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Manifestation of the van der Waals surface interaction in the spontaneous emission of atoms into an optical nanofiber

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Abstract

We study the spontaneous emission of atoms near an optical nanofiber and analyze the coupling efficiency of the spontaneous emission into a nanofiber. We also investigate the influence of the van der Waals interaction of atoms with the surface of the optical nanofiber on the spectrum of coupled light. Using, as an example, ^{85}Rb atoms we show that the van der Waals interaction may considerably extend the red wing of the spontaneous emission line and, accordingly, produce a well-defined asymmetry of the spontaneous emission spectrum coupled into an optical nanofiber.

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I. INTRODUCTION

The spontaneous emission of atoms located near nanostructures can be viewed as a unique tool for studying the interaction between an atom and a nanostructure. Such an interaction may lead to a modification of the spontaneous emission rate of an atom near nanobodies, such as dielectric nanofibers, nanospheres and nanodisks, [1–6] and it can also produce a shift of the spontaneous emission line. Therefore, an experimental observation of the shape of the spontaneous emission line can be used to obtain information on the details of the atomic interaction with nanobody surfaces. In practice, the shape of the spontaneous emission line produced by atoms near a nanofiber can be affected by many other factors, including such significant effects as the van der Waals and Casimir-Polder frequency shifts.

Recently, the ability to fabricate optical nanofibers [7, 8] has enabled a growth of experimental studies into "atom & nanofiber" systems. Some of the latest experimental observations have indicated that the spectrum of spontaneous emission can have either a well-pronounced, long red tail [6] or an asymmetry with an increased red wing of the spectral line [9]. In earlier work [6], the long red tail of the spectrum was first assigned to bound transitions of atoms in the van der Waals potential [10]. However, in later work [9], the authors noted that the long red tail was only observed when they failed to clean the surface of the nanofiber prior to data acquisition. Subsequently, on cleaning the surface by violet light, the spectrum exhibited a well-pronounced asymmetry of the spectral line with a prevailing red side [9], rather than the previously reported long red tail.

It is, therefore, of principal importance for experiments on atom-fiber interactions to determine the contributions to the asymmetry of the spontaneous emission excitation spectrum that arise from basic physical mechanisms, rather than due to using a dirty surface. For clean surfaces, such basic mechanisms should include the van der Waals interaction (see, for example, the review paper [11]). The contribution of the van der Waals interaction to the red shift of the spectral line has already been observed in selective-reflection spectroscopy of cesium vapor located near a dielectric surface [12].

The purpose of this paper is to analyze the manifestation of the van der Waals frequency shift into the shape of the spontaneous emission line for the experimentally significant case of atoms spontaneously emitting light into an optical nanofiber. Specifically, we will evaluate the spectrum of light spontaneously emitted by optically excited ^{85}Rb atoms into the

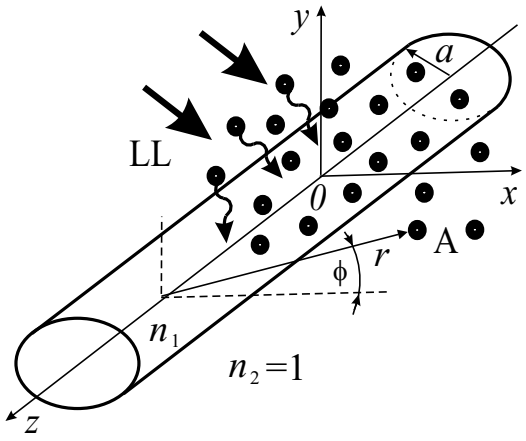

Figure 1: Atomic cloud around an optical nanofiber. Atoms are excited by laser light (LL) and spontaneously emit light into the nanofiber.

fundamental guided mode, HE_{11}, of an optical nanofiber. Results from our study show that the contribution of the van der Waals red shift leads to the appearance of a well-pronounced asymmetry of the line of spontaneous emission coupled into an optical nanofiber, for typical optical nanofiber diameters of 100-1000 nm and atomic clouds that are tightly confined around the nanofiber.

