Angular distribution and polarization of atomic radiative emission in electric and magnetic fields

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A density-matrix approach has been developed for the angular distribution and polarization of radiative emission during single-photon atomic transitions for a general set of steady-state excitation processes in an arbitrary arrangement of static (or quasistatic) electric and magnetic fields. Particular attention has been directed at spectroscopic observations in the intense fields of the high-power ion diodes on the Particle Beam Fusion Accelerator II (PBFA II) and SABRE devices at Sandia National Laboratories and at magnetic-field measurements in tokamak plasmas. The field-dependent atomic eigenstates are represented as expansions in a complete basis set of field-free bound and continuum eigenstates. Particular emphasis has been given to directed-electron collisional excitations, which may be produced by an anisotropic incident-electron velocity distribution. We have allowed for the possibility of the coherent excitation of the nearly degenerate field-dependent atomic substates, which can give rise to a complex spectral pattern of overlapping Stark-Zeeman components. Coherent excitations may be produced by a beam of electrons that are spin-polarized at an angle with respect to the propagation direction or by nonparallel electric and magnetic fields. Our main result is a general expression for the matrix elements of the photon-polarization density operator representing the total intensity, angular distribution, and polarization of the atomic radiative emission. For the observation of radiative emission in the direction of the magnetic field, the detection of linearly polarized emission, in addition to the usual circularly polarized radiation, can reveal the presence of a perpendicular electric field or a coherent excitation mechanism. [S1050-2947(99)07108-5]

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I. INTRODUCTION

Polarized radiative emission can occur as a result of a nonuniform or nonstatistical distribution of the population densities of excited atoms (or ions) among their degenerate (or nearly degenerate) angular-momentum magnetic sublevels. A nonuniform magnetic-sublevel distribution can be produced by directed electron- or ion-beam excitation, photon (especially laser-photon) excitation, or the action of electric or magnetic fields. Polarized radiative emission is anisotropic, giving rise to an angular dependence of the photon emission. The measurement and analysis of the spectral intensity, angular distribution, and polarization of the radiative emission can provide detailed information on the relative importance of the elementary excitation mechanisms and on the amplitudes and directions of the electric and magnetic fields. Using a density-matrix approach together with angular-momentum algebra techniques, we have developed a comprehensive theoretical framework for the analysis of polarized radiative emission from many-electron atomic systems undergoing a general set of steady-state excitation processes in the presence of an arbitrary arrangement of static (or quasistatic) electric and magnetic fields. It has been necessary to allow for the possibility of the coherent excitation of the degenerate (or nearly degenerate) sublevels. A brief description of our density-matrix formulation has been presented by Jacobs and Filuk [1]. In this paper, a more extensive and detailed account has been provided.

A. Spectroscopic observations

We are especially concerned with radiative emission from atomic systems in the presence of electric and magnetic fields. We are particularly motivated by the important spectroscopic observations that have been made in the intense perpendicular (crossed) electric and magnetic fields of pulsed-power applied-magnetic-field ion diodes, such as those in operation at the Particle Beam Fusion Accelerator II facility PBFA II [2,3] and at the SABRE facility [4] developed at Sandia National Laboratories. In these devices, the strength of the externally applied magnetic field has been adjusted to a value in the range of from 2 to 3 T. However, magnetic-field strengths above 10 T (100 KG) can be generated. From time-resolved and space-resolved measurements of the Stark shift of the Li I $2p \rightarrow 2s$ radiative emission from the anode-cathode (acceleration-gap) region of the applied-B ion diode, Bailey *et al.* [5] have deduced quasistatic accelerating electric-field strengths that are as high as 10^9 V/m. This radiative emission has been further analyzed by Filuk *et al.* [6] to provide significant information on the

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roles of various collisional excitation processes and spectral-line-broadening mechanisms. In order to provide a precise interpretation of these spectroscopic observations, it will be necessary to carry out reliable calculations for the relevant Stark-Zeeman spectral patterns and intensities. Separate calculations should be performed for different assumptions regarding the basic excitation mechanisms, which may include collisional excitation by both electrons and ions. In our density-matrix description of atomic radiative emission, we have allowed for an arbitrary arrangement of static (or quasistatic) electric and magnetic fields and for a general set of steady-state excitation mechanisms. We have attempted to identify the more detailed information that could be derived from an analysis of the angular distribution and polarization, in addition to the usual spectral intensity.

Important spectroscopic measurements have also been conducted in tokamak plasmas. The analysis of the circular polarization of radiative emission during magnetic-dipole transitions in heavy atomic ions has been proposed as a method for the spectroscopic determination of the internally generated (poloidal) magnetic field [7]. This field is produced by the toroidally circulating, Ohmic-heating current. It is perpendicular to the much stronger, externally applied (toroidal) magnetic field, which may have a strength in the range of from 1 to 10 T. The circular polarization of atomic radiative emission in magnetic-dipole transitions has been treated theoretically by Jacobs and Seely [8]. However, the effects of the Lorentz (or motional-Stark) electric field, which is generated in a direction perpendicular to the combined or total magnetic field, were not considered. Huang *et al.* [9] have treated theoretically the circular polarization of radiative transitions involving highly excited states of hydrogenlike ions, allowing for both the Zeeman and motional-Stark effects. Moreover, they have taken into account the individual spectral-line profiles due to thermal Doppler broadening and motional-Stark broadening. The neglect of the motional-Stark broadening is found to lead to a substantial simplification in the analysis of the magnetic-field measurements. By means of our density-matrix formulation, the effects of electric and magnetic fields can be treated on an equal footing and in a systematic manner.

