Oscillator-strength enhancement of electric-dipole-forbidden transitions in evanescent light at total reflection

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The absorption line strengths of electric-dipole-forbidden (magnetic dipole and electric quadrupole) transitions have been calculated in the evanescent light field that accompanies the total reflection of light. Owing to the complex wave and polarization vectors of the evanescent light, the apparent oscillator strength is enhanced from that in the propagating light and the enhancement for the electric quadrupole transition depends on the polarization vector of the incident light. Separation of the contribution from each component of the wave and polarization vectors is proposed with a magnetic-sublevel-resolved measurement for the (*s*-*d*) electric quadrupole transition.

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I. INTRODUCTION

The optical near field, or evanescent light field, gathers interest for applications in optical detection beyond the diffraction limit of light. Owing to the spatial localization (inhomogeneity) of the field, the evanescent light has a characteristic wave vector. For example, the wave number of the evanescent light field produced by a small aperture such as the probe of a scanning near field optical microscope (SNOM) has a distribution, the width of which corresponds to the inverse of the aperture diameter [1]. For the case of internal total reflection at an interface, the wave vector of the evanescent light together with the polarization vector is complex so as to satisfy Snell's law at the interface [2].

The vast majority of observed spectra for the optical near field arise from electric dipole transitions. However, electric-dipole-forbidden transitions such as magnetic dipole and electric quadrupole transitions may be of interest because such transitions are sensitive to field inhomogeneities. For example, the interaction between a light field and an atomic system with an electric quadrupole transition from the initial state $|i\rangle$ to the final state $|f\rangle$ depends explicitly on the unit polarization vector $\hat{\mathbf{e}}$ and the wave vector $\mathbf{k}$ of the light, and has the form of $\hat{\mathbf{e}}\cdot\langle f|\mathsf{Q}|i\rangle\cdot\mathbf{k}$, where Q is the quadrupole tensor [3]. It is expected that the electric quadrupole transition strength is enhanced in the evanescent light from that in the propagating homogenous light in free space. Recently, such enhancements have been observed for the Cs $6\,^2S_{1/2}\rightarrow 5\,^2D_{5/2}$ electric quadrupole transition by reflection spectroscopy [4], the details of which are reported in the preceding paper [5].

In this article, we report a theoretical aspect of the enhancement of the magnetic dipole and electric quadrupole transitions in the evanescent light field which accompanies the total reflection of light.

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II. ENHANCEMENT OF THE OSCILLATOR STRENGTH OF THE MAGNETIC DIPOLE AND ELECTRIC QUADRUPOLE TRANSITIONS IN AN EVANESCENT FIELD AT TOTAL REFLECTION

A. The wave vector and the electric field of an evanescent field at total reflection

Let a light beam propagate in a medium having a high refractive index n_1 to a plane interface to a medium with a low refractive index n_2, with the angle of incidence θ_1 to the interface. We call the respective mediums medium 1 and medium 2 hereafter. When the light beam is refracted at the interface and propagates into medium 2, phase matching between the propagating light waves in media 1 and 2 must be fulfilled, and then the following well-known relation, called Snell's law, holds:

$$n_1 \sin\theta_1 = n_2 \sin\theta_2, \tag{1}$$

where θ_2 is the angle of refraction. However, when θ_1 is larger than the critical angle defined by

$$\theta_c = \sin^{-1}(n_1/n_2), \tag{2}$$

phase matching is no longer fulfilled, and then total reflection occurs as shown in Fig. 1. Even in this case, we may assume that Snell's law holds and the angle of refraction is expressed by [2]

$$\sin\theta_2 = (n_1/n_2)\sin\theta_1,$$
$$\cos\theta_2 = i\sqrt{(n_1/n_2)^2\sin^2\theta_1 - 1}. \tag{3}$$

