# A method for controlling the spins of atoms using optical near-fields 

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## Summary

On the basis of the procedure for controlling the spins of atoms using circularly polarized evanescent light proposed by Hori et al. [(1996) Abstracts of the 1st Asia-Pacific Workshop on Near-field Optics] we discuss the influence of boundary conditions on the probability of spontaneous emission and thus on the spin polarization efficiency, which was not considered in the Hori et al. study. Using the Carniglia-Mandel mode expansion of electromagnetic fields, we derive the spontaneous emission and spin polarization probabilities of atoms near a dielectric surface, and show the atom-surface distance dependence and refractive index dependence. Numerical evaluation for the $6 P_{1 / 2}-6 S_{1 / 2}$ transition of a Cs atom indicates an increase in the efficiency of spin polarization by $30 \%$.

## 1. Introduction

As a result of spatial localization and a unique dispersion relation originating from interaction with matter, optical near-fields produce intriguing phenomena that cannot be obtained with propagating far-fields. Some of these phenomena have been investigated extensively, both experimentally and theoretically, and their applicability to nano-photonics and atom-photonics has been discussed (Ohtsu et al., 2002). Spin control of atoms, molecules, or quantum dots is another example that has attracted considerable attention in the scientific community. In this context, a method for controlling the spin of an atomic beam by using circularly polarized evanescent light whose angular momentum is perpendicular to the propaga-

[^0]tion direction has been proposed (Hori et al., 1996; Ohdaira et al., 2001). The authors neglected the dielectric surface effect, although it is well known that the spontaneous emission rate of atoms near a surface is modified. In addition, the polarization of emitted photons becomes anisotropic, and their directional dependence is not negligible when the atoms are very close to a surface. On the basis of their proposal, we examined the dielectric surface effects on spin-control efficiency, with the help of Carniglia-Mandel (CM) modes (Carniglia \& Mandel, 1971; Inoue \& Hori, 2001), which form a complete, orthonormal basis set for electromagnetic fields with an infinite surface boundary. Section 2 briefly outlines the spincontrol method proposed by Hori et al. In Section 3, we derive the spontaneous emission and spin polarization probabilities of atoms near a dielectric planar surface, and discuss the spin-control efficiency, using the example of caesium atoms. Conclusions are drawn in Section 4, and the definitions and useful formulae of the CM modes are presented in the Appendix.

## 2. Spin-control method by optical near-fields

Following Hori et al. (1996), we outline a method that can be used to align the spins of atoms injected close to a dielectric surface. Suppose that the media below and above the plane have refractive indexes $n$ and 1, respectively. Two s-polarized plane waves with amplitude $E_{0}$, monochromatic frequency $\omega$, and wave number $k=n \omega / c \equiv n K$ are incident from the dielectric side as $E_{0} \mathbf{e}_{x} \mathrm{e}^{i k(y \sin \theta+z \cos \theta)-i \omega t}$ and $E_{0} \mathbf{e}_{y} \mathrm{e}^{i k(x \sin \theta+z \cos \theta)-i \omega t}$, where $\mathbf{e}_{x}$ and $\mathbf{e}_{y}$ are unit polarization vectors. If the incident angle $\theta$ exceeds the critical angle $\theta_{c}$, two evanescent waves are generated above the surface. If the condition

$$
\begin{equation*}
n K x \sin \theta=n K y \sin \theta+\frac{\pi}{2}+2 l \pi, \quad(l=0,1,2,3, \ldots) \tag{1}
\end{equation*}
$$

```
    \(P_{3 / 2} E_{2} \quad(J, M)=(3 / 2,3 / 2),(3 / 2,1 / 2)\)
( \(L=1\) )
```



Fig. 1. Schematic energy levels and transition processes. Repeated pumping and relaxation processes result in the accumulation of the spin-up atoms.
is satisfied, the combined waves are converted into a circularly polarized evanescent wave as

$$
\begin{align*}
E_{t r} & =E\left(\boldsymbol{e}_{\boldsymbol{x}}+i \boldsymbol{e}_{\boldsymbol{y}}\right) \mathrm{e}^{i K y n \sin \theta-i \omega t-K z \sqrt{n^{2} \sin ^{2} \theta-1}},  \tag{2}\\
E & =\frac{2 n \cos \theta}{n \cos \theta+i \sqrt{n^{2} \sin ^{2} \theta-1}} E_{0} . \tag{3}
\end{align*}
$$

