Coherent radiation scattering by resonant nanostructures

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This paper deals with the scattering of radiation by resonant nanostructures, i.e., systems of like atoms arranged at distances much less than the wavelength of light. It is demonstrated that the quasistatic interaction of atoms in such nanostructures changes the eigenfrequency structure of the atoms forming the nanostructure and causes substantial alterations in the scattered light spectrum. With the plane polarization of the incident light wave, the spectrum can be singlet (two interacting atoms), doublet (three interacting atoms), and so on, depending on the number of atoms in the nanostructure. The potential possibility is discussed of using such resonant structures as ultrahigh-density ROM devices.

The development of various nanotechnology techniques opens up the possibility of manipulating the structure of matter on an atomic-molecular scale, i.e., to control the location of impurity atoms with a nanometer-high precision (by means of scanning tunneling-microscopy,[1] focused electron or ion beams,[2] atomic optics techniques using laser light,[3,4] etc.). This makes it possible, in particular, to create various kinds of nanostructures consisting of any number of atoms.[5] As the distance between impurity atoms is reduced, their interaction becomes substantial, and this may give rise to a number of effects some of which are considered below.

Treated in the present work is the scattering of radiation by nanostructures formed by like atoms with *coincident* resonance frequencies. The geometry of the problem is shown in Fig. 1. When acted upon by a resonant field, the atoms get polarized. The dipole moments of different atoms interact with one another. Generally speaking, this interaction is a retarding one, and the expression describing it is rather complex. However, in our case, i.e., in the case of nanostructures, the retardation can be disregarded, so that the dipole interaction can be considered to be electrostatic with the interaction energy U_{12} given by

$$U_{12} = \frac{\mathbf{d}_1 \cdot \mathbf{d}_2}{|\mathbf{r}_1 - \mathbf{r}_2|^3} - 3 \frac{(\mathbf{d}_1 \cdot (\mathbf{r}_1 - \mathbf{r}_2))(\mathbf{d}_2 \cdot (\mathbf{r}_1 - \mathbf{r}_2))}{|\mathbf{r}_1 - \mathbf{r}_2|^5}, \quad (1)$$

where $\mathbf{d}_1, \mathbf{d}_2$ and $\mathbf{r}_1, \mathbf{r}_2$ are the dipole moments and the positions of the first and the second dipole, respectively.

As a result of this interaction, the eigenfrequencies of oscillations (equal in the absence of interaction) get split, and in the spectrum of the radiation scattered by the nanostructure there appear frequencies corresponding to the split oscillation eigenfrequencies of the nanostructure. The nanostructure itself can, in that case, be treated as a ''molecule'' whose geometry determines its oscillation eigenfrequencies differing from the initially equal frequencies of the free atoms.

The spontaneous emission (super-radiation) spectrum of a system of free atoms spaced at distances shorter than the wavelength of light was analyzed by Dicke as far back as 1954 (Ref. 6) (see also Ref. 7). In Refs. 8 and 9 the reflectance from absorbed overlayers was considered. In present

paper, we will consider another effect, namely, the coherent scattering of radiation by a system of interacting dipoles located in a small volume of space, and specifically the changes in the scattered radiation spectrum by the groups of closed dipoles caused by the dipole interactions.

Considered here as a most simple model of an atom of a nanostructure will be a classical dipole consisting of a fixed oscillating charge whose oscillation axis coincides with the electric-field direction in the incident wave, which is in turn assumed to be completely linearly polarized (Fig. 1). Note that the oscillator in this model is, in principle, not isotropic, because the resultant oscillation direction in an isotropic oscillator model depends not only on the external field.

Let us first consider the case of a nanostructure consisting of *two atoms* situated in a plane perpendicular to the wave vector of the incident wave. The equations of motion of the mobile charges of the first and second atoms have the form

$$m(\delta\ddot{\mathbf{r}}_1 + \gamma^* \delta\dot{\mathbf{r}}_1 + \omega_0{}^2 \delta\mathbf{r}_1) = e[\mathbf{E}_{\text{in}}(\mathbf{r}_1, t) + \mathbf{E}^{(2)}(\mathbf{r}_1, t)],$$

$$m(\delta\ddot{\mathbf{r}}_2 + \gamma^* \delta\dot{\mathbf{r}}_2 + \omega_0{}^2 \delta\mathbf{r}_2) = e[\mathbf{E}_{\text{in}}(\mathbf{r}_2, t) + \mathbf{E}^{(1)}(\mathbf{r}_2, t)].$$
$$(2)$$