II. RATE OF SPONTANEOUS EMISSION INTO THE FIBER

We consider a cloud of cold, two-level atoms excited by a near-resonant laser field. The atoms are located near the optical nanofiber, as illustrated in Fig. 1. Excited atoms emit spontaneous radiation, which partially propagates into the guided modes of the fiber. Due to practical significance, we limit our discussion to the case where the frequency of the fluorescent light is below the cut-off frequencies of all fiber modes other than the fundamental, HE_{11}, mode, so that emitted light can only ever propagate in this mode. The lower state of the atom is the ground state and we assume that the upper state can only decay to the ground level. The two-level atom model is partially justified by the fact that, for optical transitions with degenerate states, different magnetic sublevels have very similar spontaneous decay rates [4].

As a first step, we derive the spontaneous emission rate into a guided mode of an optical fiber. For this, we represent the operator of a quantized vacuum electric field of the guided

modes of a nanofiber in a standard form

$$\mathbf{E} = \sum \boldsymbol{\mathcal{E}}_\lambda a_\lambda + \text{h.c.}, \tag{1}$$

where $\boldsymbol{\mathcal{E}}_\lambda$ is the electric field of a single vacuum guided mode, a_λ is the photon annihilation operator, and the index, λ, indicates the direction of propagation and polarization of a single vacuum guided mode. The electric field of a single guided mode can be represented by [13]

$$\boldsymbol{\mathcal{E}}_\lambda = i\sqrt{\frac{\hbar\omega_\lambda}{2\varepsilon_0 L}}\,\tilde{\boldsymbol{\mathcal{E}}}_\lambda \mathrm{e}^{i\beta_\lambda z + im\varphi}, \tag{2}$$

where ω_λ is a mode frequency, β_λ is a propagation constant, $\tilde{\boldsymbol{\mathcal{E}}}_\lambda(r,\varphi)$ is a normalized amplitude of the electric field, m is a quantum number of the mode angular momentum, and L is the length of a one-dimensional "box" defined by a spatial periodicity of the field. The electric field amplitude of a single guided mode is normalized as

$$\int_0^{2\pi}\int_0^{\infty} n^2(r)\left|\tilde{\boldsymbol{\mathcal{E}}}_\lambda\right|^2 d\varphi r dr = 1, \tag{3}$$

where $n(r)$ is the value of the refractive index and is equal to n_1 inside the fiber and $n_2 = 1$ outside the fiber.

The above representation of the vacuum field corresponds to a standard form of the vacuum field Hamiltonian

$$H_{\text{vac}} = 2\varepsilon_0\varepsilon\sum\int dV\left|\boldsymbol{\mathcal{E}}_\lambda\right|^2\left(a_\lambda^\dagger a_\lambda + \tfrac{1}{2}\right) = \sum\hbar\omega_\lambda\left(a_\lambda^\dagger a_\lambda + \tfrac{1}{2}\right). \tag{4}$$

The total Hamiltonian for a system consisting of the "two-level atom + vacuum field of the guided modes of an optical nanofiber " can, accordingly, be represented as

$$H = \hbar\omega_0 b^+ b + \sum_\lambda \hbar\omega_\lambda\left(a_\lambda^+ a_\lambda + \tfrac{1}{2}\right) - \mathbf{d}\cdot\sum_\lambda\left(\boldsymbol{\mathcal{E}}_\lambda b^+ a_\lambda + \boldsymbol{\mathcal{E}}_\lambda^* b a_\lambda^+\right), \tag{5}$$

where b^+ and b are the atomic excitation and de-excitation operators, a^+ and a the photon creation and annihilation operators, and $\mathbf{d}$ is a matrix element of the atomic dipole moment.

If we now apply the Weiskopf-Wigner approach to the considered quantized system one can write equations for the probability amplitudes for the simplest case of a vacuum field initially in the ground state, so that

$$\dot{c}_{e,0} = \frac{i}{\hbar}\sum_\lambda \mathbf{d}\cdot\boldsymbol{\mathcal{E}}_\lambda \mathrm{e}^{-i\Delta_\lambda t} c_{g,1_\lambda}, \tag{6a}$$

$$\dot{c}_{g,1_\lambda} = \frac{i}{\hbar}\mathbf{d}\cdot\boldsymbol{\mathcal{E}}_\lambda^* \mathrm{e}^{i\Delta_\lambda t} c_{e,0}, \tag{6b}$$

where $c_{g,1_\lambda}$ are the probability amplitudes of the states which include the ground atomic state and the state of the vacuum field with one photon in mode λ and $c_{e,0}$ is the probability amplitude of the state which includes the excited atomic state and the state of the vacuum field with zero photon numbers in all the modes.

