Drastic effects of damping mechanisms on the third-order optical nonlinearity

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We have investigated the optical response of superradiant atoms, which undergoes three different damping mechanisms: radiative dissipation ($\gamma_r$), dephasing ($\gamma_d$), and nonradiative dissipation ($\gamma_n$). Whereas the roles of $\gamma_d$ and $\gamma_n$ are equivalent in the linear susceptibility $\chi^{(1)}$, the third-order nonlinear susceptibility $\chi^{(3)}$ drastically depends on the ratio of $\gamma_d$ and $\gamma_n$: When $\gamma_d \ll \gamma_n$, $\chi^{(3)}$ is essentially that of a single atom. Contrarily, in the opposite case of $\gamma_d \gg \gamma_n$, $\chi^{(3)}$ suffers the size-enhancement effect and becomes proportional to the system size.

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There has been much interest in optical responses of finite-sized systems. When the eigenstates of the system are delocalized in space, the transition dipole moment between the ground state and the lowest excited state scales as $N^{1/2}$, where $N$ is a parameter representing the system size [1]. This $N^{1/2}$ scaling rule is the origin of unique optical responses of finite-sized systems, such as size-enhancement of third-order optical susceptibility, $\chi^{(3)} \gg \chi^{(1)} \chi^{(1)} \chi^{(1)}$. In conventional theories on the size-effects of optical responses, whereas the size-dependences on eigenenergies and transition dipole moments were carefully taken into account, damping effects were often treated rather crudely by simply introducing phenomenological damping constants. However, because the system size would affect not only eigenenergies and transition dipole moments but also damping rates, more rigorous treatment on the damping effects is desired. Particularly, when the material is irradiated by resonant light fields, the magnitude of the optical nonlinearity is strongly sensitive to the damping rates. It is therefore indispensable to exclude phenomenology on the damping effects for quantitative evaluation of optical nonlinearity.

Here, targeting solid-state nonlinear optical devises in mind, we investigate the optical response of finite sized systems under a situation where the system suffers three different damping mechanisms: radiative dissipation, dephasing, and nonradiative dissipation. The latter two damping mechanisms are brought about by coupling to environmental degrees of freedom such as phonons. The effects of nonradiative dampings have been considered in detail in Ref. [2], but the damping constants are introduced by hand, independently of the model. (The radiative damping is incorporated through the self-consistent Maxwell fields in their formalism [3].) Spano et al. pioneered the theories without phenomenology on the damping effects, where the damping dynamics of the system is explicitly defined in the model [4, 5]. As for nonradiative damping effects, they introduced the homogeneous dephasing alone and no dissipation was explicitly treated. Although particular aspects of damping effects on the size-dependence of nonlinearity have been considered, the interplay of different damping mechanisms is still a subject of importance. To reveal the overall effects brought about by three different damping mechanisms, it is indispensable to treat the radiative and nonradiative dampings on equal footing and to make a clear distinction between dephasing and nonradiative dissipation. It is shown here that the third-order nonlinear response is drastically dependent on the ratio of two kinds of nonradiative damping rates: the ratio determines whether the size-enhancement of nonlinear response occurs or not. This fact implies that particular damping conditions could provide a novel resource for size-enhancement of nonlinear response.

The objective of this study is to investigate the third-order nonlinear optical response, taking account of different damping mechanisms explicitly. As a simplest model of a nonlinear optical system with finite size, we consider a superradiant system composed by $N$ identical two-level systems (hereafter referred to as “atoms”) with transition frequency $\Omega$ [6], which suffers, individually at each atom, both dephasing and nonradiative dissipation. Such damping mechanisms become particularly significant if the atoms are embedded in a solid-state environment, e. g., quantum dots in a microcavity. The equation of motion for the density matrix $\rho$ of atoms is the superradiant master equation [4, 6] supplemented with the terms describing dephasing and nonradiative dissipation [10]. It is given, omitting $\hbar$ and $\mu$ (transition dipole moment) for notational simplicity, by

$$\frac{d\rho}{dt} = -i[H_0 + H_{\text{int}}(t), \rho] + (L_r + L_d + L_n)\rho,$$

(1)

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where $s_j^\dagger$ and $s_j$ are the Pauli creation and annihilation operators at $j$th atom, and $S = \sum_j s_j$ is the collective operator. $E(t)$ represents the positive frequency part of the applied electric field, and the rotating wave approximation is used in $\mathcal{H}_{\text{int}}(t)$. $\gamma_r$, $\gamma_d$ and $\gamma_n$ represent the single-atom rates of radiative decay, dephasing, and nonradiative dissipation, respectively. It is of note that the atoms interact with the electromagnetic field via the collective operator [see Eqs. (3) and (4)] whereas dephasing and nonradiative dissipations occurs independently in each atom [see Eqs. (5) and (6)].

