

Effect of recoil in resonance fluorescence*

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The effect of recoil on the frequency of the spontaneously emitted photons in resonance fluorescence processes is examined. The rate of fluorescence is calculated in the limit of a weak external field. It is found that conservation of energy and linear momentum for the entire scattering process determines the frequency spectrum of the fluorescence. A simple experiment is proposed to measure the frequency shifts due to recoil.

I. INTRODUCTION

An atom will undergo recoil when it absorbs or emits radiation. This ever-present physical phenomenon, termed "photon recoil" for short, leads to frequency shifting in spectral line shapes. Whereas it is now relatively easy to measure the deflection of an atomic beam due to radiation pressure,^{1,2} it is far more difficult to observe spectral shifts since they amount to only $\approx 10^3$ – 10^6 Hz for optical transitions. There has been some effort³ to measure these shifts in the saturation spectroscopy of molecular transitions with natural widths $\approx 10^3$ Hz, and theories of saturation spectroscopy with photon recoil have appeared.^{4,5} However, to our knowledge, there have not been efforts to measure these shifts in fluorescence experiments. With the development of tunable, nearly monochromatic sources, such measurements may now be feasible since, in the limit of a weak applied field, the spectral width of the fluorescence should mirror that of the excitation spectrum⁶ and, consequently, be much smaller than the natural width.

Many calculations of resonance fluorescence exist, but none, to our knowledge, have concentrated on the determination of these shifts. Furthermore, the photon recoil effect is certainly going to play an important role in molecular spectroscopy where the natural linewidth is smaller or has the same order of magnitude as the recoil shift, so that it might be useful to present a first-principle calculation of resonance fluorescence for moving atoms.

We should note that the calculation will be restricted to the weak-field limit. More precisely, we shall assume that the probability for an atom to fluoresce during its passage through the interaction region is much less than unity, owing to the weakness of the field. For this limit, one does not have to consider effects arising from multiple excitations.⁷ On the other hand, to arrive at a fluorescence rate, we must assume that the time an atom spends in the interaction region is much

greater than the lifetime τ of the atomic states under consideration. Consequently, the interaction time t_I must satisfy the inequality

$$\tau \ll t_I < |\vec{\mu} \cdot \vec{E}/\hbar|^{-1}, \quad (1)$$

where $|\vec{\mu} \cdot \vec{E}/\hbar|$ is the effective excitation rate produced by the field.

In Sec. II, the Schrödinger equation for the system is developed, and a perturbation solution is given in Sec. III. In Sec. IV, the results are discussed and an experiment is proposed for measuring photon recoil shifts in a fluorescence experiment.

II. SCHRÖDINGER EQUATION

We consider the two-level system shown in Fig. 1, which has nondegenerate energy states $|a\rangle$ and $|b\rangle$ separated by frequency ω_0 . The atom is subjected to a classical monochromatic applied field, and radiates spontaneously due to interaction with the vacuum radiation field. We ultimately seek the spontaneously emitted spectrum. The Hamiltonian of the complete system is given by

$$H = H_{c.m.} + H_e + H_f + H' + V \quad (2)$$

where $H_{c.m.}$ is the Hamiltonian for the free-atom center-of-mass motion, H_e is the electronic Hamiltonian for the free atom, H_f is the free-vacuum field Hamiltonian, H' is the applied classical field-atom interaction, and V is the vacuum field-atom interaction. Both H' and V have nonvanishing ma-

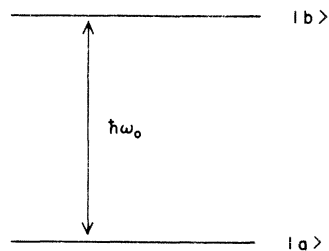


FIG. 1. The two-level system considered in this work.

trix elements only between states $|a\rangle$ and $|b\rangle$.