In the above expressions, e and m are the electronic charge and mass, $\delta\mathbf{r}_1$ and $\delta\mathbf{r}_2$ are the oscillation amplitudes of the dipoles, ω_0 and γ^* are the free oscillation frequency and damping of an isolated dipole, and $\mathbf{E}_{\text{in}}(\mathbf{r}_i, t)$ and $\mathbf{E}^{(j)}(\mathbf{r}_i, t)$
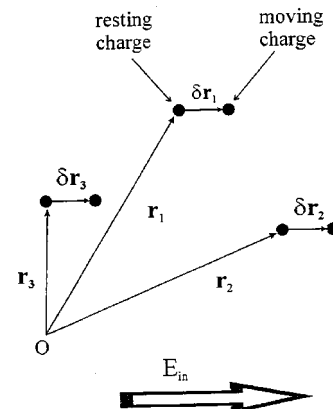

FIG. 1. Geometry of the problem (three-atom nanostructure).

are the external field at the location of the ith oscillator and the field due to the jth oscillator at the location of the ith oscillator, respectively. Note, the γ^* is total nonhomogeneous linewidth which is usually essentially higher than homogeneous linewidth and the radiative linewidth

$$\gamma_0 = 2e^2\omega_0^2/(3\,c^3m).$$

Insofar as it is nanostructures that are being considered here, use can be made of the quasistationary approximation wherein the interaction between the oscillators will be characterized by the field ($i,j=1,2$).

$$\mathbf{E}^{(j)}(\mathbf{r}_i,t) = -e\left[\frac{\delta\mathbf{r}_i}{|\mathbf{r}_i-\mathbf{r}_j|^3} - 3\frac{(\mathbf{r}_i-\mathbf{r}_j)[\,\delta\mathbf{r}_i\cdot(\mathbf{r}_i-\mathbf{r}_j)]}{|\mathbf{r}_i-\mathbf{r}_j|^5}\right]. \tag{3}$$

Since the oscillation phase shift between the atoms is in our case (the case of nanostructures) small, the emission of radiation is governed by the total dipole moment

$$d_{tot} = e(\delta r_1 + \delta r_2). \tag{4}$$

Assuming that all the variables depend on time as $e^{-i\omega t}$ and using the fact that the oscillation direction coincides with the external field direction from Eqs. (3) and (4) we find that

$$d_{tot} = \frac{2e^2 E_{in}}{m\left(-\omega^2 - i\omega\gamma^* + \omega_0^2 + \dfrac{e^2[1-3\cos^2(\theta_{12})]}{m|\mathbf{r}_1-\mathbf{r}_2|^3}\right)}. \tag{5}$$

Here

$$\cos(\theta_{12}) = [\mathbf{n}\cdot(\mathbf{r}_1-\mathbf{r}_2)]/|\mathbf{r}_1-\mathbf{r}_2|.$$

Note that expression (5) corresponds to the excitation of a single normal (symmetrical) mode, which is due to the method of excitation. When the external field is parallel to $\mathbf{r}_1-\mathbf{r}_2$, the oscillation frequency is reduced:

$$\omega_0 \rightarrow \omega_0[1 - e^2/(m\omega_0^2|\mathbf{r}_1-\mathbf{r}_2|^3)]. \tag{6}$$

When the external field is perpendicular to $\mathbf{r}_1-\mathbf{r}_2$, the oscillation frequency is increased:

$$\omega_0 \rightarrow \omega_0[1 + e^2/(2m\omega_0^2|\mathbf{r}_1-\mathbf{r}_2|^3)]. \tag{7}$$

The radiation power P of the total dipole moment is defined by the formula

$$P = (c/3)k^4|d_{tot}|^2, \tag{8}$$

where $k = 2\pi/\lambda$ and λ is the radiation wavelength. Substituting (5) into (8) and dividing the expression thus obtained by the incident flux

$$I_{in} = (c/8\pi)|E_{in}|^2, \tag{9}$$

we find the scattering cross section for such a nanostructure:

$$\sigma_2 = 4\sigma_{Thom}\left|\frac{\omega^2}{\omega^2 + i\omega\gamma^* - i\omega_0^2 - \dfrac{e^2[1-3\cos^2(\theta_{12})]}{m|\mathbf{r}_1-\mathbf{r}_2|^3}}\right|^2, \tag{10}$$

where $\sigma_{Thom} = 8\pi/3(e^2/mc^2)^2$ is the scattering cross section for a free electron. At high frequencies ($\omega\rightarrow\infty$) expression (10) naturally reduces to the coherent scattering cross section for two free particles: $\sigma_2 \rightarrow 4\sigma_{Thom}$. The maximum value of the cross section is

$$\sigma_2^{max} \simeq 4\sigma_{Thom}(\omega_0/\gamma^*)^2. \tag{11}$$

Figure 2 presents the relative scattering cross section as a function of the distance between the atoms. It can be seen that when the atoms are located sufficiently close together there takes place, as expected, a substantial shift of the radiation frequency. It is also evident that only one frequency is present in the scattered radiation spectrum, although there are, generally speaking, two eigenfrequencies in the system.