Our density-matrix formulation may be necessary for a precise description of the influence of electric and magnetic fields on the resonant electron-ion photorecombination process known as dielectronic recombination. The results of theoretical investigations by Jacobs, Davis, and Kepple [10] and by Jacobs and Davis [11] have indicated that the rate of photon emission from high-Rydberg doubly excited autoionizing states (formed by radiationless electron capture) can be significantly enhanced in the presence of a uniform, static (or quasistatic) electric field. In a recent electron-ion–beam observation of this electric-field-induced enhancement by Savin *et al.* [12], a magnetic field was applied coaxially with the electron beam and a Lorentz electric field was produced in the rest frame of the emitting ions. In order to simplify the comparison with the existing theoretical predictions, the time evolution of the electric field was not taken into account and the anisotropic nature of the radiative emission was ignored. Using our density-matrix description, the time variation of the motional electric field could be taken into account (in the quasistatic approximation), together with the nonspherical angular distribution of the polarized radiative emission. In a recent theoretical investigation by Robicheaux and Pindzola [13], the dielectronic-recombination cross section was evaluated taking into account the presence of perpendicular static electric and magnetic fields, and an additional enhancement was predicted with the inclusion of the magnetic field. However, the angular distribution and polarization of the photon emission were not considered. Our density-matrix formulation can provide the framework for a comprehensive description of field-induced dielectronic recombination, which could facilitate the resolution of the disagreement between existing theoretical predictions and a recent observation [14].

Polarized x-ray line emission from highly ionized atomic systems has been observed on the Electron Beam Ion Trap (EBIT) facility at the Lawrence Livermore National Laboratory by Beirersdorfer *et al.* [15] and also on a similar EBIT device at the National Institute of Standards and Technology by Takács *et al.* [16]. This polarized atomic radiative emission is predominately attributable to directed-electron collisional excitation processes occurring in the absence of electric and magnetic fields. The angular distribution and polarization measurements can provide detailed information on the elementary collisional interactions involving the individual magnetic sublevels of the atomic system, which would usually be unobtainable in a plasma observation. The nearly monoenergetic electron-beam distribution enables the observation of weak radiative emission processes involving forbidden transitions or narrow autoionizing resonances, which might otherwise be unobservable. External electric and magnetic fields can be expected to play an important role, even for multiply charged ions, in processes involving high-Rydberg atomic states.

B. Influence of directed excitation mechanisms

The theory of polarized atomic radiative emission following directed-electron collisional excitation was first developed by Oppenheimer [17] and subsequently extended by Percival and Seaton [18]. More recently, Inal and Dubau [19] have developed a density-matrix description for polarized x-ray line emission following directed-electron collisional excitation. Inal and Dubau [19] were primarily concerned with collisional excitation by electrons (in a high-temperature plasma) with an anisotropic distribution of velocities, and they did not consider the influence of electric and magnetic fields. Density-matrix descriptions for the polarization of the radiative emission in field-free dielectronic recombination have been presented by Inal and Dubau [20] and by Shlyaptseva and co-workers [21,22]. Our density-matrix formulation of polarized atomic radiative emission in electric and magnetic fields is a natural generalization of these earlier density-matrix descriptions.

Directed excitation can also occur during the interaction of atomic systems with coherent electromagnetic (laser) radiation. The density-matrix formulation presented in this paper is closely derived from the density-matrix descriptions that we have developed for the angular distribution and polarization of photoelectrons following single-photon ionization processes [23] and multiphoton ionization processes [24] in many-electron atoms. Adopting the formalism of Fano [25] and of Fano and Racah [26], the density operators

were represented as expansions in a basis of irreducible spherical-tensor operators. The irreducible spherical-tensor expansion coefficients or components of the final-state density operator were expressed in terms of the irreducible spherical-tensor components of the initial-state density operators (representing the initial atoms and incident photons) and the reduced matrix elements describing the atomic photoelectric transition. In this manner, a natural separation between the geometrical and dynamical factors was achieved.

We also investigated the single-photon ionization process [23] as a mechanism for the nonuniform population of the individual angular-momentum magnetic sublevels of an excited state of the residual ion. The polarization of the subsequently emitted electric-dipole decay radiation was evaluated with the assumption that the initial photon-excitation and subsequent radiative-emission processes could be treated as independent events. However, allowance was made for the possibility of the coherent excitation of the individual magnetic sublevels. The superposition of substates produced by a coherent excitation process can be represented by a density matrix that has nonzero nondiagonal elements, in addition to the diagonal elements corresponding to the familiar level-population densities. Our theory for photoionization measurements is in many respects analogous to the theory for photon–scattered-particle coincidence observations presented by Macek and Jaecks [27].

In the present investigation, the density-operator formulation and the angular-momentum algebra techniques have been employed to treat the angular distribution and polarization of atomic radiative emission in static (or quasistatic) electric and magnetic fields. Allowance has been made for a general set of steady-state atomic excitation processes, which are capable of producing a nonuniform distribution of population densities among the individual angular-momentum magnetic sublevels of the excited atomic state. We have also allowed for the possibility of the coherent excitation of the degenerate (or nearly degenerate) sublevels. The introduction of coherences or correlations can be regarded as an inevitable consequence of the general density-matrix description of interacting quantum-mechanical systems.