Based on this model, we investigate the linear and the third-order nonlinear optical responses. To the end of investigating up to third-order response, we are concerned with the following expectation values: $\langle s_i \rangle$, $\langle s_is_j \rangle$, $\langle s_j^\dagger s_j^\dagger \rangle$, and $\langle s_i^\dagger s_j^\dagger s_k^\dagger \rangle$, where the expectation value of an operator $A$ is given by $\langle A \rangle = \text{Tr} \{ \rho A \}$. Remembering the fact that all atoms are equivalent, the number of independent variables are greatly reduced. For example, it is apparent that $\langle s_i \rangle$ is independent of the site index $i$. We use the following notations:

\begin{align}
\langle s_i \rangle &= \langle s \rangle, \\
\langle s_is_j \rangle &= \begin{cases} 0 & (i = j) \\
\langle ss \rangle & (i \neq j) \end{cases}, \\
\langle s_j^\dagger s_j \rangle &= \begin{cases} \langle s's \rangle & (i = j) \\
\langle s's' \rangle & (i \neq j) \end{cases}, \\
\langle s_j^\dagger s_j s_k \rangle &= \begin{cases} 0 & (j = k) \\
\langle s'ss \rangle & (j \neq k, i = j \text{ or } k) \\
\langle s'ss' \rangle & (i \neq j, j \neq k, k \neq i) \end{cases}.
\end{align}

The equation of motion for $\langle A \rangle$ is given, using Eq. (11), by $\frac{d}{dt} \langle A \rangle = -i [\langle A \rangle, \mathcal{H}_0 + \mathcal{H}_{\text{int}}(t)] + (\gamma_r/2) \langle [S_+, A] S_- + S_+ [A, S_-] \rangle - (\gamma_d/2) \sum_j \langle [A, s_j^\dagger s_j], s_j^\dagger s_j \rangle \rangle + (\gamma_n/2) \sum_j \langle [s_j^\dagger, A] sj + s_j^\dagger [A, sj] \rangle$. Defining $\Gamma_{a,b,c}$ by

$$\Gamma_{a,b,c} = (a\gamma_r + b\gamma_d + c\gamma_n)/2,$$

the equations of motion for $\langle s \rangle$ etc are given as follows:

\begin{align}
d\langle s_1 \rangle/dt &= (-i\Omega - \Gamma_{N,1,1})\langle s_1 \rangle - iE, \\
d\langle ss \rangle/dt &= (-2i\Omega - \Gamma_{2N-2,2,2})\langle ss \rangle - 2iE\langle s_1 \rangle, \\
d\langle s's \rangle/dt &= (iE\langle s_1 \rangle + c.c.) - \Gamma_{2,0,2}(s's) - \Gamma_{2N-2,0,0}(s's'), \\
d\langle s's' \rangle/dt &= (iE^*\langle s_1 \rangle + c.c.) - \Gamma_{2,0,0}(s's) - \Gamma_{2N-2,2,2}(s's'), \\
d\langle s_3 \rangle/dt &= (-i\Omega - \Gamma_{N,1,1})\langle s_3 \rangle + 2iE\langle s_1 \rangle + \Gamma_{2N-2,0,0}(s's), \\
d\langle s's \rangle/dt &= (-i\Omega - \Gamma_{N+2,1,3})(s'ss) - \Gamma_{2N-4,0,0}(s's's') + iE\langle ss \rangle - iE(\langle s's \rangle + \langle s's' \rangle), \\
d\langle s's' \rangle/dt &= (-i\Omega - \Gamma_{3N-6,3,3})(s's's') - \Gamma_{4,0,0}(s'ss) + iE^*\langle ss \rangle - 2iE\langle s's' \rangle,
\end{align}

where $\langle s_1 \rangle$ and $\langle s_3 \rangle$ denotes the first- and third-order components of $\langle s \rangle$. Although not explicitly indicated, $\langle ss \rangle$, $\langle s's \rangle$ and $\langle s's' \rangle$ ($\langle s'ss \rangle$ and $\langle s's's' \rangle$) in the above equations are the second- (third-)order quantities. It is of note that, in the above equations of motion, the dependence on the system size $N$ appears only through the enhancement of $\gamma_r$.

