Collective dipole moments in a random medium. Retarded luminescence

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The spectrum of optical excitations of a system of atoms that are resonantly coupled by dipole–dipole interactions can exhibit collective states whose coherence extends over some finite region. The subject of this paper is how the formation of these states is affected by finiteness of the size of this coherence region. To treat this problem, a distribution function is constructed for the collective dipole moments as a function of the size of the coherence region. It is shown that when light is scattered by such a medium, the time decay of the intensity of retarded luminescence has a universal character that is sensitive only to the dimensionality of the space of dipole moments. © *1995 American Institute of Physics.*

1. INTRODUCTION

The case in which the amplitude of coherent excitation transport between impurity centers falls off according to a $1/R^3$ law (or $1/R^d$ in the general case of a d-dimensional space) plays a special role in the general problem of localization.[1] In a disordered system, more rapid falloff than this gives rise to localization; conversely, a slower falloff gives rise to delocalization (see, e.g., Ref. 2). In Ref. 3, Levitov presented a proof that a $1/R^3$ law corresponds to weak delocalization, leading to anomalous diffusion.

In this case, excitation transport over large distances involves the participation of collective states formed by a large number of coherently coupled centers. This is a direct consequence of the increase in the volume and number of centers, which is proportional to R^d and therefore compensates the falloff in the transport amplitude. In the general case, the clusters that form should possess different dipole moments. This leads to a change in the optical properties of the system, which occasionally can be quite radical in character. In this case all the correlation properties are sensitive to the distribution function of the emergent collective dipole moments. Levitov found the limiting distribution function for an infinite system, assuming only that coherent coupling is maintained at infinite distances; in fact, coherent exchange is in general limited by inelastic processes (e.g., due to interaction with phonons, see Ref. 4, or because of radiative decay of the excited states), and by the finite dimensions of the system.

The fundamental task of this paper is to find the distribution function of dipole moments of the collective states as a function of the size of the coherence region R_c. Let us consider a system of identical atoms randomly distributed in a medium, each with an isolated resonance level corresponding to a dipole transition from the ground state. The transport of an excitation from one atom to another takes place due to dipole–dipole interactions, and the transition amplitude has a typical dependence for this interaction, $f_{12} \propto 1/R_{12}^3$.

As we will see below, the portion of the distribution that is most sensitive to the size of the coherence volume turns out to be that portion associated with small dipole moments; this is natural from a physical point of view. As a rule, it is this region that dictates the peculiarities in the temporal behavior of the intensity of the light scattering for a random medium of this kind. The resulting distribution function allows us to solve the problem of retarded luminescence in such a medium. The analysis presented in this paper demonstrates the existence of a slow component of the emission, whose intensity falls off asymptotically according to a specific power law.

2. DISTRIBUTION OF COLLECTIVE DIPOLE MOMENTS IN A FINITE COHERENT REGION

Let us consider a medium with randomly distributed resonant centers whose excitation energy is distributed over a certain interval with a density function $g(\varepsilon)$ normalized to 1. Let all the centers have the same absolute value of the dipole moment d_0, and be distributed with spatial density n. We will assume that

$$\chi = 2\pi n d_0^2 g(\varepsilon) \ll 1. \tag{1}$$

The parameter χ determines the probability of finding a resonant partner for an up–down dipole–dipole transition at the average separation (the energy difference for the resonant states $\varepsilon_1 - \varepsilon_2$ is at most of the same order as their effective coupling f_{12}). Since the dipole interaction falls off according to $1/R^d$, where d is the dimension of the space, this probability increases as the size of the region increases:

$$w(R) \approx \chi \ln(R/\bar{R}). \tag{2}$$

This implies that the probability will be close to 1 when the size of the region reaches

$$R_* = \bar{R} \exp(1/\chi). \tag{3}$$

Choosing a volume of size R_1,

$$\bar{R} \ll R_1 \ll R_*, \tag{4}$$

we can state that the probability of simultaneously finding two or more (n) resonance partners is small, namely $w^2(R)$ [or $w^n(R)$]. This enables us to treat only the pairwise formation of resonance configurations. If we successively add layers of thickness R_1, then at each step the argument that the coupling is pairwise remains entirely correct, although the objects that enter into the resonance coupling may even be clusters (in which case the coupling will involve one of the

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states in the cluster; it is not difficult to see that the statistical distance between energies of the states in the cluster is large compared to the energy of the new coupling).