Taking a formal solution of the second equation in the above set

$$c_{g,1_\lambda} = \frac{i}{\hbar}\mathbf{d}\cdot\boldsymbol{\mathcal{E}}_\lambda^* \int_{t_0}^{t} \mathrm{e}^{i\Delta_\lambda t'} c_{e,0}(t')dt', \tag{7}$$

and substituting it into the first equation one obtains an equation describing the spontaneous decay of the upper atomic state

$$\dot{c}_{e,0} = -\frac{1}{\hbar^2}\sum_\lambda |\mathbf{d}\cdot\boldsymbol{\mathcal{E}}_\lambda|^2 \int_{t_0}^{t} \mathrm{e}^{i\Delta_\lambda (t'-t)} c_{e,0}(t')dt'. \tag{8}$$

To apply Eq. (8) to the fundamental guided mode of an optical nanofiber one can consider the vacuum field of a single guided mode as being periodic with spatial period, L. The periodicity condition can be written as $\beta_\alpha L = 2\pi n_\alpha$, where the integer numbers, $n_\alpha = 1, 2, 3, ...$, define different values of the propagation constant, β_α. By making use of the periodicity condition, the sum over discrete numbers, n_α, entering Eq. (8) can be replaced by an integral where

$$\sum \to \frac{L}{2\pi c}\int d\beta.$$

Next, if we consider a one-to-one correspondence between values of the propagation constant and frequencies of the vacuum modes, $\beta = \beta(\omega)$, one can replace the differential, $d\beta$, by $d\beta = \beta' dk = \beta' d\omega/c$. This reduces the summation to an integral over frequency where

$$\sum \to \frac{L}{2\pi c}\int \beta' d\omega.$$

The integral over frequency can be reduced to a delta-function such that

$$\int \mathrm{e}^{i(\omega-\omega_0)(t'-t)} d\omega = 2\pi\delta(t-t').$$

Finally, integrating over time and taking into account that any guided mode has two directions of propagation Eq. (8) can be rewritten as

$$\dot{c}_{e,0} = -\gamma^{(\mathrm{g})} c_{e,0}, \tag{9}$$

where $\gamma^{(\mathrm{g})}$ is half the spontaneous decay rate into the guided mode of an optical nanofiber, i.e.

$$W_{\mathrm{sp}}^{(\mathrm{g})} = 2\gamma^{(\mathrm{g})} = \frac{\omega_0 \beta'}{\varepsilon_0 \hbar c}\left|\mathbf{d}\cdot\tilde{\boldsymbol{\mathcal{E}}}\right|^2. \tag{10}$$

In the last equation $\tilde{\boldsymbol{\mathcal{E}}}$ is the amplitude of the guided mode with a specific direction of propagation. Note that Eq. (10) is similar to the equation developed in [4] but differs from it by the absence of a factor of two. As we however see in next section this discrepancy has no effect on the numerical data which differ by less than 10%.

III. ELECTRIC FIELD OF THE FUNDAMENTAL MODE, HE_{11}

For the fundamental guided mode, HE_{11}, defined by angular index $m = 1$ the propagation constant, β, is defined by the eigenvalue equation as [14]

$$\frac{J_0(ha)}{haJ_1(ha)} = -\left(\frac{n_1^2+n_2^2}{2n_1^2}\right)\frac{K_1'(qa)}{qaK_1(qa)} + \frac{1}{h^2a^2} - \left[\left(\frac{n_1^2-n_2^2}{2n_1^2}\right)^2\left(\frac{K_1'(qa)}{qaK_1(qa)}\right)^2 + \left(\frac{\beta}{n_1k}\right)^2\left(\frac{1}{h^2a^2}+\frac{1}{q^2a^2}\right)^2\right]^{1/2},$$

where J_m are Bessel functions of the first kind, K_m are modified Bessel functions of the second kind, $k = \omega/c$, $h = \sqrt{n_1^2k^2 - \beta^2}$ and $q = \sqrt{\beta^2 - n_2^2k^2}$.

It should be noted that there are four different field distributions for the fundamental mode, HE_{11}, two of which have opposite directions of propagation and two of which have opposite circular polarizations. In what follows, we write the field distribution for a guided mode with positive propagation constant and positive circular polarization using a decomposition over cylindrical unit vectors, $\tilde{\boldsymbol{\mathcal{E}}} = \mathbf{e}_r\tilde{\mathcal{E}}_r + \mathbf{e}_\varphi\tilde{\mathcal{E}}_\varphi + \mathbf{e}_z\tilde{\mathcal{E}}_z$.