C. Effects of electric and magnetic fields

In electric or magnetic fields, normally overlapping magnetic components of atomic spectral lines may be substantially shifted from their field-free positions and split into spectroscopically resolvable features. Due to the breakdown of the field-free angular-momentum and parity selection rules, otherwise forbidden components can be generated. Spectral patterns and polarizations produced by either a uniform static electric or magnetic field have been extensively investigated, and the individual Stark and Zeeman effects have been treated in both the weak-field (perturbative) and strong-field (nonperturbative) regimes. More complex Stark-Zeeman patterns have been investigated primarily for simple atomic systems, including hydrogenlike or high-Rydberg atoms, and for special applied-field configurations, including parallel and perpendicular electric and magnetic fields. The energy-level structure of atomic hydrogen in perpendicular (crossed) electric and magnetic fields was investigated by Pauli [28]. Energy-level structures and photoabsorption spectra for high-Rydberg states of nonhydrogenic atoms in perpendicular electric and magnetic fields have been treated by Rao and Taylor [29]. Spectra for high-Rydberg bound states of sodium atoms in perpendicular electric and magnetic fields have been investigated by Korevaar and Littman [30], while continuous photoabsorption (photoionization) spectra of lithium atoms in crossed electric and magnetic fields have been studied by Crosswhite *et al.* [31]. McGuire [32] has calculated Stark-Zeeman energy-level structures and spectral-line shifts, as well as radiative transition probabilities, for alkali-metal atoms in uniform static electric and magnetic fields with arbitrary directions.

In our density-matrix formulation, we shall assume that the energy eigenvalues and eigenstates of the many-electron atomic system have been determined in the presence of an arbitrary arrangement of uniform static (or quasistatic) electric and magnetic fields. The field-dependent atomic eigenstates will be represented as expansions in a field-free (angular-momentum-and-parity) basis set, consisting of bound and continuum eigenstates. The emitted-photon and atomic-system density operators can be represented in terms of their irreducible spherical-tensor components [25,26]. However, the irreducible spherical-tensor representation is not a requirement and may not be advantageous for an arrangement of electric and magnetic fields with neither spherical nor axial symmetry. The total (frequency-integrated) photon intensities, angular distributions, and polarizations of the individual Stark-Zeeman components are evaluated for spontaneous single-photon transitions, treating the electromagnetic interaction in the lowest nonvanishing order of quantum-electrodynamical perturbation theory. The detailed (frequency-dependent) spectral-line shapes can be investigated within the framework of the Liouville-space resolvent-operator version of our density-matrix formulation [33–35], where the elementary collisional and radiative broadening mechanisms are described (in a nonperturbative manner) by means of self-energy-operator corrections.

In the theory of spectral-line broadening by plasmas [36,37], it is a common practice to treat slowly varying (e.g., ion-produced) electric microfields and magnetic fields in the quasistatic approximation. Sarid, Maron, and Troyansky [38] investigated the individual polarization components of hydrogen lines emitted in an applied-B ion-diode plasma under the influence of fluctuating (collective) anisotropic electric fields, which they treated in the quasistatic approximation. In an investigation of the influence of a strong, static uniform magnetic field on the broadening of helium lines produced by a plasma, Deutsch [39] evaluated the spectral patterns produced by the combined static Stark and Zeeman effects. He employed the generalized impact theory of Griem *et al.* [40] to treat the additional broadening of the quasistatic Stark-Zeeman components due to collisions with the plasma electrons, which are assumed to be completed in a relatively short time. Using the quasistatic approximation for the line wings and the impact approximation for the line center, Savchenko and Fisch [41] evaluated the frequency-dependent, polarized atomic radiative emission due to laser-photon excitation. Adopting a density-matrix description for the combined atom and radiation-field system, they determined the atomic-state coherences associated with the different magnetic sublevels. The polarization at the line center,

which corresponds to relatively long radiation times, was found to be reduced due to the effects of relaxation processes, which tend to destroy the atomic-state coherences.

Transition probabilities (or cross sections) for elementary atomic radiative and collisional interactions can be substantially altered by electric and magnetic fields. Consequently, atomic-level population densities can be appreciably modified. Laming [42] investigated radiative emission from hydrogenic ions with a nonstatistical distribution of exited-level population densities produced by directed-electron capture into Stark eigenstates in a strong electric field. Demkin [43] treated coherent atomic excitation in a strong electric field. In our density-matrix description of collisional and radiative interactions in the presence of electric and magnetic fields, the atomic-level population densities (or diagonal density-matrix elements) and the atomic-state coherences (or nondiagonal elements) can be determined self-consistently. In the traditional collisional-radiative model for atomic processes in a plasma [44], only the population densities are determined and only the field-free atomic transition rates are normally employed. In the conventional description of spectral-line broadening in plasmas [36], the electron densities are often assumed to be sufficiently high for the establishment of local-thermodynamic-equilibrium (LTE) population densities, and any atomic-state coherences are assumed to be destroyed. In the single-particle approximation of plasma kinetic theory [45], electric and magnetic fields are frequently included in the (Boltzmann or Vlasov) equation for the nonequilibrium electron-velocity distribution function. For a comprehensive description of atomic processes in a plasma, it is necessary to provide a self-consistent treatment of the effects of electric and magnetic fields on the atomic-state and plasma-electron kinetics.

