PHYSICAL REVIEW A VOLUME 5, NUMBER 5 MAY 1972

Collisional Transfer of Coherence: Polarization of Sensitized Fluorescence

Lue-Yung Chow Chiu*

Department of Chemistry, Howard University, Washington, D. C. 20001

(Received 20 September 1971)

Energy transfer and coherence transfer between two atoms of different species are studied theoretically. The electric dipole-dipole interaction is the mechanism considered for the excitation transfer. A generalized perturbation method for a damping system is used. A general formula is derived for the polarization of the sensitized fluorescence resulting from the excitation transfer. This polarization is shown to be dependent on the angular momenta of the two atoms, and to be independent of their relative velocity as well as of their difference in the energy of excitation. The polarizations are found to be nonzero in many cases which demonstrate the transfer of coherence through collision processes. A general and compact formula is also derived for the polarization of pure fluorescence where excitation exchange has not taken place. The polarization of the sensitized fluorescence is in general smaller than that of the pure fluorescence. This means that the coherence is transferred only partially. Finally, the magnetic field effect has been included. This field depolarization depends on the g_j values and the lifetimes of the excited states of both atoms. The general feature of the field dependence also differs considerably from that of the Hanle effect for resonance fluorescence.

INTRODUCTION

The polarization of resonance fluorescence has long been a known phenomenon.[1] For example, when ${}^1S_0 \rightarrow {}^1P_1$ excitation is induced by a linearly polarized light with y polarization, the resulting resonance fluorescence observed in the z direction is expected to be 100% polarized in the y direction. If a slowly increasing magnetic field is applied in the z direction, the coherence among the magnetic sublevels of the excited state will be slowly destroyed because of the splitting of the sublevels, and the polarization of the scattered light decreases accordingly. The magnetic field depolarization also depends on the g_j value and the lifetime of the excited state. This is the well-known Hanle[2] effect for measuring the lifetime of the excited state. Early theoretical works on damping and polarization of radiation was done by Weisskopf[3] and by Breit.[4] It has since been applied to the level-crossing[5] and optical-pumping[6] problems and has also been applied to the polarization of the electron-atom collision light.[7]

The polarization of the sensitized fluorescence was observed recently by Gough.[8] In his experiment a mixture of mercury and cadmium vapour was irradiated by a linearly polarized resonant light of mercury, and the fluorescence of cadmium was found to be 2–5% polarized. This implies[9] that part of the coherence among the sublevels of the excited mercury was transferred to the excited cadmium through a collision of the second kind.

An optically excited atom A may transfer its excitation resonantly to atom B of the same element through electrostatic dipole-dipole interaction and radiative electromagnetic interaction. These interactions were considered by Hutchinson and Hameka,[10] Fontana and Hearn,[11] and Stephen[12] in their theoretical studies of the lifetime and the linewidth of the emitted fluorescence due to interacting atoms. Happer and Soloman[13] studied these effects in connection with signal shapes in optical double resonance. Chiu[14] has studied the collisional depolarization of the resonance fluorescence by considering these interactions. In all these studies, there is no net energy transfer between the excitation energy and the kinetic energy, and the quasistatic approximation was used.

In the present work, we study theoretically the polarization of sensitized fluorescence. Here atoms A and B are assumed to be of different species and, therefore, possess different energies of excitation. Atom A can be excited by the incident light, which is linearly polarized. The transfer of excitation from A to B is accompanied by converting the excess energy of excitation $\Delta\epsilon$ into the kinetic energy of the relative motion. The probability (or cross section) for atom B to emit light of polarization parallel or perpendicular to that of incident light is calculated. The polarization of sensitized fluorescence is then obtained. Due to a proper rotational transformation, one is able to integrate the relative motion of the final states and to yield a compact expression which was not previously obtained for the probability of sensitized fluorescence. A general formula (in terms of Clebsch-Gordan coefficients) for the polarization is derived in Sec. I. The examples in Sec. III indicate that the emitted sensitized fluorescence is polarized in many cases. This is the theoretical verification that coherence can be transferred through collision of the second kind. Although the cross section for energy transfer depends on $\Delta\epsilon$ and the relative velocity, the polarization is found to depend only on the quantum numbers of the excited and the ground states of atoms A and B.

With a magnetic field present in the quantization direction, the polarization decreases. The effect of magnetic field on polarization can be expressed as a multiplicative factor which (as shown in Sec. III) will depend on the natural lifetimes and g_j values of the excited states of both atoms. The experiment on the magnetic-field-dependent line shape, which is similar to the Hanle effect of resonance fluorescence, will be able to determine the lifetimes of both atoms. This may be a useful method to measure the lifetimes of certain atomic states which are not measurable otherwise. Finally, the results of sensitized fluorescence and that of pure fluorescence (where excitation-exchange has not taken place) are compared in Sec. III. Not only that the field-free polarization of sensitized fluorescence is always smaller than the case of pure fluorescence, the magnetic field depolarizations in the two cases are also distinctively different. The results of present work (as will be discussed at the end of Sec. III) can be applied to atoms with nuclear spin when one can isolate and follow the optical transitions between a pair of hyperfine levels, one belonging to the upper and the other to the lower state. The general treatment on the nuclear-spin effect will be presented in a later work.

The method of present work can be extended to calculate the cross section of energy transfer[15] where different fine-structure components are involved. Zare and co-workers[16] have recently measured the cross-section of excitation transfer between various fine-structure components of alkali-atom–alkali-atom collision. Melton and Klemperer[17] have measured the same between different vibrational levels of the NO molecule. In all these works the results are obtained by measuring the relative intensities of sensitized fluorescence. Following the present method, we can calculate[18] directly the intensities of sensitized fluorescence, hence making possible direct comparison with experimental results.

I. GENERAL FORMALISM

Atoms A and B of different elements are initially in their ground states $(J_a M_a)$ and $(J_b M_b)$, respectively. The incident light (see Fig. 1) of energy $\hbar c\kappa_\nu$ and of polarization $\vec{\sigma}_\nu$ can excite atom A into one of its excited states $(j_a m_a)$. The excitation of A is then transferred to B such that B is excited to state $(j_b m_b)$ and A goes to the lower state $(J'_a M'_a)$. Finally atom B goes to its final state $(J'_b M'_b)$ by emitting sensitized fluorescence radiation of energy $\hbar c\kappa_\mu$ and polarization $\vec{\sigma}_\mu$. Throughout this work, the angular momentum quantum numbers J and M in capital letter are used for the lower-energy states (i.e., initial and final states) and j and m in small letter for the optically excited states. Our purpose is to calculate the polarization $\vec{\sigma}_\mu$ of the sensitized fluorescence knowing that the polarization of the incident radiation is $\vec{\sigma}_\nu$. Such information will indicate whether the coherence among the excited states of atom A is transferred to atom B. The total angular momenta $\vec{J}\hbar$ (of the ground state) and $\vec{j}\hbar$ (of the excited state) are quite arbitrary except that they can be connected through electric dipole transition. In other words $\vec{J}\hbar$ (or $\vec{j}\hbar$) will be the electronic orbital angular momentum if the atom possesses neither electronic nor nuclear spin. J (or j) will be the fine-structure or hyperfine-structure quantum number depending on whether the atom possesses electronic spin or both electronic and nuclear spin. The states of different J (or j) are assumed to be well resolved in comparison with the linewidth of the incident and the emitted radiation. Neglecting the electron-exchange effect, the initial $(\Psi_{i\nu})$, intermediate (Ψ_A and Ψ_B), and final $(\Psi_{f\mu})$ states are simple products of atomic wave functions (ϕ), wave function due to internuclear motion (χ), and wave function of the photon field (ψ). The wave functions and their corresponding energies are as follows:

$$\Psi_{i\nu}=\phi_{J_aM_a}(\vec{r}_{Ai})\,\phi_{J_bM_b}(\vec{r}_{Bj})\,\chi_i(\vec{R})\,\psi(n_\nu)\ , \tag{1.1a}$$

$$E_{i\nu}=\epsilon_{J_aM_a}+\epsilon_{J_bM_b}+T_i+n_\nu\hbar c\kappa_\nu\ , \tag{1.1b}$$

$$\Psi_A=\phi_{j_am_a}(\vec{r}_{Ai})\,\phi_{J_bM_b}(\vec{r}_{Bj})\,\chi_a(\vec{R})\,\psi(n_\nu-1)\ , \tag{1.2a}$$

$$E_A=\epsilon_{j_am_a}+\epsilon_{J_bM_b}+T_A+(n_\nu-1)\hbar c\kappa_\nu\ , \tag{1.2b}$$

$$\Psi_B=\phi_{J'_aM'_a}(\vec{r}_{Ai})\,\phi_{j_bm_b}(\vec{r}_{Bj})\,\chi_b(\vec{R})\,\psi(n_\nu-1)\ , \tag{1.3a}$$

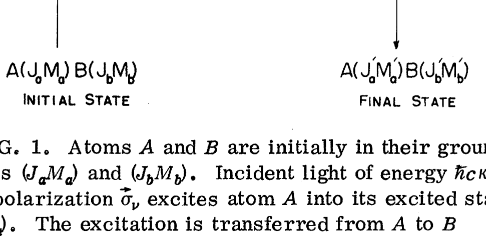

FIG. 1. Atoms A and B are initially in their ground states $(J_a M_a)$ and $(J_b M_b)$. Incident light of energy $\hbar c\kappa_\nu$ and polarization $\vec{\sigma}_\nu$ excites atom A into its excited state $(j_a m_a)$. The excitation is transferred from A to B accompanied by changes in kinetic energy of the relative motion. Sensitized fluorescence of $\hbar c\kappa_\mu$ and $\vec{\sigma}_\mu$ is emitted by atom B resulting in the lower-energy final state $(J'_b M'_b)$ for B.

