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The Doppler shifts of resonant fluorescence spectrum for a two-level ^{85}Rb atom via multiphoton Compton scattering

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Abstract. Usually, it is difficult to observe Compton scattering in an atom. One way to overcome this difficulty is to allow a multiphoton to collide with an atom. This phenomenon is called multiphoton Compton scattering (MCS) phenomenon. Thus, we can investigate the MCS in the visible light region. The cluster atoms move as a whole, namely atomic Dicke states, and the multiphoton interacts with cluster atoms. We can observe a significant Doppler shift of resonant fluorescence spectrum (RFS) in a room-temperature two-level ^{85}Rb atomic system. In this paper, we present a detailed analysis of the physics mechanism of the Doppler shift and propose a method to measure the component of the Dicke states (the atomic polymers with different masses) by using the Doppler shift of the RFS.

Keywords. Doppler shift; resonant fluorescence spectrum; multiphoton Compton scattering; two-level ^{85}Rb atomic system.

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1. Introduction

The Compton scattering phenomenon was first observed when a γ-ray collides with an electron [1]. The Compton wavelength of the electron is $\lambda_c = h/m_e c$, where m_e is the electron mass, h is the Planck constant, c is the speed of light. Substituting γ-ray by photon, we may investigate the collision of photon with an atom, where m is the atomic mass. As $h/mc \ll h/m_e c$, it is very difficult to observe single photon Compton scattering process. The only way to overcome this difficulty is to allow multiphoton to collide with an atom. The Doppler shift is dependent on the direction of observation and the mass of atoms. The physics mechanism and magnitude of the Doppler shift are quite different from that of the Doppler broadening. The Doppler broadening is independent of the direction of observation due to isotropy.

Since the invention of the laser in the early 1960s, we came to know that the laser beam is a type of electromagnetic wave which has high density of photons that may be in coherent states, the photon degeneracy up to $n = 5 \times 10^7$ [2–4]. Thus, we can observe the multiphoton Compton scattering (MCS) process in the visible light range. Considering many photons collide simultaneously with cluster atoms, namely the Dicke states [5,6], which are moving as a whole, the MCS process produce energy $nh\nu$ and momentum $n\hbar\vec{k}'$. The MCS can be represented by the Feynman diagram where the electron is treated as an undressed one, and the result can be applied only to a low intensity laser. In order to investigate the effect of coherency of the photon on Compton scattering, a formula based on coherent-state formalism is required.

When a two-level atom interacts with a single photon, the atom absorbs one photon moving from the ground state to the excited state, and then returns to the ground state again, radiating resonance fluorescence spectrum (RFS). The power spectrum of the light scattered by a two-level atom in the presence of a coherent continuous-pulse-train driving field is analysed [7]. Separate expressions for the coherent and incoher-

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ent components of the spectra are obtained [8,9]. The coherent part of the spectrum consists of a very narrow peak, without energy and momentum exchange during the collision of atoms and photon. The incoherent spectrum has energy–momentum exchange. That is, the spectrum consists of a series of triplets in RFS [10,11]. We consider a dense two-level ^{85}Rb atomic gas, supposed to be in equilibrium at room temperature T. Two kinds of phenomena are happening in RFS: (1) the random thermal motion of the radiating atoms induces Doppler broadening (isotropic), the magnitude of which is independent of the direction of observation. The Boltzmann constant $k = 1.380649\times10^{-23}$ J/K, the room temperature $T = 300$ K, the mass of proton $m_p = 1.6726231 \times 10^{-24}$ g, according to the atomic periodic table, the mass of rubidium atom $m_{\mathrm{Rb}} = 85 \times m_p = 1.4217296 \times 10^{-22}$ g, $c = 3.0 \times 10^8$ m/s, $(\Delta\nu/\nu)_{\mathrm{Db}} = \sqrt{2kT/m_{\mathrm{Rb}}}/c \approx 8.04 \times 10^{-7}$ is very small, may be neglected. (2) Collision of atoms with photons produces Doppler shift, the magnitude of which depends on the direction of observation and the atomic mass. Guo first investigated the Doppler shift and observed the recoil-induced resonances [12]. Stark first detected the Doppler shift for the light emitted by the moving hydrogen atoms [13]. Recently, Utsunomiya *et al* produced the quasimonochromatic γ-ray by the collision of photons with relativistic electrons [14]. Although the RFS has a quite complicated structure [15,16], a significant Doppler shift $(\bar{\nu}_{\mathrm{D}} - \nu)/\nu \approx 1.06 \times 10^{-3}$ can be observed. In this paper, we shall investigate the Doppler shift and the Dicke states using the MCS process.

