Nonlinear optical response of cold atoms

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(Received 30 June 1995; revised manuscript received 18 September 1995)

The third-order optical susceptibility $\chi^{(3)}$ is calculated for a system of N cold atoms trapped in a three-dimensional harmonic potential. The dipole-dipole interaction between atoms contributes to $\chi^{(3)}$ reflecting quantum statistical properties of atoms. This contribution in Bose condensed systems is proportional to N^2 and is much larger than that in the system of Fermi atoms.

PACS number(s): 42.65.An, 05.30.−d, 32.80.Lg

In a system of laser-cooled atoms, which has been of great interest in the field of quantum optics recently, the thermal de Broglie wavelength can exceed the interatomic distance at very low temperatures and atoms behave as fully quantum-mechanical objects under such an extreme condition. We may therefore expect different behavior for ultracold Bose and Fermi atoms. In fact, the Bose-Einstein condensation in dilute Rb atoms has been observed [1]. So far, several theoretical investigations on optical responses of cold atoms such as light scattering and fluorescence have been reported [2–12]. The nonlinear optical probe is also expected to reveal the quantum statistical properties of cold atoms.

In this paper we calculate the third-order optical susceptibility $\chi^{(3)}$ of ultracold bosonic and fermionic atoms. We consider a system of N identical two-level atoms. For simplicity, we assume that the atoms are spinless. It is the nature of quantum statistics the atoms obey that plays an essential role and not the spin of the atoms. The relevant electronic transition of the atom is the transition from an s state (ground state $|g\rangle$) to threefold-degenerate p states [excited states $|e^{\alpha}\rangle$, $\alpha=(x,y,z)$] and the transition energy is $\hbar\omega_{eg}$. The magnitude of the dipole moment for the electronic transition of the atom is denoted by d. Atoms are trapped in an isotropic and harmonic-oscillator potential of the frequency ν. The states of an atom in the trapping potential are therefore denoted as $|g\rangle|\mathbf{n}\rangle$ for the ground-state potential and $|e^{\alpha}\rangle|\mathbf{n}\rangle$ for the excited-state potential, where $|\mathbf{n}\rangle$ is the eigenstate of the quantized center-of-mass motion of an atom with mass M in the trap. The index $\mathbf{n}$ is a triple index $\mathbf{n}=(n_x,n_y,n_z)$. The eigenenergy of $|\mathbf{n}\rangle$ is given by $\varepsilon_{\mathbf{n}}\hbar\nu(n_x+n_y+n_z+\frac{3}{2})$. Here we have assumed that the trapping potential is the same for ground- and excited-state atoms, although this is not the case in general. Atoms are coherently driven by a strong classical laser field of frequency Ω, wave vector $\mathbf{k}=(k,0,0)$, magnitude of the electric field E, and polarization parallel to $\hat{z}$. Atoms interact also with the electromagnetic field of the vacuum.