$$E_B = \epsilon_{J'_a M'_a} + \epsilon_{j_b m_b} + T_B + (n_\nu - 1)\hbar c \kappa_\nu , \qquad (1.3b)$$

$$\Psi_{f\mu} = \phi_{J'_a M'_a}(\vec{\mathrm{r}}_{Ai})\, \phi_{J'_b M'_b}(\vec{\mathrm{r}}_{Bj})\, \chi_f(\vec{\mathrm{R}})\psi(n_\nu - 1, \mu) , \qquad (1.4a)$$

$$E_{f\mu} = \epsilon_{J'_a M'_a} + \epsilon_{J'_b M'_b} + T_f + (n_\nu - 1)\hbar c\kappa_\nu + \hbar c \kappa_\mu . \qquad (1.4b)$$

The intermediate state Ψ_A (or Ψ_B) refers to atom A (or B) being excited. $\psi(n_\nu)$ is the phonon state where n photons of energy $\hbar c\kappa_\nu$ and polarization $\vec{\sigma}_\nu$ are present; $\psi(n_\nu - 1)$ is the state when a photon ν is absorbed; and $\psi(n_\nu - 1, \mu)$ is the state where a photon ν is absorbed and a photon μ is emitted. $\chi(\vec{\mathrm{R}})$, the wave function of the relative internuclear motion, is approximated by a plane wave as follows:

$$\chi(\vec{\mathrm{R}}) = (1/L)^{3/2}\, e^{i\vec{\mathrm{k}}\cdot\vec{\mathrm{R}}} . \qquad (1.5)$$

The above $\chi(\vec{\mathrm{R}})$ is normalized in a large box of dimension L. ϵ_{jm} and ϵ_{JM} (or $\epsilon_{J'M'}$) are the energies of the atomic excited and ground states, respectively. T's are the kinetic energies of the internuclear motion. Since the optical excitation and deexcitation are much faster than the nuclear motion, we have

$$T_A = T_i = \hbar^2 k_i^2 / 2M \qquad (1.6)$$

and

$$T_B = T_f = \hbar^2 k_f^2 / 2M , \qquad (1.7)$$

where $M = m_A m_B/(m_A + m_B)$ is the reduced mass of A and B.

At a given time t, the state wave function $u(t)$ is a linear combination of the above Ψ's with time-dependent coefficient $b(t)$,

$$U(t) = b_{i\nu}(t)\,\Psi_{i\nu}\, e^{-iE_{i\nu} t/\hbar} + b_A(t)\,\Psi_A e^{-iE_A t/\hbar} + b_B(t)\,\Psi_B e^{-iE_B t/\hbar} + b_{f\mu}(t)\,\Psi_{f\mu}\, e^{-iE_{f\mu} t/\hbar} . \qquad (1.8)$$

The perturbations H' considered here are the electrostatic interaction V between atoms A and B, and the radiative electromagnetic interaction H between the atomic system and the radiation field. By multipole expansion of the electrostatic interaction between two neutral atoms, the following dipole-dipole interaction,[14,19] which is the lowest-order term to connect two differently excited atoms, will be considered here:

$$V = -(\sqrt{6}\, e^2/R^3) \sum_{ij} \sum_{uv} (-)^v C(112; u, v-u, v) \times r_u(Ai)\, r_{v-u}(Bj)\, C_{2,-v}(\hat{R}) , \qquad (1.9)$$

where

$$C_{l,m}(\hat{R}) = [4\pi/(2l+1)]^{1/2}\, Y_{l,m}(\hat{R}) , \quad C(112; u, v-u, v)$$

is the Clebsch-Gordan coefficient, e is the electronic charge, $r_u(Ai)$ is the uth component of the spherical vector $\vec{\mathrm{r}}(Ai)$ of electron i centered on nucleus A, and $r_{v-u}(Bj)$ is the corresponding one centered on B. Neglecting the second-order terms, i.e., A_i^2 terms, the radiative interaction[18] becomes

$$H = -(e/mc) \textstyle\sum_i \vec{\mathrm{A}}_i \cdot \vec{\mathrm{P}}_i , \qquad (1.10)$$

and

$$\vec{\mathrm{A}}_i = (4\pi)^{1/2} c \textstyle\sum_\lambda \vec{\sigma}_\lambda (q_\lambda e^{i\vec{\kappa}_\lambda \cdot \vec{\gamma}_i} + q_\lambda^* e^{-i\vec{\kappa}_\lambda \cdot \vec{\gamma}_i}) , \qquad (1.11)$$

where $\vec{\mathrm{P}}_i$ is the linear momentum of electron i, $\vec{\mathrm{r}}_i$ is the electronic radius vector refers to the space-fixed center, m is the electronic mass, and c is the velocity of light. The operator q_λ annihilates a photon λ and q_λ^* creates a photon λ. The matrix elements of these operators over the photon states are as follows:

$$(n_\lambda - 1 | q_\lambda | n_\lambda) = (n_\lambda | q_\lambda^* | n_\lambda - 1) = (\hbar n_\lambda / 2c\kappa_\nu L^3)^{1/2} . \qquad (1.12)$$

Substituting (1.8) into the time-dependent Schrödinger equation, we obtain equations for the time-dependent coefficient $b_n(t)$'s:

$$i\hbar \dot{b}_n(t) = \textstyle\sum_{n'} H'_{nn'}\, b_{n'}(t)\, e^{i(E_n - E_{n'})t/\hbar} . \qquad (1.13)$$

Following the time-dependent method for damping systems,[14,20,21] $b_n(t)$ is expressed in terms of a Fourier energy transform,

$$b_n(t) = -\frac{1}{2\pi i} \int_{-\infty}^{\infty} dE\, U_n(E) \frac{e^{i(E_n - E)t/\hbar} - 1}{[E - E_i + \frac{1}{2} i\hbar\Gamma(E)](E - E_n)} \qquad (n \neq i) \qquad (1.14)$$

where

$$U_n(E) = H'_{n,i} + \sum_{n' \neq i} H'_{nn'}\, \zeta(E - E_{n'})\, U_{n'}(E) , \qquad (1.15)$$

$$\tfrac{1}{2}\hbar\Gamma(E) = i \sum_{n' \neq i} H'_{in'}\, \zeta(E - E_{n'}) U_{n'}(E) , \qquad (1.16)$$

and

$$\zeta(E - E_{n'}) = \lim_{\epsilon \to 0} (E - E_{n'} + i\epsilon)^{-1} = P(E - E_{n'})^{-1} - i\pi\delta(E - E_{n'}) . \qquad (1.17)$$

The index i refers to the initial state which is $\Psi_{i\nu}$ in the present work. Since we are interested in the intensity of the sensitized fluorescence, we only need to compute $|b_{f\mu}(t \to \infty)|^2$, which is the probability amplitude of reaching the final state by emitting a photon of energy $\hbar c\kappa_\mu$ and polarization $\vec{\sigma}_\mu$ after the system has been excited by a photon characterized by $\hbar c\kappa_\mu$ and $\vec{\sigma}_\nu$. For $t \to \infty$, the time dependent factor in (1.14) becomes a ζ function and we have[20]

$$b_{f\mu}(\infty) = U_{f\mu}(E_{f\mu})[E_{f\mu} - E_{i\nu} + \tfrac{1}{2} i\hbar\Gamma(E_{f\mu})]^{-1} , \qquad (1.18)$$

where

$$\Gamma(E) = (2i/\hbar) \textstyle\sum_{\mathrm{A}} H_{i\nu,\mathrm{A}}\, \zeta(E - E_{\mathrm{A}}) U_{\mathrm{A}}(E) , \qquad (1.19)$$

$$U_{f\mu}(E) = \textstyle\sum_{B'} H_{f\mu,B'}\, U_{B'}(E)\, \zeta(E - E_{B'}) , \qquad (1.20)$$

$$U_{\mathrm{A}}(E) = H_{\mathrm{A},i\nu} + \textstyle\sum_{f'\lambda} H_{\mathrm{A},f'\lambda}\, U_{f'\lambda}(E)\, \zeta(E - E_{f'\lambda})$$

$$+\sum_{B'} V_{A,B'}U_{B'}(E)\zeta(E-E_{B'}) \,, \tag{1.21a}$$

$$U_B(E)=\sum_{f'\lambda} {}_\lambda H_{B,f'\lambda}\,U_{f'\lambda}(E)\zeta(E-E_{f'\lambda})$$

$$+\sum_{A'} V_{B,A'}\,U_{A'}(E)\zeta(E-E_{A'}) \,, \tag{1.21b}$$

and

$$U_{f'\lambda}(E)=\textstyle\sum_{A'} H_{f'\lambda,A'}\,U_{A'}(E)\zeta(E-E_{A'})$$

$$+\textstyle\sum_{B'} H_{f'\lambda,B'}\,U_{B'}(E)\zeta(E-E_{B'}) \,. \tag{1.22}$$

As defined in (1.19), Γ is the total probability of absorption from the initial state, and it is proportional to the intensity of the incident light. For incident light that is not too strong, Γ is a very small quantity which after averaging over the initial states of atom A becomes (derivation will be shown later in Appendix A)

$$\Gamma=(2\pi\rho_\nu/\hbar^2c)\,d\Omega_\nu|\bar{H}(\nu)|^2 \,, \tag{1.23}$$

where ρ_ν is the density function of the incident light and is assumed to be constant over the absorption width. $d\Omega_\nu$ is the solid angle of the incident beam on the system and

$$|\bar{H}(\nu)|^2 \equiv (2J_a+1)^{-1}\sum_{M_a m_a}(j_a m_a|H|J_a M_a)(J_a M_a|H|j_a m_a) \,. \tag{1.24a}$$

The right-hand side of Eq. (1.24a) (can be shown later) reduces to

$$(\tfrac{2}{3}\pi)e^2\hbar c n_\nu \kappa_\nu L^{-3}|(J_a||\textstyle\sum_i \vec{\mathrm{r}}(Ai)||j_a)|^2 \,. \tag{1.24b}$$