The paper is organised as follows: In §2 we discuss MCS process and in §3 we investigate the RFS and DRFS (RFS with Doppler shift) of the two-level atomic system. In §4, we investigate the RFS and DRFS of atom Dicke state. Finally, in §5 conclusions are given.

2. The multiphoton compton scattering process

The incident photons move along the x-axis, the photon number states $|n\rangle$ and the eigenvalue n unchanged during the scattering process. We make the assumption that the atom with mass m stays in the static state, the initial velocity of atom is zero, the energy and momentum exchange satisfy the conservation equations of the participating particles [17]. Figure 1 shows the single-photon Compton scattering process. Suppose ν is the frequency of the photon before the collision, $\bar{\nu}$ is the frequency of the scattering atom, α is the angle between the direction of motion of the photon and the direction of motion of the scattered photon after the collision, the angle β is the angle between the direction of motion of the photon before and after the collision, that is, the scattering angle.

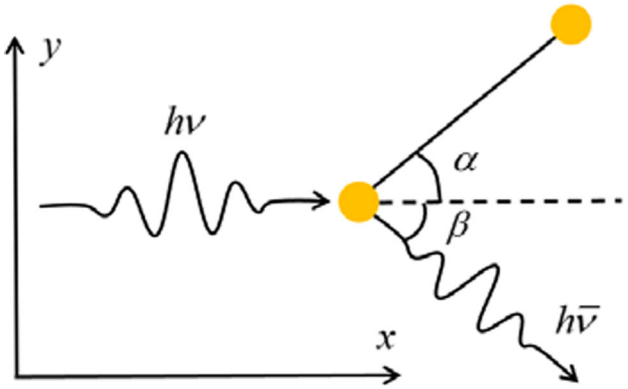

Figure 1. The single-photon Compton scattering process.

Einstein thought that the quantum $h\nu$ always carried a momentum $h\nu/c$, the emission of a quantum $h\nu$ by an atom produced a 'jump' in its velocity and that this jump was responsible for the Doppler shift. A detailed account of the Doppler effect as a photon phenomenon is presented in [18,19]. Schrödinger thought that the emission of a light quantum by a moving atom satisfied the conservation laws of the energy [20], n photons collided simultaneously with atoms, which were moving with the same velocity as the system as a whole along the x-axis, substituting $h\nu$ by $nh\nu$.

When a laser field is applied to an atomic system, nonlinear Compton effect will appear, in which several photons are absorbed in a single point, not only a single high-energy photon is emitted, but also an atom possibly scatters twice or more as it traverses the laser focus. Assuming that atom and photons produce nonlinear MCS, the frequency of Compton scattering photons decreases with the number of photons n. The single photon energy of the laser is far less than the static energy of the atom. The laser field can be regarded as the classical wave field, and we can deal with the effect of atom and light field in the range of relativistic electrodynamics. At the beginning, as the atom is moving, it is difficult to determine the behaviour of the scattering atom. So, we assume that the atom with mass m is stationary before collision. The energy of the atom and n-photon before scattering is mc^2 and $nh\nu$, respectively. After scattering, the energy of the atom and the emitted high-frequency photon are $mc^2/\sqrt{1-u^2/c^2}$ and $nh\bar{\nu}$, respectively. The individual probabilities of emissions are not independent of each other, because the initial atom momentum at each emission is different. In the case of MCS, the physics has been explained in the preceding section, we substitute $nh\nu$ for $h\nu$ in the incident beam, and $(n-k)h\bar{\nu}$,

$k \ll n$ for $h\bar{\nu}$ in the scattered beam. The physics is taking account of k photons being absorbed by the atom to excite the electron from the ground state to the excited state, thus only $(n-k)$ photons being scattered. In fact, the emission of each photon modifies the atom state and consequently the next emissions. If the polarisation of the photon is not considered, according to the conservation of energy and momentum, when the photon is scattered by the atom

$$\begin{aligned} mc^2 + nh\nu &= \frac{mc^2}{\sqrt{1-u^2/c^2}} + (n-k)h\bar{\nu}, \\ \frac{nh\nu}{c} &= (n-k)\frac{h\bar{\nu}}{c}\cos[\beta] + \frac{mu}{\sqrt{1-u^2/c^2}}\cos[\alpha], \\ 0 &= -(n-k)\frac{h\bar{\nu}}{c}\sin[\beta] + \frac{mu}{\sqrt{1-u^2/c^2}}\sin[\alpha]. \end{aligned} \tag{1}$$