The pairwise character of the interaction leads to important consequences.

Each stage of the coherent interaction of two states leads to two new states. This implies that the overall number of states per unit volume remains equal to the number of original resonance centers n. On the other hand, the discrete character of the transformation of the states, and along with it the transition dipole moments

$$\tilde{\mathbf{d}}_1=\cos(\varphi/2)\mathbf{d}_1+\sin(\varphi/2)\mathbf{d}_2,$$
$$\tilde{\mathbf{d}}_2=-\sin(\varphi/2)\mathbf{d}_1+\cos(\varphi/2)\mathbf{d}_2 \tag{5}$$

preserves the sum of squares of the dipole moments of the system. Referenced to a single state, it remains equal to d_0^2. The mixing angle of the states φ in (5) is given by

$$\mathrm{tg}(\varphi)=V_{12}/(\varepsilon_1-\varepsilon_2), \tag{6}$$

$$V_{12}=(\mathbf{d}_1\mathbf{d}_2-3(\mathbf{d}_1\mathbf{s})(\mathbf{d}_2\mathbf{s}))/R_{12}^3. \tag{7}$$

By representing the interaction in pairwise form, Levitov[3] was able to formulate a renormalization-group equation with a transparent structure for the distribution function of collective dipole moments $P(\mathbf{d},R)$, which describes the evolution of this function as the interaction V is turned on with increasingly more distant states. This equation can be written in the form

$$\partial P(\mathbf{d},\xi)/\partial\xi=(\chi/d_0^2)\int d\mathbf{d}_1 d\mathbf{d}_2 P(\mathbf{d}_1,\xi)P(\mathbf{d}_2,\xi)d_1d_2F$$
$$\times(\mathbf{n}_1\mathbf{n}_2)\int_{-\pi/2}^{+\pi/2}(d\varphi/\sin^2\varphi)\{\delta(\tilde{\mathbf{d}}_1-\mathbf{d})$$
$$+\delta(\tilde{\mathbf{d}}_2-\mathbf{d})-\delta(\mathbf{d}_1-\mathbf{d})-\delta(\mathbf{d}_2-\mathbf{d})\}, \tag{8}$$

$$F(\mathbf{n}_1\mathbf{n}_2)=\int(d\mathbf{s}/4\pi)|\mathbf{n}_1\mathbf{n}_2-3(\mathbf{n}_1\mathbf{s})(\mathbf{n}_2\mathbf{s})|, \tag{9}$$

where

$$\xi=\ln(R/\bar{R}). \tag{10}$$

In principle, the distribution function P depends on energy. However, under condition (1) only states with nearby energies can interact effectively, i.e., states for which the energy difference is small compared to $g^{-1}(\varepsilon)$. The energy density function itself remains essentially constant. Therefore, all quantities in (8) are defined at the same energy, which dictates the value of the parameter χ in (1).

We are interested in "nonstationary" solutions to Eq. (8) for finite values of ξ. The structure of the kernel of the integral equation, which contains the product d_1d_2, is responsible for a peculiarity in the behavior of the function $P(\mathbf{d},\xi)$ when $d\ll d_0$. Actually, as $d\to 0$, the outgoing term tends to zero, whereas the incoming term remains finite. This type of nonequilibrium is maintained until the accumulation of small dipole moments leads to a dependence $P(\mathbf{d},\xi)\sim 1/d$. The smaller d is, the larger the distances ξ that must be included in order to form states with small dipole moments. The finiteness of the coherent region freezes the distribution for small d in its nonequilibrium form. As $\xi\to+\infty$, the "equilibrium" distribution function, taking into account that the sum of the squares of the dipole moments is conserved in any resonant restructuring of the states, has the form[3]