The time-dependent Schrödinger equation of the system is

$$i\hbar \frac{\partial |\Psi t\rangle}{\partial t} = H |\Psi t\rangle, \quad (3)$$

and the wave function $|\Psi t\rangle$ can be written as a superposition of product states involving the eigenstates $|\vec{p}\rangle$ of $H_{c.m.}$ (\vec{p} is the atomic center-of-mass linear momentum), $|\alpha\rangle$ of H_e (α is an energy eigenstate of the free atom), and $|\{n\}\rangle$ of H_f (n repre-

sents all possible states of photons with wave vector \vec{k}_i and polarization λ_i , i.e., $|\{n\}\rangle = \prod_{i=1}^{\infty} |n_{\vec{k}_i, \lambda_i}\rangle$). Explicitly, one writes

$$|\Psi t\rangle = \sum_{\alpha=a,b} \sum_{\{n\}} \int d^3p |\vec{p}\alpha\{n\}\rangle \langle \vec{p}\alpha\{n\} | \Psi t\rangle.$$

We will be interested only in terms containing, at most, one photon, and if the interaction representation is introduced, $|\Psi t\rangle$ can be written in the form

$$\begin{aligned} |\Psi t\rangle = & \sum_{\alpha=a,b} \int d^3p |\vec{p}\alpha 0\rangle A_{\alpha 0}(\vec{p}, t) \exp[-i(E_{\alpha} + E_p)t/\hbar] \\ & + \sum_{\alpha=a,b} \sum_{\vec{k}\lambda} \int d^3p |\vec{p}\alpha\vec{k}\lambda\rangle A_{\alpha\vec{k}\lambda}(\vec{p}, t) \exp[-i(E_{\alpha} + E_p + \hbar\omega_k)t/\hbar] \\ & + (\text{terms involving two or more photons}), \end{aligned} \quad (4)$$

where

$$A_{\alpha\{n\}}(\vec{p}, t) = \langle \vec{p}\alpha\{n\} | \Psi t\rangle \exp[i(E_{\alpha} + E_p + \hbar\omega_{\{n\}})t/\hbar]. \quad (5)$$

E_{α} is the energy of $|\alpha\rangle$, $E_p = p^2/2M$ (M being the mass of the atom), and $\hbar\omega_{\{n\}}$ is the energy of the emitted photon of type $\{n\}$.

Substituting Eq. (4) into Eq. (3), and performing appropriate inner products, we have

$$i\hbar \frac{\partial}{\partial t} A_{a0}(\vec{p}', t) = \int d^3p A_{b0}(\vec{p}, t) \langle \vec{p}' a 0 | H' | \vec{p} b 0\rangle \exp[i(E_{p',p} - \hbar\omega_0)t/\hbar], \quad (6a)$$

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} A_{b0}(\vec{p}', t) = & \int d^3p A_{a0}(\vec{p}, t) \langle \vec{p}' b 0 | H' | \vec{p} a 0\rangle \exp[i(E_{p',p} + \hbar\omega_0)t/\hbar] \\ & + \sum_{\vec{k}\lambda} \int d^3p A_{a\vec{k}\lambda}(\vec{p}, t) \langle \vec{p}' b 0 | V | \vec{p} a \vec{k}\lambda\rangle \exp[i(E_{p',p} + \hbar\omega_0 - \hbar\omega_k)t/\hbar], \end{aligned} \quad (6b)$$

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} A_{a\vec{k}'\lambda}(\vec{p}', t) = & \int d^3p A_{b0}(\vec{p}, t) \langle \vec{p}' a \vec{k}'\lambda | V | \vec{p} b 0\rangle \exp[i(E_{p',p} - \hbar\omega_0 + \hbar\omega_k)t/\hbar] \\ & + \int d^3p A_{b\vec{k}'\lambda}(\vec{p}, t) \langle \vec{p}' a \vec{k}'\lambda | H' | \vec{p} b \vec{k}'\lambda\rangle \exp[i(E_{p',p} - \hbar\omega_0)t/\hbar], \end{aligned} \quad (6c)$$

and so on. The energy difference $E_{p',p}$ is defined as

$$E_{p',p} = E_{p'} - E_p.$$

The initial condition is taken as $|\Psi 0\rangle = |\vec{p}_0 a 0\rangle$, implying that the atom is in its ground state, has center-of-mass momentum \vec{p}_0 , and no photons are present. The corresponding initial amplitude is obtained from Eq. (5) as

$$A_{\alpha\{n\}}(\vec{p}, 0) = \delta_{\alpha a} \delta_{\{n\} 0} \delta(\vec{p} - \vec{p}_0). \quad (7)$$

Hence, the hierarchy of differential equations plus initial condition form a well-posed problem.