Before the collision, the photons move parallel to the x-axis. After the collision, the photons move in a direction making an angle β with the x-axis and the atom moves with a velocity u in a direction making an angle α with the x-axis.

Following ref. [17], eq. (1) implies

$$\begin{aligned} &m[n\nu - (n-k)\bar{\nu}] \\ &\quad -\frac{h}{c^2}(1-\cos[\beta])n(n-k)\nu\bar{\nu} = 0, \\ &\bar{\lambda} = \frac{c}{\bar{\nu}}, \quad \lambda = \frac{c}{\nu}. \end{aligned} \tag{2}$$

Equation (2) shows that the MCS wavelength $\lambda_{nc} = nh/mc = n\lambda_c$. The photon degeneracy is increased n times compared to that of the single-photon Compton wavelength λ_c. There are two special solutions: One solution $\beta = \pi$ represents the mirror image reflection, the wavelength difference $\bar{\lambda} - \lambda = 2(n-k)\lambda_c$ attains a maximum after colliding with an atom. The other solution $\beta = 0$ represents the photons being absorbed, initiating resonant florescence. In our model, a vast majority of photons are used for initiating mirror image reflection, only a small part of photons for initiating the resonate fluorescence.

When $\beta = \pi$, we have the following relations:

$$\begin{aligned} &n - (n-k)\,\frac{\bar{\nu}}{\nu} - 2n(n-k)\frac{\bar{\nu}}{\nu_c} = 0, \\ &\bar{\nu} = \frac{n}{(n-k)(1/\nu + 2n/\nu_c)} = \frac{n\nu}{(n-k)(1+2n\nu/\nu_c)}, \\ &\bar{\lambda} = \lambda(n-k)\left(1+2n\frac{\lambda_c}{\lambda}\right)\frac{1}{n} = \frac{n-k}{n}\lambda \\ &\quad +2(n-k)\lambda_c \simeq \lambda + 2(n-k)\lambda_c, \\ &\frac{\bar{\nu}}{\nu} = \frac{\lambda}{\lambda + 2(n-k)\lambda_c} = \frac{1}{1+2(n-k)\nu/\nu_c}. \end{aligned} \tag{3}$$

The conservation of momentum in eq. (1),

$$\begin{aligned} &n\frac{h\nu}{c} + (n-k)\frac{h\bar{\nu}}{c} = \frac{mu_{n,k}}{\sqrt{1-u_{n,k}^2/c^2}}, \\ &n\frac{h\nu}{c}\left[1 + \frac{(n-k)}{n}\frac{\bar{\nu}}{\nu}\right] \\ &= n\frac{h\nu}{c} + (n-k)\frac{h\nu}{1+2(n-k)\nu/\nu_c}, \\ &A_{n,k} = n\frac{h\nu}{mc^2} + (n-k)\frac{h\nu}{mc^2}\frac{1}{1+2(n-k)\nu/\nu_c} \\ &\quad = \frac{u_{n,k}/c}{\sqrt{1-u_{n,k}^2/c^2}}, \\ &\frac{u_{n,k}}{c} = \frac{A_{n,k}}{\sqrt{1+A_{n,k}^2}} \simeq A_{n,k} \\ &\quad = \frac{\nu}{\nu_c}\left[n + (n-k)\frac{1}{1+2(n-k)\nu/\nu_c}\right], \end{aligned} \tag{4}$$

When the observer's moving velocity is zero, the Doppler effect takes the form [21]

$$\frac{\bar{\nu}_{\mathrm{D}} - \nu}{\nu} = \frac{u_{n,k}/c}{1-u_{n,k}/c} \simeq \frac{A_{n,k}}{1-A_{n,k}} \simeq A_{n,k}. \tag{5}$$

In general, the multiphoton interacts with Dicke states, then interacts with a single-mode radiation field. The number of Dicke states r, here $r = 1, 2, 3, \ldots$ represents monomer (single atom $r = 1$), dimer (two atoms $r = 2$) ,..., polymer (multiatoms $r = n$), respectively. The mass of proton $m_p = 1.6726231 \times 10^{-24}$g, for a monomer, $m_{\mathrm{Rb}} = 85m_p = 1.4217296 \times 10^{-22}$g; for a dimer, $2m_{\mathrm{Rb}} = 2.84345927 \times 10^{-22}$ g. In general, r atoms in the polymer state, moving as a whole, $rm_{\mathrm{Rb}} = 85m_p r$, multiphoton interacts with r atoms.