$$P(\mathbf{d})=(a/d)\exp(-d^2/b). \tag{11}$$

This result is exact if the dipole moments are oriented only along one direction (e.g., due to anisotropy of the crystal field), and the factor (9) has the fixed value $F=4/3^{3/2}\approx 0.75$. In the general case, F varies weakly[3] over the range 0.65 to 0.75; replacing it by a certain average value, we once again obtain relation (11). Direct numerical calculations show that as $d\to 0$, the solution approaches a value very close to (11) when the functional dependence (9) is preserved.

The coefficients a and b in (11) are found directly from the normalization condition

$$\int d\mathbf{d}P(\mathbf{d},\xi)=1 \tag{12}$$

and the conservation of the mean square of the dipole moment

$$\int d\mathbf{d}P(\mathbf{d},\xi)d^2=d_0^2. \tag{13}$$

Equation (8) automatically ensures that these conditions are satisfied for arbitrary ξ.

Let us turn to the solution of Eq. (8) in the region $d\ll d_0$. We will verify below that in this case angles φ that are small compared with unity are important. As follows from (5), here one of the two interacting states must possess a small dipole moment (for definiteness we choose $\mathbf{d}_1$), while the other can have a relatively arbitrary value $\tilde{d}_0$. We use the symmetry of the expression under the integral sign in (8) with respect to the interchange $\mathbf{d}_1\leftrightarrow\mathbf{d}_2$, and replace F by a certain average value $\bar{F}$ to simplify the analysis. If we include $\bar{F}$ in the definition of χ, then Eq. (8) can be converted to the form (see Ref. 5):

$$\partial P(\mathbf{d},\xi)/\partial\xi=(2\chi/d_0^2)\int d\mathbf{d}_2P(\mathbf{d}_2,\xi)d_2\int_{-\infty}^{+\infty}(d\varphi/\varphi^2)$$
$$\times[|\mathbf{d}-\mathbf{d}_2\varphi/2|P(\mathbf{d}-\mathbf{d}_2\varphi/2,\xi)-dP(d,\xi)]. \tag{14}$$

Further simplifications can be made by introducing a new variable $\phi=d_2\varphi/2$ and using Eq. (13):

$$\partial P(\mathbf{d},\xi)/\partial\xi=\chi\int(d\mathbf{n}_2/\Omega)\int_{-\infty}^{+\infty}(d\phi/\phi^2)$$
$$\times[|\mathbf{d}-\mathbf{n}_2\phi|P(\mathbf{d}-\mathbf{n}_2\phi,\xi)-dP(d,\xi)]. \tag{15}$$

Here $\Omega=4\pi$ when the dimensionality of the dipole moment space $n=3$; $\Omega=2\pi$ for $n=2$; and in the case $n=1,\Omega=2$, we replace the integral over $d\mathbf{n}_2$ with a sum over the two directions of the dipole moment.

Equation (15), which is now linear, has a particular solution with variables separable. This solution can be written in the form

$$\exp(\alpha\ln(d_0/d))\exp(\beta\xi), \tag{16}$$

where α and β are related by

$$\beta=\chi\int (d\mathbf{n}_2/\Omega_n)\int_{-\infty}^{+\infty} dy/y^2[|\mathbf{n}-\mathbf{n}_2 y|^{1-\alpha}-1]. \tag{17}$$