The important assumptions and limits in our model are as follows. First, the size of the trapping potential a is smaller than the wavelength of the incident laser field $\lambda_{\mathrm{inc}}=2\pi/k$ and that of the electronic transition of the atom $\lambda_{eg}=2\pi c/\omega_{eg}$. Namely, we take the Lamb-Dicke approximation. Second, the density of atoms is low, i.e., the atomic density $n=N/a^3$ satisfies $na_0^3\ll 1$, where a_0 is a typical atomic size. The atom-atom collisions can be neglected for the low-density atom gas in the Lamb-Dicke limit. The characteristic energy of the atom-atom collisions may be written as $E_c=na_s\hbar^2/M$, where a_s denotes the s-wave scattering length and is on the order of 10–100 Å for alkali-metal atoms. In order to neglect atom-atom collisions in the present model, E_c should be smaller than $\hbar\nu$ and the energy shift produced by the dipole-dipole interaction between atoms E_d. The energy E_d may be roughly estimated as $E_d=N\hbar\gamma_{\mathrm{rad}}$, where $\gamma_{\mathrm{rad}}=d^2\omega_{eg}^3/6\pi\hbar\varepsilon_0c^3$ is the natural linewidth of the electronic transition of the atom. For example, if we choose $a=1000$ Å, $a_s=100$ Å, $N=100$, and $M=100\ m_{\mathrm{H}}$, with m_{H} being the mass of a hydrogen atom, $E_c\approx 0.4\times 10^{-6}$ meV. This is much smaller than $\hbar\nu=\hbar^2/(Ma^2)\approx 0.4\times 10^{-2}$ meV and $E_d\approx 10^{-1}$ meV for $\hbar\gamma_{\mathrm{rad}}\approx 10^{-3}$ meV. Furthermore, the intensity of the incident laser field should be strong enough to satisfy $E_c\ll\hbar dE$. This condition can be easily achieved for off-resonant conditions ($\Omega<\omega_{eg}$). In the actual Bose condensate, however, the atom-atom collisions are crucial to prevent the fragmentation of the condensate into several nearly degenerate states of the trapping potential and to initiate the nucleation of the phase transition from a normal Boltzmann distribution [13]. Although the realistic model taking into account atom-atom collisions is preferable, our simple model may capture the essential feature of the dilute Bose and Fermi atom gases at zero or very low temperatures.

The system without the external laser field is described by the following master equation for the density matrix of the total system, i.e., the atomic system coupled to the electromagnetic field of the vacuum:

$$\frac{d}{dt}\rho(t)=\frac{1}{i\hbar}[H_A+H_B+H_I,\rho(t)], \tag{1}$$

where

$$H_A=\sum_{\mathbf{n}}\ \varepsilon_n c_{g\mathbf{n}}^{\dagger}c_{g\mathbf{n}}+\sum_{\mathbf{n},\alpha}\ (\varepsilon_n+\hbar\omega_{eg})c_{e\mathbf{n}}^{\alpha\dagger}c_{e\mathbf{n}}^{\alpha}, \tag{2}$$

$$H_B=\sum_{\mathbf{q},\lambda}\ \hbar\omega\mathbf{q}a_{\mathbf{q}\lambda}^{\dagger}a_{\mathbf{q}\lambda}, \tag{3}$$

and

$$H_I = -\kappa \sum_{\mathbf{n},\mathbf{n}'} \sum_{\mathbf{q}\mu,\alpha} \zeta^{\alpha}_{\mathbf{n}'\mathbf{n},\mu}(\mathbf{q}) c^{\alpha\dagger}_{e\mathbf{n}'} c_{g\mathbf{n}} a_{\mathbf{q}\mu} - \text{H.c.}, \tag{4}$$

with the Franck-Condon factor

$$\zeta^{\alpha}_{\mathbf{n}'\mathbf{n},\mu}(\mathbf{q}) = d\left(\frac{\hbar w_{\mathbf{q}}}{2\varepsilon_0 V}\right)^{1/2} e^{\alpha}_{\mathbf{q}\mu} \int d\mathbf{r}\, \exp(i\mathbf{q}\cdot\mathbf{r}) \psi^*_{\mathbf{n}'}(\mathbf{r}) \psi_{\mathbf{n}}(\mathbf{r}). \tag{5}$$