D. Organization of this paper

The remainder of this paper has been organized in the following manner: Our density-matrix description is presented in Sec. II. Our main result is an expression for the photon-polarization density-matrix elements, which determines the total intensities, angular distributions, and polarizations for single-photon spontaneous emission from a many-electron atomic system in the presence of an arbitrary arrangement of uniform static (or quasistatic) electric and magnetic fields. We have allowed for a general steady-state balance of collisional and radiative excitation and deexcitation processes. The coherent excitation of the degenerate (or nearly degenerate) field-dependent atomic substates can be taken into account. Our results for the polarization emission observed in the magnetic-field direction are presented in Sec. III. Finally, a summary and our conclusions are given in Sec. IV. Subjects of future investigations include field-dependent atomic eigenstate expansions, coherent atomic-state kinetics, and polarized spectral-line shapes.

II. DENSITY-MATRIX DESCRIPTION

We describe atomic radiative emission by means of the density-matrix formalism [46–48]. The excitation and spontaneous radiative emission processes will be treated as independent events. The initial excited atomic states in the presence of electric and magnetic fields will be represented by the density operator $\rho_i(t=0)$, with diagonal matrix elements giving the level population densities and nondiagonal elements corresponding to the coherences. At an arbitrary time $t \geqslant 0$ the final states of the (entangled) atom and radiation-field system can be represented by the density operator $\rho_f(t)$. Our basic starting point is the relationship $\rho_f(t) = U(t)\rho_i(t=0)U(t)^\dagger$, where $U(t)$ is the time evolution operator. In our time-independent or steady-state description the asymptotic final-state density operator ρ_f (at $t=+\infty$) is given by $\rho_f = S\rho_i S^\dagger$, where S denotes the scattering operator describing the electromagnetic interaction. The frequency-integrated photon intensities, angular distributions, and polarizations may be evaluated in the lowest nonvanishing order of quantum-electrodynamical perturbation theory [49]. The corresponding frequency-dependent properties should be investigated by means of a higher-order (or nonperturbative) density-matrix formulation [35,50]. The reduced photon-polarization density operator ρ^R is defined as an average (trace) of the full asymptotic final-state density operator ρ_f over the quantum states of the decayed atomic system.

A. Photon density matrix in lowest-order perturbation theory

The steady-state frequency-integrated intensity, angular distribution, and polarization of the radiation emitted in the atomic transition $\gamma_i \rightarrow \gamma_f$ can be expressed in terms of the matrix elements of the reduced photon-polarization density operator ρ^R. In lowest-order perturbation theory, these density-matrix elements are given by

$$\langle \lambda|\rho^R|\lambda'\rangle = \sum_{f,i,i'}{}' \langle \gamma_f,\vec{k}\lambda|V|\gamma_i,0\rangle\langle \gamma_i|\rho^A|\gamma_{i'}\rangle \times \langle \gamma_{i'},0|V|\gamma_f,\vec{k}\lambda'\rangle. \quad (1)$$

The operator V represents the entire interaction between the many-electron atomic system and the quantized electromagnetic field. The angular frequency ω of the observed radiation is given in terms of the magnitude of the photon wave vector k by means of the free-space relation $\omega = kc$. The photon helicity quantum numbers, which represent the projections of the intrinsic spin of the spin-1 quanta along the propagation vector $\vec{k}$, may have only the values λ, $\lambda' = \pm 1$, corresponding to right and left circular polarization. The restricted summations over f, i, and i' (indicated by the prime above the summation symbol) include only the quantum numbers specifying degenerate or nearly degenerate sublevels of the field-dependent final and initial atomic states. When field effects are neglected, the degenerate sublevels can be specified in terms of the usual angular-momentum projection (or magnetic) quantum numbers. In the presence of perpendicular (crossed) electric and magnetic fields, for which there is only a twofold degeneracy associated with a plane of reflection symmetry, the magnetic substates are no longer exactly degenerate. The description of single-photon emission processes based on Eq. (1) is appropriate for narrow, isolated emission lines or blended emission features, whose individual spectral-line profiles may not be resolvable.

We shall assume that the reduced initial-state atomic density matrix ρ^A has been obtained from a steady-state master

equation of the form $R\rho^A=0$. The Liouville-space relaxation operator R [47] (whose matrix elements are specified by four indices) describes, in the Markov (or short-memory-time) approximation, the influence of the elementary collisional and radiative interactions in the presence of electric and magnetic fields. In the equation for the diagonal matrix elements of ρ^A (corresponding to the level populations), the tetradic matrix elements of R represent the familiar (time-independent) rates for all possible collisional and radiative transitions between pairs of field-dependent atomic levels. The electron collision rates can be evaluated in terms of the electron-impact excitation or deexcitation cross sections and the single-electron velocity distribution function, while the radiative contributions may include both the spontaneous radiative emission rates and the radiation-intensity-dependent absorption or induced emission rates. The collisional and radiative relaxation processes can be most systematically treated within the framework of an open-system reduced-density-matrix description [47], starting with the equation of motion for the density operator representing the enlarged system consisting of the relevant atomic system together with the quantized-electromagnetic-field and charged-particle environment.