Solutions cannot be obtained until some kind of approximation is introduced. The Weisskopf-Wigner scheme is found to be useful, since it leads to a partial truncation of the infinite system. In this scheme, the net effect of the vacuum field on

the upper state $|b\rangle$ is to approximate the term involving $A_{a\vec{k}\lambda}$ in Eq. (6b) by $-i\hbar\Gamma A_{b0}(\vec{p}', t)/2$, where Γ is the decay rate of the upper state. In this limit, Eq. (6b) is replaced by

$$\begin{aligned} \frac{\partial}{\partial t} A_{b0}(\vec{p}'; t) + \frac{1}{2}\Gamma A_{b0}(\vec{p}', t) \\ = \frac{1}{i\hbar} \int d^3p A_{a0}(\vec{p}, t) \langle \vec{p}' b 0 | H' | \vec{p} a 0\rangle \\ \times \exp[i(E_{p',p} + \hbar\omega_0)t/\hbar]. \end{aligned} \quad (6b')$$

In the next section, Eqs. (6a), (6b'), and (6c) are solved in perturbation theory for a given applied field.

III. PERTURBATION SOLUTION

We wish to solve Eqs. (6a), (6b'), and (6c) in a perturbation scheme for an applied monochromatic laser field of the form

$$\vec{E}_L = \vec{E}_0 \cos(\vec{k}_L \cdot \vec{R} - \omega_L t),$$

where \vec{R} is the quantized center-of-mass coordinate. One starts with the atom in its ground state, and the laser field gives rise to an upper-state probability amplitude $A_{b0}(\vec{p}, t)$. This process is followed by action of the vacuum field leading to the one photon state amplitude $A_{a\vec{k}\lambda}(\vec{p}, t)$. The object of the calculation is to determine the probability per unit time for the emission of a photon of type $\vec{k}\lambda$. Physically, a time-independent rate can be achieved if one considers times for which $\Gamma t \gg 1$. This limiting condition implies that the entire process of resonance fluorescence has been completed for each atom. In this time domain, a balance is achieved between the pumping due to the applied field and the fluorescence due to the vacuum field.

In the dipole approximation, the applied-field-atom interaction Hamiltonian is given by

$$H' = -\vec{\mu} \cdot \vec{E}_0 \cos(\vec{k}_L \cdot \vec{R} - \omega_L t), \quad (8)$$

where $\vec{\mu}$ is the dipole moment of the atom. The matrix elements of H' can be easily calculated as

$$\langle \vec{p}' a \{n\} | H' | \vec{p} b \{n\} \rangle = \frac{1}{2} \chi_{ab} e^{i\omega_L t} \delta(\vec{p} - \vec{p}' - \hbar \vec{k}_L), \quad (9)$$

where

$$\chi_{ab} = -\langle a | \vec{\mu} | b \rangle \cdot \vec{E}_0 = -\vec{\mu}_{ab} \cdot \vec{E}_0,$$

and antiresonant terms have been neglected (rotating-wave approximation) since the incoming radiation is assumed to be tuned near resonance.

The vacuum-field-atom interaction is, in the dipole approximation of the form

$$V = -(e/mc) \vec{P} \cdot \vec{A}(\vec{R}), \quad (10)$$

where \vec{P} is the generalized momentum,

$$\vec{A}(\vec{R}) = \sum_{\vec{k}\lambda} c \left(\frac{2\pi}{\mathcal{V} \omega_k} \right)^{1/2} (a_{\vec{k}\lambda} \hat{\epsilon}_\lambda e^{i\vec{k} \cdot \vec{R}} + a_{\vec{k}\lambda}^\dagger \hat{\epsilon}_\lambda e^{-i\vec{k} \cdot \vec{R}}).$$

$a_{\vec{k}\lambda}$ and $a_{\vec{k}\lambda}^\dagger$ are annihilation and creation operators, respectively, $\hat{\epsilon}_\lambda$ ($\lambda = 1, 2$) is the unit polarization vector of the emitted photon and \mathcal{V} is the normalization volume. The matrix element of the vacuum-field-atom interaction can be computed in a straightforward manner as

$$\langle \vec{p}' a \vec{k}\lambda | \mathcal{V} | \vec{p} b 0 \rangle = i(2\pi/\mathcal{V} \omega_k)^{1/2} \vec{\mu}_{ab} \cdot \hat{\epsilon}_\lambda \times \delta(\vec{p}' - \vec{p} - \hbar \vec{k}) \omega_0. \quad (11)$$