For the HE_{11} mode, the cylindrical components of a normalized electric field amplitude in the core region are given by [14]

$$\begin{aligned}
\tilde{\mathcal{E}}_r &= iA\frac{q}{h}\frac{K_1(qa)}{J_1(ha)}\left[(1-s)J_0(hr) - (1+s)J_2(hr)\right],\\
\tilde{\mathcal{E}}_\varphi &= -A\frac{q}{h}\frac{K_1(qa)}{J_1(ha)}\left[(1-s)J_0(hr) + (1+s)J_2(hr)\right],\\
\tilde{\mathcal{E}}_z &= 2A\frac{q}{\beta}\frac{K_1(qa)}{J_1(ha)}J_1(hr),
\end{aligned}$$

while those outside of the core region are given by

$$\begin{aligned}
\tilde{\mathcal{E}}_r &= iA\left[(1-s)K_0(qr) + (1+s)K_2(qr)\right],\\
\tilde{\mathcal{E}}_\varphi &= -A\left[(1-s)K_0(qr) - (1+s)K_2(qr)\right],\\
\tilde{\mathcal{E}}_z &= 2A\left(q/\beta\right)K_1(qr).
\end{aligned}$$

In the above equations s is a dimensionless parameter such that

$$s = \frac{1/h^2a^2 + 1/q^2a^2}{J_1'(ha)/haJ_1(ha) + K_1'(qa)/qaK_1(qa)}.$$

The normalization constant, defined from Eq. (3), is

$$A = \frac{\beta}{2q} \frac{J_1(ha)/K_1(qa)}{\sqrt{2\pi a^2 (n_1^2 N_1 + n_2^2 N_2)}}, \tag{11}$$

where

$$N_1 = \frac{\beta^2}{4h^2} \left\{ (1-s)^2 \left[J_0^2(ha) + J_1^2(ha) \right] + (1+s)^2 \left[J_2^2(ha) - J_1(ha)J_3(ha) \right] \right\} + \frac{1}{2} \left[J_1^2(ha) - J_0(ha)J_2(ha) \right],$$

$$N_2 = \frac{J_1^2(ha)}{2K_1^2(qa)} \left\{ \frac{\beta^2}{2q^2} \left[(1-s)^2 \left[K_1^2(qa) - K_0^2(qa) \right] - (1+s)^2 \left[K_2^2(qa) - K_1(qa)K_3(qa) \right] \right] - K_1^2(qa) + K_0(qa)K_2(qa) \right\}.$$

The intensity distribution of the electric field outside the core is defined by the quantity

$$|\tilde{\boldsymbol{\mathcal{E}}}(r)|^2 = 2A^2 \left[(1-s)^2 K_0^2(qr) + (1+s)^2 K_2^2(qr) + \frac{2q^2}{\beta^2} K_1^2(qr) \right]. \tag{12}$$

IV. POWER OF COUPLED LIGHT

Taking into account the electric field distribution outside the fiber described by Eq. (12), one can rewrite the spontaneous decay rate into the fundamental guided mode, HE_{11}, as

$$W_{\rm sp}^{(\rm g)}(r) = 2\gamma^{(\rm g)} = 2A^2 \frac{d^2\omega_0\beta'}{\varepsilon_0\hbar c} \left[(1-s)^2 K_0^2(qr) + (1+s)^2 K_2^2(qr) + \frac{2q^2}{\beta^2} K_1^2(qr) \right], \tag{13}$$

where $d = |\boldsymbol{d}|$, $\beta' = d\beta/dk = cd\beta/d\omega$ and A is a constant as defined by Eq. (11). We can rewrite Eq. (13) in a convenient form if we introduce the spontaneous decay rate into free space,

$$W_{\rm sp} = 2\gamma_0 = \frac{1}{4\pi\varepsilon_0} \frac{4d^2\omega_0^3}{3\hbar c^3} \tag{14}$$

and use the wavelength of the light λ. This yields an equation of the form

$$W_{\rm sp}^{(\rm g)}(r) = 2\gamma^{(\rm g)} = \gamma_0 \frac{3A^2\lambda^2\beta'}{\pi} \left[(1-s)^2 K_0^2(qr) + (1+s)^2 K_2^2(qr) + \frac{2q^2}{\beta^2} K_1^2(qr) \right]. \tag{15}$$