The refracted light showing exponential decay, called the evanescent light, is localized in medium 2 at the interface and does not propagate into medium 2. The wave vector of the evanescent light is given as

$$\mathbf{k} \equiv (k_x,k_y,k_z) = (n_2k_0\sin\theta_2, 0, n_2k_0\cos\theta_2)$$
$$= \left(n_1k_0\sin\theta_1, 0, ik_0\sqrt{n_1^2\sin^2\theta_1 - n_2^2}\right), \tag{4}$$

where k_0 is the wave number of the propagating light in

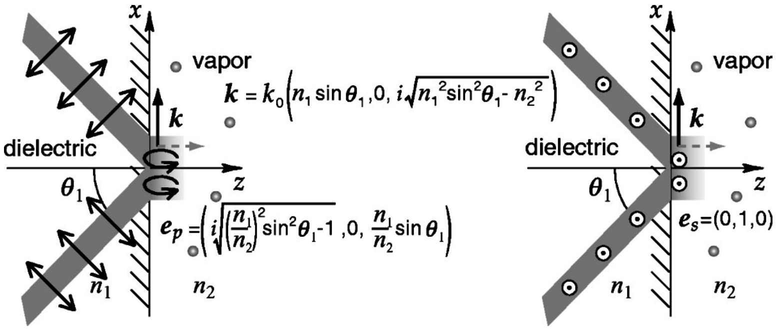

FIG. 1. A schematic illustration of the internal total reflection and the relevant evanescent field with the wave and polarization vectors for p (left) and s (right) polarizations.

vacuum. Here we define the x axis parallel to the interface in the incident plane and the z axis perpendicular to the interface as shown in Fig. 1. The real k_x, which is larger than k_0, is known as the pseudomomentum and the imaginary k_z leads to a short penetration depth of the order of the light wavelength.

From the boundary condition for Maxwell's equations at the interface, the electric field amplitudes of the refracted light are given from the Fresnel formulas as

$$T_{2s} = \frac{2n_1 \cos\theta_1}{n_1 \cos\theta_1 + \sqrt{n_2^2 - n_1^2 \sin^2\theta_1}} E_{1s}, \tag{5}$$

$$T_{2p} = \frac{2n_1 \cos\theta_1}{n_2 \cos\theta_1 + (n_1/n_2)\sqrt{n_2^2 - n_1^2 \sin^2\theta_1}} E_{1p}, \tag{6}$$

for s and p polarizations of the incident light, respectively. $E_{1s,1p}$ are the corresponding electric field amplitudes of the incident light. From these formulas together with Eq. (3), the electric field of the evanescent light for s polarization is given as

$$\mathbf{E}_{2s} = (0,1,0)|T_{2s}| \equiv \mathbf{e}_s|T_{2s}| \tag{7}$$

and that for p polarization is

$$\mathbf{E}_{2p} = (-|T_{2p}|\cos\theta_2, 0, |T_{2p}|\sin\theta_2) = \left(-i\sqrt{\left(\frac{n_1}{n_2}\right)^2 \sin^2\theta_1 - 1}, 0, \frac{n_1}{n_2}\sin\theta_1\right)|T_{2p}| \equiv \mathbf{e}_p|T_{2p}|, \tag{8}$$

where $\mathbf{e}_s$ and $\mathbf{e}_p$ are the polarization vectors of the evanescent light. In contrast to the wave vector, the polarization vector depends on the polarization of the incident light. The characteristics of the wave and polarization vectors of the evanescent light are schematically summarized in Fig. 1.

The magnitude of the polarization vector for the p polarization is known to be larger than unity as

$$|\mathbf{e}_p| = \frac{1}{n_2}\sqrt{2n_1^2 \sin^2\theta_1 - n_2^2} \geqslant 1. \tag{9}$$

Here we note the effect of this polarization vector. Let medium 2 be an atomic vapor. The strength of the generated evanescent field for p polarization is considered to be $|\mathbf{E}_{2p}|$, not $|T_{2p}|$ [2]. Therefore, the induced atomic polarization may be enhanced by a factor of the magnitude of the polarization vector and the enhancement may be detected by light scattering or fluorescence measurement from the vapor side. This phenomenon is due to the enhancement of the light field, and therefore takes place in any optical transitions such as the electric dipole transition and the electric quadrupole transition. In total reflection spectroscopy, however, the enhancement is canceled by the generation process of radiation into medium 1 from the induced atomic polarization because the radiation generation is reduced by $1/|\mathbf{e}_p|$, which is the reverse process of evanescent light generation. This is the physical origin of the absolute square of the polarization vector in the denominator of Eq. (4.6) in Ref. [6].