Consider a nontrivial group of *three atoms*. In that case, the dynamics of the moving charges of the nanostructure is defined by the set of equations, which is similar to (2):

$$m(\delta\ddot{\mathbf{r}}_1 + \gamma^*\delta\dot{\mathbf{r}}_1 + \omega_0^2\delta\mathbf{r}_1) = e[\mathbf{E}_{in}(\mathbf{r}_1,t) + \mathbf{E}^{(2)}(\mathbf{r}_1,t) + \mathbf{E}^{(3)}(\mathbf{r}_1,t)],$$

$$m(\delta\ddot{\mathbf{r}} + \gamma^*\delta\dot{\mathbf{r}}_2 + \omega_0^2\delta\mathbf{r}_2) = e[\mathbf{E}_{in}(\mathbf{r}_2,t) + \mathbf{E}^{(1)}(\mathbf{r}_2,t) + \mathbf{E}^{(3)}(\mathbf{r}_2,t)], \tag{12}$$

$$m(\delta\ddot{\mathbf{r}}_3 + \gamma^*\delta\dot{\mathbf{r}}_3 + \omega_0^2\delta\mathbf{r}_3) = e[\mathbf{E}_{in}(\mathbf{r}_3,t) + \mathbf{E}^{(1)}(\mathbf{r}_3,t) + \mathbf{E}^{(2)}(\mathbf{r}_3,t)].$$

The solution of this system can be found in full analogy with the case of two-atom nanostructures. As a result, we get the following expression for the scattering cross-section:

$$\sigma_3 = \sigma_{Thom}\left|\frac{\omega^2[3\alpha_0^2 - 2\alpha_0(\alpha_{12}+\alpha_{13}+\alpha_{23}) - 2(\alpha_{12}^2+\alpha_{13}^2+\alpha_{23}^2) + (\alpha_{12}+\alpha_{13}+\alpha_{23})^2]}{\alpha_0^3 - \alpha_0(\alpha_{12}^2+\alpha_{13}^2+\alpha_{23}^2) + 2\alpha_{12}\alpha_{13}\alpha_{23}}\right|^2. \tag{13}$$

Here

$$\alpha_{ij} = \{e^2[1-3\cos^2(\theta_{ij})]\}/m|\mathbf{r}_i-\mathbf{r}_j|^3$$

and $\alpha_0 = (-\omega^2 - i\omega\gamma^* + \omega_0^2)$.

At high frequencies ($\omega\rightarrow\infty$) (13) naturally reduces to the coherent scattering cross section for three free particles: $\sigma_3 \rightarrow 9\sigma_{Thom}$. The maximum value of the cross section is

$$\sigma_3^{max} \simeq 9\sigma_{Thom}(\omega_0/\gamma^*)^2 \quad \text{for} \quad \Delta/\lambda \simeq 0.01 \tag{14a}$$

and

$$\sigma_3^{max} \simeq (3)\sigma_{Thom}(\omega_0/\gamma^*)^2 \quad \text{for} \quad \Delta/\lambda \simeq 0.001. \tag{14b}$$

Generally speaking, in such a system all eigenfrequencies are present. But in actuality with the dipoles being excited in

a synchronous fashion, there arise two modes only. Figure 3 shows the effective scattering cross section of a structure of three atoms arranged at the vertices of an equilateral triangle with a side of Δ as a function of the interatomic spacing Δ and the incident radiation frequency ω. It can be seen that in the case of three-atom nanostructures the radiation spectra are more complex. Where the external field and oscillation direction are parallel with one of the sides of the triangle, it is only two out of the three possible modes that effectively get excited. This is perhaps a general rule: in a planar structure, a normally incident plane wave excites all the normal modes save one.

The scattering of radiation by more complex nanostructures is studied in a similar way. Thus, the present work considers natural oscillations in nanostructures made up of like atoms interacting electrostatically and the spectra of radiation scattered by them. It is demonstrated that the radiation spectra depend materially on the geometry of the nanostructure and the orientation of the latter with respect to the polarization of the incident radiation. In the case of plane geometry of the nanostructure and normal incidence upon it of the external field, one of the possible natural frequencies practically fails to get excited and is absent in the scattered radiation spectrum.