H_A and H_B represent, respectively, the Hamiltonian for the free atomic system and that for the electromagnetic field of the vacuum. H_I describes the interaction between them and κ ($\rightarrow 1$) denotes a formal expansion parameter. In Eqs. (3) and (4), $a_{\mathbf{q}\mu}$ ($a^{\dagger}_{\mathbf{q}\mu}$) denotes the annihilation (creation) operator for the photon of momentum $\mathbf{q}$, energy $\omega_{\mathbf{q}} = c|\mathbf{q}|$, and linear transverse polarization $\mathbf{e}_{\mathbf{q}\mu}$ ($\mu = 1,2$). In Eqs. (2) and (4), $c_{g\mathbf{n}}$ ($c^{\dagger}_{\mathbf{gn}}$) denotes atomic annihilation (creation) operator for the state $|g\rangle|\mathbf{n}\rangle$ and $c^{\alpha}_{e\mathbf{n}}$ ($c^{\alpha\dagger}_{e\mathbf{n}}$) denotes the atomic annihilation (creation) operator for the state $|e^{\alpha}\rangle|\mathbf{n}\rangle$. They satisfy the usual commutation or anticommutation relations depending on whether atoms are bosons or fermions. In Eq. (5), V is the quantization volume for the electromagnetic field of the vacuum and $\psi_{\mathbf{n}}(\mathbf{r}) = \phi_{n_x}(x)\phi_{n_y}(y)\phi_{n_z}(z)$ is the eigenfunction corresponding to the state $|\mathbf{n}\rangle$, where $\phi_n(x) = H_n(x/a)\exp(-x^2/2a^2)/\sqrt{a\sqrt{\pi}2^n n!}$ with the nth-order Hermite polynomial H_n and $a = \sqrt{\hbar/M\nu}$.

We follow the standard procedure of treating the vacuum as a reservoir and eliminating the degree of freedom of the vacuum field mode [14]. Taking the trace over variables of the vacuum field mode (B), we obtain the following master equation for the reduced density matrix $\sigma(t) = \mathrm{Tr}_{(B)}\rho(t)$:

$$\frac{d}{dt}\sigma(t) = \frac{1}{i\hbar}[H_A, \sigma(t)] + \kappa^2 \Lambda\sigma(t), \tag{6}$$

where the "damping term" $\Lambda\sigma(t)$ is calculated in the Born-Markov approximation as

$$\begin{aligned}\Lambda\sigma(t) = {} & i \sum_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3,\mathbf{n}_4} \sum_{\alpha,\beta} \Delta_{\alpha\beta}(\mathbf{n}_1,\mathbf{n}_2;\mathbf{n}_3,\mathbf{n}_4) \\ & \times \{c^{\alpha\dagger}_{e\mathbf{n}_2} c_{g\mathbf{n}_1} c^{\dagger}_{g\mathbf{n}_3} c^{\beta}_{\mathbf{en}_4}, \sigma(t)\}_- \\ & - \sum_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3,\mathbf{n}_4} \sum_{\alpha,\beta} \Gamma_{\alpha\beta}(\mathbf{n}_1,\mathbf{n}_2;\mathbf{n}_3,\mathbf{n}_4) \\ & \times [\{c^{\alpha\dagger}_{e\mathbf{n}_2} c_{g\mathbf{n}_1} c^{\dagger}_{g\mathbf{n}_3} c^{\beta}_{e\mathbf{n}_4}, \sigma(t)\}_+ \\ & - 2 c^{\dagger}_{g\mathbf{n}_1} c^{\alpha}_{e\mathbf{n}_2} \sigma(t) c^{\beta\dagger}_{e\mathbf{n}_4} c_{g\mathbf{n}_3}].\end{aligned} \tag{7}$$

Here $\{A,B\}_{\pm} = AB \pm BA$,

$$\begin{aligned}\Delta_{\alpha\beta}(\mathbf{n}_1,\mathbf{n}_2;\mathbf{n}_3,\mathbf{n}_4) = {} & \frac{1}{16\pi}\frac{d^2}{\hbar c^3 \varepsilon_0} \mathrm{P}\int_0^{\infty} d\omega \frac{\omega^3}{\omega - \omega_{eg}} \\ & \times \int d\mathbf{r} \int d\mathbf{r}'\ \Phi_{\alpha\beta}(\omega, \mathbf{r}-\mathbf{r}') \psi_{\mathbf{n}_1}(\mathbf{r}) \\ & \times \psi^*_{\mathbf{n}_2}(\mathbf{r}) \psi^*_{\mathbf{n}_3}(\mathbf{r}') \psi_{\mathbf{n}_4}(\mathbf{r}'),\end{aligned} \tag{8}$$