B. Oscillator strength of the magnetic dipole and the electric quadrupole transitions in evanescent light at total reflection

With the approximation of $e^{i\mathbf{k}\cdot\mathbf{r}} \approx 1 + i\mathbf{k}\cdot\mathbf{r}$ for the light field, the oscillator strength for a transition from the initial state $|i\rangle$ to the final state $|f\rangle$ is expressed as [3,7]

$$f_{\text{total}} = \frac{4\pi m_e \nu_0}{g\hbar}\left[\sum_m |\hat{\mathbf{e}} \cdot \langle f|\mathbf{r}|i\rangle|^2 + \frac{c^2}{4\pi^2 e^2 \nu_0^2} \times \sum_m |(\mathbf{k}\times\hat{\mathbf{e}}) \cdot \langle f|\boldsymbol{\mu}|i\rangle|^2 + \frac{1}{4}\sum_m |\hat{\mathbf{e}} \cdot \langle f|\mathsf{Q}|i\rangle \cdot \mathbf{k}|^2\right], \tag{10}$$

where m_e is the electron mass, ν_0 is the resonance frequency of the transition, g is the degeneracy of the initial state, $\hbar$ is the Planck constant, m shows the magnetic sublevels, e is the elementary charge, c is the speed of light, $\mathbf{r}$ is the position operator of the electron, $\hat{\mathbf{e}}$ is the unit polarization vector of the light field, and $\boldsymbol{\mu}$ is the magnetic moment operator. Since the oscillator strength corresponds to the efficiency of polarization generation by the field at the atom and is independent of the field intensity, we adopted the unit polarization vector for the oscillator strength in the evanescent field in Eq. (10) [6]. The electric quadrupole tensor Q is expressed as [3]

$$\mathsf{Q} = \begin{pmatrix} \frac{r^2}{3}(3\sin^2\theta\cos^2\phi - 1) & r^2\sin^2\theta\sin\phi\cos\phi & r^2\sin\theta\cos\theta\cos\phi \\ r^2\sin^2\theta\sin\phi\cos\phi & \frac{r^2}{3}(3\sin^2\theta\sin^2\phi - 1) & r^2\sin\theta\cos\theta\sin\phi \\ r^2\sin\theta\cos\theta\cos\phi & r^2\sin\theta\cos\theta\sin\phi & \frac{r^2}{3}(3\cos^2\theta - 1) \end{pmatrix}, \quad (11)$$

in the polar coordinate (r, θ, ϕ).

The first, second, and third terms on the right-hand side of Eq. (10) correspond to the electric dipole, magnetic dipole, and electric quadrupole transitions, respectively. For the oscillator strength of the electric dipole transition, there is no modification owing to the wave and polarization vectors of the evanescent light. Since the polarization vector and the wave vector of the evanescent light at total reflection are orthogonal with each other for both the s and p polarizations, the oscillator strength of the magnetic dipole transition is modified with the enhancement factor of $|\mathbf{k}\times\hat{\mathbf{e}}|^2/k_0^2=(k_x^2+k_z^2)/k_0^2$ from that in the propagating light. For the electric quadrupole transition, the modification is not trivial. In the following we calculate the oscillator strength for the (s-d) electric quadrupole transition of an alkali-metal atom taken as an example.