The fact that the scattered radiation spectra depend on the geometry of the nanostructure suggests that such nanostructures can be used as information encoding elements. Many methods have already been proposed to increase optical data recording density (see, for example, Ref. 10). One of the most interesting methods is the persistent spectral hole burning technique.[11–13] With this technique, a narrow-band laser is used to burn a set of holes in an inhomogeneously broadened line of impurity or color centers, the width of the holes corresponding to an individual impurity molecule. In that case, it is proposed to use *noninteracting* dipoles with *different* natural frequencies. And we consider an entirely different effect, namely, *interacting* pairs (triplets) of closely spaced dipoles with *equal* frequencies which become, because of interaction, oscillators with *different* natural frequencies, frequency splitting being dependent on the distance between the elementary dipoles in the group.

Resonant nanostructures of interacting atoms can be used as logical elements in memory registers even at room temperature because the distances between the atoms forming the nanostructure on a solid surface (and governing the struc-

ture of the scattered radiation spectrum) remain practically unchanged with time. It should be emphasized once more that the scattering or absorption spectra in the given case depend solely on the mutual arrangement of the atoms forming the nanostructure.

The appropriate choice of the geometry of the nanostructure can provide for some or other type of scattered radiation spectrum and thus allow encoding some amount of information. This amount can be estimated as follows. The spectrum of radiation scattered by a nanostructure can be treated as a register of N binary digits whose number is governed by the relationship between the total linewidth γ^* and the frequency band $\Delta\omega \sim \omega_0$ of the incident radiation:

$$N \sim \Delta\omega / \gamma^* \propto 10^3. \qquad (15)$$

Thus, if we could manage to produce a nanostructure having 1000 eigenfrequencies with a width of the order of γ^*, we would then be able to encode with its aid some 100 bytes of information on a surface with an area of the order of λ^2. Note that in contrast to the persistent hole burning technique wherein one bit is associated with a hole of the order of the homogeneous width γ_0 of an individual center, one bit in our case is associated with the inhomogeneous linewidth γ^* which is several orders of magnitude greater than the homogeneous and radiative width γ_0.

Consider briefly some specifics of realizing nanostructures capable of high-density information storage. A most simple way is to accommodate on a surface with an area of the order of λ^2 a set of two-atom nanostructures whose interatomic distances will provide for the desired frequencies in the radiation spectrum (Fig. 4). The mutual arrangement of the individual two-atom nanostructures are naturally unimportant. Each binary nanostructure corresponds to a hole in the absorption spectrum in the persistent hole burning technique.

As the characteristic number of lines in the scattered radiation spectrum is defined by expression (15), the number N_1 of the binary nanostructures accommodated on a surface with an area of the order of λ^2 should not exceed the number of resolved lines:

$$N_1 \leq N = \Delta\omega / \gamma^*. \qquad (16)$$

Otherwise their spectra will be superimposed on one another, and encoding will be erroneous.
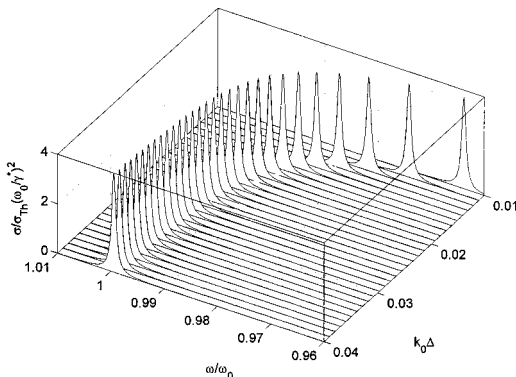

FIG. 2. Spectrum of radiation scattered by a two-atom nanostructure (radiation polarized perpendicular to the axis of the nanostructure, $k_0 = \omega_0/c$, distance between atoms Δ).
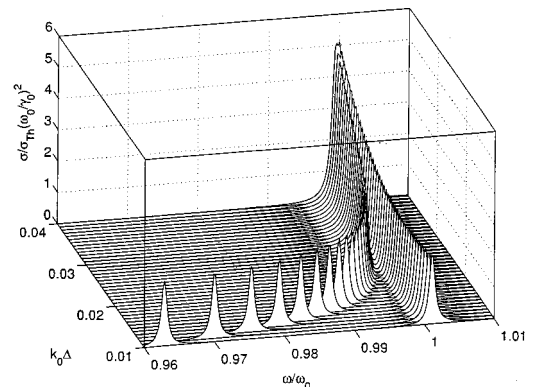