We can easily obtain the stationary solutions of these simultaneous equations. Assuming that $E(t)$ is monochromatic as $E(t) \sim e^{-i\omega t}$, and introducing $f_{a,b,c}(\omega)$ by

$$f_{a,b,c}(\omega) = (\omega - \Omega + i\Gamma_{a,b,c})^{-1},$$

$\langle s_1 \rangle$, $\langle ss \rangle$, $\langle s's \rangle$ and $\langle s's' \rangle$ are given as follows:
\[ \langle s_1 \rangle = f_{N,1,1}(\omega)E, \]
\[ \langle ss \rangle = f_{N,1,1}(\omega)f_{N-1,1,1}(\omega)E^2, \]
\[ \frac{\langle s^\dagger s \rangle}{\langle s^\dagger s' \rangle} = \frac{N\gamma_d + \gamma_n + \gamma_n}{\gamma_d N + \gamma_n} \left( \frac{\gamma_d + \gamma_n}{\gamma_n} \right) \left| f_{N,1,1}(\omega) \right|^2 |E|^2. \]

Eq. \((22)\) shows that, in determining time-independent quantities such as \(\langle s^\dagger s \rangle\) and \(\langle s^\dagger s' \rangle\), the ratio of damping constants play crucial roles. This feature was also observed in conventional theories on the nonlinear susceptibilities using phenomenological damping constants. In terms of the second-order quantities, \(\langle s^\dagger ss \rangle\) and \(\langle s^\dagger ss' \rangle\) are given by
\[ \left( \begin{array}{cc}
\langle s^\dagger ss \rangle & \langle s^\dagger ss' \rangle \\
\langle s^\dagger ss' \rangle & \langle s^\dagger ss'' \rangle
\end{array} \right) = \left( \begin{array}{cc}
f_{N+1,1,3}(\omega) & \Omega_0 \\
\Omega_0 & \Omega_0
\end{array} \right) \left( \begin{array}{cc}
\langle s^\dagger s \rangle & \langle s^\dagger s' \rangle \\
\langle s^\dagger s' \rangle & \langle s^\dagger s'' \rangle
\end{array} \right)^{-1} \left( \begin{array}{cc}
-\Omega^2 \langle ss \rangle + \Omega^2 \langle s^\dagger s \rangle + \Omega^2 \langle s^\dagger s' \rangle \\
-\Omega^2 \langle s^\dagger s' \rangle + 2\Omega^2 \langle s^\dagger s'' \rangle
\end{array} \right), \]
and \(\langle s_3 \rangle\) is given, in terms of \(\langle s^\dagger s \rangle\) and \(\langle s^\dagger ss \rangle\), by
\[ \langle s_3 \rangle = f_{N,1,1}(\omega) \left[ -2\Omega^2 \langle s^\dagger s \rangle + i\Omega_0 \langle s^\dagger ss \rangle \right]. \]

Thus, we obtain the linear and third-order susceptibilities per one atom as follows:
\[ \chi^{(1)}(\omega) = \frac{\langle s_1 \rangle}{E} = f_{N,1,1}(\omega), \]
\[ \chi^{(3)}(\omega) = \frac{\langle s_3 \rangle}{|E|^2 E} = f_{N,1,1}(\omega) \left[ -2 \langle s^\dagger s \rangle \frac{\langle s^\dagger s \rangle}{|E|^2 E} + i\Omega_0 \langle s^\dagger ss \rangle \frac{\langle s^\dagger s \rangle}{|E|^2 E} \right], \]
both of which are free from phenomenological treatment on the damping effects. In the following part of this study, we discuss how \(\chi^{(3)}\) depends on the relaxation parameters \(\gamma_d, \gamma_n\) and the number \(N\) of atoms. Recent nanotechnologies aim to fabricate clean quantum systems with long coherence times, and extensive efforts on suppressing \(\gamma_d\) and \(\gamma_n\) are being made. In the following part of this study, we restrict our attention to a case where \(\gamma_d\) and \(\gamma_n\) are well suppressed and satisfy \(\gamma_d, \gamma_n \ll \gamma_r\).