In Eq. (17) we have introduced the new variable

$$y=\phi/d. \tag{18}$$

The even function under the integral sign ensures that there is no singularity at $y=0$ in (17). By going into the complex plane and treating the integral as a principal value, we find for the case $n=1$

$$\beta=(\pi\chi)(1-\alpha)\mathrm{ctg}(\pi\alpha/2). \tag{19}$$

For $n=3$, carrying out first the integration over $d\mathbf{n}_2$, we obtain

$$\beta=(2-\alpha)\pi\ \mathrm{ctg}(\pi\alpha/2)/2. \tag{20}$$

It is not difficult to verify directly that even for $\chi\xi\gtrsim 1$, the distribution $P(\mathbf{d},\xi)$ will be close to uniform. Therefore, in treating the problem for $\chi\xi\gg 1$, for simplicity we can choose as our initial condition $P(\mathbf{d},\xi)=P_0$. Then the solution to Eq. (17) can be written in the form

$$P(\mathbf{d},\xi)=\oint (d\alpha/2i\pi\alpha)\exp(\alpha\ \ln(d_0/d))\exp(\beta(\alpha)\xi), \tag{21}$$

where the integral is taken along a contour in the complex domain that includes the coordinate origin ($|\alpha|<2$). We emphasize that this is the solution to the general Eq. (8) when $d\ll d_0$.

As we will verify below, regions where the value of d is sufficiently small correspond to the inequality $\alpha\ll 1$. In this case

$$\beta\approx 2\chi/\alpha. \tag{22}$$

By substituting this relation into (21) and making the change of variables

$$z=\alpha[\ln(d_0/d)/2\chi\xi]^{1/2}, \tag{23}$$

we can transform Eq. (21) to the form

$$P(\mathbf{d},\xi)=\oint (dz/2\pi iz)\exp\{\eta(z+1/z)\}. \tag{24}$$

Here

$$\eta[2\ \ln(d_0/d)\chi\xi]^{1/2}. \tag{25}$$

Choosing the contour of integration in (23) to correspond to the unit circle $z=e^{i\theta}$, we find

$$P(\mathbf{d},\xi)=(P_0/\pi)\int_0^{\pi} d\theta e^{2\eta^{\cos(\theta)}}=P_0 I_0(2\eta), \tag{26}$$

where I_0 is the Bessel function of imaginary argument. Keeping in mind that $\eta\gg 1$, and using the well-known asymptotic form of the Bessel function, we are led to the following final expression for the distribution function:

$$P(\mathbf{d},\xi)=(P_0/2\pi^{1/2}\eta^{1/2})e^{2\eta}. \tag{27}$$

The condition $|\alpha|\ll 1$, which we used in deriving (26), corresponds to the inequality

$$\ln(d_0/d)\gg\chi\xi, \tag{28}$$

which in turn corresponds to the region of small dipole moments whose upper limit decreases as ξ increases. In this region, the distribution of dipole moments differs radically from the distribution (22). The increase in the distribution function with decreasing d reflects the fact mentioned above that for $d\ll d_0$ the outgoing probability is $\sim d$, whereas the incoming probability does not depend on d. Let us now verify the assumption that the effective values of the mixing angle φ are small.

In calculating $\beta(\alpha)$ from (17) we find that the integral builds up for $y>1$, $\ln(y)\approx 1/\alpha$ for small α. From the definition (23) it follows that on the contour $|z|=1$ chosen to calculate the integral in (24),

$$|\alpha|=\eta/\ln(d_0/d).$$

Returning to the definition (18) and keeping in mind that $d_2\sim d_0$, we find

$$\phi\sim yd/d_0\approx\exp[-\ln(d_0/d)+(1/\eta)\ln(d_0/d)].$$

Since $d\ll d_0$ and $\eta\gg 1$, from this it quickly follows that $\varphi\ll 1$.

At the limit of applicability, where $\ln(d/d_0)\simeq 4\chi\xi$, the solution (22) approaches the "equilibrium" value $P\sim 1/d$. It is natural to assume that the transition to the equilibrium distribution is realized in the limit opposite to (27). In order to make the discussion as transparent as possible, let us investigate the case $n=1$ in detail.