Using the matrix elements (9) and (11) in Eqs. (6a), (6b'), and (6c), one can perform a straightforward perturbation calculation and obtain the rate of emission of a photon of type $\vec{k}\lambda$ as⁸

$$I_{\vec{k}\lambda}(\vec{p}) = \lim_{\Gamma t \gg 1} \frac{|A_{a\vec{k}\lambda}(\vec{p}, t)|^2}{t} = \frac{|\chi_{ab}|^2 |\vec{\mu}_{ab} \cdot \hat{\epsilon}_\lambda|^2 (2\pi)^2 \omega_0^2}{4\hbar^4 \mathcal{V} \omega_k} \frac{\delta(\vec{p}_0 + \hbar \vec{k}_L - \vec{p} - \hbar \vec{k}) \delta[(E_{p_0, p_0} - \hbar \omega_L + \hbar \omega_k)/\hbar]}{(\frac{1}{2}\Gamma)^2 + (E_{\vec{p}_0 + \hbar \vec{k}_L, p_0} - \hbar \omega_L + \hbar \omega_0)^2/\hbar^2}. \quad (12)$$

It is worthwhile to note that the two δ functions imply conservation of linear momentum and energy for the entire scattering process. This result agrees with Heitler's argument⁶ that absorption and subsequent emission of photons cannot be treated as two separate processes.

The spectral distribution of emitted photons can be determined by integrating Eq. (12) over all final momenta of the atoms, i.e.,

$$I_{\vec{k}\lambda}^* = \int d^3 p I_{\vec{k}\lambda}(\vec{p}) = \frac{|\chi_{ab}|^2 |\vec{\mu}_{ab} \cdot \hat{\epsilon}_\lambda|^2 (2\pi)^2 \omega_0^2}{4\hbar^4 \mathcal{V} \omega_k} \frac{\delta[(E_{p_0 + \hbar \vec{k}_L, p_0} - \hbar \omega_L + \hbar \omega_k)/\hbar]}{(\frac{1}{2}\Gamma)^2 + (E_{\vec{p}_0 + \hbar \vec{k}_L, p_0} - \Delta)^2/\hbar^2}, \quad (13)$$

where the detuning

$$\Delta = \hbar \omega_L - \hbar \omega_0.$$

The argument of the δ function is a quadratic equation in ω_k . By using the identity

$$\delta((\omega - \omega_1)(\omega - \omega_2)) = \frac{\delta(\omega - \omega_1) + \delta(\omega - \omega_2)}{|\omega_1 - \omega_2|},$$

and keeping only the positive frequency part, Eq. (13) becomes

$$I_{\vec{k}\lambda} = \frac{|\chi_{ab}|^2 |\vec{\mu}_{ab} \cdot \hat{\epsilon}_\lambda|^2 (2\pi)^2 \omega_0^2}{4\hbar^4 \mathcal{V} \omega_k} \mathcal{L}(\Delta, |\vec{p}_0 + \hbar \vec{k}_L|) N(p_0, \hbar \vec{k}_L, \beta_1, \beta_2, \phi_1, \phi_2) \delta(\omega_k - \bar{\omega}), \quad (14)$$

where

$$\mathcal{L}(\Delta, |\vec{p}_0 + \hbar\vec{k}_L|) = \left[\left(\frac{\Gamma}{2} \right)^2 + \frac{(E_{\vec{p}_0 + \hbar\vec{k}_L} - \Delta)^2}{\hbar^2} \right]^{-1},$$

$$N(p_0, \hbar k_L, \beta_1, \beta_2, \phi_1, \phi_2) = \left[\left(1 - \frac{p_0 [\cos\beta_1 \cos\beta_2 + \sin\beta_1 \sin\beta_2 \cos(\phi_1 - \phi_2) + \hbar k_L \cos\beta_2]}{Mc} \right)^2 \right. \\ \left. - \frac{2}{Mc^2} \left(\frac{\hbar^2 k_L^2}{2M} + \frac{p_0 \hbar k_L \cos\beta_1}{M} - \hbar\omega_L \right) \right]^{-1/2},$$