Substituting (1.22) into (1.21) and using the relations (which will be proved in Appendix B)

$$\sum_{f'\lambda} H_{A,f'\lambda}H_{f'\lambda,B'}\zeta(E_{f\mu}-E_{f'\lambda})=0 \tag{1.25}$$

and

$$\sum_{f'\lambda} H_{A,f'\lambda}H_{f'\lambda,A'}\zeta(E_{f\mu}-E_{f'\lambda})=(\hbar/2i)\gamma_A\delta_{A,A'} \,, \tag{1.26a}$$

where

$$\gamma_A \equiv (4e^2/3\hbar)(\hbar c)^{-3}$$

$$\times\textstyle\sum_{J'_a}|(j_a||\sum\vec{\mathrm{r}}(Ai)||J'_a)|^2(\epsilon_{j_a}-\epsilon_{J'_a})^3 \,, \tag{1.26b}$$

and a similar relation for γ_B, Eqs. (1.21a) and (1.21b) now read

$$(E_{f\mu}-E_A+\tfrac{1}{2}i\hbar\gamma_A)U_A(E_{f\mu})\zeta(E_{f\mu}-E_A)$$

$$=H_{A,i\nu}+\textstyle\sum_{B'}V_{A,B'}U_{B'}(E_{f\mu})\zeta(E_{f\mu}-E_{B'}) \tag{1.27a}$$

and

$$(E_{f\mu}-E_B+\tfrac{1}{2}i\hbar\gamma_B)U_B(E_{f\mu})\zeta(E_{f\mu}-E_B)$$

$$=\textstyle\sum_{A'}V_{B,A'}\,U_{A'}(E_{f\mu})\zeta(E_{f\mu}-E_{A'}) \,, \tag{1.27b}$$

respectively. The identity

$$(E-E_n)\zeta(E-E_n)=1 \tag{1.28}$$

was used in deriving the above equations. The quantity γ_A (similarly for γ_B) as defined in (1.26) is the total transition probability from state A to all the final states via radiative interaction, and the reciprocal of γ_A is the lifetime of the excited state of atom A. Neglecting the second term on the right-hand side of (1.27a), we obtain a first-order expression for U_A which is then substituted into (1.27b) to give a second-order expression for U_B. This second-order expression of U_B is then substituted into (1.20) to give a third-order expression for $U_{f\mu}(E)$,

$$U_{f\mu}(E)=\sum_{AB}\frac{H_{f\mu B}V_{B,A}H_{A,i\nu}}{(E-E_A+\frac{1}{2}i\hbar\gamma_A)(E-E_B+\frac{1}{2}i\hbar\gamma_B)} \,. \tag{1.29}$$

The probability $W(\nu\rightarrow\mu)$ of absorbing a photon of energy $\hbar c\kappa_\nu$ and polarization $\vec{\sigma}_\nu$ (at solid angle $d\Omega_\nu$) and then emitting a photon of energy $\hbar c\kappa_\mu$ and polarization $\vec{\sigma}_\mu$ (at solid angle $d\Omega_\mu$) is obtained by summing $|b_{f\mu}(\infty)|^2$ over the final states and averaging over the initial states as follows:

$$W(\nu\rightarrow\mu)=\bar{\sum_i}\sum_f\sum_{\nu\mu}|b_{f\mu}(\infty)|^2 \,, \tag{1.30}$$

where the summation over ν and μ means the summation over the incident and the emitted photon energy, and the $\bar{\sum}_i$ sign means averaging over the initial states. From (1.18) we have

$$\textstyle\sum_\nu|b_{f\mu}(\infty)|^2=\sum_\nu|U_{f\mu}(E_{f\mu})|^2[(E_{f\mu}-E_{i\nu})^2+\frac{1}{4}\hbar^2\Gamma^2]^{-1} \,. \tag{1.31}$$

Since Γ is very small, the denominator has practically the property of a $\delta(E_{f\mu}-E_{i\nu})$ function. Replacing $\sum_\nu$ by $d\Omega_\nu\int d\kappa_\nu\rho_\nu$ and using (1.23) for Γ, Eq. (1.31) becomes

$$\textstyle\sum_\nu|b_{f\mu}(\infty)|^2=|U_{f\mu}(\infty)|^2/|\bar{H}(\nu)|^2 \,. \tag{1.32}$$

The integration of the δ function implies the energy conservation $E_{f\mu}=E_{i\nu}$. Next, we replace $\sum_\mu$ by $(L/2\pi)^3 d\Omega_\mu\int d\kappa_\mu\kappa_\mu^2$ and keep all factors which vary slowly with κ_μ outside the integral. The following is obtained:

$$\sum_\mu\sum_\nu|b_{f\mu}(\infty)|^2=\sum_{ABA'B'}\frac{d\Omega_\mu\kappa_\mu^2L^3GH_{f\mu,B}V_{B,A}H_{A,i\nu}H^*_{f\mu,B'}V^*_{B',A'}H^*_{A',i\nu}}{(2\pi)^3|\bar{H}(\nu)|^2\hbar c} \,, \tag{1.33}$$

where

$$G \equiv \int d(\hbar c\kappa_\mu)[(E_{f\mu} - E_A + \tfrac{1}{2} i\hbar\gamma_A)(E_{f\mu} - E_B + \tfrac{1}{2} i\hbar\gamma_B) \times (E_{f\mu} - E_{A'} - \tfrac{1}{2} i\hbar\gamma_A)(E_{f\mu} - E_{B'} - \tfrac{1}{2} i\hbar\gamma_B)]^{-1} . \tag{1.34}$$

The denominator on the right-hand side of the above equation can be fractionated into a linear combination of four terms. Due to the smallness of γ_A and γ_B, each term is a representation of a ξ function, e.g.,

$$(E_{f\mu} - E_B + \tfrac{1}{2} i\hbar\gamma_B)^{-1} = P(E_{f\mu} - E_B)^{-1} - i\pi\delta(E_{f\mu} - E_B) , \tag{1.35}$$

where

$$E_{f\mu} - E_B = \hbar c\kappa_\nu + \epsilon_{J_b' M_b'} - \epsilon_{j_b m_b} . \tag{1.36}$$

The integration over the principal-value parts of the four terms cancels each term whereas the integration over the δ-function parts gives

$$G = -\frac{2\pi i}{[\Delta_{AB'} + \hbar^2(k_i^2 - k_f^2)(2M)^{-1} - i\hbar\gamma_{AB}][\Delta_{A'B} + \hbar^2(k_i^2 - k_f^2)(2M)^{-1} + i\hbar\gamma_{AB}]} \left(\frac{1}{\Delta_{AA'} - i\hbar\gamma_A} + \frac{1}{\Delta_{BB'} - i\hbar\gamma_B}\right) , \tag{1.37}$$

where

$$\gamma_{AB} = \tfrac{1}{2}(\gamma_A + \gamma_B) , \tag{1.38}$$

$$\Delta_{AA'} = \epsilon_{j_a m_a} - \epsilon_{j_a m_a'} , \tag{1.39}$$

$$\Delta_{BB'} = \epsilon_{j_b m_b} - \epsilon_{j_b m_b'} , \tag{1.40}$$

$$\Delta_{AB'} = (\epsilon_{j_a m_a} - \epsilon_{j_b m_b'}) - (\epsilon_{J_a' M_a'} - \epsilon_{J_b M_b}) , \tag{1.41}$$

and

$$\Delta_{A'B} = (\epsilon_{j_a m_a'} - \epsilon_{j_b m_b}) - (\epsilon_{J_a' M_a'} - \epsilon_{J_b M_b}) . \tag{1.42}$$

Fractionating the denominator, Eq. (1.37) becomes

$$G = \left(\frac{1}{\Delta_{AB'} + \hbar^2(k_i^2 - k_f^2)(2M)^{-1} - i\hbar\gamma_{AB}} - \frac{1}{\Delta_{A'B} + \hbar^2(k_i^2 - k_f^2)(2M)^{-1} + i\hbar\gamma_{AB}}\right) \frac{2\pi i}{(\Delta_{AA'} - i\hbar\gamma_A)(\Delta_{BB'} - i\hbar\gamma_B)} . \tag{1.43}$$

Now we consider the special case where the magnetic field along the quantization axis (z axis) is zero (the case of nonzero magnetic field will be considered in Sec. II). Magnetic sublevels are degenerate and we have

$$\Delta_{AA'} = \Delta_{BB'} = 0 \quad \text{and} \quad \Delta_{AB'} = \Delta_{A'B} = \Delta . \tag{1.44}$$

Since γ_{AB} is also a small value, each term inside the curly bracket of (1.43) is a ξ function. By Eq. (1.44), Eq. (1.43) becomes

$$G = \frac{4\pi^2}{\hbar^2 \gamma_A \gamma_B} \delta[\hbar^2(k_f^2 - k_i^2)/(2M) - \Delta] . \tag{1.45}$$

We substitute the above expression for G into (1.33), then sum (1.33) over the final states and average it over the initial states. Since

$$\overline{\sum_i} \sum_f = (2J_a + 1)^{-1} (2J_b + 1)^{-1} (L/2\pi)^3 \times \sum_{M_a M_b} \sum_{M_a' M_b'} \int dk_i \, d\Omega_i \, \rho(k_i) \int dk_f \, d\Omega_f \, k_f^2 \tag{1.46}$$

we integrate the G of Eq. (1.45) over dk_f to give

$$\int dk_f \, k_f^2 G = (4\pi^2 M k_f)/(\hbar^4 \gamma_A \gamma_B) \tag{1.47}$$

and to obtain the following expression for (1.30):

$$W(\nu \div \mu) = \frac{d\Omega_\mu \, M\kappa^2{}_\mu L^6}{(2\pi\hbar)^4 \gamma_A \gamma_B |\overline{H}(\nu)|^2 \hbar c (2J_a + 1)(2J_b + 1)} \sum \int dk_i \, d\Omega_i \, \rho(k_i) \int d\Omega_f \, H_{f\mu,B} V_{B,A} H_{A,i\nu} H^*_{f\mu,B'} V^*_{B',A'} H^*_{A',i\nu} . \tag{1.48}$$

In Eq. (1.48), the summation sign on the right-hand side sums over the intermediate states A, B, A', B' and the magnetic sublevels (M_a, M_b, M_a', M_b') of the initial and the final states of atoms A and B.