Let us use Eq. (19), and rewrite the solution (21) in the form

$$P(\mathbf{d},\xi)=P_0(d_0/d)\oint (d\alpha/2i\pi\alpha)\exp\{-(1-\alpha)\ln(d_0/d)+\xi\chi\pi(1-\alpha)\mathrm{tg}[\pi(1-\alpha)/2]\}. \tag{29}$$

We will choose the contour of integration corresponding to the unit circle $\alpha=e^{i\phi}$, and compute the integral in the limit $\xi\chi\gg\ln(d_0/d)\gg 1$. Direct analysis shows that the real part of the exponent in (29) is negative over the entire contour and has a minimum for $\phi=0$. The extremum lies in the region where $\phi\ll 1$. In this limit, the exponent equals

$$-\xi\chi(\pi^2/2)\phi^2-i\phi\ \ln(d_0/d).$$

Calculating the integral in (29), we find

$$P(\mathbf{d},\xi)=P_0(d_0/d)(1/\pi^{3/2})(2\chi\xi)^{-1/2}\times\exp[-\ln^2(d_0/d)/2\pi^2\chi\xi]. \tag{30}$$

Since $|\alpha|=1$, the characteristic values of y in the integral (17) are of order 1. Therefore, the characteristic values of the mixing angle φ [see (18)] are

$$\varphi\simeq yd/d_0\ll 1,$$

and the original assumption of smallness of φ remains correct.

If we put the factor d_0/d back into the exponential, its exponent takes the form

$$\ln(d_0/d)[1-\ln(d_0/d)/2\pi^2\chi\xi].$$

From this it is clear that the transition to equilibrium behavior $P \propto 1/d$ is characterized by the parameter $\ln(d_0/d)/\pi^2\chi\xi$. The transition is actually complete for $d \gg d_0 \times \exp(-\pi\sqrt{\chi\xi})$.

Thus, strictly speaking, the $1/d$ law is found to be violated for the range of dipole moments

$$d<d_1, \quad d_1=d_0 \exp(-\pi\sqrt{\chi\xi}). \tag{31}$$

This range has two intervals separated by a value d_*, given by

$$d_*=d_0 e^{-4\chi\xi}. \tag{32}$$

For $d<d_*$ the distribution of dipole moments is determined by Eqs. (27) and (25), while for $d_*<d<d_1$ it is determined by Eq. (30). It is not difficult to write down an interpolation expression that encompasses both intervals:

$$P(\mathbf{d},\xi)=P_0(1/2\pi^{1/2})(2\chi\xi)^{-1/2}[1+\ln(d_0/d)/2\chi\xi]^{-1/4} \times \exp\{\ln(d_0/d)/(1+\ln(d_0/d)/8\chi\xi)^{1/2}\} \tag{33}$$

(the limiting expressions can be distinguished from those found above by the replacement of $\pi^2/4$ by 2).

For $d>d_1$ the distribution function has the equilibrium structure (11). When the spaces of the dipole moments are three-dimensional or two-dimensional, the small-d phase volume in Eqs. (12) and (13) causes the normalization to be essentially determined by the region $d \sim d_0$. Therefore, the coefficients in "a" and "b" in (11) are close to their values for the case of complete equilibrium:

$$n=3: \quad a=1/(2\pi d_0^2), \quad b=d_0^2; \quad n=2:$$
$$a=1/(\pi^{3/2}d_0), \quad b=2d_0^2. \tag{34}$$

For $n=1$ the situation turns out to be more complicated, since the equilibrium distribution function predetermines that the normalization interval (12) will diverge logarithmically at its lower limit. Because of this divergence, it is necessary to treat the system as one with a coherence region of finite size ξ_L from the outset; that size, in particular, is associated with the finite geometric dimensions (there is no problem with this in the integral (13), which, as before, is determined by the region of large d).