and

$$\begin{aligned}\Gamma_{\alpha\beta}(\mathbf{n}_1,\mathbf{n}_2;\mathbf{n}_3,\mathbf{n}_4) & \\ = {} & \frac{1}{16\pi^4}\frac{d^2}{\hbar c^3 \varepsilon_0} \omega_{eg}^3 \int d\mathbf{r} \int d\mathbf{r}' \\ & \times \Phi_{\alpha\beta}(\omega_{eg}, \mathbf{r}-\mathbf{r}') \psi_{\mathbf{n}_1}(\mathbf{r}) \psi^*_{\mathbf{n}_2}(\mathbf{r}) \psi^*_{\mathbf{n}_3}(\mathbf{r}') \psi_{\mathbf{n}_4}(\mathbf{r}').\end{aligned} \tag{9}$$

In Eqs. (8) and (9),

$$\Phi_{\alpha\beta}(\omega, \mathbf{r}) = \oint d\omega(\mathbf{q}) \exp(-i\mathbf{q}\cdot\mathbf{r}) \sum_{\mu=1,2} e^{\alpha}_{\mathbf{q}\mu} e^{\beta}_{\mathbf{q}\mu}, \tag{10}$$

where $d\omega(\mathbf{q})$ is the element of solid angle about $\mathbf{q}$ and P in Eq. (8) stands for the principle value. Here we assumed that the equilibrium temperature of the vacuum is zero. Furthermore, we neglected the difference between different $\varepsilon_{\mathbf{n}}$'s because ν is much smaller than ω_{eg}.

In order to calculate $\chi^{(3)}$, we add a perturbation term $[H_{\text{ext}}(t), \sigma(t)]/i\hbar$ to the right-hand side of Eq. (6) and solve the resulting equation for $\sigma(t)$ iteratively with respect to $H_{\text{ext}}(t)$ under the initial condition $\sigma(t \rightarrow -\infty) = |\Phi_0\rangle\langle\Phi_0|$; $\sigma(t) = |\Phi_0\rangle\langle\Phi_0| + \sigma^{(1)}(t) + \sigma^{(2)} + \cdots$. Here $|\Phi_0\rangle$ is the initial state of the system of N atoms. Since we are in the Lamb-Dicke limit, the incident laser field excites atoms coherently over the entire volume of the atomic system. Therefore, the interaction between the atomic system and the external incident laser field is written as

$$H_{\text{ext}}(t) = -\tfrac{1}{2}\hat{P}^{\dagger} E \exp(-i\Omega t) - \text{H.c.}, \tag{11}$$

with

$$\hat{P} = d \sum_{\mathbf{n}_1,\mathbf{n}_2} p^*(\mathbf{n}_1,\mathbf{n}_2) c^{\dagger}_{g\mathbf{n}_2} c^{z}_{e\mathbf{n}_1} \tag{12}$$

the polarization operator and $p(\mathbf{n}_1,\mathbf{n}_2) = \langle\mathbf{n}_1|e^{ikx}|\mathbf{n}_2\rangle$ the matrix element for the transition from the state $|g\rangle|\mathbf{n}_2\rangle$ to the state $|e^z\rangle|\mathbf{n}_1\rangle$. We assume that for the system of Bose atoms $|\Phi_0\rangle$ is the state where the lowest-energy state $|g\rangle|\mathbf{n}_1\rangle$ with $\mathbf{n}_1 = (0,0,0)$ is occupied by all N atoms coherently, i.e., a perfectly Bose condensed state, and for the system of Fermi atoms $|\Phi_0\rangle$ is the state where each of lowest N states is occupied by one atom. In both cases $|\Phi_0\rangle$ is the lowest-energy state for the system of N atoms. The "damping term" $\kappa^2\Lambda\sigma(t)$ in Eq. (6) is also treated perturbatively: $\sigma^{(n)} = \sigma^{(n)}_{(0)} + \kappa^2\sigma^{(n)}_{(1)} + \kappa^4\sigma^{(n)}_{(2)} + \cdots$. This turns out to be the expansion with respect to $N\gamma_{\text{rad}}/\delta$ with $\delta = \omega_{eg} - \Omega$, the off-resonance frequency. Therefore, this expansion can be carried out when the number of atoms is moderate or the off resonance is large.

