. Gong, Phys. Rev. A **35**, 1595 (1987).

²H. Dekker, Physica A **95**, 311 (1979).

³L. S. Zhang, Physica A **117**, 355 (1983).

⁴M. Sargent, M. O. Scully, and W. E. Lamb, *Laser Physics* (Addison-Wesley, Reading, Mass., 1974), Chap. 16.

⁵X.-s. Li and C.-d. Gong, J. Phys. B **21**, 1429 (1988).

⁶X.-s. Li and C.-d. Gong, Phys. Lett. A **131**, 138 (1988).

⁷X.-s. Li and C.-d. Gong, Phys. Rev. B **39**, 8284 (1989).

⁸X. Y. Huang, J. T. Lin, and T. F. George, J. Chem. Phys. **80**, 893 (1984); X. Y. Huang and T. F. George, *ibid.* **88**, 4801 (1984); X. Y. Huang, K. T. Lee, and T. F. George, *ibid.* **85**, 567 (1986).

⁹J. T. Lin, X. Y. Huang, and T. F. George, J. Opt. Soc. Am. B **4**, 219 (1987).

¹⁰H. F. Arnoldus and T. F. George, J. Phys. B **21**, 431 (1988).

¹¹H. F. Arnoldus and T. F. George, Phys. Rev. A **37**, 761 (1988).

¹²See B. Simon, Adv. Appl. Math. **3**, 463 (1982), and references therein.

¹³J. P. Lu, Odagaki, and J. L. Birman, Phys. Rev. B **33**, 4809 (1986).

¹⁴M. Kohmoto and J. R. Banavar, Phys. Rev. B **34**, 563 (1986); F. Nori and J. P. Rodriguez, *ibid.* **34**, 2207 (1986); J. M. Luck and D. Petretis, J. Stat. Phys. **42**, 289 (1986); M. Kohmoto, B. Sutherland, and C. Tang, Phys. Rev. B **35**, 1020 (1987); A. H. MacDonald and G. C. Aers, *ibid.* **36**, 9142 (1987).

¹⁵D. Schectman, I. Blech, D. Gratias, and J. W. Cahn, Phys. Rev. Lett. **53**, 1951 (1984).

¹⁶G. S. Agarwal, Phys. Rev. A **11**, 230 (1975); **11**, 243 (1975); **11**, 253 (1975); **12**, 1475 (1975).

¹⁷G. S. Agarwal and S. V. Oneil, Phys. Rev. B **28**, 487 (1983); G. S. Agarwal, *Quantum Electrodynamics and Quantum Optics* (Plenum, New York, 1984), p. 1.