Firstly, we discuss the limiting case of \(\gamma_d \to 0\). In this limit, it is easily confirmed that \(\langle s^\dagger s \rangle = \langle s^\dagger s' \rangle = \langle s^\dagger ss \rangle = \langle s^\dagger ss' \rangle = \langle s^\dagger ss'' \rangle\). These equalities imply that \(N\) atoms respond to the electric field cooperatively, as a spin \(N/2\) object. \(\chi^{(3)}(\omega)\) is reduced to the following form:
\[ \chi^{(3)}_{\gamma_d=0}(\omega) = -2f_{N,0,1}(\omega)^2 f_{N-1,0,1}(\omega) f_{0,0,1}(\omega). \]

This equation reveals that \(\chi^{(3)}_{\gamma_d=0}\) depends on the system size \(N\) only through the enhancement of \(\gamma_r\). In the off-resonant frequency regions, \(\chi^{(3)}_{\gamma_d=0} \sim -2/\omega^3\), which is independent of \(N\). Thus, in the limit of \(\gamma_d \to 0\), the optical nonlinearity is essentially that of a single atom, except for minor corrections around the resonant frequency region.

Next, we consider a more general case of \(\gamma_d \neq 0\). We should remark that, when \(\gamma_d, \gamma_n \ll \gamma_r\) is satisfied, \(\langle s^\dagger s \rangle\) and \(\langle s^\dagger s' \rangle\) are reduced to the following forms:
\[ \langle s^\dagger s \rangle \simeq \frac{N\gamma_d + \gamma_n + \gamma_n}{\gamma_d N + \gamma_n} \left| f_{N,1,1}(\omega) \right|^2 |E|^2, \]
\[ \langle s^\dagger s' \rangle \simeq \frac{N\gamma_n}{\gamma_d + \gamma_n} \left| f_{N,1,1}(\omega) \right|^2 |E|^2. \]

In case of \(\gamma_d \ll \gamma_n\), all of \(\langle ss \rangle\), \(\langle s^\dagger s \rangle\), and \(\langle s^\dagger s' \rangle\) are of the same order \(\sim \left| f \right|^2 |E|^2\), and the nonlinear susceptibility is given by Eq. \((27)\). Contrarily, in the opposite case of \(\gamma_d \gg \gamma_n\), \(\langle s^\dagger s \rangle\) becomes much larger than \(\langle ss \rangle\) and \(\langle s^\dagger s' \rangle\). \(\langle s^\dagger s \rangle \sim N\left| f \right|^2 |E|^2\), whereas \(\langle ss \rangle\) and \(\langle s^\dagger s' \rangle \sim \left| f \right|^2 |E|^2\). Then, Eqs. \((28)\) and \((29)\) suggest that \(\langle s^\dagger ss \rangle\), \(\langle s^\dagger ss' \rangle\), and \(\chi^{(3)}\) become almost proportional to \(\langle s^\dagger s \rangle\). Using the fact that \(\langle s^\dagger s \rangle\) is magnified by a factor \((N\gamma_d + \gamma_n)/\gamma_d + \gamma_n\) in comparison with the \(\gamma_d \to 0\) case, we obtain the following approximate expression of the third-order nonlinear susceptibility:
\[ \chi^{(3)}(\omega) \simeq \frac{N\gamma_d + \gamma_n}{\gamma_d + \gamma_n} \chi^{(3)}_{\gamma_d=0}(\omega). \]
In Fig. 1, the approximate susceptibility $\chi^{(3)}$ is compared with the rigorous susceptibility $\chi^{(3)}$. The figure demonstrates that $\tilde{\chi}^{(3)}$ serves as a good approximation of $\chi^{(3)}$.