The third-order induced polarization is calculated through $\sigma^{(3)}(t)$, the third-order part of $\sigma(t)$ with respect of $H_{\text{ext}}(t)$,

$$P^{(3)}[\Omega]=\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\ \exp(i\Omega t)\mathrm{Tr}[\hat{P}\sigma^{(3)}(t)]$$
$$=\sum_{\phi_0}\langle\phi_0|\hat{P}\sigma^{(3)}[\Omega]|\phi_0\rangle+\sum_{\phi_1}\langle\phi_1|\hat{P}\sigma^{(3)}[\Omega]|\phi_1\rangle, \tag{13}$$

where ϕ_0 denotes the state where no atoms are excited and ϕ_1 the state where only one atom is excited and the summation is done over all possible ϕ_0 and ϕ_1. Here we have assumed that the wave vector and the polarization of the probe field are the same as those of the pump (external incident) field and used the same notation $\hat{P}$ for the polarization operator for the pump field defined by Eq. (12) and that for the probe field appearing in Eq. (13). The third-order optical susceptibility is given by $\chi^{(3)}[\Omega]=P^{(3)}[\Omega]/(\varepsilon_0 E^3)$. In this paper, we calculate $\chi^{(3)}[\Omega]$ up to the first order of κ^2. It should be noted that $\chi^{(3)}[\Omega]$ thus defined is the third-order optical susceptibility for the entire N atom system and is not normalized by the volume of the atomic system.

First, we calculate the third-order optical susceptibility for Bose condensed atoms $\chi_B^{(3)}[\Omega]$. Most of the procedure is straightforward; however, the calculation of the matrix element $\langle\Phi_0|\sigma^{(2)}[0]|\Phi_0\rangle$ requires some caution. This is determined as shown below. It is easy to show that

$$\frac{d}{dt}\langle 0|\sigma_{(0)}^{(2)}(t)|0\rangle+\sum_{\mathbf{n}}\frac{d}{dt}\langle(\mathbf{n})_e|\sigma_{(0)}^{(2)}|(\mathbf{n})_e\rangle=0 \tag{14}$$

in the zeroth order of κ^2 and

$$\frac{d}{dt}\langle 0|\sigma_{(1)}^{(2)}(t)|0\rangle+\sum_{\mathbf{n}\,(\neq\mathbf{n}_1)}\frac{d}{dt}\langle(\mathbf{n})_g|\sigma_{(1)}^{(2)}(t)|(\mathbf{n})_g\rangle$$
$$+\sum_{\mathbf{n}}\frac{d}{dt}\langle(\mathbf{n})_e|\sigma_{(1)}^{(2)}(t)|(\mathbf{n})_e\rangle=0 \tag{15}$$

in the first order of κ^2. Here $|0\rangle=|\Phi_0\rangle=(c_{g\mathbf{n}_1}^{\dagger})^N|\mathrm{vac}\rangle/\sqrt{N!}$, $|(\mathbf{n})_g\rangle=c_{g\mathbf{n}}^{\dagger}(c_{g\mathbf{n}_1}^{\dagger})^{N-1}|\mathrm{vac}\rangle/\sqrt{(N-1)!}$, and $|(\mathbf{n})_e\rangle=c_{e\mathbf{n}}^{\dagger}(c_{g\mathbf{n}_1}^{\dagger})^{N-1}|\mathrm{vac}\rangle/\sqrt{(N-1)!}$ with $|\mathrm{vac}\rangle$ being a state with no atoms. Equations (14) and (15) yield, respectively,

$$\langle 0|\sigma_{(0)}^{(2)}[0]0\rangle=-\sum_{\mathbf{n}}\langle(\mathbf{n})_e|\sigma_{(0)}^{(2)}[0]|(\mathbf{n})_e\rangle \tag{16}$$

and

$$\langle 0|\sigma_{(1)}^{(2)}[0]|0\rangle=-\sum_{\mathbf{n}(\neq\mathbf{n}_1)}\langle(\mathbf{n})_g|\sigma_{(1)}^{(2)}[0]|(\mathbf{n})_g\rangle$$
$$-\sum_{\mathbf{n}}\langle(\mathbf{n})_e|\sigma_{(1)}^{(2)}[0]|(\mathbf{n})_e\rangle. \tag{17}$$