The wave function is written as $R_{nL}(r)Y_{Lm_L}(\theta,\phi)$ with the radial function $R_{nL}(r)$ and the spherical harmonics $Y_{Lm_L}(\theta,\phi)$, where n, L, and m_L are the principal, azimuthal, and magnetic quantum numbers, respectively. The radial integral of the matrix elements is independent of m_L. The oscillator strength f_Q is calculated as

$$f_Q = \frac{\pi m_e \nu_0}{\hbar}|\langle R_{n'2}|r^2|R_{n0}\rangle|^2 \sum_{m_L}\left|\hat{\mathbf{e}}\cdot\left\langle Y_{2m_L}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}\right|^2. \quad (12)$$

Let the propagating (homogeneous) light beam pass through the atomic vapor (medium 2) along the x direction, and then $\mathbf{k}_0=(k_0,0,0)$. The polarization of the light is assumed to be parallel to the y direction, and then $\mathbf{e}_0=(0,1,0)$. The matrix elements for the angular part are

$$\left|\hat{\mathbf{e}}_0\cdot\left\langle Y_{20}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}_0\right|^2 = 0, \quad (13)$$

$$\left|\hat{\mathbf{e}}_0\cdot\left\langle Y_{2\pm1}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}_0\right|^2 = 0, \quad (14)$$

$$\left|\hat{\mathbf{e}}_0\cdot\left\langle Y_{2\pm2}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}_0\right|^2 = \frac{1}{30}k_0^2, \quad (15)$$

and then the oscillator strength is

$$f_Q^0 = \frac{\pi m_e \nu_0}{\hbar}|\langle R_{n'2}|r^2|R_{n0}\rangle|^2\frac{k_0^2}{15}. \quad (16)$$

In the evanescent light for s polarization, the unit polarization vector is $\hat{\mathbf{e}}_s=(0,1,0)$. The matrix elements are

$$\left|\hat{\mathbf{e}}_s\cdot\left\langle Y_{20}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}\right|^2 = 0, \quad (17)$$

$$\left|\hat{\mathbf{e}}_s\cdot\left\langle Y_{2\pm1}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}\right|^2 = \frac{|k_z|^2}{30} = \frac{n_1^2\sin^2\theta_1 - n_2^2}{30}k_0^2, \quad (18)$$

$$\left|\hat{\mathbf{e}}_s\cdot\left\langle Y_{2\pm2}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}\right|^2 = \frac{|k_x|^2}{30} = \frac{n_1^2\sin^2\theta_1}{30}k_0^2. \quad (19)$$

At $\theta_1=\theta_c$, these equations coincide with Eqs. (13)–(15) from their identical symmetry.

The oscillator strength for s polarization, f_Q^s, is written as

$$f_Q^s = \frac{k_x^2+k_z^2}{k_0^2}f_Q^0 = (2n_1^2\sin^2\theta_1 - n_2^2)f_Q^0 \equiv g_s(\theta_1)f_Q^0; \quad (20)$$

here $g_s(\theta_1)$ is called the enhancement factor. The enhancement factor simply depends on the magnitude of the wave vector of the evanescent light as the magnetic dipole transition does. Equations (18) and (19) show that each matrix element corresponds to a component of the wave vector of the evanescent field. This suggests that it may be possible to deduce the magnitude of each wave vector component directly from an observation of the contribution from the relevant matrix element.

In the case of p polarization, the unit polarization vector is $\hat{\mathbf{e}}_p\equiv(\epsilon_x^p,\epsilon_y^p,\epsilon_z^p)=(2n_1^2\sin^2\theta_i-n_2^2)^{-1/2}(-i\sqrt{n_1^2\sin^2\theta_i-n_2^2}, 0, n_1\sin\theta_i)$. The matrix elements are

$$\left|\hat{\mathbf{e}}_p\cdot\left\langle Y_{20}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}\right|^2 = \frac{|-\epsilon_x^p k_x + 2\epsilon_z^p k_z|^2}{45} = \frac{n_1^2\sin^2\theta_1(n_1^2\sin^2\theta_1 - n_2^2)}{5(2n_1^2\sin^2\theta_1 - n_2^2)}k_0^2, \quad (21)$$

$$\left|\hat{\mathbf{e}}_p\cdot\left\langle Y_{2\pm1}\left|\frac{\mathsf{Q}}{r^2}\right|Y_{00}\right\rangle\cdot\mathbf{k}\right|^2 = \frac{|\epsilon_x^p k_z + \epsilon_z^p k_x|^2}{30} = \frac{2n_1^2\sin^2\theta_1 - n_2^2}{30}k_0^2, \quad (22)$$