For a monomer,

$$\begin{aligned} \lambda_c &= \frac{h}{m_{\mathrm{Rb}}c} = 1.553 \times 10^{-15}\,\mathrm{cm}, \\ \frac{\nu}{\nu_c} &= \nu\frac{\lambda_c}{c} = \nu\frac{1.5535 \times 10^{-15}\,\mathrm{cm}}{3 \times 10^{10}\,\mathrm{cm/s}} \\ &= 0.5178 \times 10^{-25}\nu s. \end{aligned} \tag{6}$$

The frequency of the visible light $\nu = 10^{15}\,\mathrm{s}^{-1}$, and we have $\nu/\nu_c = 0.5178 \times 10^{-10}$. If the photon degeneracy $n = 10^7$ [10,11], we have $n\nu/\nu_c = 5.178 \times 10^{-4}$.

Employing eqs (3)–(5), we have the MCS Doppler shift

$$\begin{aligned} &\frac{\bar{\nu}_{\mathrm{D}} - \nu}{\nu} \simeq A_{n,k} \\ &= n\frac{\nu}{\nu_c} + (n-k)\frac{\nu}{\nu_c}\frac{1}{1+2(n-k)\nu/\nu_c}. \end{aligned} \tag{7}$$

For $k = 1, n = 10^7, \nu/\nu_c = 0.5178 \times 10^{-10}$, we have

$$A_{n,1} = n\frac{\nu}{\nu_c} + (n-1)\frac{\nu}{\nu_c}\frac{1}{1+2(n-1)\nu/\nu_c} \simeq 1.06 \times 10^{-3}.$$

When the atom moves towards the observer, the RF frequency ν radiated by the atom shows a blue Doppler shift

$$\nu_{\rm ob} = \nu\left(1 + \frac{\bar{\nu}_{\rm D} - \nu}{\nu}\right). \tag{8}$$

The Doppler shift is significant. We note that the MCS Doppler shift in eqs (6) and (7) contains two terms in the bracket, the first term represents the incident photon momentum and the second term represents the reflected photons.

When $\Delta t > 1/\Delta\nu$ [22], according to eq. (7), the curves are essentially indistinguishable. When transition happens, the atom will stay in the excited state until $\Delta t > 1/\Delta\nu$, i.e., out-of-photon-state, where $\Delta\nu$ is the spontaneous emission bandwidth of the atom. Here, we consider a two-level system and the MCS is a coherent scattering. When the atom transits to the ground state, it absorbs the second photon, and then transits to the excited state. But the longitude mode $\Delta t\Delta\nu \leq 1$ restricts transition to the next mode, which violates coherent scattering. Therefore, two-photon absorption never happens in the same photon state ($\Delta t\Delta\nu \leq 1$). One way to absorb more photons is to change two-level to multilevel system. Thus, more photons are absorbed, for example, $k \geq 2$. In this case, the second term may be replaced by

$$\frac{\nu}{\nu_c}\frac{(n-k)}{1+2(n-k)\nu/\nu_c}.$$

When $n \gg k$, the influence of the second term on the calculation results is still small.

Another way to absorb more photons is to substitute the coherent MCS longitude mode $\Delta t\Delta\nu \leq 1$ restriction by the incoherent MCS. Before and after collision, not in the same photon state, the conservation of energy and momentum does not hold, and the photon degeneration is different. It is very difficult to get an analytical solution for the incoherent MCS. The strong nonlinear MCS occurs in a high atoms and ions density laser plasma, collision between atoms and ions play important roles also. The physics mechanism is very complex beyond the coherent MCS considered in the present paper. For the coherent MCS, the atom density is very low, and therefore the collision between atoms can be neglected.