The matrix elements over H and V are

$$H_{f\mu,B} = ie(2\pi\hbar c\kappa_\mu L^{-3})^{1/2} (J_b' M_b' | \textstyle\sum_j \vec{\sigma}_\mu \cdot \vec{r}(B_j) | j_b m_b) \times (\chi_f | e^{-i\vec{\kappa}_\mu \cdot \vec{r}_B} | \chi_b) , \tag{1.49}$$

$$H_{A,i\nu} = -ie(2\pi\hbar c n_\nu \kappa_\nu L^{-3})^{1/2} (j_a m_a | \textstyle\sum_i \vec{\sigma}_\nu \cdot \vec{r}(Ai) | J_a M_a) \times (\chi_a | e^{i\vec{\kappa}_\nu \cdot \vec{r}_A} | \chi_i) , \tag{1.50}$$

$$V_{B,A} = (-\sqrt{6}\, e^2) \sum_{uv} (-)^v C\,(112; u, v-u, v)$$

$$\times (J_a' M_a') | \textstyle\sum_i r_u(Ai) | j_a m_a)$$
$$\times (j_b m_b | \textstyle\sum_j r_{v-u}(Bj) | J_b M_b)$$
$$\times (\chi_b | C_{2,-v}(\hat{R}) R^{-3} | \chi_a) , \qquad (1.51)$$

where $\vec{r}_A$ and $\vec{r}_B$ are the position vectors of nuclei A and B, respectively, measured from the space-fixed center. Substituting the matrix elements (1.49)–(1.51) into (1.48), we perform the summations over the intermediate states A, B, A', B'. These summations imply the summations over the nuclear relative motions as well as the magnetic sublevels of the atomic excited states, i.e.,

$$\sum_A = \sum_{m_a} \sum_a , \qquad (1.52)$$

where

$$\textstyle\sum_a = (L/2\pi)^3 \int dk_a \, d\Omega_a \, k_a^2 , \qquad (1.53)$$

and by Eq. (1.5) of the nuclear wave functions, we have

$$\textstyle\sum_a \chi_a^* (\vec{R}) \chi_a(\vec{R}') = \delta(\vec{R} - \vec{R}') . \qquad (1.54)$$

Having performed the above-mentioned summations, (1.48) now becomes

$$W(\nu \to \mu) = F_1(\hbar^2 \gamma_A \gamma_B)^{-1} \sum \mathcal{L}(\nu\mu) \int d\Omega_i \, dk_i \, \rho(k_i) k_f$$
$$\times \int d\Omega_f \, C_v C_v^* , \qquad (1.55)$$

where

$$F_1 = \frac{6 d\Omega_\mu \kappa_\mu^3 c M e^8 n_\nu \kappa_\nu}{(2\pi)^2 \hbar | \overline{H}(\nu) |^2 (2J_a + 1)(2J_b + 1)} , \qquad (1.56)$$

$$C_v = (-)^v (\chi_f | e^{-i\vec{\kappa}_\mu \cdot \vec{r}_B} c_{2,-v}(\hat{R}) R^{-3} e^{i\vec{\kappa}_\nu \cdot \vec{r}_A} | \chi_i) , \qquad (1.57)$$

and

$$\mathcal{L}(\nu\mu) = C(112; u, v-u, v) C(112; u', v'-u', v') (J_b' M_b' | \textstyle\sum_j \vec{\sigma}_\mu \cdot \vec{r}(B_j) | j_b m_b')^* (j_b m_b' | \textstyle\sum_j r_{v'-u'}(Bj) | J_{b'} M_{b'})^*$$
$$\times (J_a' M_a' | \textstyle\sum_i r_{u'}(Ai) | j_a m_a')^* (j_a m_a' | \textstyle\sum_i \vec{\sigma}_{\nu'} \cdot \vec{r}(Ai) | J_a M_a)^* (J_b' M_b' | \textstyle\sum_j \vec{\sigma}_\mu \cdot \vec{r}(Bj) | j_b m_b)(j_b m_b | \textstyle\sum_j r_{v-u}(B_j) | J_b M_b)$$
$$\times (J_a' M_a' | \textstyle\sum_i r_u(Ai) | j_a m_a)(j_a m_a | \textstyle\sum_i \vec{\sigma}_\nu \cdot \vec{r}(Ai) | J_a M_a) . \qquad (1.58)$$

The summation sign in Eq. (1.55) now stands for the summation to sum over all the magnetic quantum numbers (i.e., $u, v, u', v', m_a, m_b, m_a', m_b', M_a, M_b, M_a', M_b'$). Substituting (1.5) into (1.57) we have

$$C_v = (-)^v L^{-3} e^{i(\vec{\kappa}_\nu - \vec{\kappa}_\mu) \cdot \vec{R}_c} \int d^3R \, e^{i\vec{\rho} \cdot \vec{R}} C_{2,-v}(\hat{R}) R^{-3} , \qquad (1.59)$$

where

$$\vec{R}_c = (m_A + m_B)^{-1} (m_A \vec{r}_A + m_B \vec{r}_B) , \qquad (1.60a)$$

$$\vec{R} = \vec{r}_A - \vec{r}_B , \qquad (1.60b)$$

and

$$\vec{\rho} = \vec{K} - \vec{k}_f , \qquad (1.61)$$

with

$$\vec{K} = (m_A + m_B)^{-1} (m_B \vec{\kappa}_\nu + m_A \vec{\kappa}_\mu) + \vec{k}_i . \qquad (1.62)$$

By partial-wave expansion

$$e^{i\vec{\rho} \cdot \vec{R}} = 4\pi \sum_{l=0}^{\infty} \sum_{m=-l}^{l} i^l j_l(\rho R) Y_{lm}(\hat{R}) Y_{lm}^*(\hat{\rho}) \qquad (1.63)$$

and integration over the angular part, we have

$$\int d^3\vec{R} \, e^{i\vec{\rho} \cdot \vec{R}} \, C_{2,-v}(\hat{R}) R^{-3}$$
$$= -4\pi (\tfrac{4}{5}\pi)^{1/2} Y_{2,-v}(\hat{\rho}) \int_0^{R_0} dR \, j_2(\rho R) R^{-1} . \qquad (1.64)$$

The integration limit R_0 practically equals the dimension of the box L, which is very large. We therefore have[22]

$$\int_0^{R_0 \to \infty} dR \, j_2(\rho R) R^{-1} = \tfrac{1}{3} . \qquad (1.65)$$

Substituting (1.65) into (1.64), the latter is then in turn substituted into (1.59) to give an expression for C_v; we obtain

$$C_v C_{v'}^* = L^{-6} (\tfrac{4}{3}\pi)^2 (\tfrac{4}{5}\pi) Y_{2,v}^*(\hat{\rho}) \, Y_{2,v'}(\hat{\rho}) . \qquad (1.66)$$

The unit vector $\hat{\rho}$ that appears in $Y_{lm}(\hat{\rho})$ implies the polar and azimuth angles $(\theta_\rho, \varphi_\rho)$ of vector $\hat{\rho}$ with respect to the space-fixed z axis. By a rotational transformation (see Fig. 2), $Y_{lm}(\theta_\rho \varphi_\rho)$ transforms into the following:

$$Y_{lm}(\theta_\rho \varphi_\rho) = \textstyle\sum_{m'} D_{mm'}^{l*}(\theta_f, \varphi_f, 0) Y_{lm'}(\chi\psi) , \qquad (1.67)$$

where (χ, ψ) are the corresponding angles of $\vec{\rho}$ with respect to vector $\vec{k}_f$ (recall $\vec{\rho} = \vec{K} - \vec{k}_f$), $D_{mm'}^{l*}(\theta_f, \varphi_f, 0)$ is the rotational matrix,[23] and (θ_f, φ_f) are the angles of vector $\vec{k}_f$ with respect to the fixed z axis. Integrating (1.67) over $d\Omega_f$, we have

$$\int Y_{lm}(\theta_\rho \varphi_\rho) \, d\Omega_f = 4\pi \delta_{l,0} \delta_{m,0} \textstyle\sum_{m'} \delta_{m',0} Y_{l,m'}(\chi\psi)$$
$$= (4\pi)^{1/2} \delta_{l,0} \delta_{m,0} . \qquad (1.68)$$

Since the product of two spherical harmonics gives a linear combination of spherical harmonics with appropriate coefficients,[23] the integration of (1.66) over $d\Omega_f$ by using the above formula (1.68) becomes

$$\int d\Omega_f\, C_v C^*_{v'} = L^{-6}(4\pi)^3(9)^{-1}(-)^v C(220;\ -vv0)\,C(220;\ 000)\,\delta_{v,v'} = L^{-6}(4\pi)^3(45)^{-1}\delta_{v,v'}\ . \tag{1.69}$$

Substituting this into (1.55), we have

$$W(\nu\rightarrow\mu) = F_2(\hbar^2\gamma_A\gamma_B)^{-1}\sum \mathfrak{L}(\nu\mu)\delta_{v,v'}\ , \tag{1.70}$$

where

$$F_2 = F_1 L^{-6}(4\pi)^3(45)^{-1}\int d\Omega_i\, dk_i\,\rho(k_i)k_f\ ,\qquad k_f = [k_i^2 + (\epsilon_{j_a} - \epsilon_{j_b})(2M/\hbar^2)]^{1/2}\ . \tag{1.71}$$

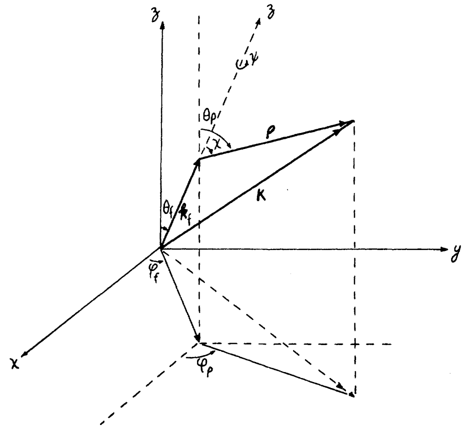

FIG. 2. Rotational transformation: $Y_{lm}(\theta_\rho\varphi_\rho)$ is expressed in terms of $Y_{lm'}(\chi\psi)$ [see Eq. (1.67)] by rotating the space-fixed z axis into the z' axis. The latter is parallel to vector $\vec{k}_f$.