$$\bar{\omega} = -\frac{Mc^2}{\hbar} + \frac{Mc^2}{\hbar} N^{-1}(p_0, \hbar k_L, \beta_1, \beta_2, \phi_1, \phi_2) + \frac{c}{\hbar} (p_0 [\cos\beta_1 \cos\beta_2 + \sin\beta_1 \sin\beta_2 \cos(\phi_1 - \phi_2)] + \hbar k_L \cos\beta_2),$$

where β_1 is the angle between \vec{p}_0 and $\hbar\vec{k}_L$, β_2 is the angle between $\hbar\vec{k}$ and $\hbar\vec{k}_L$, and ϕ_1 and ϕ_2 are the azimuthal angles as shown in Fig. 2. Equation (15) can, in the optical range of frequencies, be approximated as

$$\bar{\omega} = \omega_L \left(1 - \frac{\hbar k_L}{Mc} (1 - \cos\beta_2) \right. \\ \left. + \frac{p_0}{Mc} [-\cos\beta_1 + \cos\beta_1 \cos\beta_2 \right. \\ \left. + \sin\beta_1 \sin\beta_2 \cos(\phi_1 - \phi_2)] \right). \quad (16)$$

Equation (14) for the spectral profile is the basic result of this work. The fluorescence at any scattering angle is monochromatic, mirroring the spectrum of the excitation. Within the restriction placed by conservation of energy and linear momentum in Eq. (14), it follows that all scattering directions are permissible. Moreover, the probability for emission [as given by Eq. (14)] does not depend on the angle of emission to order v/c . The requirement of the conservation laws are such that photons scattered in various directions will have

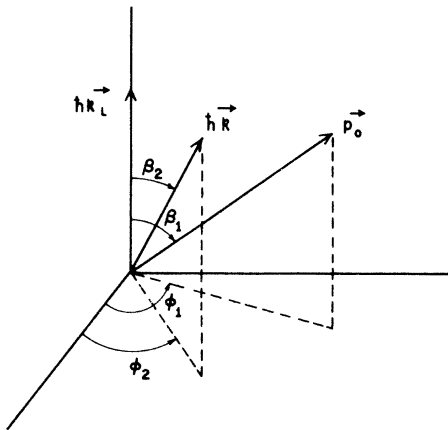


FIG. 2. A view of the scattering process showing the relative orientation of the excitation field $\hbar\vec{k}_L$, the fluorescence $\hbar\vec{k}$, and the initial atomic momentum \vec{p}_0 .

their frequencies shifted and there will be a correlated change in the momentum and energy of the scatterer. The physics becomes more transparent if some examples are considered.

Forward scattering ($\beta_2 = 0^\circ$). For any β_1 the frequency of emitted photon, given by Eq. (15), is the same as the frequency of incoming radiation.

Backward scattering ($\beta_2 = 180^\circ$). The frequency of the emitted photon is shifted by an amount [see Eq. (16)]

$$\Delta_S \approx -2\omega_L (\hbar k_L + p_0 \cos\beta_1) / Mc. \quad (17)$$

The second term in Eq. (17) simply represents the Doppler shift accrued in the backscattering of light from a moving "reflector," while the first term represents the photon recoil effect, i.e., an additional shift due to the change of atomic velocity resulting from the scattering process. If $\cos\beta_1 < 0$ and $p_0 \gg \hbar k_L$, the shift is positive and, correspondingly, the atom will lose energy to the field. This type of effect, using a laser tuned below an atomic resonance so that only those atoms moving towards the laser ($\cos\beta_1 < 0$) will be in resonance with the field, has been proposed as a method of laser cooling of gases.⁹

90° scattering ($\beta_2 = 90^\circ$). From Eq. (16), one finds the frequency shift of the emitted photon is

$$\Delta_S \approx \frac{-\omega_L \{ \hbar k_L + p_0 [\cos\beta_1 - \sin\beta_1 \cos(\phi_1 - \phi_2)] \}}{Mc}. \quad (18)$$