As shown in figure 2, after scattering, the atom moves towards the observer, the RFS frequency ν radiated by the atom. The RFS frequency is observed perpendicular to the direction of the atoms' motion and the Doppler shift DRFS frequency $\nu_{\rm ob}$ is observed in the direction of the atoms' motion.

The multiphoton laser is focussed by the microscopy objective, and consequently produces MCS with the hydrogen atom, then emits resonant fluorescence which is transparent through the container and hit the observed sample, and we can realise an ultraresolving power beyond the Rayleigh limit.

3. The RFS for two-level atomic system driven by the coherent light

As we all know, the RFS of a single atom system contains three resonance peaks [8–12], while the RFS of a multiatom system contains many side peaks, besides the resonance peak. For example, Agarwal *et al* [23–25] studied the RFS of two-atom system. When the light intensity is not very high, the RFS possesses three peaks, but when the light intensity is very high, the side peaks appear. Tan and Gu noticed that the emission of one atom modifies the electron states of the neighbourhood atom's emission [16] and the RFS gradually transits from three peaks to five peaks, a violation of the definition of the Dicke states. In our calculation model, the corresponding reference spectrum (without Doppler shift) and moving spectrum (with Doppler shift) will change, but the relative shifts between two spectra remain unchanged. We assume that the incident field frequency ν is in resonance with the atomic transition frequency ω_{10}, i.e., $\nu = \omega_{10}$, the coherent spectrum $g(\nu)_{\rm coh} \propto \delta(\nu - \omega_{10})$ (where δ is the Dirac function), and the incoherent spectrum $g(\nu)_{\rm incoh}$ namely the RFS [8,9]

$$g(\nu)_{\rm incoh} = \frac{\kappa\bar{n}_\infty\Omega^2((\nu-\omega_{10})^2+(\Omega^2/2+\kappa^2))}{((\nu-\omega_{10})^2+s_0^2)((\nu-\omega_{10})^2+s_1^2)((\nu-\omega_{10})^2+s_2^2)}$$

and the two roots are given by

$$s_0 = -\kappa/2,\ s_1 = -3\kappa/4 - \sqrt{\kappa^2/16 - \Omega^2},$$
$$s_2 = -3\kappa/4 + \sqrt{\kappa^2/16 - \Omega^2}, \tag{9}$$

where the average value of photons $\bar{n}_\infty = (\Omega^2/4)/(\Omega^2/2 + \kappa^2/4)$ approaches equilibrium value [1], κ is the spontaneous emission coefficient. For the definition of notations $\bar{n}_\infty$ and κ, please refer to ref. [9]. $\Omega = 2\mu E_0/\hbar$ is the Rabi frequency of the pump field, E_0 is the two-level atom energy, μ is the electric dipole moment. As long as we know the spectral distribution, we can determine the statistical distribution law of n-

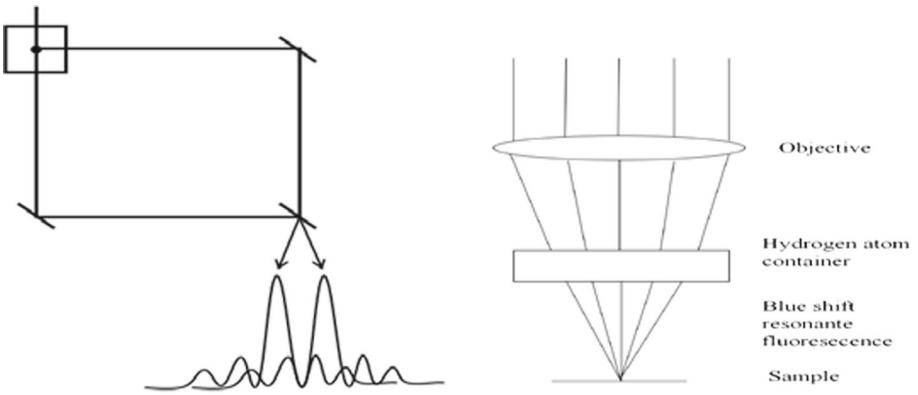

Figure 2. The scheme of multiphoton Compton scattering.

order correlation function and the state function of the resonance fluorescence field.