B. Field-free electronic eigenstate representation

We will assume that the field-dependent initial atomic eigenstates $|\gamma_i\rangle$ can be expanded in a basis set of field-free angular-momentum eigenstates $|\Delta_i J_i M_i\rangle$ as follows:

$$|\gamma_i\rangle=\sum_{\Delta_i J_i M_i}|\Delta_i J_i M_i\rangle\langle\Delta_i J_i M_i|\gamma_i\rangle. \tag{2}$$

The field-dependent final atomic eigenstates $|\gamma_f\rangle$ will also be represented by an expansion in the same form as Eq. (2). Here J_i is the total electronic angular momentum and M_i is the projection along the atomic quantization axis. The symbol Δ_i denotes the set of remaining quantum numbers required to completely specify the field-free many-electron atomic states, including the principal quantum number and parity. Hyperfine structure will be ignored. The complete basis set contains both bound and continuum field-free atomic eigenstates. In weak-field atomic-structure calculations, the field-induced mixing of only a relatively small set of low-lying field-free bound eigenstates is normally considered. For an accurate determination of the shifts and splittings of the spectral patterns in a strong field, highly excited bound and continuum field-free atomic eigenstates must be taken into account [51].

The many-electron atomic Hamiltonian operator H^A can be expressed in the form $H^A=H^A(0)+H^A(E)+H^A(B)$, where $H^A(0)$ describes the isolated, field-free atomic system and $H^A(E)$ and $H^A(B)$ represent the electric-field and magnetic-field interactions, respectively. We shall assume that the field-dependent eigenvalues and eigenstates of H^A have been determined as expansions in the field-free angular-momentum representation, employing a relativistic multiconfiguration atomic-structure calculation. For sufficiently weak fields, a perturbation-theory analysis may provide a preliminary approximation for the complex Stark-Zeeman spectral patterns. The Zeeman-effect spectral patterns, which are produced by a weak magnetic field acting alone, are characterized by the (conserved) total electronic angular-momentum projections M_i and M_f. For a magnetic field sufficiently strong to uncouple the orbital and spin angular momenta, the Paschen-Back representation may be more appropriate. In either magnetic-field regime, a preliminary perturbation-theory analysis may be used to investigate the influence of a relatively weak electric field on the Zeeman patterns.

C. Multipole expansion of the electromagnetic interaction

The photon polarization, which is most naturally defined with respect to the propagation direction (along $\vec{k}$), can be related to the atomic quantization axis by expanding the electromagnetic-interaction matrix elements (connecting the field-free eigenstates) in terms of the matrix elements of the electric and magnetic multipole-moment operators. The photon eigenstate representation characterized by linear momentum and intrinsic spin is thereby replaced by the alternative representation based on angular momentum and parity. This transformation may be expressed by means of the expansion [49]

$$\begin{aligned}\langle\Delta_f J_f M_f,\vec{k}\lambda|V|\Delta_i J_i M_i,0\rangle&\\ =\sum_j\sum_m\left(\frac{2j+1}{4\pi}\right)^{1/2}A(j)D^{(j)}_{\lambda m}(\hat{k})&\\ \times(-1)^m\langle\Delta_f J_f M_f|Q^{(j)}_{-m}|\Delta_i J_i M_i\rangle.&\end{aligned} \tag{3}$$

Here $Q^{(j)}_m$ denotes the irreducible spherical-tensor form of the multipole-moment operator associated with the photon angular momentum j and projection quantum number m, and $D^{(j)}_{\lambda m}(\hat{k})$ designates the Wigner rotation-matrix element $D^{(j)}_{\lambda m}(\varphi,\theta,0)$. In order to include both the electric and the magnetic multipole contributions corresponding to a given value of j, the multipole-moment operator $Q^{(j)}_m$ should be taken as a sum of the individual contributions corresponding to the permissible values of the photon parity. The overall multiplying factor $A(j)$ depends on the particular type of multipole radiation. The field-free matrix elements of $Q^{(j)}_m$ can be evaluated, in terms of the Wigner 3-j symbols and the reduced matrix elements $(\Delta_f J_f\|Q^{(j)}\|\Delta_i J_i)$, by means of the Wigner-Eckart theorem [52]

$$\begin{aligned}\langle\Delta_f J_f M_f|Q^{(j)}_m|\Delta_i J_i M_i\rangle=(-1)^{J_f-M_f}\begin{pmatrix}J_f & j & J_i\\ -M_f & m & M_i\end{pmatrix}&\\ \times(\Delta_f J_f\|Q^{(j)}\|\Delta_i J_i).&\end{aligned} \tag{4}$$

In contrast with the photon helicity λ, which can have only the values ±1, the angular-momentum projection m is noted limited to ±1. The symmetry (or angular-momentum conservation) information is completely represented by the 3-j symbols, while the dynamical information is entirely contained in the reduced matrix elements.