Then, the stimulated absorption probability of atoms moving with velocity $V$ along the line given by Eq. (1) can be written as

$$
\begin{equation*}
\left.w_{i \rightarrow f}=|E|^{2} \mathrm{e}^{-2 K Q_{2} \sqrt{n^{2} \sin ^{2} \theta-1}}\left|\langle f| T_{+}\right| i\right\rangle\left.\right|^{2} 2 \pi \delta\left(\omega_{f i}+K V n \sin \theta-\omega\right) \tag{4}
\end{equation*}
$$

where $Q_{z}$ is the $z$ component of the position of the centre of mass of each atom whose resonance frequency, initial and final states are denoted as $\omega_{f i},|i\rangle$, and $|f\rangle$, respectively, and $T_{+}$is
the spherical harmonic tensor that increases the $z$ component of the total angular momentum of each atom by one. Let the initial state $|i\rangle$ be $\left|S_{1 / 2}\right\rangle$ for an alkali atom; then the valence electron is either in the spin-up state $(J, M)=(1 / 2,1 / 2)$ or the spin-down state $(J, M)=(1 / 2,-1 / 2)$. The final state $|f\rangle$ can be either $\left|P_{1 / 2}\right\rangle$ or $\left|P_{3 / 2}\right\rangle$. However, we can selectively eliminate the $\left|P_{3 / 2}\right\rangle$ state by red detuning, and all the atoms in the spindown state $(J, M)=(1 / 2,-1 / 2)$ are excited to the $\left|P_{1 / 2}\right\rangle$ state by stimulated absorption, whereas all the atoms in the spin-up state $(J, M)=(1 / 2,1 / 2)$ remain in the same state. Because atoms in the $\left|P_{1 / 2}\right\rangle$ state relax to either the $(J, M)=(1 / 2,1 / 2)$ or $(J, M)=(1 / 2,-1 / 2)$ state by spontaneous emission, repetitions of the pumping process lead to spin polarization of the final atoms, as depicted in Fig. 1. When the emitted photons are unpolarized, i.e. $\left|e_{x}\right|^{2}=\left|e_{y}\right|^{2}=\left|e_{z}\right|^{2}$, the ratio of relaxation probability $w_{u p}$ to $w_{\text {down }}$ is given as

$$
\begin{equation*}
R=\frac{w_{u p}}{w_{\text {down }}}=\frac{\left|e_{z}\right|^{2}}{\left|e_{x}\right|^{2}+\left|e_{y}\right|^{2}}=\frac{1}{2}, \tag{5}
\end{equation*}
$$

where $w_{u p}$ means the relaxation probability from the $\left|P_{1 / 2}\right\rangle$ state to the spin-up state, and $w_{\text {down }}$ stands for the relaxation probability from the $\left|P_{1 / 2}\right\rangle$ state to the spin-down state. This indicates that one can efficiently align the spins of atoms as the ratio $R$ increases.

## 3. Spontaneous emission under planar boundary conditions

In order to examine the effects of a planar boundary on the transition probabilities $w_{u p}$ and $w_{\text {down }}$, and their ratio $R$, discussed above, we use the CM modes (see Appendix), which form a complete, orthonormal basis set for the quantization of electromagnetic fields with an infinite planar boundary. Starting with the following total Hamiltonian of the system,

$$
\begin{align*}
\mathscr{H} & =\mathscr{H}_{0}+\mathscr{H}^{\prime},  \tag{6}\\
\mathscr{H}_{0} & =\mathscr{H}_{\text {elec }}+\mathscr{H}_{\text {rad }}+\mathscr{T},  \tag{7}\\
\mathscr{H}^{\prime} & =-\frac{e}{m} \boldsymbol{p} \cdot \boldsymbol{A}(\boldsymbol{Q}, t), \tag{8}
\end{align*}
$$

the spontaneous emission rate per unit time is investigated. Here $\mathscr{H}_{0}$ is the unperturbed part, and consists of three parts, $\mathscr{H}_{\text {elec }}, \mathscr{H}_{\text {rad }}$ and $\mathscr{T}$, which describe a valence electron, the electromagnetic fields expressed as Eq. (A9) in terms of the CM modes, and the translational motion of an atom, respectively. The interaction of the valence electron with the electromagnetic fields $\mathscr{H}^{\prime}$ is given by Eq. (8), where $e, m, p$ and $A(Q, t)$ represent the electric charge, mass, momentum of the electron, and the vector potential at position $\boldsymbol{Q}$ of the centre of mass of the atom at time $t$, respectively. The explicit form of the vector potential is given as Eq. (A8) in the Appendix.