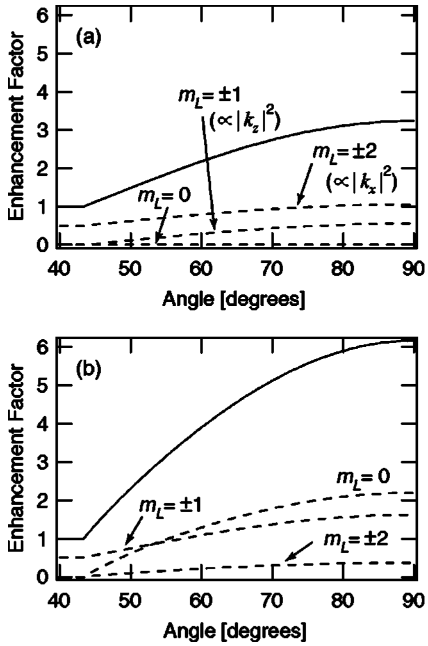

FIG. 2. The θ_1 dependence of the enhancement factor for the (s-d) electric quadrupole transition in the m_L basis for s (a) and p (b) polarizations (solid lines). The dotted lines in both figures show the contribution to the enhancement factor from the relevant matrix element; the magnetic sublevels are shown in the figure.

$$\left|\hat{\mathbf{e}}_p \cdot \left\langle Y_{2\pm 2}\left|\frac{\mathrm{Q}}{r^2}\right|Y_{00}\right\rangle \cdot \mathbf{k}\right|^2 = \frac{|\epsilon_x^p k_x|^2}{30} = \frac{n_1^2 \sin^2\theta_1 (n_1^2\sin^2\theta_1 - n_2^2)}{30(2n_1^2\sin^2\theta_1 - n_2^2)} k_0^2. \quad (23)$$

In contrary to the case of s polarization, these matrix elements depend on the components of the wave and polarization vectors. However, with the use of the information on the wave vector components from the measurement for s polarization, we may extract information about the polarization vector components from Eqs. (21)–(23).

The oscillator strength for p polarization f_Q^p is written as

$$f_Q^p = \frac{4|\epsilon_x^p k_x|^2 + 4|\epsilon_z^p k_z|^2 + 3|\epsilon_x^p k_z|^2 + 3|\epsilon_z^p k_x|^2 + 10|\epsilon_x^p \epsilon_z^p k_x k_z|}{3k_0^2} f_Q^0 = \left[\frac{8n_1^4\sin^4\theta_1 - 8n_1^2n_2^2\sin^2\theta_1 + n_2^4}{2n_1^2\sin^2\theta_1 - n_2^2}\right] f_Q^0 \equiv g_p(\theta_1) f_Q^0, \quad (24)$$

with the enhancement factor $g_p(\theta_1)$ for p polarization [8]. The θ_1 dependences of the enhancement factors and the contributions to the factors are shown in Figs. 2(a) and 2(b). Here, we use $n_1 = 1.456$ [5]. The factor for p polarization even reaches 6 at the angle of incidence $\pi/2$ rad.

C. Zeeman effects on the cesium ($6\,^2S_{1/2} \to 5\,^2D_{5/2}$) electric quadrupole transition in an evanescent field

In the above subsection, we suggested the possibility of separating the contributions from the components of the wave and polarization vectors. We consider here its feasibility with the model system of the Cs ($6\,^2S_{1/2} \to 5\,^2D_{5/2}$) electric quadruple transition, with which the enhancement of the oscillator strength in the evanescent light was observed [4].