The normalization integral for the distribution function (27) gives a contribution $\sim(\xi_L\chi)\exp(-4\xi_L\chi)$ on the interval $(0, d_*)$. This contribution is small compared to 1. We must keep in mind that the region where the solution (30) is correct overlaps the equilibrium region. Therefore, the coefficient "a" must be equal to

$$a \approx (1/(2\pi)^{3/2})(1/(\chi\xi_L)^{1/2}), \tag{35}$$

since $2P_0d_0=1$. Estimating the contribution of the interval $(0,d_0)$ to the integral (13), we see that it is of order $ad_0^2 \ll d_0^2$. The main contribution comes from the region $(d_0,+\infty)$; accordingly, we have

$$ab \approx d_0^2.$$

From this

$$b \approx d_0^2(2\pi)^{3/2}(\chi\xi_L)^{1/2}. \tag{36}$$

Thus, for $n=1$, in addition to the peculiarities in the behavior for small d, a quasi-equilibrium blurring of the distribution function appears in the direction of dipole moments that are large compared with d_0 when $\chi\xi_L \gg 1$.

Let us make one remark here. As a direct analysis of Eq. (8) for $n=1$ shows, when

$$d>d_0/(\chi\xi)^{3/4} \gg d_1$$

the "incoming" term becomes important due to pairwise interactions between states with large dipole moments. In other words, the assumption that small angles φ play a decisive role in (8) is violated. This in no way changes the results given above, since the distribution reaches its equilibrium value even for $d>d_1$. For $n=3$, the small-angle approximation breaks down, in fact, for $d>d_*$. Therefore, in order to describe the transitional region, we must find the solution to the general Eq. (8).

3. RETARDED LUMINESCENCE

The time dependence of secondary emission that accompanies light scattering by a medium with randomly distributed resonant centers depends on the distribution of dipole moments of the collective states. Assume that a light pulse is incident on such a medium with a frequency distribution $\Phi(w)$ for the field. Let us consider a medium with a high density of centers, i.e., that satisfies the condition

$$n\lambda\!\!\!^{-3} \gg 1. \tag{37}$$

In this case, an excited state that is delocalized due to the resonant dipole–dipole interaction possesses a set of dipole moments with the distribution found in the previous section. If we consider single scattering only, we have a natural bound on the thickness of the medium $L<l$, where l is the mean free path of the photons. The scatter of resonance levels Γ in the absence of foreign defects is given by $\Gamma \sim nd_0^2$. If the frequency distribution of the incident pulse lies in this interval, then the mean free path will turn out to be of the same order as the wavelength $\lambda\!\!\!^{-}$. As we detune from resonance, the mean free path can become considerably longer; however, in this case it is necessary to include spatial phase in the coherent part of the scattering. Let us consider the case $L \lesssim \lambda\!\!\!^{-}$, and then discuss a straightforward generalization to the case $L \sim l \gg \lambda\!\!\!^{-}$. We will assume that in all cases $\chi\xi_L \gg 1$ while $\chi<1$.

Up to an unimportant coefficient, the time dependence of the intensity of the scattered component can be written in the form

$$I(t)=A\left|\int_{-\infty}^{+\infty} d\omega\Phi(\omega)\sum_\alpha \frac{\gamma_\alpha e^{-i\omega t}}{\omega-\varepsilon_\alpha+i\gamma_\alpha}\right|^2. \tag{38}$$

Here α labels a collective state with characteristic frequency ω_α and dipole moment d_α, $\gamma_\alpha=(2/3)d_\alpha^2\omega_0^3$ is the radiative width of the state α.