Using the time-dependent perturbation theory with respect to $\mathscr{H}^{\prime}$ in Eq. (8), we can obtain the desired emission rate as

$$
\begin{align*}
w_{k, \boldsymbol{\varepsilon}} & =\lim _{t \rightarrow \infty} \frac{1}{t}|\langle f \mid \Psi(t)\rangle|^{2} \\
& \left.=\frac{e^{2} \omega_{\beta \alpha}}{2 \pi^{2} \varepsilon_{0} \hbar} \delta\left(\omega-\omega_{\beta \alpha}\right) \mathrm{e}^{-i\left(\boldsymbol{K}^{*}-\boldsymbol{K}\right) \cdot \boldsymbol{Q}}|\langle\beta| \boldsymbol{\varepsilon} \cdot \boldsymbol{r}| \alpha\right\rangle\left.\right|^{2}|a(\lambda)|^{2}, \tag{9}
\end{align*}
$$

where the final state $|f\rangle$ is assigned to state $\left|\beta, 1_{k, \boldsymbol{\varepsilon}}\right\rangle$ consisting of atomic state $\beta$ and one photon state with wave vector $k$ and polarization vector $\boldsymbol{\varepsilon}$, whereas $|\psi(t)\rangle$ is a time-evolved state from the initial state $|i\rangle=|\alpha, v a c\rangle$ constructed from atomic state $\alpha$ and no photon state (vacuum), and the atomic resonance frequency is denoted as $\omega_{\beta, \alpha}$. Note that the explicit form of $\boldsymbol{\varepsilon}_{L(R)}(\boldsymbol{k}, \boldsymbol{\lambda}, r)$ in Eq. (A4) is used as well as the conversion from
$\langle\beta| \boldsymbol{e} \cdot \boldsymbol{p}|\alpha\rangle$ to $\langle\beta| \boldsymbol{e} \cdot \boldsymbol{r}|\alpha\rangle$, and that the Doppler shift due to atomic motion at velocity $V$ is neglected.

For further evaluation, the following four cases of $w_{k, \varepsilon}$ in Eq. (9) with respect to emitted photons are considered:

Emitted photon $=\left\{\begin{array}{l}\text { homogeneous }\left(\theta<\theta_{c}\right)\left\{\begin{array}{l}\boldsymbol{\varepsilon}=s \text {-polarization } \\ \boldsymbol{\varepsilon}=p \text {-polarization }\end{array}\right. \\ \text { evanescent }\left(\theta>\theta_{c}\right)\end{array}\left\{\begin{array}{l}\boldsymbol{\varepsilon}=s \text {-polarization } \\ \boldsymbol{\varepsilon}=p \text {-polarization }\end{array}\right.\right.$

As illustrated in Fig. 2, homogeneous waves are described by the $R$-mode, whereas evanescent waves are expressed in terms of the $L$-mode. From this classification, we can obtain the directional-dependent emission rate as

$$
\begin{align*}
& w_{u p}^{R}(\boldsymbol{K}, s)=0,  \tag{10}\\
& w_{u p}^{R}(\boldsymbol{K}, p)=\frac{1}{4} F(k) \sin ^{2} \theta^{\prime},  \tag{11}\\
& w_{\text {up }}^{L}(\boldsymbol{k}, s)=0,  \tag{12}\\
& w_{u p}^{L}(\boldsymbol{k}, p)=F(k) \frac{n^{2} \cos ^{2} \theta \sin ^{2} \theta}{\cos ^{2} \theta+n^{2}\left(n^{2} \sin ^{2} \theta-1\right)} \mathrm{e}^{-20_{2} \frac{\omega}{c} \sqrt{n^{2} \sin ^{2} \theta-1}},  \tag{13}\\
& w_{\text {down }}^{R}(\boldsymbol{K}, s)=\frac{1}{4} F(k), \\
& w_{\text {down }}^{R}(\boldsymbol{K}, p)=\frac{1}{4} F(k) \cos ^{2} \theta^{\prime}, \\
& W_{\text {down }}^{L}(\boldsymbol{k}, s)=F(k) \frac{\cos ^{2} \theta}{n^{2}-1} \mathrm{e}^{-2 Q_{z} \frac{\omega}{c} \sqrt{n^{2} \sin ^{2} \theta-1}}, \\
& w_{\text {down }}^{L}(\boldsymbol{k}, p)=F(k) \frac{\left(n^{2} \sin ^{2} \theta-1\right) \cos ^{2} \theta}{\cos ^{2} \theta+n^{2}\left(n^{2} \sin ^{2} \theta-1\right)} \mathrm{e}^{-2 \mathrm{Q}_{2} \frac{\theta}{c} \sqrt{n^{2} \sin ^{2} \theta-1}},  \tag{17}\\
& \left.F(k)=\frac{(e c)^{2}}{4 \pi^{2} \hbar \omega} \omega_{\alpha \beta}^{2}\left|\left\langle a \frac{1}{2}\left\|T^{(1)}\right\| b \frac{1}{2}\right\rangle\right|^{2} \right\rvert\,\left\langle\left.\left\langle\frac{1}{2}, 1 ; \frac{1}{2},-1 \left\lvert\, \frac{1}{2}-\frac{1}{2}\right.\right\rangle\right|^{2} \delta\left(\omega-\omega_{\alpha \beta}\right),\right. \tag{18}
\end{align*}
$$