Although our general formulation is applicable to an arbitrary arrangement of static (or quasistatic) electric and

magnetic fields, we will be particularly concerned with perpendicular (crossed) electric and magnetic fields. The geometry for spectroscopic observations in crossed electric and magnetic fields is illustrated in Fig. 1. The total magnetic field is often the sum of an applied (external) magnetic field and a dynamical (internal) magnetic field. In PBFA II or SABRE, a dynamical magnetic field (nearly parallel to the applied magnetic field) is generated in the anode-cathode region of the applied-B ion diode. In a tokamak plasma, a dynamical (poloidal) magnetic field is generated perpendicular to the applied (toroidal) magnetic field. The viewing angle θ is parallel to the applied magnetic field in PBFA II and SABRE and perpendicular to the applied (toroidal) magnetic field in a tokamak plasma. In the EBIT facility at the Lawrence Livermore National Laboratory, the emitted photons are detected at an angle θ perpendicular to the (parallel) magnetic-field and electron-beam directions.

D. Photon-polarization density matrix allowing for coherent excitation processes in a general arrangement of electric and magnetic fields

The product of two Wigner rotation matrices, of rank j and j', can be expressed as a summation of Wigner rotation matrices corresponding to the total angular momentum J, each multiplied by two new 3-j symbols. The matrix elements of the photon-polarization density operator ρ^R, which are defined by Eq. (1), can then be expressed in the form

$$\begin{aligned}\langle\lambda|\rho^R|\lambda'\rangle=&\sum_{i,i',f}{}' \langle\gamma_i|\rho^A|\gamma_{i'}\rangle\sum_{\Delta_f J_f M_f}\sum_{\Delta_f' J_f' M_f'}\langle\gamma_f|\Delta_f J_f M_f\rangle\langle\gamma_f|\Delta_f' J_f' M_f'\rangle^*\sum_{\Delta_i J_i M_i}\sum_{\Delta_i' J_i' M_i'}\langle\Delta_i J_i M_i|\gamma_i\rangle\\ &\times\langle\Delta_i' J_i' M_i'|\gamma_{i'}\rangle^*\sum_{j,m}\sum_{j',m'}\sum_{J,M,M'}(\Delta_f J_f\|Q^{(j)}\|\Delta_i J_i)(\Delta_f' J_f'\|Q^{(j')}\|\Delta_i' J_i')^*\left(\frac{1}{4\pi}\right)(2j+1)^{1/2}(2j'+1)^{1/2}(2J+1)\\ &\times A(j)A(j')^*(-1)^{J_f+J_f'-M_f-M_f'-m-m'+\lambda'-m'+2j-2j'-M-M'}\begin{pmatrix}J_f & j & J_i\\ -M_f & -m & M_i\end{pmatrix}\begin{pmatrix}J_f' & j' & J_i'\\ -M_f' & -m' & M_i'\end{pmatrix}\\ &\times\begin{pmatrix}j & j' & J\\ \lambda & -\lambda' & -M\end{pmatrix}\begin{pmatrix}j & j' & J\\ m & -m' & -M\end{pmatrix}D^{(J)}{}_{MM'}(\hat{k}).\end{aligned}\tag{5}$$

The summations over f, i, and i' include only the quantum numbers specifying degenerate or nearly degenerate field-dependent atomic sublevels.

Equation (5) is valid for any single-photon emission process from a many-electron atomic system under the influence of a general steady-state balance of elementary collisional and radiative excitation and deexcitation processes in an arbitrary arrangement of static (or quasistatic) electric and magnetic fields. The dependence on the electric and magnetic fields, which may be quite complex and not expressible in a simple analytical form, is introduced by the field transformation or mixing coefficients $\langle\Delta_i J_i M_i|\gamma_i\rangle$ and $\langle\gamma_f|\Delta_f J_f M_f\rangle$ and by the initial-state atomic density-matrix elements $\langle\gamma_i|\rho^A|\gamma_{i'}\rangle$. The nondiagonal elements of ρ^A describe coherent excitation of the field-dependent initial-state atomic sublevels. Coherences between atomic levels with different energy eigenvalues, which can be produced by short-pulse laser-photon excitation, generate quantum beats in a time-resolved photon-detection process. With the incorporation of relaxation processes [47], the time-dependent density-matrix description provides a rigorous starting point for the introduction of the time-integrated, reduced photon density matrix. The coherences for excited atomic states with different energy eigenvalues are found to be important when the energy-level separations are small in comparison with the spectral linewidths.

E. Photon-polarization density matrix in the absence of coherent excitation processes and electric and magnetic fields

If we neglect all effects of the field-induced mixing of the initial and final atomic eigenstates, as well as the coherences corresponding to the nondiagonal matrix elements of the initial-state atomic density operator ρ^A, the general expression for the photon-polarization density-matrix elements given by Eq. (5) can be reduced to the form

$$\begin{aligned}\langle\lambda|\rho^R|\lambda'\rangle=&\sum_{M_i}N(\Delta_i J_i M_i)\sum_{j,j',J}(-1)^{J_f+j+j'+\lambda'-M_i}\\ &\times[(2j+1)(2j'+1)(2J+1)/4\pi]^{1/2}\\ &\times A(j)A(j')^*(\Delta_f J_f l\|Q^{(j)}\|\Delta_i J_i)\\ &\times(\Delta_f J_f\|Q^{(j')}\|\Delta_i J_i)^*\begin{pmatrix}J & j & j'\\ \lambda'-\lambda & \lambda & -\lambda'\end{pmatrix}\\ &\times\begin{pmatrix}J & J_i & J_i\\ 0 & -M_i & M_i\end{pmatrix}\begin{Bmatrix}J & J_i & J_i\\ J_f & j' & j\end{Bmatrix}Y^{(J)}{}_{\lambda'-\lambda}(\hat{k}).\end{aligned}\tag{6}$$