As we all know, a laser can be well described by a coherent state $|\alpha\rangle$. When the phase of a coherent state is randomised, it is equivalent to a mixed state of Fock states [26]

$$|\alpha\rangle = \sum_{n=0}^{\infty} \mathrm{e}^{-\frac{\alpha^2}{2}} \frac{\alpha^n}{\sqrt{n!}} |n\rangle = \sum_{n=0}^{\infty} \sqrt{p(n)}\, |n\rangle\,, \tag{10}$$

where α is the coherence coefficient, the probability of n-atom obeying the binomial probability distribution $p(n)$. The normalisation condition reads as $\sum_{n=0}^{\infty} p(n) = 1$.

The number states are the eigenstates of the number operator $\hat{a}^\dagger\hat{a} : \hat{a}^\dagger\hat{a}\,|n\rangle = n\,|n\rangle$, $n = 1, 2, 3, \ldots$, where $\hat{a}$ and $\hat{a}^\dagger$ are the annihilation and creation operators satisfying the commutation relation. The photon number of the coherent state follows Poisson distribution [26] with a mean of $|\alpha|^2$ [27].

$$\langle n\rangle = \left\langle \alpha \left| \hat{a}^\dagger\hat{a} \right| \alpha \right\rangle$$

$$= \mathrm{e}^{-\alpha^2} \sum_{n=0}^{\infty} \frac{\alpha^{2n}}{(n-1)!} = \alpha^2 \sum_{n=0}^{\infty} p(n) = \alpha^2. \tag{11}$$

According to eqs (6)–(10), the Doppler shift of the RFS can be rewritten as

$$I(\nu) = g(\nu_{\mathrm{ob}}) = \sum_{n=0}^{\infty} p(n) g_{\mathrm{incoh}} \left(\left(1 - \frac{u_n}{c} \right) \nu \right)$$

$$= \sum_{n=0}^{\infty} p(n) g_{\mathrm{incoh}}((1 - A_{n,k})\nu). \tag{12}$$

In the following numerical simulation, we take the following parameters: the transition frequency $\omega_{10} =$ 386 THz, the emission frequency $\nu =$ 385.5–386.5 THz, the spontaneous emission coefficient $\kappa = 0.05$ THz and the Rabi frequency $\Omega = 3\kappa$ [27,28].

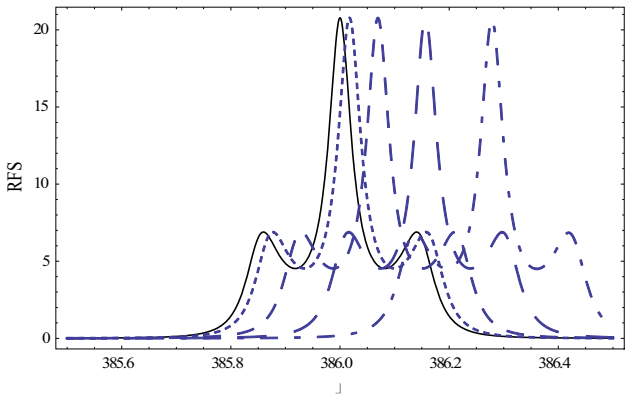

Figure 3. When the number of cluster atoms $r = 1$ (monomer), the RFS of ^{85}Rb atom (without the Doppler shift) with the coherence coefficient $\alpha = 0$ (solid line), the DRFS of ^{85}Rb atom (with the Doppler shift) with the coherence coefficient $\alpha = 10^3$ (dotted line), $\alpha = 2 \times 10^3$ (short-dashed line), $\alpha = 3 \times 10^3$ (the long-dashed line), $\alpha = 4 \times 10^3$ (the dash dotted line), respectively.

In figure 3, we investigate the RFS and DRFS of ^{85}Rb atom for the monomer via MCS. We find that the curve moves to the right (blue shift) with increasing coherence coefficient α. According to eq. (10), the degeneration of the coherent state is $\langle n\rangle = \alpha^2$.