Let the incident radiation have a Lorentzian distribution with center ω_* and width γ_*. Then by calculating the integral in (38) we find

$$J_1=\sum_\alpha \int_{-\infty}^{+\infty} d\omega\Phi(\omega)\frac{\gamma_\alpha e^{-i\omega t}}{\omega-\varepsilon_\alpha+i\gamma_\alpha}=-\sum_\alpha \left\{e^{-i\omega_* t-\gamma_* t}/[\varepsilon_\alpha-\omega_*-i(\gamma_\alpha+\gamma_*)]\right.$$

$$-i\frac{2\gamma_\alpha\gamma_*}{[\varepsilon_\alpha-\omega_*-i(\gamma_\alpha+\gamma_*)]\,[\varepsilon_\alpha-\omega_*+i(\gamma_\alpha+\gamma_*)]}$$

$$\left.\times\exp(-i\varepsilon_\alpha t-\gamma_\alpha t)\right\}. \tag{39}$$

Substituting this expression into (38), we can determine the individual incoherent I_{nc} (diagonal in mode index) and coherent I_c components of the scattered radiation. We will assume that

$$\gamma_\alpha \ll \gamma_* . \tag{40}$$

Let us first consider the incoherent radiation, keeping only the component that decays slowly with time:

$$I_{nc}=A\sum_\alpha \frac{4\gamma_\alpha^2\gamma_*^2}{(\varepsilon_\alpha-\omega_*)^2+\gamma_*^2}\exp(-2\gamma_\alpha t). \tag{41}$$

For $\chi\xi_L \gg 1$ the sum over α reduces in fact to an integration over the distribution of levels $g(\varepsilon)$ and to an independent average over the dipole moments. If the scatter is determined only by the dipole–dipole interaction, then its characteristic width is dictated by the quantity $\Gamma \simeq nd_0^2$, and the distribution is Lorentzian in character. Let us keep the assumption of this character for the distribution in the general case. We will deal with the two cases $\gamma_* \ll \Gamma$ and $\gamma_* \gg \Gamma$.

In the first case we have

$$I_{nc}(t)=A(2\Gamma/\gamma_*)(nL^3)\{\gamma_0^2/[(\omega_0-\omega_*)^2+\Gamma^2]\} \times \int d\mathbf{d}P(d,\xi_L)(d/d_0)^4\exp(-2\gamma_0 t(d/d_0)^2), \quad \gamma_* \ll \Gamma. \tag{42}$$

In the second case,

$$I_{nc}(t)=A(nL^3)4\gamma_0^2\gamma_*^2/[(\omega_0-\omega_*)^2+\gamma_*^2]^2 \times \int d\mathbf{d}P(d,\xi_L)(d/d_0)^4\exp(-2\gamma_0 t(d/d_0)^2), \quad \gamma_* \gg \Gamma. \tag{43}$$

In considering the coherent component, the average must be carried out over amplitudes (39). In this case we immediately discover that deexcitation in the coherent channel takes place rapidly with the characteristic time $1/\Gamma$ or $1/\gamma_*$. Since both of these quantities are independent of d, we can use Eq. (13). Then, once more examining the two cases, we have finally

$$I_c(t)=A(nL^3)^2\{\gamma_0^2/[(\omega_0-\omega_*)^2+\Gamma^2]\}\exp(-2\gamma_* t), \quad \gamma_* \ll \Gamma. \tag{44}$$

$$I_c(t)=A(nL^3)^2(4\gamma_0^2\gamma_*^2/[(\omega_0-\omega_*)^2+\gamma_*^2]^2\exp(-2\Gamma t), \quad \gamma_* \gg \Gamma. \tag{45}$$

Comparing these expressions with (42) and (43), we see that coherent deexcitation takes place much faster than incoherent deexcitation. Nevertheless, the integrals over time turn out to have the same order of magnitude for both components.

Thus, the retarded emission at times large compared to $\tau_0=1/2\gamma_0$ is associated entirely with incoherent scattering. As t increases, smaller and smaller values of the collective dipole moments are found to be responsible for secondary emission.

For $t > \tau_0(d_0/d_*)^2$ [see (32)] the integral in (42) and (43) builds up at dipole moments whose distribution is determined by Eqs. (27) and (25).

The asymptotic time dependence of the intensity in this case has a power-law structure of the form

$$I(t)\propto(\tau_0/t)^{(4+n)/2}\exp\{(2\ln(t/\tau_0)\chi\xi_L)^{1/2}\}, \tag{46}$$

where η, as before, is determined by the dimension of the dipole moment space.