where $\theta^{\prime}$ represents the incident or transmitted angle on the vacuum side, while $\left\langle\frac{1}{2}, 1 ; \frac{1}{2},-1 \left\lvert\, \frac{1}{2}-\frac{1}{2}\right.\right\rangle$ and $\left\langle a \frac{1}{2}\left\|T^{(1)}\right\| b \frac{1}{2}\right\rangle$ denote the Clebsh-Gordan coefficient and the reduced matrix element, respectively. The notation for each expression on the left hand side of Eqs $(10-17)$ is summarized in Table 1. Corresponding to the probability ratio $R$ defined in Eq. (5), we integrate each directional-dependent emission rate in Eqs (10-17) with respect to the emission direction and sum them regarding the polarization as


Fig. 2. Photon emission from an atom. The $R$-mode corresponds to homogeneous waves emitted by the atom, and the $L$-mode corresponds to evanescent waves. Integration range of $\theta^{\prime}$ for the $R$-mode varies from 0 to $\pi / 2$ and $\theta$ for the $L$-mode ranges from $\theta_{c}$ to $\pi / 2$.


Fig. 3. Transition probability ratio $R(n, z)=w_{u p} / w_{\text {down }}$ as a function of (a) atom-surface distance and (b) refractive index. We assume $n=1.5$ in (a), and $z=$ 10 nm in (b).

Table 1: Transition probabilities to the spin-up and spin-down states depending on the polarization for the $R$-mode and $L$-mode.

| transition | homogeneous ! JRmode! K |  | evanescent!JLmode! K |  |
| :--- | :---: | :---: | :---: | :---: |
| $\left\|P_{J}, M\right\rangle \rightarrow\left\|S_{J^{\prime}}, M^{\prime}\right\rangle$ | $s$-polarization | $p$-polarization | $s$-polarization | $p$-polarization |
| $\left\|P_{\frac{1}{2}}, \frac{1}{2}\right\rangle \rightarrow\left\|S_{\frac{1}{2}}, \frac{1}{2}\right\rangle$ |  |  |  |  |
| $\left(w_{u p}\right)$ | $w_{u p}^{R}(\boldsymbol{K}, s)$ | $w_{u p}^{R}(\boldsymbol{K}, p)$ | $w_{u p}^{L}(\boldsymbol{k}, s)$ | $w_{u p}^{L}(\boldsymbol{k}, p)$ |
| $\left\|P_{\frac{1}{2}}, \frac{1}{2}\right\rangle \rightarrow\left\|S_{\frac{1}{2}},-\frac{1}{2}\right\rangle$ |  |  |  |  |
| $\left(w_{\text {down }}\right)$ | $w_{\text {down }}^{R}(\boldsymbol{K}, s)$ | $w_{\text {down }}^{R}(\boldsymbol{K}, p)$ | $w_{d o w n}^{L}(\boldsymbol{k}, s)$ | $w_{d o w n}^{L}(\boldsymbol{k}, p)$ |

$$
\begin{equation*}
w_{u p}=\int d \Omega\left[w_{u p}^{R}(\boldsymbol{K}, s)+w_{u p}^{R}(\boldsymbol{K}, p)\right]+\int d \Omega^{\prime}\left[w_{u p}^{L}(\boldsymbol{k}, s)+w_{u p}^{L}(\boldsymbol{k}, p)\right], \tag{19}
\end{equation*}
$$