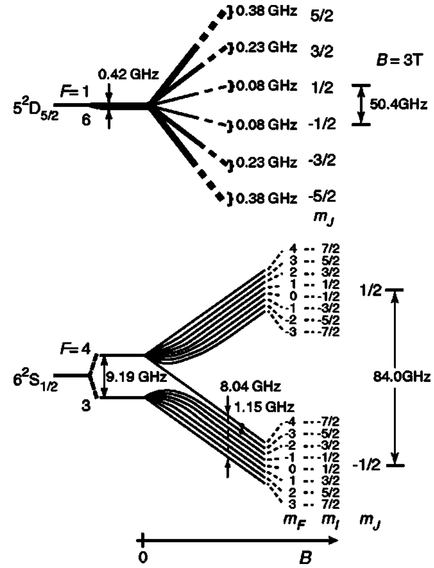

FIG. 3. Zeeman effects on the energy levels relevant to the Cs ($6\,^2S_{1/2} \to 5\,^2D_{5/2}$) electric quadrupole transition (the nuclear spin $I = 7/2$) up to 3 T.

Let a magnetic field is applied along the z direction, the intensity of which is assumed small enough so that the Zeeman effect is much smaller than the spin-orbit interaction. The Zeeman shift from the first-order perturbation theory under the condition $\mu_B B \gg \hbar A_{\mathrm{HF}}$ is given as [9]

$$\Delta E_{m_J, m_I} = 2\pi\hbar A_{\mathrm{HF}} m_J m_I + g_J \mu_B m_J B, \quad (25)$$

where g_J is the Landé g factor on the m_J basis, μ_B is the Bohr magneton, B is the magnetic flux density, and A_{HF} is the HFS splitting constant. The respective Zeeman splittings are calculated to be 28.0 and 16.8 GHz/T for the $6\,^2S_{1/2}$ and $5\,^2D_{5/2}$ states [10]. The Zeeman splittings are shown in Fig. 3.

For the s state, the Clebsch-Gordan coefficients for the m_J basis from the m_L basis are unity or null depending on the spin state, while some m_J states in the d state are mixtures of two m_L states. From the orthogonality of the electron spin wave functions, the matrix elements on the m_J basis are obtained to be

$$\left|\hat{\mathbf{e}} \cdot \left\langle d_{m_{J'}}\left|\frac{\mathrm{Q}}{r^2}\right|s_{m_J}\right\rangle \cdot \mathbf{k}\right|^2 = \beta\xi_{m_{J'},m_J}^2 \left|\hat{\mathbf{e}} \cdot \left\langle d_{m_{L'}}\left|\frac{\mathrm{Q}}{r^2}\right|s_{m_L}\right\rangle \cdot \mathbf{k}\right|^2 \quad (26)$$

where J and J' are the total angular momentum quantum numbers of the initial and final states, respectively, $\beta = f_Q / f_Q^{6S\text{-}5D}$ is the ratio of the oscillator strength of the $6\,^2S_{1/2} - 5\,^2D_{5/2}$ transition f_Q to the total oscillator strength of the $6S$-$5D$ transition $f_Q^{6S\text{-}5D}$ [11], $\xi_{m_{J'},m_J}$ is the relevant Clebsch-Gordan coefficient, $|d_{m_{L'}}\rangle$ and $|s_{m_L}\rangle$ are the atomic wave functions on the m_L basis, and $|d_{m_{J'}}\rangle$ and $|s_{m_J}\rangle$ are those

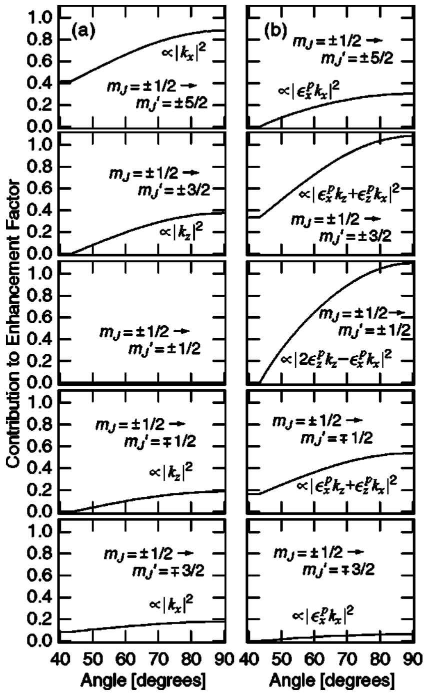

FIG. 4. Contribution to the enhancement factor for the ($S_{1/2} \rightarrow D_{5/2}$) electric quadrupole transition in the m_J basis for s (a) and p (b) polarizations.