The diagonal matrix elements of ρ^A, which correspond to the usual level population densities, are now denoted by $N(\Delta_i J_i M_i)$. The condition $M'=0$ follows from neglecting the nondiagonal matrix elements of ρ^A, together with ne-

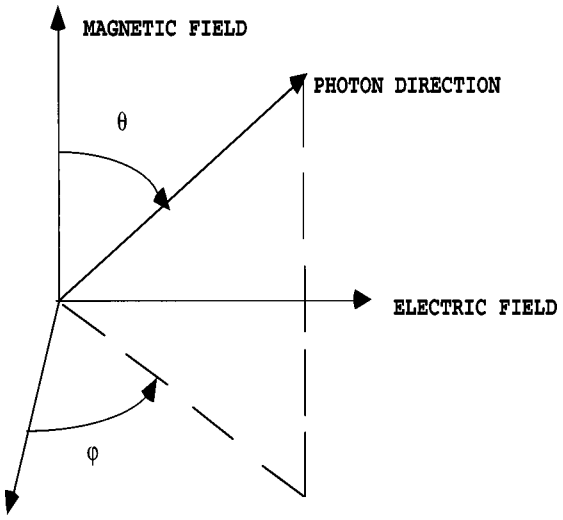

FIG. 1. Viewing angles for spectroscopic observations of polarized atomic radiative emission in perpendicular (crossed) electric and magnetic fields.

glecting the field-induced mixing of the upper, initial-state sublevels. The Wigner rotation matrix elements $D^{(J)}_{MM'}(\hat{k})$ can then be reduced to the spherical harmonic functions $Y^{(J)}_{\lambda'-\lambda}(\hat{k})$, for which the angular momentum J can now assume only integer values. The 6-j symbol is introduced after neglecting the field-induced mixing of the lower, final-state sublevels, which is expected to be valid for low-lying bound levels. The dominant radiative emission process is usually assumed to involve only a single multipole component of the electromagnetic field, in which case $j=j'$. Equations (5) and (6) are valid for interfering multipole components, corresponding to $j\neq j'$. This interference can have a more important effect on the angular distribution and polarization than on the total photon intensity.

The simplified expression given by Eq. (6) is in agreement with the result obtained by Inal and Dubau [19], who investigated directed electron-beam excitation of polarized radiative emission from atomic systems in the absence of electric and magnetic fields. Only even values of J can contribute for excitation by a beam of unpolarized electrons, due to the reflection symmetry in the plane perpendicular to the atomic quantization axis (which is taken as the electron-beam propagation direction). The usual axial symmetry can be broken in the coherent excitation of the magnetic sublevels by a spin-polarized electron beam for which the spin projection is defined at an angle with respect to the atomic quantization axis [53]. The axial symmetry can also be broken during atomic excitation in perpendicular (crossed) electric and magnetic fields, due to coherent excitation of the magnetic sublevels.

F. Irreducible spherical-tensor representation of the density operators

The reduced photon-polarization density operator may be expanded, in terms of the complete set of irreducible spherical-tensor operators [25,26,46,54,55], in the form

$$\rho^R=\sum_{j,j'}\sum_{J,M,M'}\rho^R(j,j';J,M')T^{(J)}{}_M(j,j')D^{(J)}{}_{MM'}(\hat{k}). \tag{7}$$

In the Liouville-space Dirac notation, this expansion corresponds to a transformation from the representation of the uncoupled states $|jm,j'm'\rangle\rangle$ to the representation of the coupled states $|j,j';J,M\rangle\rangle$. The photon-helicity quantum numbers are defined, with respect to the photon propagation direction, by means of the Wigner rotation matrices $D^{(J)}_{MM'}(\hat{k})$. The ordinary Hilbert-space matrix elements of the irreducible spherical-tensor operator $T^{(J)}_M(j,j')$ can be evaluated using the Wigner-Eckart theorem

$$\langle j\lambda|T^{(J)}{}_M(j,j')|j'\lambda'\rangle=(-1)^{j-\lambda}\times(2J+1)^{1/2}\begin{pmatrix} j & J & j' \\ -\lambda & M & \lambda' \end{pmatrix}. \tag{8}$$

The reduced matrix element of $T^{(J)}_M(j,j')$ is given by the factor $(2J+1)^{1/2}$. The general expression for the irreducible spherical-tensor components $\rho^R(j,j';J,M)$ or state multipoles can be obtained by comparing Eqs. (5) and (7), employing Eq. (8) and the symmetry properties of the 3-j symbols. This irreducible spherical-tensor-operator representation is often advantageous, because only a few multipole components of the electromagnetic field are usually required in the description of an atomic radiative-emission process.