For intermediate times

$$\tau_0 < t \ll \tau_0(d_0/d_*)^2 \tag{47}$$

the characteristic dipole moments in (42) and (43) correspond to the distribution $P(\mathbf{d},\xi_L)\propto 1/d$, and accordingly,

$$I(t)\sim(\tau_0/t)^{(3+n)/2}. \tag{48}$$

Thus, there is a change in the asymptotic time behavior, which is more marked the smaller the dimensionality.

At significant offsets from resonance (i.e., $|\omega_*-\omega_0|$ large), or for $\Gamma \gg nd_0^2$, the mean free path increases compared to λ; moreover, the extent L of the medium may be enlarged. In this case we can retain all the results for the incoherent component of the scattering. In calculating the coherent part it is now necessary to take into account the phase factor $\exp[i(\mathbf{k}-\mathbf{k}')\mathbf{r}_\alpha]$ in the scattering amplitude. From this it follows that the coherent scattering will be concentrated over a narrow range of angles near the forward direction. In this case the extent of this concentration is a function of the total particle number; however, the statement regarding the relation between the intensities of coherent and incoherent remains unchanged.

Another example of retarded luminescence relates to pulsed excitation involving a third level. Decomposing the local state (i) that appears in terms of the collective modes according to $|i\rangle=\Sigma_\alpha c_{i\alpha}|\alpha\rangle$, we have for the probability that the excitation will not relax before time t

$$W_i(t)=\sum_{\alpha} |c_{i\alpha}|^2 \exp(-2\gamma_\alpha t).$$

Let us average over the position of the excited center and determine the intensity of the emission, taking the derivative of $W(t)$ with respect to time. Assuming that the size of the system does not exceed the mean free path of the photon, we find

$$I(t)\sim\gamma_0 \int d(\mathbf{d})P(d,\xi_L)(d/d_0)^2 \exp\{-2\gamma_0 t(d/d_0)^2\}. \tag{49}$$

For $t>\tau_0(d_0/d_*)^2$ the intensity will be determined by Eq. (46), in which we make the following replacement of exponents:

$$(4+n)/2\to(2+n)/2,$$

for the intermediate asymptotic form (47) we use an expression of the form (48) with the replacement

$$(3+n)/2\to(1+n)/2.$$

4. CONCLUDING REMARKS

The results given here indicate that when the density of resonant scatterers is large ($n\chi^3\gg1$), a universal distribution of collective dipole moments is formed. This distribution, and the optical properties of the medium determined by it, depends only on the spectral density of scatterers (through the parameter χ), the effective dimensionality of the space of dipole moments, and the size of the coherent region. This universality is absent when the density of scatterers is small, i.e., when $n\chi^3\ll1$. In this case, the light is actually scattered independently by isolated centers, and the optical properties of the medium are determined by the original parameters of the scatterers.

The role of universality is strongly evident in the example discussed here, i.e., the temporal behavior of the retarded luminescence. In fact, for scattering in a disordered material the power-law decrease in the retarded luminescence with time turns out to be sensitive only to the dimensionality of the dipole moment space.

The power-law nature of the time dependence of retarded luminescence was observed in experiments by Watanabe *et al.*[6] and Sturge *et al.*[7]

Note one important circumstance. When the space of dipole moments is one-dimensional, the spectrum of relaxation times ($1/\tau=\alpha d^2$) of the collective states becomes logarithmic:

$$P(\tau)\propto \int d(d)/d\,\delta(\tau-1/[\alpha d^2])=1/2\tau. \tag{50}$$

It is well known that the presence of a logarithmic spectrum of relaxation times is a sufficient condition to explain phenomena such as $1/f$ noise. Since interactions between defect centers (for example, mediated by a strain field) in a medium are, as a rule, subject to a $1/R^3$ law, we cannot dismiss the possibility that this result could be nonrandom.

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