on the m_J basis. From this relation, the contributions to the enhancement factors can be readily calculated. Figure 4 shows the results. For s polarization, the contributions from $m_J = \pm 1/2$ to $m_{J'} = \pm 5/2$ and from $m_J = \pm 1/2$ to $m_{J'} = \mp 3/2$ are proportional to $|k_x|^2$ while those from $m_J = \pm 1/2$ to $m_{J'} = \pm 3/2$ and from $m_J = \pm 1/2$ to $m_{J'} = \mp 1/2$ are proportional to $|k_z|^2$.

Figure 5 shows the calculated spectra at 3 T, which show the attenuation spectra in the reflection for $\theta_1 = \theta_c$ (a),(b), $\theta_1 = 45°$ (c),(d), and $\theta_1 = 70°$ (e),(f). Figures 5(a), 5(c), and 5(e) are for s polarization and Figs. 5(b), 5(d), and 5(f) for p polarization. In the calculation we assume the same experimental conditions as those in the preceding experimental paper except for the presence of the magnetic field [5]. All the observable transitions between the magnetic sublevels on the m_J basis are clearly resolved. At $\theta_1 = 70°$, a slight overlap of the wings of the absorption lines is seen, which is due to the θ_1-dependent transit-time broadening and Doppler broadening in the evanescent field. The attenuations in the reflection spectrum at the peak are estimated to be on the order of 10^{-7} to 10^{-8}, which is less than our detection sensitivity of 10^{-6}–10^{-7} in the preceding experimental paper. In order to overcome the difficulty, it may be possible to increase the sensitivity with the increase in the frequency modulation amplitude up to the width of the spectra. The increase in the atom density and the use of the multiple total reflection method [12] may be other candidates.

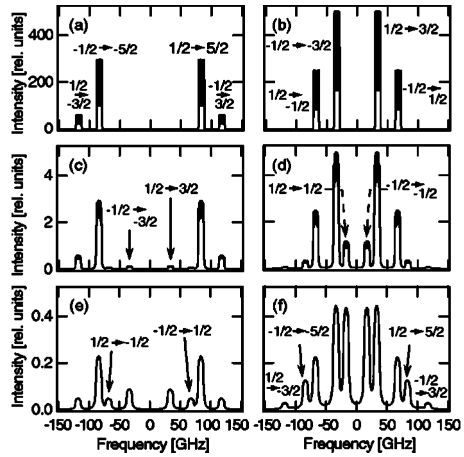

FIG. 5. Calculated spectra of the Cs ($6\,^2S_{1/2} \rightarrow 5\,^2D_{5/2}$) electric quadrupole transition at 3 T for s polarization (a),(c),(e) and for p polarization (b),(d),(f). (a),(b) $\theta_1 = \theta_c$ (43.38°); (c),(d) $\theta_1 = 45°$; (e),(f) $\theta_1 = 70°$.

III. CONCLUSION

We considered the enhancement of the apparent oscillator strength in the evanescent light field at total reflection for both a magnetic dipole transition and an electric quadrupole transition. The matrix elements of the electric quadrupole interaction between the evanescent light field and the ($s-d$) atomic system are presented. Each matrix element corresponds to a component of the wave vector of the evanescent light for s polarization. The components of the wave and polarization vectors are mixed in the matrix elements for p polarization, which results in larger enhancement of the oscillator strengths than that for s polarization. We also show the possibility of separating the contribution from each component of the wave vector and that of the polarization vector in the reflection spectra with a model system of the Cs ($6\,^2S_{1/2} \rightarrow 5\,^2D_{5/2}$) electric quadrupole transition in the strong magnetic field.

Since the enhancement of an electric quadrupole transition reflects the wave and polarization vectors of the evanescent light, it may be useful to characterize the evanescent light even at a small aperture such as the probe of a scanning near field optical microscope. When the size of the aperture is much smaller than the light wavelength, a substantial enhancement is expected in the evanescent light localized near the aperture.

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