Now we discuss the implications of Eq. (30). As far as the linear optical response is questioned, Eq. (25) indicates that the roles of dephasing ($\gamma_d$) and nonradiative dissipation ($\gamma_n$) are equivalent in determining the linear optical response. In contrast, when one questions the nonlinear optical response, the roles of two damping mechanisms are no more equivalent: The prefactor of the RHS of Eq. (30) indicates that magnitude of $\chi^{(3)}$ is sensitive to the ratio $\gamma_d/\gamma_n$, even if both $\gamma_d$ and $\gamma_n$ are much smaller than $\gamma_r$. When $\gamma_d \ll \gamma_n$, $\chi^{(3)}$ is essentially independent of the system size $N$. Contrarily, when $\gamma_d \gg \gamma_n$, $\chi^{(3)}$ suffers the size-enhancement effect. This observation demonstrates that it is indispensable for quantitative evaluation of nonlinear susceptibility to discriminate two damping mechanisms and to treat them non-phenomenologically.

One might feel uneasy about the fact that $\chi^{(3)}$ is indefinite at $\gamma_d = \gamma_n = 0$. In order to resolve this problem, we investigate the transient optical response by considering a situation where monochromatic field, $E(t) = E e^{-i\omega t}$, is switched on at $t = 0$. By inspecting Eqs. (14) and (15), we can find that $\chi^{(3)}$ has two relaxation rates, $N \gamma_c (= \tau_1^{-1})$ and $2 \gamma_d/N + 2 \gamma_n (= \tau_2^{-1})$, before attaining to its stationary value. The temporal behavior of $|\chi^{(3)}|$ is numerically
pursued in Fig. 2. The figure clarifies that $\chi^{(3)}$ first relaxes to the unenhanced value within a short time ($t < \tau_1$), and the size-enhancement effect emerges gradually in the later stage ($t \sim \tau_2$). When $\gamma_d = \gamma_n = 0$ (solid line in Fig. 2), the size-enhancement does not take place, and $\chi^{(3)} = \chi^{(3)}_{\gamma_d=0}$ forever. More generally, when one is concerned with a transient behavior ($t \sim \tau_2$), the size-enhancement effect is not expected; it is expected only after a long time, $t \gtrsim \tau_2$.

Finally, we comment on the relevance to previous studies. Mathematically, $\chi^{(3)}$ should be evaluated by the stationary solutions of equations of motion for $\langle s_i \rangle$, $\langle s^\dagger_i s_j \rangle$, etc. [5]. Because these quantities generally have dependence on the site index $i$, it is usually difficult to obtain analytic expression of $\chi^{(3)}$. (By taking the eigenstates of the system Hamiltonian $H_0$ as the basis, one may diagonalize the unitary part of Eq. (1). However, the basis generally does not diagonalize the damping part of Eq. (1) simultaneously [4]. The conventional expansion for $\chi^{(3)}$ [11] is obtained by approximately neglecting the off-diagonal part.)

In the model of our study, an analytic form of $\chi^{(3)}$ is obtained without any approximation by virtue of symmetry of the system, and it is revealed that tiny difference in the damping rates in Eqs. (14) and (15) may result in drastically different optical response, as observed in Fig. 2. A novel prediction in the present study is that the size-enhancement may take place, even when there is no transfer of excitations among the atoms.

In summary, we have analyzed the third-order nonlinear susceptibility of superradiant atoms, which undergoes three different damping mechanisms: radiative dissipation ($\gamma_r$), dephasing ($\gamma_d$), and nonradiative dissipation ($\gamma_n$). The analysis is based on the superradiant master equation supplemented with effects of dephasing and nonradiative dissipation [Eqs. (1)-(6)]. The linear susceptibility $\chi^{(1)}$ and the third-order susceptibility $\chi^{(3)}$ per one atom are given by Eqs. (25) and (26), and $\chi^{(3)}$ is well approximated by Eq. (31). Whereas the roles of $\gamma_d$ and $\gamma_n$ are equivalent in $\chi^{(1)}$ [see Eq. (25)], they are no more equivalent in $\chi^{(3)}$ [see Eq. (31)]: $\chi^{(3)}$ depends on the ratio $\gamma_d/\gamma_n$. When $\gamma_d \ll \gamma_n$, $\chi^{(3)}$ is essentially that of a single atom. Contrarily, when $\gamma_d \gg \gamma_n$, $\chi^{(3)}$ suffers the size-enhancement effect and becomes proportional to the system size $N$. These observations indicate that, for qualitative evaluation of $\chi^{(3)}$, it is indispensable to distinguish $\gamma_d$ and $\gamma_n$ clearly, and to handle them in a non-phenomenological manner.

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