$w_{\text {down }}=\int d \Omega\left[w_{\text {down }}^{R}(\boldsymbol{K}, s)+w_{d o w n}^{R}(\boldsymbol{K}, p)\right]$

$$
\begin{equation*}
+\int d \Omega^{\prime}\left[w_{d o w n}^{L}(\boldsymbol{k}, s)+w_{d o w n}^{L}(\boldsymbol{k}, p)\right] . \tag{20}
\end{equation*}
$$

Here the integration range of $\theta^{\prime}$ for the $R$-mode ranges from 0 to $\pi / 2$, whereas $\theta$ for the $L$-mode ranges from $\theta_{c}$ to $\pi / 2$. The ratio $R(n, z)=w_{u p} / w_{\text {down }}$ with the refractive index $n$ and the atom-surface distance $z$ gives the final result, including the surface boundary effects.

As an example, we consider the transition from the $6 P_{1 / 2}$ level to the $6 S_{1 / 2}$ level of a Cs atom, which has a resonance energy of $\approx 1.38 \mathrm{eV}$. Figure 3(a) shows the atom-surface distance dependence of the ratio $R(n, z)$ calculated from Eqs (19 and 20). When the refractive index $n$ is taken as 1.5 , it follows from the figure that the ratio is larger than the free space value as the atoms come close to the surface, i.e. Cs atoms are more efficiently converted to the spin-up state. In addition, the ratio becomes $1 / 2$ in Eq. (5) at the limit of an infinite atom-surface distance. Figure 3(b) shows the refractive index dependence of the ratio $R(n, z)$ when the atom-surface distance $z$ is taken as 10 nm . We find that $R(n, z)$ has a maximum value at $n \sim 1.35$ and is enhanced by $30 \%$ compared with the value of $1 / 2$ given in Eq. (5). This implies that the surface effects increase the spontaneous emission rate to the spin-up state. Although it is not shown in the figure, this result remains valid if the atomsurface distance is $<10 \mathrm{~nm}$.

The results can be qualitatively understood as follows. The relaxation rate to the spin-up or spin-down state is approximated by

$$
\begin{equation*}
\left.\left.w_{u p} \sim\left|\langle f| T_{0}\right| i\right\rangle\left.\right|^{2}\left|e_{z}\right|^{2}, \quad w_{\text {down }} \sim\left|\langle f| T_{-}\right| i\right\rangle\left.\right|^{2}\left|e_{-}\right|^{2}, \quad e_{-}=\left(e_{x}-i e_{y}\right) / \sqrt{2} . \tag{21}
\end{equation*}
$$

With the help of the Wigner-Eckart theorem, we obtain the following relation

$$
\begin{equation*}
\langle f| T_{0}|i\rangle=\langle f| T_{-}|i\rangle / \sqrt{2} \tag{22}
\end{equation*}
$$

Thus, we can approximate the ratio $R$ as

$$
\begin{equation*}
R(n, z) \sim \frac{\left|e_{z}\right|^{2}}{2\left|e_{-}\right|^{2}} . \tag{23}
\end{equation*}
$$

The increase in the relaxation rate to the spin-up state comes from the increase in $\left|e_{z}\right|$, i.e. the $z$-component of the electro-
magnetic field, which can be produced by the image dipole due to the planar surface effect.

## 4. Conclusions

In terms of the CM mode expansion of electromagnetic fields, we investigated the effects of a dielectric planar surface on the spin-control method proposed by Hori et al. (1996). Using the derived formulae, we showed the dependence of the relaxation probability ratio $R(n, z)=w_{u p} / w_{\text {down }}$ on the atom-surface distance and the refractive index for the $6 P_{1 / 2}-6 S_{1 / 2}$ levels of a Cs atom numerically. The main result indicates that the ratio has a maximum value at $n \sim 1.35$, which is enhanced by $30 \%$ over the free space value. This means that the surface effects increase the spontaneous emission rate to the spin-up state. Therefore, one can efficiently align the spins of atoms in repeated laser cycles of state-selective excitation and spontaneous emission.