Consider now a single, motionless, two-level atom placed near the optical fiber and excited by an external laser field near-resonant to the dipole optical transition. The probability of finding the atom in the upper excited state is given by

$$p_{\mathrm{e}} = \frac{1}{2}\frac{\Omega^2}{(\omega-\omega_0)^2+\gamma^2+\Omega^2}, \tag{16}$$

where $\Omega = dE_0/2\hbar$ is the Rabi frequency defined by the atomic dipole matrix element, d, and amplitude, E_0, of the exciting laser field, ω is the frequency of the laser light, ω_0 is the position-dependent atomic transition frequency, and γ is half the position-dependent total spontaneous decay rate, $W_{\mathrm{sp}} = 2\gamma$. For the case of interest, the spontaneous decay rate consists of the position-dependent decay rate, $\gamma^{(\mathrm{g})}$, into the guided modes of the fiber and the position-dependent decay rate, $\gamma^{(\mathrm{r})}$, into the radiation modes of the fiber, such that

$$W_{\mathrm{sp}} = 2\gamma = 2\gamma^{(\mathrm{g})} + 2\gamma^{(\mathrm{r})}. \tag{17}$$

For a single atom the probability of spontaneous photon emission per unit time into a guided fiber mode is proportional to the population, p_{e}, of the excited atomic state and half the rate of spontaneous emission, $\gamma^{(\mathrm{g})}$, into the guided mode propagating in one direction,

$$W(r) = \gamma^{(\mathrm{g})}(r)p_{\mathrm{e}}(r) = \frac{1}{2}\frac{\gamma^{(\mathrm{g})}(r)\Omega^2}{(\omega-\omega_0(r))^2+\gamma^2(r)+\Omega^2}. \tag{18}$$

In Eq. (18) we explicitly use the fact that both the atomic transition frequency and the spontaneous emission rates are functions of the atom's position, r. For an ensemble of motionless, two-level atoms distributed near the fiber with density $n(\mathbf{r})$, the light power coupled into the fundamental guided mode is defined, therefore, by the volume integral

$$P = \frac{1}{2}\hbar\omega \int \frac{\gamma^{(\mathrm{g})}(r)\,\Omega^2}{(\omega-\omega_0(r))^2+\gamma^2(r)+\Omega^2} n(\mathbf{r})\, dV. \tag{19}$$

Hence, the power coupled into the optical fiber depends on the position of the atomic cloud with respect to the fiber axis and the atomic cloud shape.

In the following, we consider weak optical saturation and we neglect the Rabi frequency in the denominator of the excitation probability. For weak saturation the atoms are mainly in the ground state and the atomic transition frequency is shifted primarily due to a ground state shift. If we take into account that a contribution to the shifts of the atomic states comes from the van der Waals interaction, the atomic transition frequency shift can be evaluated as [11, 15–18]

$$\omega_0(r) = \omega_0^0 - \frac{C_{3\mathrm{g}}}{(r-a)^3}. \tag{20}$$

In the above equation ω_0^0 is the transition frequency, C_{3g} is the van der Waals constant for the ground atomic state, and $r - a$ is the distance between the atom and the fiber surface.

Finally, the fluorescent light power coupled into the guided fiber mode for weak optical saturation can be written as

$$P = \frac{1}{2}\hbar\omega \int \frac{\gamma^{(g)}(r)\,\Omega^2}{\left[\omega - \omega_0^0 + \delta\omega(r)\right]^2 + \gamma^2(r)} n(\mathbf{r})\, dV. \tag{21}$$

In our basic Eq. (21) there are two unknown quantities: the spontaneous emission rate into the guided mode and the spontaneous emission rate into the radiation modes. Of these two quantities, the most important for our analysis is the rate of spontaneous emission into the guided mode. This quantity varies sharply near the surface of the fiber and, therefore, strongly influences the coupling rate for spontaneously emitted light into the fiber. The rate of spontaneous emission into the radiation modes changes weakly near the fiber and its value is approximately equal to the rate of spontaneous emission into free space. In the following analysis we will neglect the weak spatial dependence of the spontaneous emission rate into the radiation modes and consider only the position dependence of the spontaneous emission rate into the guided mode.