4. The physical diagnosis of the Dicke state

The single-photon Compton wavelength λ_c is inversely proportional to the atomic mass m. The mass of the cluster atoms should be rm, the cluster atoms move together with a single mode radiation field. For the Dicke states, the Compton wavelength $\lambda_c = h/mc$, $\nu_c = c/\lambda_c$ in eq. (3) should be modified to

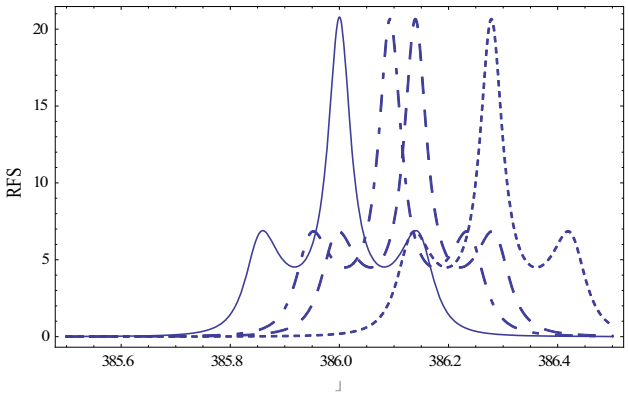

Figure 4. The RFS of ^{85}Rb with the coherence coefficient $\alpha = 0$ (without the Doppler shift) and the number of cluster atoms $r = 1$ (monomer, the solid line). Given the coherence coefficient $\alpha = 4 \times 10^3$, the DRFS of ^{85}Rb (with the Doppler shift) with the number of cluster atoms $r = 1$ (monomer, the short-dashed line), $r = 2$ (dimer, the long-dashed line), $r = 3$ (trimer, the dashed dotted line), respectively.

$$\lambda_{rc} = h/rmc = \lambda_c/r,\ \nu_{rc} = c/\lambda_{rc} = r\nu_c,$$
$$\frac{u_{rn,k}}{c}\frac{1}{\sqrt{1-u_{rn,k}^2/c^2}} = \frac{n-k}{r}\frac{\nu}{\nu_c}$$
$$\times\left(1+\frac{1}{1+2(n-k)\nu/r\nu_c}\right) = A_{rn,k}.$$

Accordingly, eq. (12) turns to

$$I(\nu) = g(\nu_{\rm rob}) = \sum_{n=0}^{\infty} p(n) g_{\rm incoh}(\nu(1-A_{rn,k})). \quad (13)$$

Figure 4 shows the RFS and the DRFS of ^{85}Rb via MCS with the same coherence coefficient $\alpha = 4 \times 10^3$ and different number of cluster atoms $r = 1-3$.

5. Conclusion and discussion

In this work, our aim was to determine the Compton process in Dicke states and analyse the generated Doppler shift in the RFS. The Dicke state is a coherent state of multiple particles, which is fundamentally responsible for various intriguing collective behaviours of many-body systems. The model we have adopted as the basis of our analysis consists of a cluster of two-level atoms driven by a laser field. Numerical solutions have been used to show the Doppler shift induced by MCS in the proposed model. In order to use MCS for the diagnosis of the Dicke states, the numerical solutions are depicted in figures 3 and 4. These are some of the key points to be mentioned:

(i) For a two-level ($|0\rangle$ and $|1\rangle$) atomic system, we choose the resonant transition line $\omega_{10} = 386$ THz in figure 3 [28], the RFS is represented as $g_{\rm incoh}(\nu)$ in eq. (8) and the corresponding DRFS is expressed by $g_{\rm incoh}(\nu_{\rm ob})$ in eq. (13). For a three-level ($|0\rangle$, $|1\rangle$ and $|2\rangle$) atomic system, the influence of the neighbour energy level $|2\rangle$ must be considered, and the corresponding resonant transition line turns to $\omega_{20} = 388$ THz [28,29]. Equation (8) should be modified by $\bar{g}_{\rm incoh}(\nu)$, and the corresponding eq. (13) turns to

$$\bar{g}_{\rm incoh}(\nu_{\rm ob}) = \sum_{n=0}^{\infty} p(n)\bar{g}_{\rm incoh}(\nu(1-A_{n,k})).$$

(ii) In figures 3 and 4, the RFS (the solid line) is the reference spectrum. Besides the difference between the Doppler shift, we may investigate the difference in resonant spectra between the monomer and the polymer [15,16]. In experiment, without the Doppler shift, the observed resonant spectrum actually is the superposition of different composition ratios from monomer to polymer. If we consider the Doppler shift, for Dicke state, the resonant spectrum separates from each other [5,6]. The resonant spectra of the Dicke states have been studied for monomer, dimer and polymer [13,15]. Thus, the observation of a Doppler-shifted resonant spectra can help us to diagnosis the Dicke states, the constitution of the Dicke states and the associated resonant spectrum.

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