Inoue \& Hori (2001) recently discussed the spontaneous emission of atoms near a dielectric surface, using the CM modes, and showed the quantum interference effects on the angular dependence of the spontaneous emission with the help of the detector modes. It will be interesting to include these quantum effects in our studies in the near future. Because the CM modes form a basis set of the quantization of the electromagnetic fields, we can discuss the quantum properties of the evanescent field such as the possibility of the squeezed state of the evanescent field.
It will also be interesting to find a normal mode for other types of boundary, such as a fibre tip, and to discuss atom manipulation using such a probe (Kobayashi et al., 2001), in the same manner as the CM mode for a planar boundary.

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## Appendix

Here we present some useful expressions related to the CM mode of electromagnetic fields. In order to describe the electromagnetic field with a planar interface, we use the solutions $\boldsymbol{\varepsilon}_{L}$ and $\boldsymbol{\varepsilon}_{R}$ of the Helmholtz equation

$$
\begin{gather*}
\nabla^{2} \boldsymbol{\varepsilon}_{L(R)}(\boldsymbol{k}, \lambda, \boldsymbol{r})+\frac{n^{2}(\boldsymbol{r}) \omega^{2}}{c^{2}} \boldsymbol{\varepsilon}_{L(R)}(\boldsymbol{k}, \lambda, \boldsymbol{r})=0, \text { where } \\
n(\boldsymbol{r})= \begin{cases}1, & z>0, \\
n, & z<0,\end{cases} \tag{A1}
\end{gather*}
$$

where $\lambda$ stands for $s$ - or $p$-polarization. In addition, the suffix $R$ indicates the $R$-mode, in which plane waves are incident from the vacuum side, and $L$ designates the $L$-mode, in which plane waves are incident from the dielectric side (see Fig. A1). From incident $\boldsymbol{\varepsilon}^{I}$, reflected $\boldsymbol{\varepsilon}^{R}$, and transmitted $\boldsymbol{\varepsilon}^{T}$ waves as solutions of Eq. (A1), the CM mode functions are defined as

$$
\begin{gather*}
\boldsymbol{\varepsilon}_{L}(\boldsymbol{k}, \boldsymbol{\lambda}, \boldsymbol{r})=\boldsymbol{\varepsilon}_{L}^{I}(\boldsymbol{k}, \boldsymbol{\lambda}, \boldsymbol{r})+\boldsymbol{\varepsilon}_{L}^{R}(\boldsymbol{k}, \boldsymbol{\lambda}, \boldsymbol{r})+\boldsymbol{\varepsilon}_{L}^{T}(\boldsymbol{k}, \boldsymbol{\lambda}, \boldsymbol{r}),  \tag{A2}\\
\boldsymbol{\varepsilon}_{R}(\boldsymbol{K}, \boldsymbol{\lambda}, \boldsymbol{r})=\boldsymbol{\varepsilon}_{R}^{I}(\boldsymbol{K}, \boldsymbol{\lambda}, \boldsymbol{r})+\boldsymbol{\varepsilon}_{R}^{R}(\boldsymbol{K}, \boldsymbol{\lambda}, \boldsymbol{r})+\boldsymbol{\varepsilon}_{R}^{T}(\boldsymbol{K}, \boldsymbol{\lambda}, \boldsymbol{r}) . \tag{A3}
\end{gather*}
$$

Because these mode functions form a complete orthonormal set for transverse fields (Bialynicki-Birula \& Brojan, 1972; Carniglia \& Mandel, 1971), we can expand an arbitrary electric field using the CM modes as follows:


Fig. A1. CM triplet modes: the R-mode (left) and the $L$-mode (right). Only the $L$-mode includes evanescent waves.

$$
\begin{align*}
\hat{E}(\boldsymbol{r}, t)= & \frac{1}{(2 \pi)^{3}} \sum_{\lambda}\left\{\int_{k_{3}>0} d^{3} k \sqrt{\frac{\hbar \omega}{\varepsilon_{0}}}\left(\boldsymbol{\varepsilon}_{L}(\boldsymbol{k}, \lambda, \boldsymbol{r}) \hat{u}(\boldsymbol{k}, \boldsymbol{\lambda}) \mathrm{e}^{-i \omega t}+\text { h.c. }\right)\right. \\
& \left.+\int_{K_{3}<0} d^{3} K \sqrt{\frac{\hbar \omega}{\varepsilon_{0}}}\left(\boldsymbol{\varepsilon}_{R}(\boldsymbol{K}, \boldsymbol{\lambda}, \boldsymbol{r}) \hat{v}(\boldsymbol{K}, \boldsymbol{\lambda}) \mathrm{e}^{-i \omega t}+\text { h.c. }\right)\right\},(\mathrm{A} 4 \tag{A4}
\end{align*}
$$