The incident light, propagating along the x axis, is assumed to be y polarized, i.e., $\vec{\sigma}_\nu\cdot\vec{r}=y$. The emitted sensitized fluorescence observed in the z direction can be either x or y polarized, i.e., $\vec{\sigma}_\mu\cdot\vec{r}=x$ or y. If the intensity of the scattered light of x polarization equals to that of y polarization, the sensitized fluorescence is not polarized. On the other hand we will have 100% polarization if the intensity of x polarization equals to zero. Both x and y can be expressed in terms of spherical tensors r_1 and r_{-1},

$$x = -(1/\sqrt{2})(r_1 - r_{-1}) \quad\text{and}\quad y = (i/\sqrt{2})(r_1 + r_{-1})\ . \tag{1.72}$$

The matrix elements of r_1 and r_{-1} can easily be evaluated as[23]

$$(jm|r_u|JM) = C(J1j;\ Mum)(j||\vec{r}||J)\ . \tag{1.73}$$

Using the relations in (1.72) and (1.73), we now sum $\mathfrak{L}(\nu\mu)$ of Eq. (1.58) over v' and all the magnetic quantum numbers, i.e., M_a, M'_a, M_b, M'_b, of the ground states and obtain

$$\sum_{\substack{M_aM'_a\\ M_bM'_bv'}} \mathfrak{L}(\nu=y,\ \mu=y)\delta_{vv'} = q_0\{q_1\delta_{m'_a,m_a}\delta_{m'_b,m_b}\delta_{u',u} + q_2\delta_{m'_a,m_a-2}\delta_{m'_b,m_b-2}\delta_{u',u+2} + q_3\delta_{m'_a,m_a+2}\delta_{m'_b,m_b+2}\delta_{u',u-2}\}R(J;\ j)\ , \tag{1.74}$$

where

$$R(J;\ j) = |(J'_b||\textstyle\sum_j\vec{r}(Bj)||j_b)|^2|(j_b||\sum_j\vec{r}(Bj)||J_b)|^2 \times|(j_a||\sum_i\vec{r}(Ai)||J_a)|^2|(J'_a||\sum_i\vec{r}(Ai)||j_a)|^2\ , \tag{1.75}$$

$$q_0 = \tfrac{1}{4}C(112;\ u,\ v-u,\ v)\,C(J_b1j_b;\ m_b-v+u,\ v-u,\ m_b)\,C(j_a1J'_a;\ m_a,\ u,\ m_a+u)\ , \tag{1.76}$$

$$q_1 = C(112;\ u,\ v-u;\ v)[C(j_b1J'_b;\ m_b,\ 1,\ m_b+1)^2 + C(j_b1J'_b;\ m_b,\ -1,\ m_b-1)^2][C(J_a1j_a;\ m_a-1,\ 1,\ m_a)^2 + C(J_a1j_a;\ m_a+1,\ -1,\ m_a)^2]\,C(J_b1j_b;\ m_b-v+u,\ v-u,\ m_b)\,C(j_a1J'_a;\ m_a,\ u,\ m_a+u)\ , \tag{1.77}$$

$$q_2 \text{ or } q_3 = C(112;\ u\pm 2,\ v-u\mp 2,\ v)\,C(j_b1J'_b;\ m_b\mp 2,\ \pm 1,\ m_b\mp 1)\,C(j_b1J'_b;\ m_b,\ \mp 1,\ m_b\mp 1) \times C(J_b1j_b;\ m_b-v+u,\ v-u\mp 2,\ m_b\mp 2)\,C(J_a1j_a;\ m_a\mp 1,\ \mp 1,\ m_a\mp 2)\,C(J_a1j_a;\ m_a\mp 1,\ \pm 1,\ m_a) \times C(j_a1J'_a;\ m_a\mp 2,\ u\pm 2,\ m_a+u)\ . \tag{1.78}$$

In Eq. (1.78), the upper sign gives q_2 and the lower sign gives q_3. Again summing (1.74) over m'_a, m'_b, and u', we have

$$\sum_{m'_am'_bu'}(1.74) = [Q_1(m_am_buv) + Q_2(m_am_buv) + Q_3(m_am_buv)]R(J;\ j)\ , \tag{1.79}$$

where

$$Q_1(m_am_buv) = q_1q_0\ , \tag{1.80a}$$

$$Q_2(m_am_buv) = q_2q_0\ , \tag{1.80b}$$

$$Q_3(m_am_buv) = q_3q_0\ . \tag{1.80c}$$

Since

$$\sum_{m_am_buv} Q_2(m_am_buv) = \sum_{m_am_buv} Q_3(m_am_buv)\ , \tag{1.81}$$

the summation of (1.79) over the remaining magnetic quantum numbers, i.e., m_a, m_b, u, v, reduces to

$$\sum_{\text{all}} \mathcal{L}(yy)\delta_{vv'}$$

$$= \sum_{m_a m_b uv} [Q_1(m_a m_b uv) + 2Q_2(m_a m_b uv)] R(J;\, j) \,, \tag{1.82}$$

where the $\sum_{\text{all}}$ sign on the left-hand side sums over all the magnetic quantum numbers. Substituting (1.82) into (1.70), the probability of observing y-polarized light becomes

$$W(y \to y) = F_2(\hbar^2\gamma_A\gamma_B)^{-1} \sum_{m_a m_b uv} [Q_1(m_a m_b uv) + 2Q_2(m_a m_b uv)] R(J;\, j) \,. \tag{1.83}$$

Following similar procedures, the probability for observing x-polarized scattered light after the excitation by y-polarized light becomes

$$W(y \to x) = F_2(\hbar^2\gamma_A\gamma_B)^{-1} \sum_{m_a m_b uv} [Q_1(m_a m_b uv) - 2Q_2(m_a m_b uv)] R(J;\, j). \tag{1.84}$$

The polarization P_0 of the sensitized fluorescence therefore becomes

$$P_0 = \frac{W(y \to y) - W(y \to x)}{W(y \to y) + W(y \to x)} = \frac{2\sum_{m_a m_b uv} Q_2(m_a m_b uv)}{\sum_{m_a m_b uv} Q_1(m_a m_b uv)} \,. \tag{1.85}$$

The polarization P_0 expressed in the above Eq. (1.85) is the polarization in the absence of magnetic field. When the field is zero, $W(\nu \to \mu)$ and, consequently, P_0 are invariant with respect to the rotation of the coordinates. As a result y can be replaced by z and x can be replaced by y, i.e., $W(y \to y) \to W(z \to z)$ and $W(y \to x) \to W(z \to y)$. The matrix elements are transformed accordingly and $W(\nu \to \mu)$ becomes easier to evaluate in many cases. The polarization now takes the following form though it has the same value as that of (1.85):

$$P_0 = \frac{W(z \to z) - W(z \to y)}{W(z \to z) + W(z \to y)} = \frac{\sum_{m_a m_b uv}[Q_\parallel(m_a m_b uv) - Q_\perp(m_a m_b uv)]}{\sum_{m_a m_b uv}[Q_\parallel(m_a m_b uv) + Q_\perp(m_a m_b uv)]} \,, \tag{1.86}$$

where

$$Q_\parallel(m_a m_b uv) = C(112;\, u,\, v-u,\, v)^2 C(j_b 1 J_b';\, m_b 0 m_b)^2\, C(J_a 1 j_a;\, m_a 0 m_a)^2 C(J_b 1 j_b;\, m_b - v + u,\, v-u,\, m_b)^2 \times C(j_a 1 J_a';\, m_a,\, u,\, m_a + u)^2 \,, \tag{1.87}$$

$$Q_\perp(m_a m_b uv) = \tfrac{1}{2} C(112;\, u,\, v-u,\, v)^2 [C(j_b 1 J_b';\, m_b,\, 1,\, m_b+1)^2 + C(j_b 1 J_b';\, m_b,\, -1,\, m_b - 1)^2] C(J_a 1 j_a;\, m_a 0 m_a)^2 \times C(j_a 1 J_a',\, m_a,\, u,\, m_a + u)^2 C(J_b 1 j_b;\, m_b - v + u,\, v-u,\, m_b)^2 \,. \tag{1.88}$$

Both Eq. (1.85) and (1.86) can be used to calculate the (magnetic) field-free polarization.