If one chooses a totally orthogonal geometry ($\beta_1 = 90^\circ$, $\phi_1 - \phi_2 = 90^\circ$), the shift arises solely from the photon-recoil effect and is given by

$$\Delta_S \approx -(\hbar\omega_L / Mc^2)\omega_L. \quad (19)$$

IV. DISCUSSION

A perturbation calculation of the resonance fluorescence from atoms moving initially with momentum \vec{p}_0 and excited by a monochromatic field has been presented. In the weak-field limit considered in this paper, the fluorescence is

monochromatic and is shifted from the incident frequency due to atomic recoil in the scattering process. The monochromatic nature of the fluorescence is a consequence of the fact that the initial and final states of the atom are both the ground state, with no associated natural width. The frequency shift of the fluorescence and the corresponding change in atomic velocity can be calculated from conservation of momentum and energy. As stressed by Heitler, the scattering must be viewed as a single process.

The calculation can be easily extended to include incident radiation of a finite bandwidth or a distribution of atomic velocities. There has been no attempt to include multiple scattering or collective effects.

Given a lifetime τ much less than the interaction time t_I , the restriction expressed by Eq. (1) sets an upper bound for the field strength, i.e., $|\vec{\mu} \cdot \vec{E}/\hbar| \ll 1/\tau$. Some typical values of the physical parameters in question are $|\vec{\mu}| \approx 10^{-30}$ C m, $\tau \approx 10^{-9}$ sec, giving $|\vec{E}| \ll 10^5$ V/m. A violation of such a criterion will certainly introduce additional effects such as the dynamic Stark shift. This generalization will further complicate the simple picture of recoil in resonance fluorescence.

In closing, we would like to propose an experiment that might be used to measure the photon recoil shift. The experiment is shown schematically in Fig. 3. A laser strikes an atomic beam at right angles and the fluorescence is detected perpendicular to both the laser and the atomic beam. A part of the laser is sent to the detector to permit a heterodyne determination of the beat frequency between the laser and the fluorescence. A suitable detection scheme requires that the laser beam be locked to the atomic beam and that the laser and the fluorescence be collinear inside the detector. Such a technique has been discussed in detail by Forrester.¹⁰ Since the geometry is mutually orthogonal, the frequency shift as given by Eq. (19) is a direct measure of the photon recoil.

If the system is not perfectly aligned, there will be a correction term. In Fig. 2, if $\beta_1 = 90^\circ + \epsilon_1$, $\beta_2 = 90^\circ + \epsilon_2$, $\phi_1 - \phi_2 = 90^\circ + \epsilon_3$, with all ϵ 's much less than unity, then the shift calculated from Eq. (16) to first order in ϵ is

$$\Delta_s = -\frac{\hbar \omega_L^2}{Mc^2} + \frac{p_0 \omega_L}{Mc} (\epsilon_1 - \epsilon_3).$$

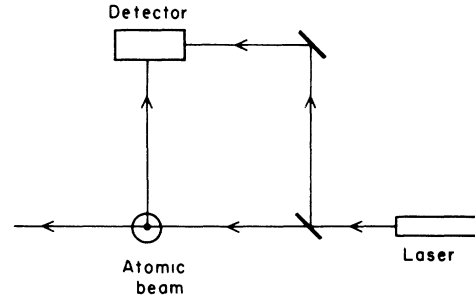


FIG. 3. A schematic setup of the proposed experiment with the atomic beam pointing out of the paper. The laser is assumed to be locked to the atomic beam. The fluorescence and the laser must be collinear inside the detector.

Since p_0/Mc is typically 10^4 times greater than $\hbar \omega_L/Mc^2$, small errors can be significant. A new method for checking for alignment errors would be to vary the laser frequency ω_L . The photon recoil term is quadratic, but the error term is linear in ω_L , allowing for a separation of the two contributions.

An error ϵ_1 is intrinsically present in any experiment because of the limit of beam collimation, and ϵ_3 is intrinsically present because of the finite solid angle of detection. If the atomic beam is not initially collimated, we might expect a certain amount of Doppler broadening in the line shape. A rough estimate can be given for the upper limit of the beam divergence such that it does not obscure the recoil shift. If the photon recoil is of the order of 10^5 – 10^6 Hz, the thermal velocity is $\sim 10^3$ m/sec, and a wave number of the order of 10^7 m⁻¹; then the maximum degree of divergence must be of the order of 10^{-3} – 10^{-4} rad. With a similar detection angle one might expect a counting rate of fluorescence quanta of the order of 1 photon/sec.

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