where $\hat{u}$ and $\hat{v}$ obey the following commutation relations:

$$
\begin{equation*}
\left[\hat{u}(\boldsymbol{k}, \boldsymbol{\lambda}), \hat{u}^{\dagger}\left(\boldsymbol{k}^{\prime}, \lambda^{\prime}\right)\right]=(2 \pi)^{3} \hbar \delta\left(\boldsymbol{k}-\boldsymbol{k}^{\prime}\right) \delta_{\lambda \lambda^{\prime}}, \tag{A5}
\end{equation*}
$$

$$
\begin{equation*}
\left[\hat{v}(\boldsymbol{K}, \boldsymbol{\lambda}), \hat{v}^{\dagger}\left(\boldsymbol{K}^{\prime}, \lambda^{\prime}\right)\right]=(2 \pi)^{3} \hbar \delta\left(\boldsymbol{K}-\boldsymbol{K}^{\prime}\right) \boldsymbol{\delta}_{\lambda \lambda^{\prime}} \tag{A6}
\end{equation*}
$$

and the CM mode functions are described as

$$
\begin{equation*}
\boldsymbol{\varepsilon}_{R(L)}(\boldsymbol{k}, \boldsymbol{\lambda}, \boldsymbol{r})=\boldsymbol{e}(\boldsymbol{\lambda}) a_{R(L)}(\lambda) \mathrm{e}^{\mathrm{i} \boldsymbol{k} \cdot \boldsymbol{r}}, \tag{A7}
\end{equation*}
$$

with the polarization vector $\boldsymbol{e}(\lambda)$ and Fresnel's coefficient $a_{R(L)}(\lambda)$. Using the relation $E=-\partial A / \partial t$ for Eq. (A4), we can also express the vector potential in terms of the CM mode as

$$
\begin{align*}
A(\boldsymbol{r}, t)= & -\frac{i}{(2 \pi)^{3}} \sum_{\lambda}\left\{\int_{k_{3}>0} d^{3} k \sqrt{\frac{\hbar}{\varepsilon_{0} \omega}}\left(\boldsymbol{\varepsilon}_{L}(\boldsymbol{k}, \boldsymbol{\lambda}, \boldsymbol{r}) \hat{u}(\boldsymbol{k}, \boldsymbol{\lambda}) \mathrm{e}^{-i \omega t}-\text { h.c. }\right)\right. \\
& \left.+\int_{K_{3}<0} d^{3} K \sqrt{\frac{\hbar}{\varepsilon_{0} \omega}}\left(\boldsymbol{\varepsilon}_{R}(\boldsymbol{K}, \lambda, \boldsymbol{r}) \hat{v}(\boldsymbol{K}, \boldsymbol{\lambda}) \mathrm{e}^{-i \omega t}-\text { h.c. }\right)\right\} .(\mathrm{A} 8 \tag{A8}
\end{align*}
$$

Owing to the relation $\nabla \times E=-\partial B / \partial t$, analogous formulas hold for magnetic fields, with the annihilation operators $\hat{u}$ and $\hat{v}$ in common. Therefore, the Hamiltonian for the electromagnetic field is described by the CM modes as:

$$
\begin{align*}
\hat{H}= & \frac{1}{2} \int d^{3} r\{\boldsymbol{D} \cdot \boldsymbol{E}+\boldsymbol{B} \cdot \boldsymbol{H}\} \\
= & \frac{1}{(2 \pi)^{3}} \sum_{\lambda}\left\{\int_{K_{3}>0} d^{3} k \hbar \omega \hat{u}^{\dagger}(\boldsymbol{k}, \boldsymbol{\lambda}) \hat{u}(\boldsymbol{k}, \boldsymbol{\lambda})\right. \\
& \left.+\int_{K_{3}<0} d^{3} K \hbar \omega \hat{v}^{\dagger}(\boldsymbol{K}, \boldsymbol{\lambda}) \hat{v}(\boldsymbol{K}, \boldsymbol{\lambda})\right\} . \tag{A9}
\end{align*}
$$

It follows that the CM modes form normal modes for the planar boundary condition and $\hat{u}, \hat{u}^{\dagger}$ and $\hat{v}, \hat{v}^{\dagger}$ are the annihilation and creation operators for $L$-mode and $R$-mode photons, respectively.


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