II. MAGNETIC FIELD EFFECT

If the magnetic field H_z along the quantization z axis is not zero, the magnetic sublevels of the atomic states will not be degenerate and hence the conditions of (1.44) and the expression (1.45) for G will not be satisfied. We now reexpress the original expression for G in (1.43) as follows:

$$G = [\zeta^*(\Delta_{AB'} + \hbar^2(k_i^2 - k_f^2)(2M)^{-1}) - \zeta(\Delta_{A'B} + \hbar^2(k_i^2 - k_f^2)(2M)^{-1})] \times \frac{2\pi i}{(\Delta_{AA'} - i\hbar\gamma_A)(\Delta_{BB'} - i\hbar\gamma_B)} \,. \tag{2.1}$$

The ζ functions above, as shown in (1.35), are linear combination of the principal-value part and a δ function. The principal values arriving from ζ^* and ζ above will cancel each other after integrating G over dk_f, and the integration over the δ functions gives

$$\int dk_f\, k_f^2 G = -\frac{4\pi^2 M k_f}{\hbar^2(\Delta_{AA'} - i\hbar\gamma_A)(\Delta_{BB'} - i\hbar\gamma_B)} \,. \tag{2.2}$$

Following the same argument as in Sec. I, the summation and averaging over (1.30) now gives

$$W_H(\nu \to \mu) = \overline{\sum_i} \sum_f \sum_\mu \sum_\nu |b_{f\mu}(\infty)|^2 = -\frac{d\Omega_\mu M \kappa_\mu^2 L^6 \int d\Omega_i\, dk_i\, \rho(k_i)\, k_f}{(2\pi\hbar)^2(4\pi^2)|\overline{H}(\nu)|^2 \hbar c(2J_a+1)(2J_b+1)} \times \sum \frac{\int d\Omega_f H_{f\mu,B} V_{B,A} H_{A,i\nu} H^*_{f\mu,B'} V^*_{B',A'} H^*_{A',i\nu}}{(\Delta_{AA'} - i\hbar\gamma_A)(\Delta_{BB'} - i\hbar\gamma_B)} \,. \tag{2.3}$$

The right-hand-side summation again sums over all the intermediate states A, B, A', B', and the sublevels (M_a, M_b, M_a', M_b') of the initial and final atomic states. A subscript H is added here to the above probability $W_H(\nu \to \mu)$ to indicate its magnetic field dependence. Let

$$\beta_a = \mu_B g_{j_a} H_z \quad \text{and} \quad \beta_b = \mu_B g_{j_b} H_z \,, \tag{2.4}$$

where $\mu_B = e\hbar/2mc$ is the Bohr magneton, then

(1.39) and (1.40) can be written as

$$\Delta_{AA'} = \beta_a(m_a - m_a') \text{ and } \Delta_{BB'} = \beta_b(m_b - m_b') \ . \qquad (2.5)$$

After substituting (1.49)–(1.51) for all the matrix elements and performing the necessary summations, (2.3) now becomes

$$W_H(\nu \to \mu) = -F_2 \sum\nolimits_{\text{all}} \mathcal{L}(\nu\mu)\delta_{vv'}[\beta_a(m_a - m_a') - i\hbar\gamma_A]^{-1}$$

$$\times [\beta_b(m_b - m_b') - i\hbar\gamma_B]^{-1} \ . \qquad (2.6)$$

$\mathcal{L}(\nu\mu)$ above is defined in (1.58) and the summation sign sums over all the magnetic quantum numbers. Equation (2.6) differs from the previous expression (1.70) for the field-free case by this extra magnetic-field-dependent denominator.

Similar to Sec. I, the incident light is assumed to be y polarized, i.e., $\vec{\sigma}_\nu \cdot \vec{r} = y$ and the emitted sensitized fluorescence either be x or y polarized. Let us first consider the case $\vec{\sigma}_\mu \cdot \vec{r} = y$ and find $W(y \to y)$ the probability of observing y-polarized light. Since the denominator in (2.6) depends on the magnetic quantum numbers m_a, m_a', m_b, m_b' of the excited states only we can sum $\mathcal{L}(yy)\delta_{vv'}$ over the other quantum numbers, i.e., M_a, M_a', M_b, M_b', and v', and this is the result of (1.74). Substituting Eq. (1.74) into (2.6) and performing the summation on m_a', m_b', and u', we have

$$W_H(y \to y) = F_2 \sum_{m_a m_b u v} \left(\frac{Q_1(m_a m_b u v)}{\hbar^2 \gamma_A \gamma_B} - \frac{Q_2(m_a m_b u v)}{(2\beta_a - i\hbar\gamma_A)(2\beta_b - i\hbar\gamma_B)} - \frac{Q_3(m_a m_b u v)}{(2\beta_a + i\hbar\gamma_A)(2\beta_b + i\hbar\gamma_B)} \right) R(J;\ j) \ , \qquad (2.7)$$

where Q_1, Q_2, Q_3, and $R(J;j)$ are defined in (1.80) and (1.75), respectively. Since the summation indices are contained in Q's only and the denominators are independent of them, by applying the result of (1.81) we obtain

$$W_H(y \to y) = F_2 \sum_{m_a m_b u v} \left(\frac{Q_1(m_a m_b u v)}{\hbar^2 \gamma_A \gamma_B} + \frac{2Q_2(m_a m_b u v)(\hbar^2\gamma_A\gamma_B - 4\beta_a\beta_b)}{(4\beta_a^2 + \hbar^2\gamma_A^2)(4\beta_b^2 + \hbar^2\gamma_B^2)} \right) R(J;\ j) \ . \qquad (2.8)$$

Following similar procedures, the probability for observing x-polarized light becomes

$$W_H(y \to x) = F_2 \sum_{m_a m_b u v} \left(\frac{Q_1(m_a m_b u v)}{\hbar^2 \gamma_A \gamma_B} - \frac{2Q_2(m_a m_b u v)(\hbar^2\gamma_A\gamma_B - 4\beta_a\beta_b)}{(4\beta_a^2 + \hbar^2\gamma_A^2)(4\beta_b^2 + \hbar^2\gamma_B^2)} \right) R(J;\ j) \ . \qquad (2.9)$$

Substituting (2.4) for β_a and β_b the polarization of the sensitized fluorescence in the presence of the magnetic field is as follows:

$$P(H_z) = P_0 \frac{(\hbar^2\gamma_A\gamma_B - 4g_{j_a}g_{j_b}\mu_B{}^2H_z{}^2)\hbar^2\gamma_A\gamma_B}{(4g_{j_a}^2\mu_B{}^2H_z{}^2 + \hbar^2\gamma_A{}^2)(4g_{j_b}^2\mu_B{}^2H_z{}^2 + \hbar^2\gamma_B{}^2)} \ , \qquad (2.10)$$

where P_0, expressed in either (1.85) or (1.86), is the polarization for the field-free case. When $H_z = 0$, the magnetic-field-dependent factor in (2.10) becomes unity, or $P(H_z) = P_0$. As H_z increases, $P(H_z)$ decreases and becomes zero at $H_z = \pm(\hbar^2\gamma_A\gamma_B/4g_{j_a}g_{j_b}\mu_B{}^2)^{1/2}$. After the zero points $P(H_z)$ becomes negative and approaches zero again when H_z becomes very large.

III. EXAMPLES AND COMPARISON WITH PURE FLUORESCENCE

Let us review briefly the polarization of pure fluorescence (without sensitizer) when the excitation of the system was induced by linearly polarized light. In this case the optical excitation is not transferred between two atoms. The radiation is absorbed by and then reemitted from the same atom. We use the same notations, namely (JM) and $(J'M')$ to specify the initial and the final states, respectively, and (jm) and (jm') the excited states. J and J' need not be the same. However, if $J = J'$ and they belong to the same state, we have the case of "resonance fluorescence." Following the result of Ref. 12 and expressing it in the notation of the present work, the probability of observing μ-polarized fluorescence after the system was excited by light of ν polarization is as follows:

$$W_H(\nu \to \mu) = \overline{\sum_i} \sum_f \sum_\mu \sum_\nu |b_{f\mu}(\infty)|^2$$

$$= 2\pi i \overline{F}_1 \sum_{mm'MM'} (J'M' | \textstyle\sum_i \vec{\sigma}_\mu \cdot \vec{r}_i | jm)^*$$

$$\times (jm | \textstyle\sum_i \vec{\sigma}_\nu \cdot \vec{r} | JM)^* (J'M' | \textstyle\sum_i \vec{\sigma}_\mu \cdot \vec{r}_i | jm')$$

$$\times (jm' | \textstyle\sum_i \vec{\sigma}_\nu \cdot \vec{r}_i | JM)(\epsilon_m - \epsilon_{m'} + i\hbar\gamma)^{-1} \ , \qquad (3.1)$$

where

$$\overline{F}_1 = (2\pi)^2 c\hbar e^4 d\Omega_\mu\ \kappa_\mu{}^3 n_\nu \kappa_\nu / |\overline{H}(\nu)|^2 \ , \qquad (3.2)$$

and γ, similar to γ_A (or γ_B) in (1.25), is the damping constant of the excited state. The incident light is again assumed to be y polarized, i.e., $\vec{\sigma}_\nu \cdot \vec{r} = y$. After carrying out the summation procedures in (1.31), the probability of observing either y- or x-

polarized fluorescence becomes

$$W_H(y\rightarrow\mu)=\bar{F}_2\sum_m\left(\frac{\bar{Q}_1(m)}{\hbar\gamma}\pm\frac{2\hbar\gamma\bar{Q}_2(m)}{(2\mu_B g_j H_z)^2+\hbar^2\gamma^2}\right), \tag{3.3}$$

where the "+" sign refers to $W(y\rightarrow y)$, i.e., y polarization, and the "−" sign refers to $W(y\rightarrow x)$, i.e., x polarization,

$$\bar{F}_2=2\pi\bar{F}_1\left|(J'\,||\textstyle\sum_i\vec{\mathrm{r}}_i||\,j)\right|^2\left|(j\,||\textstyle\sum_i\vec{\mathrm{r}}_i||\,J)\right|^2, \tag{3.4}$$

$$\bar{Q}_1(m)=\tfrac{1}{4}[C(J1j;m-1,1,m)^2+C(J1j;m+1,-1,m)^2]\times[C(j1J';m,1,m+1)^2+C(j1J';m,-1,m-1)^2], \tag{3.5}$$

$$\bar{Q}_2(m)=\tfrac{1}{4}C(J1j;m-1,1,m)C(J1j;m-1,-1,m-2)\times C(j1J';m,-1,m-1)C(j1J';m-2,1,m-1). \tag{3.6}$$