We consider the spontaneous emission for ^{85}Rb atoms. We assume the atoms emit spontaneous light into an optical fiber made of fused silica, with permittivity, $\varepsilon = 2.1$. The refractive index of the fiber is $n_1 = 1.45$, while the refractive index of the outside medium is $n_2 = 1$. The rubidium atoms are assumed to be excited at the 5S-5P optical dipole transition, with a wavelength of 780 nm and a spontaneous decay rate of the 5P state, $2\gamma_0 = 2\pi \cdot$ 6 MHz [19–21]. For the ground state of rubidium the van der Waals constant is given by $C_{3g} = 2\pi \cdot 3$ kHz$(\mu$m$)^3$ [12, 22, 23].

Figure 2 shows the position dependence of the spontaneous decay rate for the two-level atom which we considered as a model for ^{85}Rb atoms. The decay rate is evaluated numerically from Eq. (15). It is worth noting that our case of a nanofiber of radius $a = 200$ nm can be compared with a similar case considered for ^{133}Cs atoms in paper [4]. In our case of ^{85}Rb atoms maximum spontaneous emission rate at a surface of the nanofiber is 0.53 while in case of ^{133}Cs atoms maximum value is 0.56 [4].

We assume that the cold atoms are distributed in a spherically symmetric cloud centered on the axis of the optical fiber. The cloud is also assumed to have a Gaussian density

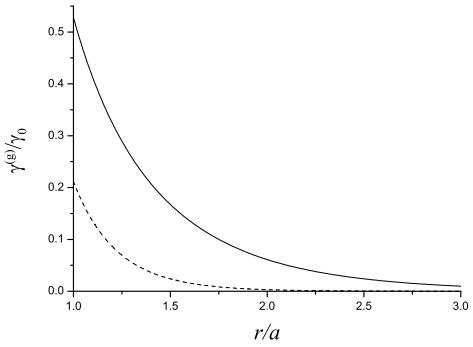

Figure 2: Normalized spontaneous decay rate of a ^{85}Rb atom into the fundamental guided mode, HE_{11}, as a function of distance between the atom and the axis of the optical nanofiber with radius $a = 200$ nm (solid line) and 300 nm (dashed line).

distribution, $n(r)$, in the radial direction with half width, R, such that

$$n(\mathbf{r}) = n(r) = \frac{N}{\pi\sqrt{\pi}R^3} \exp\left[-\left(\frac{r}{R}\right)^2\right], \tag{22}$$

where N is the total number of atoms and is given by

$$N = 4\pi \int n(r) r^2 dr. \tag{23}$$

Figure 3 shows the coupled fluorescence spectrum calculated from Eq. (19) taking the van der Waals shift into account. As one can see, the asymmetry of the fluorescence lineshape increases when the radius of the atomic cloud decreases. In other words, the tighter the cloud around the fiber the more pronounced the asymmetry becomes. As the radius of the cloud increases the atoms located further from the nanofiber are less influenced by the change in the van der Waals frequency shift and, hence, the shape of the fluorescence spectrum approaches that of the symmetrical, free space distribution.

V. CONCLUSION

We conclude that the van der Waals interaction of atoms with the surface of an optical nanofiber can produce well-pronounced asymmetry in the frequency dependence of spontaneous emission coupled into the guided mode of an optical nanofiber. For typical diameters of nanofibers 100-1000 nm the van der Waals red shift is found to increase the red half-width of the spontaneous emission line by 10-30% while keeping the blue half-width of the

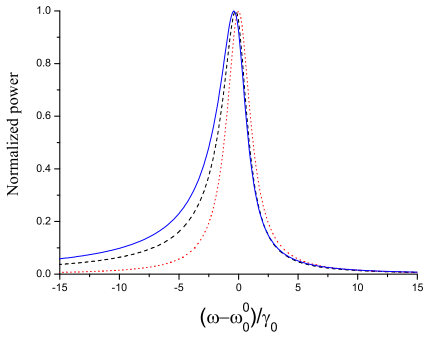

Figure 3: Frequency dependence of the normalized spontaneous emission power from a ^{85}Rb cloud coupled into the optical nanofiber for a fiber radius $a = 200$ nm and an atomic cloud radius $R = 400$ nm (solid line) and 1000 nm (dashed line). The dotted line shows, for comparison, the lineshape for the artificial case where the van der Waals shift is absent.

spectral line unchanged. Therefore, the results of our evaluations show that the van der Waals frequency shifts should be taken into account in any experimental observations of the spontaneous emission line which deal with atomic ensembles that are tightly confined around an optical nanofiber or other type of nanobody. In our opinion it is also desirable to study possible influences of the Casimir-Polder effect on the spontaneous emission coupled into nanobodies.

Acknowledgments

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