The matrix element $\langle(\mathbf{n})_g|\sigma_{(1)}^{(2)}[0]|(\mathbf{n})_g\rangle$ does not contribute to $P^{(3)}[\Omega]$ and $\langle(\mathbf{n})_e|\sigma^{(2)}[0]|(\mathbf{n})_e\rangle$ is given by

$$\langle(\mathbf{n})_e|\sigma^{(2)}[0]|(\mathbf{n})_e\rangle$$
$$=-\frac{N}{4}\frac{(dE)^2}{\hbar^2\delta^2}|p(\mathbf{n},\mathbf{n}_1)|^2+\frac{N}{4}\frac{(dE)^2}{\hbar^2\delta^3}$$
$$\times\sum_{\mathbf{n}'}\Bigg[p(\mathbf{n},\mathbf{n}_1)p^*(\mathbf{n}',\mathbf{n}_1)\Bigg((N-1)[\Delta_{zz}(\mathbf{n}_1,\mathbf{n};\mathbf{n}_1,\mathbf{n}')$$
$$-i\Gamma_{zz}(\mathbf{n}_1,\mathbf{n};\mathbf{n}_1,\mathbf{n}')]$$
$$+\sum_{\mathbf{n}''}[\Delta_{zz}(\mathbf{n}'',\mathbf{n};\mathbf{n}'',\mathbf{n}')-i\Gamma_{zz}(\mathbf{n}'',\mathbf{n};\mathbf{n}'',\mathbf{n}')]\Bigg)+\mathrm{c.c.}\Bigg]. \tag{18}$$

Here we assumed $\hbar\delta\gg\varepsilon_n$. By using Eqs. (16) and (17), $\chi_B^{(3)}[\Omega]$ is calculated as

$$\chi_B^{(3)}[\Omega]=-\frac{N}{4}\frac{d^4}{\varepsilon_0\hbar^3\delta^3}-\frac{N}{4}\frac{d^4}{\varepsilon_0\hbar^3\delta^4}\sum_{\mathbf{n}'}\sum_{\mathbf{n}''}p^*(\mathbf{n}',\mathbf{n}_1)p(\mathbf{n}'',\mathbf{n}_1)$$
$$\times\Bigg(2(N-1)[2\Delta_{zz}(\mathbf{n}_1,\mathbf{n}';\mathbf{n}_1,\mathbf{n}'')+i\Gamma_{zz}(\mathbf{n}_1,\mathbf{n}';\mathbf{n}_1,\mathbf{n}'')]+3\sum_{\mathbf{n}}\Delta_{zz}(\mathbf{n}_1,\mathbf{n};\mathbf{n}_1,\mathbf{n})+i\sum_{\mathbf{n}}\Gamma_{zz}(\mathbf{n}_1,\mathbf{n};\mathbf{n}_1,\mathbf{n})\Bigg). \tag{19}$$

The second term in large parentheses on the right-hand side of Eq. (19) exhibits the ultraviolet divergence. This term originates from Lamb shift, which can be formally renormalized to ω_{eg} so that we neglect it. We further simplify the above expression by assuming $a/\lambda_{eg},a/\lambda_{\mathrm{inc}}\to 0$ and $N\gg 1$. The result is

$$\chi_B^{(3)}[\Omega]=-\frac{Nd^4}{4\varepsilon_0(\hbar\delta)^3}\left[1+i\frac{2N}{\pi^2}\frac{\gamma_{\mathrm{rad}}}{\delta}+\frac{\gamma_{\mathrm{rad}}}{\delta}\left(\frac{\lambda_{eg}}{a}\right)^3\xi_B\right], \tag{20}$$