The polarization of the observed fluorescence therefore is

$$P'(H_z)=\left(\frac{\hbar^2\gamma^2}{(2\mu_B g_j H_z)^2+\hbar^2\gamma^2}\right)P_0', \tag{3.7}$$

where

$$P_0'=\frac{2\sum_m\bar{Q}_2(m)}{\sum_m\bar{Q}_1(m)}=\frac{W_{H=0}(y\rightarrow y)-W_{H=0}(y\rightarrow x)}{W_{H=0}(y\rightarrow y)+W_{H=0}(y\rightarrow x)} \tag{3.8}$$

is the polarization in the absence of a magnetic field. The prime on P and P_0 is introduced to distinguish it from the case of sensitized fluorescence. Similar to the discussion in Sec. I, the polarization P_0' [as well as $W(y\rightarrow y)$ and $W(y\rightarrow z)$] in the absence of the magnetic field is rotationally invariant. By rotating the coordinates such that $W(y\rightarrow y)\rightarrow W(z\rightarrow z)$ and $W(y\rightarrow x)\rightarrow W(z\rightarrow y)$, P_0' can also be expressed as follows:

$$P_0'=\frac{W_{H=0}(z\rightarrow z)-W_{H=0}(z\rightarrow y)}{W_{H=0}(z\rightarrow z)+W_{H=0}(z\rightarrow y)}$$

$$=\frac{\sum_m\bar{Q}_{\parallel}(m)-\sum_m\bar{Q}_{\perp}(m)}{\sum_m\bar{Q}_{\parallel}(m)+\sum_m\bar{Q}_{\perp}(m)}, \tag{3.9}$$

where

$$\bar{Q}_{\parallel}(m)=C(J1j;m0m)^2C(j1J;m0m)^2 \tag{3.10}$$

and

$$\bar{Q}_{\perp}(m)=\tfrac{1}{2}[C(j1J';m,1,m+1)^2+C(j1J';m,-1,m-1)^2]C(J1j;m0m)^2. \tag{3.11}$$

The expressions (3.8) and (3.9) for field-free polarization P_0' are identical. Of course, they are different from that of sensitized fluorescence, which is expressed in (1.85) and (1.86). The numerical values of field-free polarization for both pure fluorescence and sensitized fluorescence are listed in Table I for different transitions. The polarization of sensitized fluorescence is in general smaller than that of pure fluorescence. Since the former differs from the latter by its energy-transfer process, these results indicate that coherence is only partially transferred during the excitation transfer between atoms.

The magnetic field depolarization of sensitized fluorescence as shown in (2.10) is very different from that of pure fluorescence which is expressed in (3.7). For direct comparison, we consider a special case where $\gamma_A=\gamma_B=\gamma$ and $g_{j_a}=g_{j_b}=g_j$. Equation (2.10) of sensitized fluorescence now becomes

$$P(H_z)=P_0\left(\frac{1}{(\xi H_z)^2+1}-\frac{2(\xi H_z)^2}{[(\xi H_z)^2+1]^2}\right), \tag{3.12}$$

where

$$\xi=2\mu_B g_j/\hbar\gamma. \tag{3.13}$$

We also express (3.7), the expression for pure fluorescence, in terms of ξ,

$$P'(H_z)=\frac{P_0'}{(\xi H_z)^2+1}. \tag{3.14}$$

Equations (3.12) and (3.14) are plotted against

TABLE I. Polarizations in the absence of magnetic field.

Transitions	P_0' (pure fluorescence)[a] (%)	Transitions			P_0 (sensitized fluorescence)[b] (%)
${}^1S_0\rightarrow{}^1P_1\rightarrow{}^1S_0$	100	${}^1S_0(A)\rightarrow{}^1P_1(A)$	⋯	${}^1P_1(B)\rightarrow{}^1S_0(B)$	14.29
${}^1P_1\rightarrow{}^1D_2\rightarrow{}^1P_1$	44.68	${}^1P_1(A)\rightarrow{}^1D_2(A)$	⋯	${}^1D_2(B)\rightarrow{}^1P_1(B)$	1.83
${}^2S_{1/2}\rightarrow{}^2P_{1/2}\rightarrow{}^2S_{1/2}$	0	${}^2S_{1/2}(A)\rightarrow{}^2P_{1/2}(A)$	⋯	${}^2P_{1/2}(B)\rightarrow{}^2S_{1/2}(B)$	0
${}^2S_{1/2}\rightarrow{}^2P_{3/2}\rightarrow{}^2S_{1/2}$	60	${}^2S_{1/2}(A)\rightarrow{}^2P_{3/2}(A)$	⋯	${}^2P_{3/2}(B)\rightarrow{}^2S_{1/2}(B)$	3.70

[a] Results are obtained from Eq. (3.8) or (3.9). Capital J and J' refer there to the total electronic angular momenta of the initial and final states, respectively, and small j refers to that of the excited state. For all the transitions considered here we have $J=J'$.

[b] Results are obtained from Eq. (1.85) or (1.86). For all the transitions considered here we have $J_a(=J_b)=J_a'(=J_b')$ for the initial and final states and $j_a=j_b$ for the excited states.

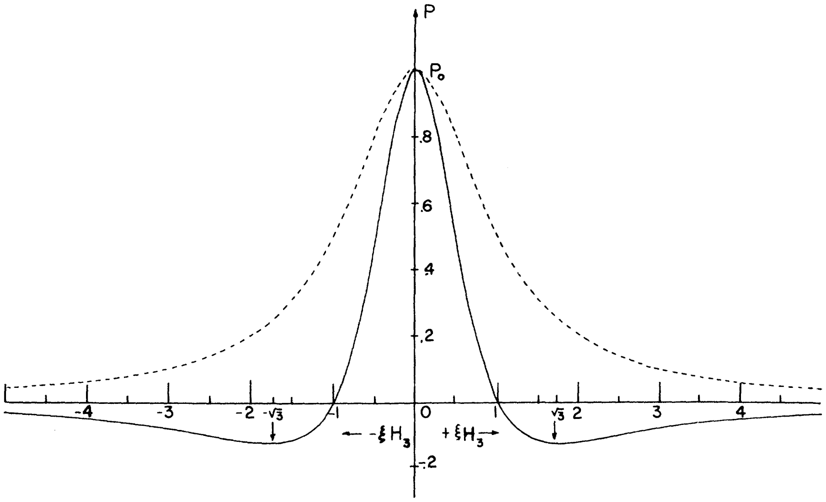

FIG. 3. Magnetic field depolarization: Polarizations [see Eqs. (3.12) and (3.14)] are plotted against ξH_z where $\xi = 2\mu_0 g_j/\hbar\gamma$. Solid line represents a special case (i.e., $g_{ja} = g_{jb} = g_j$ and $\gamma_A = \gamma_B = \gamma$) of sensitized fluorescence, and $\xi H_z = \pm\sqrt{3}$ corresponds to a minimum value of $-\frac{1}{8}P_0$ of the polarization. Dashed line represents the pure fluorescence where no excitation transfer has taken place. P_0 is the field-free polarization for both cases.

ξH_z in Fig. 3. At $\xi H_z = 0$ both $P(H_z)$ and $P'(H_z)$ are at their maximum values, namely, their field-free values P_0 and P_0', respectively. The polarization of pure fluorescence $P'(H_z)$ drops to half of its maximum value at $\xi H_z = \pm 1$, and it approaches zero as $\xi H_z \rightarrow \infty$. On the other hand the polarization of sensitized fluorescence $P(H_z)$ becomes zero at $\xi H_z = \pm 1$, and it reaches a negative minimum, namely, $-\frac{1}{8}P_0$, at $\xi H_z = \pm\sqrt{3}$. When $\xi H_z \rightarrow \pm\infty$, $P(H_z)$ again approaches zero, however, from the negative side. For the general case of sensitized fluorescence, where $\gamma_A \neq \gamma_B$ and $g_{j_a} \neq g_{j_B}$, the depolarization will depend on γ_A, γ_B, g_{j_a}, and g_{j_b}. The experimental line shape of depolarization can therefore determine the lifetimes of the excited states of both atoms A and B.

The radiative interaction and the electrostatic interaction considered in this work are purely electronic, thus the nuclear spin of the atom will not have a direct effect on them. However if the atom possesses nuclear spin $\vec{I}$, the spin angular momentum will couple with the electronic angular momentum $\vec{J}\hbar$ to give total angular momentum $\vec{F}\hbar$ (i.e., $\vec{F} = \vec{I} + \vec{J}$) of the atom. The hyperfine interaction $\vec{I}\cdot\vec{J}$ requires that our eigenstate be represented by F instead of J. The nuclear spin, therefore, affects the polarization of the fluorescence indirectly. Such nuclear spin effect has been studied in the cases of optical-pumping signals[24] and Hanle-effect signals.[25] However, the results of the present work can be applied to atoms with nuclear spin provided one can isolate and follow (e.g., by optical laser) the optical transitions between a hyperfine level F of the ground state and a hyperfine level f of the excited state. It was mentioned in Sec. I that $\vec{J}\hbar$ (of the ground state) and $\vec{j}\hbar$ (of the excited state) refer to the total angular momenta of the atom. When atoms under consideration possess nuclear spin, J and j are therefore to be replaced by the conventional quantum numbers F (of the ground state) and f (of the excited state) for the hyperfine levels, respectively. The magnetic quantum numbers M and m now refer to the magnetic sublevels of F and f. With these proper substitutions, all the formulas derived in this work are directly applicable to atoms with nuclear spin. Often one cannot isolate the optical transitions between a pair of hyperfine levels F and f, and instead one observes the superposition of all the transitions between different pairs of hyperfine levels. When a magnetic field is present, the intensity due to each individual transition will be weighted by its own field-depolarization factor which depends on the g_f value of the excited level involved. When this is the case, a proper summation over all the hyperfine levels involved will be needed. A general treatment on this nuclear-spin effect will be presented in a later work.