FIG. 3. Spectrum of radiation scattered by a three-atom nanostructure (radiation polarized parallel to one of the sides of the equilateral triangle, $k_0 = \omega_0/c$, side of the triangle Δ).
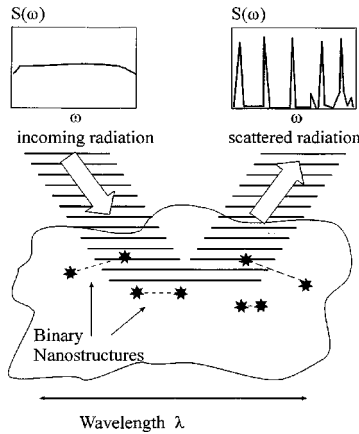
FIG. 4. Possible geometry of a memory element built around resonant nanostructures.

On the other hand, the distance between the individual two-atom components of a memory cell should be large enough in order that their interaction should not cause any additional frequency shift making the encoding process difficult to accomplish. In other words, this distance should be such that the frequency shift due to it does not exceed the total linewidth γ^*. It is not very difficult to see that the minimum distance between two-atom components of a nanostructure can be estimated by means of (5) and (6) as

$$\delta\omega_0 \propto e^2/(m\omega_0|\mathbf{r}_1-\mathbf{r}_2|^3_{\min}) \lesssim \gamma^*, \qquad (17)$$

whence we get the following estimate for the minimum permissible distance between two-atom components:

$$|\mathbf{r}_1-\mathbf{r}_2|_{\min} \lesssim 3\left[\frac{r_0\lambda_0^2}{4\pi^2}\left(\frac{\omega_0}{\gamma^*}\right)\right]^{1/2}, \qquad (18)$$

where r_0 is the classical radius of the electron.

So, the maximum possible number of negligibly interacting nanostructures on a surface with an area of the order of λ^2 will be defined by the expression

$$N_2 = \frac{\lambda^2}{|\mathbf{r}_1-\mathbf{r}_2|_{\min}^2} = \left(4\pi^2\frac{\lambda_0}{r_0}\frac{\gamma^*}{\omega_0}\right)^{2/3}. \qquad (19)$$

Figure 5 presents the numbers N_1 and N_2 as a function of the relative linewidth γ^*/ω_0. It can be seen that at $\gamma^*/\omega_0 < 10^{-4}$ the decisive factor is the requirement that the interaction between individual nanostructures should be weak. In
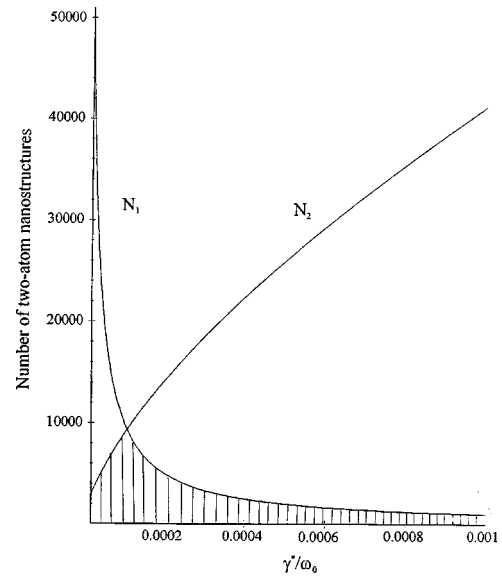

FIG. 5. Illustrating the determination of the optical memory element geometry: N_1–the maximum number of lines in the scattered radiation spectrum; N_2–the maximum number of noninteracting two-atom nanostructures accommodated on a surface with an area of λ^2 (the region of permissible numbers of two-atom nanostructures is shown by hatching).

the opposite case, the decisive factor is the limited character of the bandwidth of the incident light.

Under optimal conditions where $\gamma^*/\omega_0 \approx 10^{-4}$, it is possible to accommodate on a surface with an area of the order of λ^2 around 10^4 two-atom nanostructures capable of carrying, in principle, over 1 KB of information (400 GB/cm^2 at $\lambda = 0.5\,\mu$m). Of course, various practical restrictions that inevitably arise in the course of realization will reduce this amount, but we hope that the final actual figures will be high enough.

It should be emphasized that we only discuss here the principal possibility of using the effect under consideration in high-density memory devices without considering the actual possibilities of their realization, and particularly without discussing the information reading system and many other practical matters.

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