with $\xi_B=N\Delta(\mathbf{n}_1,\mathbf{n}_1)/4\pi^7$. The dimensionless quantity Δ is given by

$$\Delta(\mathbf{n},\mathbf{n}')=\prod_{\alpha=x,y,z}\frac{1}{2^{n_\alpha}n_\alpha!}\frac{1}{2^{n'_\alpha}n'_\alpha!}\int_0^\infty dt\ t^2\int d\mathbf{r}\int d\mathbf{r}'\ \exp(-|\mathbf{r}|^2-|\mathbf{r}'|^2)H_{n_x}^2(x)H_{n_y}^2(y)$$
$$\times H_{n_z}^2(z)H_{n'_x}^2(x')H_{n'_y}^2(y')H_{n'_z}^2(z')\left[2j_0(t|\mathbf{r}-\mathbf{r}'|)+j_2(t|\mathbf{r}-\mathbf{r}'|)\left(3\frac{|z-z'|^2}{|\mathbf{r}-\mathbf{r}'|^2}-1\right)\right], \tag{21}$$

the $j_n(x)$ being spherical Bessel function of the first kind.

Second, the third-order optical susceptibility for the system of Fermi atoms $\chi_f^{(3)}[\Omega]$ is obtained in the same way:

$$\chi_F^{(3)}[\Omega]=-\frac{Nd^4}{4\varepsilon_0(\hbar\delta)^3}\left[1+i\frac{2N}{\pi^2}\frac{\gamma_{\rm rad}}{\delta}+\frac{\gamma_{\rm rad}}{\delta}\left(\frac{\lambda_{eg}}{a}\right)^3\xi_F\right], \tag{22}$$

with

$$\xi_F=\frac{1}{4\pi^7}\frac{1}{N}\sum_{i=1}^{N}\sum_{j=1}^{N}\Delta(\mathbf{n}_i,\mathbf{n}_j),$$

where $\Delta(\mathbf{n}_i,\mathbf{n}_j)$ is also given by Eq. (21). The third-order optical susceptibility $\chi^{(3)}$ is composed of three terms. The first leading-order term $-0.25Nd^4/[\varepsilon_0(\hbar\delta)^3]$ originates from the simple fact that in the course of coherent excitation the degree of freedom to excite one atom in the ground-state potential is reduced from N to $N-1$. Therefore, this term does not reflect quantum statistical properties. The second term is the contribution of the spontaneous decay. The third term is the contribution of the dipole-dipole interaction induced by the electromagnetic field of the vacuum and explicitly depends on quantum statistics. For the system of Bose condensed atoms, ξ_B is calculated as $3\sqrt{2}N/4\pi^{7/2}\simeq 0.0193N$, while for the system of Fermi atoms ξ_F is estimated as 0.0368 for $N=35$, 0.0462 for $N=56$, and 0.0565 for $N=84$. Although ξ_F is also approximately proportional to N, it is much smaller than ξ_B. The reduction of ξ_F comes from the oscillating behavior of the single-particle wave function $\psi_{\mathbf{n}}(\mathbf{r})$ for large $\mathbf{n}$. When we choose $\delta=20N\gamma_{\rm rad}$, the contribution of spontaneous decay $2N\gamma_{\rm rad}/\pi^2\delta\approx 1\times 10^{-2}$, while that of the dipole-dipole interaction in Bose condensed system $\xi_B(\lambda_{eg}/a)^3\gamma_{\rm rad}/\delta\approx 1\times 10^{-3}(\lambda_{eg}/a)^3$, which becomes of the order of 1 for $a\approx 0.1\lambda_{eg}$. The second and third terms of $\chi^{(3)}$ are proportional to N^2 and this enhancement factor is due to the collective response of atoms located within a distance smaller than $\lambda_{\rm inc}$ and λ_{eg}, i.e., the superradiative character of atoms.

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