APPENDIX A

In this Appendix, we prove Eq. (1.23), which is

$$\bar{\Gamma} = (4\pi^2/3\hbar L^3)\, e^2 n_\nu \kappa_\nu \rho_\nu d\Omega_\nu \left| (J_a \left| \textstyle\sum_i \vec{\mathrm{r}}(Ai) \right| j_a) \right|^2 . \tag{A1}$$

After substituting Eq. (1.15) into (1.16), the definition for the ground-state damping constant becomes

$$\Gamma(E) = (2i/\hbar) \textstyle\sum_A H_{i\nu,A} H_{A,\,i\nu}\, \zeta(E - E_A) \ . \qquad \text{(A}$$

Since the transition-matrix elements here involve atom A only, the summation in (A2) reduces to $\sum_{m_a}$ and the summation over the photon states of absorption, which for linearly polarized light incident at a solid angle $d\Omega_\nu$ is simply

$$d\Omega_\nu \int d\kappa_\nu \rho_\nu \ . \tag{A3}$$

The principal-value part of the ζ function in (A2) is neglected because it contributes to a small energy shift only. After integrating the δ-function part and substituting (1.49) and (1.50) for the matrix elements, (A2) becomes

$$\Gamma=(4\pi^2/\hbar L^3)\,e^2 n_\nu\,\kappa_\nu\,\rho_\nu\,d\Omega_\nu \sum_{m_a} (J_a M_a|\textstyle\sum_i \vec{\sigma}_\nu\cdot\vec{\mathrm{r}}(Ai)\,|j_a m_a)(j_a m_a|\sum \vec{\sigma}_\nu\cdot\vec{\mathrm{r}}(Ai)\,|J_a M_a)\ . \tag{A4}$$

Since the averaging over the initial states of atom A is defined as

$$\bar{\Gamma}=(2J_a+1)^{-1}\sum_{m_a}\Gamma \tag{A5}$$

and for linearly polarized light (i.e., $\vec{\sigma}_\nu\cdot\vec{\mathrm{r}}=x, y$, or z) we have

$$\begin{aligned}\sum_{M_a m_a} &(J_a m_a|\textstyle\sum_i \vec{\sigma}_\nu\cdot\vec{\mathrm{r}}(Ai)\,|j_a m_a)\\ &\times(j_a m_a|\textstyle\sum_i \vec{\sigma}_\nu\cdot\vec{\mathrm{r}}(Ai)\,|J_a M_a)\\ &\qquad=\tfrac{1}{3}(2J_a+1)\,|(J_a|\,|\textstyle\sum_i\vec{\mathrm{r}}(Ai)|\,|j_a)|^2,\end{aligned} \tag{A6}$$

the resulting $\bar{\Gamma}$, therefore, takes the expression in (A1).

APPENDIX B

Here we verify the following equation:

$$\sum_{f'\lambda} H_{A,f'\lambda}H_{f'\lambda,A'}\,\zeta\,(E_{f\mu}-E_{f'\lambda})=(\hbar/2i)\,\gamma_A\,\delta_{AA'}, \tag{B1}$$

which appears as Eq. (1.26) in the text. The $\zeta(E_{f\mu}-E_{f'\lambda})$ on the left-hand side of (B1) is again replaced by $-i\pi\delta(E_{f\mu}-E_{f'\lambda})$. The summation in (B1) sums over all the final states, i.e.,

$$\sum_{f'}=(L/2\pi)^3\sum_{J_a'M_a'}\int d\Omega_{f'}\,dk_{f'}\,k_{f'}^2\ , \tag{B2}$$

and the photon states of emission, i.e.,

$$\textstyle\sum_\lambda=(L/2\pi)^3\displaystyle\sum_{\vec{\sigma}_\lambda}\int d\Omega_\lambda dk_\lambda K_\lambda^2\,, \tag{B3}$$

where $\sum_{\vec{\sigma}_\lambda}$ includes the two transverse polarizations. The matrix elements over H, which can be derived from (1.49) and (1.50), involves the optical transitions of atom A only. Consequently, the summation over the nuclear motion [by (1.54)] gives rise to the following orthogonal relation:

$$(L/2\pi)^3\int d\Omega_{f'}\,dk_{f'}\,k_{f'}^2\,(\chi_{f'}|\,e^{-i\vec{k}_\lambda\cdot\vec{\mathrm{r}}_A}\,|\chi_{a'})(\chi_a|\,e^{i\vec{k}_\lambda\cdot\vec{\mathrm{r}}_A}\,|\chi_{f'})=\delta_{a',a}\,. \tag{B4}$$

The summation over the atomic matrix elements and the photon polarizations gives

$$\begin{aligned}\sum_{J_a'M_a'}\sum_{\sigma_\lambda}&\int d\Omega_\lambda\,(j_a m_a|\textstyle\sum_i\vec{\sigma}_\lambda\cdot\vec{\mathrm{r}}(Ai)|J_a'M_a')(J_a'M_a'|\sum_i\vec{\sigma}_\lambda\cdot\vec{\mathrm{r}}(Ai)|j_a'm_a')\\ &=(\tfrac{8}{3}\pi)\sum_{J_a'M_a'}\sum_{\mu}(-)^\mu(j_a m_a|\textstyle\sum_i r_\mu(Ai)|J_a'M_a')(J_a'M_a'|\sum_i r_{-\mu}(Ai)|j_a'm_a')\\ &=(\tfrac{8}{3}\pi)\sum_{J_a'}\,|(j_a|\,|\textstyle\sum_i\vec{\mathrm{r}}(Ai)|\,|J_a')|^2\delta_{m_a,m_a'}\,\delta_{j_a,j_a'}\,.\end{aligned} \tag{B5}$$

Integrating the δ function over $d\kappa_\lambda$ and letting

$$\delta_{m_a,m_a'}\,\delta_{j_a,j_a'}\,\delta_{a,a'}=\delta_{A,A'}\,, \tag{B6}$$

Eq. (B1) becomes

$$\begin{aligned}\sum_{f'\lambda}&H_{A,f'\lambda}H_{f'\lambda,A'}\,\zeta(E_{f\mu}-E_{f'\lambda})\\ &=(-\tfrac{2}{3}i)\,e^2(\hbar c)^{-3}\,\delta_{AA'}\\ &\qquad\times\sum_{J_a'}\,|(j_a|\,|\textstyle\sum_i\vec{\mathrm{r}}(Ai)|\,|\,J_a')\,|^2(\epsilon_{j_a}-\epsilon_{J_a'})^3\ ,\end{aligned} \tag{B7}$$

where

$$\epsilon_{j_a}-\epsilon_{J_a'}=\hbar c\,\kappa_\lambda \tag{B8}$$

is the energy of the emitted photon when atom A is making a transition from the upper state j_a to the lower state J_a'. Defining

$$\begin{aligned}\gamma_A&\equiv(4e^2/3\hbar)(\hbar c)^{-3}\\ &\qquad\times\sum_{J_a'}\,|(j_a|\,|\textstyle\sum\vec{\mathrm{r}}(Ai)|\,|\,J_a')\,|^2\,(\epsilon_{j_a}-\epsilon_{J_a'})^3\ ,\end{aligned} \tag{B9}$$

which is the total transition probability from the excited state j_a to all lower states, and substituting (B9) into (B7), we thus prove Eq. (B1). The damping constant γ_A here in (B9) is more general than that introduced in (2.9) of Ref. 12 where only a single lower state was considered.

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*Work supported by Atomic Energy Commission under Contract No. AT-(40-1)-4048.

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PHYSICAL REVIEW A VOLUME 5, NUMBER 5 MAY 1972

Absolute Elastic and Inelastic Differential Cross Sections for the Scattering of Low-Energy Electrons by Potassium*†

James A. Slevin,‡ Paul J. Visconti, and Kenneth Rubin
City College of The City University of New York, New York, New York 10031

(Received 11 October 1971)

A new method for obtaining absolute differential excitation cross sections in low-energy electron-atom scattering is employed. The method utilizes certain of the kinematic properties of a recoiling-atom beam in an electron-atom crossed-beam configuration to distinguish between elastic scattering events and excitation to various atomic levels. Differential cross sections for elastic scattering and excitation of the $4p$ state of potassium are presented at 1.0, 3.0, 4.4, and 5.2 eV. A combined cross section for the $4s$-$5s$, $3d$ excitation integrated over a restricted angular range is also presented.

I. INTRODUCTION

The need for experimental information on the excitation of atoms by collisions with electrons has been outlined in a recent article by Moiseiwitsch and Smith.[1] The techniques which have been employed thus far to study such excitation processes involve using either (i) radiation detectors, (ii) metastable detectors, or (iii) scattered-electron detectors.

It is the purpose of this paper to introduce another method for studying inelastic processes in which observations are made on the scattered atoms and to present elastic and inelastic differential cross sections for the $4s \rightarrow 4p$ transitions in electron-potassium scattering at low energies (< 6 eV). This method has certain advantages over the first two techniques in that it is more general, i.e., not limited to a specific type of excitation, and is not complicated by such problems as cascading and uncertainties in detector efficiencies. Compared to the third technique, it has the advantage of not requiring a sophisticated electron source and detection system but suffers to some extent from the disadvantage of not having as good resolution.

II. RECOIL METHOD

The method used here is an extension of the atom-beam recoil technique first introduced by Rubin *et al.*,[2] and subsequently used by a number of investigators[3-6] to measure total, differential, spin-exchange, and spin-flip cross sections. The basic method has been described in detail in a recent paper.[7] We present here a brief summary of the recoil technique and a detailed account of the application of this method to the measurement of differential elastic and inelastic cross sections.

The technique utilizes certain of the kinematic properties of the recoiling atoms in an electron-atom crossed-beam configuration, to distinguish between collisions involving elastic scattering and excitation to various atomic levels. In the recoil method, observations are made on the scattered atoms and the determination of absolute cross sections requires a measurement of the ratio of the scattered to the unscattered atom-beam currents. Since both quantities are measured with the same detector, the detector efficiency does not enter into