The density operator ρ^A, representing the field-dependent initial excited atomic states, may be expanded in the basis set of irreducible spherical-tensor operators $T^{(K)}_N(J_iJ_i')$ in the form

$$\rho^A=\sum_{\Delta_i\Delta_i'}\sum_{J_iJ_i'}\sum_{K,N}\rho^A(\Delta_i\Delta_i'J_iJ_i';K,N)T^{(K)}{}_N(J_iJ_i'). \tag{9}$$

In order to express the irreducible spherical-tensor components of the photon-polarization density operator in terms of the atomic irreducible spherical-tensor components $\rho^A(\Delta_i\Delta_i'J_iJ_i';K,N)$, it is necessary to introduce into Eq. (5) the transformation

$$\langle\gamma_i|\rho^A|\gamma_{i'}\rangle=\sum_{\Delta_iJ_iM_i}\sum_{\Delta_i'J_i'M_i'}\langle\Delta_iJ_iM_i|\gamma_i\rangle^*\langle\Delta_i'J_i'M_i'|\gamma_{i'}\rangle\times\sum_{K,N}\rho^A(\Delta_i\Delta_i'J_iJ_i';K,N)(-1)^{J_i-M_i}\times(2K+1)^{1/2}\begin{pmatrix} J_i & K & J_i' \\ -M_i & N & M_i' \end{pmatrix}. \tag{10}$$

While the irreducible spherical-tensor expansion of the photon-polarization density-operator usually involves only a relatively small set of electromagnetic multipole contributions, the corresponding expansion of the field-dependent atomic density operator contains two additional summations over the complete set of unperturbed, field-free angular-momentum eigenstates. Equation (10) could be advantageous in a general arrangement of relatively weak electric and mag-

netic fields, for which only manageably small sets of unperturbed initial and final atomic substates are appreciably mixed, or in parallel electric and magnetic fields, for which the axial symmetry can be exploited. The simplified density-matrix representation for axially symmetric systems has been based on the concepts of orientation (corresponding to odd values K) and alignment (involving even, nonzero, values of K) [56,57].

If field-induced mixing of atomic eigenstates and atomic-state coherences are neglected, the irreducible spherical-tensor components of the photon-polarization density operator can be simply related to the irreducible spherical-tensor components of the initial-state atomic density operator as follows:

$$\begin{aligned}\rho^R(j,j';J,0) = {} & \rho^A(\Delta_i\Delta_i J_i J_i;J,0)(-1)^{J_i+J_f+J+j+j'} \\ & \times(1/4\pi)[(2j+1)(2j'+1)]^{1/2}A(j)A(j')^* \\ & \times(\Delta_f J_f\|Q^{(j)}\|\Delta_i J_i)(\Delta_f J_f\|Q^{(j')}\|\Delta_i J_i)^* \\ & \times\begin{Bmatrix} J & J_i & J_i \\ J_f & j' & j \end{Bmatrix}, \end{aligned} \tag{11}$$

which is equivalent to Eq. (6). Coherences between initial atomic states with different magnetic quantum numbers can be taken into account only by including the irreducible spherical-tensor components $\rho^A(\Delta_i\Delta_i J_i J_i;K,N)$ with $N\neq 0$.

For systems with spherical or axial symmetry, the steady-state master equation $R\rho^A=0$ governing the atomic density operator can be advantageously reexpressed in the irreducible spherical-tensor representation [56,57]. The steady-state equation may be solved, taking into account the relevant set of elementary atomic collisional and radiative relaxation processes in the presence of the electric and magnetic fields. The spherical-tensor components $\rho^A(\Delta_i\Delta_i' J_i J_i';K,N)$ thereby obtained could then be transformed into the field-dependent representation by means of Eq. (10). However, the spherical-tensor representation may not retain its characteristic advantage in the presence of a general arrangement of electric and magnetic fields, for which the field-free angular-momentum and parity selections rules may no longer be rigorously valid.

G. Stokes-parameter representation of the photon density operator

The photon-polarization density matrix may be specified, in terms of the Stokes parameters, in the form [47]

$$\rho^R=\left(\frac{I}{2}\right)\begin{pmatrix} 1+\eta_2 & -\eta_3+i\eta_1 \\ -\eta_3-i\eta_1 & 1-\eta_2 \end{pmatrix}. \tag{12}$$

The total intensity I of the emitted electromagnetic radiation is related to the diagonal helicity matrix elements of ρ^R by the normalization condition

$$I=\mathrm{Tr}\rho=\langle 1|\rho^R|1\rangle+\langle -1|\rho^R|-1\rangle. \tag{13}$$

The parameters η_1 and η_3 specify linear polarization, while η_2 represents circular polarization.

In the directed excitation of an atomic system by a beam of electrons that are not spin polarized, axial symmetry is preserved and reflection symmetry is maintained in the plane perpendicular to the quantization axis, which is taken to be the electron-beam direction. Under these conditions, the atomic magnetic sublevels cannot be coherently excited. It follows that the Stokes parameters η_1 and η_2 must vanish. The only nonzero polarization parameter η_3, which is one of the two linear-polarization parameters, can be conveniently defined by means of the relationship

$$\eta_3=(I_\parallel-I_\perp)/(I_\parallel+I_\perp), \tag{14}$$

where $I_\parallel$ and $I_\perp$ refer to the emitted photon intensities with polarization directions parallel and perpendicular, respectively, to the plane that is defined by the photon propagation direction and the atomic quantization axis. In terms of the helicity matrix elements of the photon-polarization density operator ρ^R, this linear-polarization parameter may be expressed in the form

$$\eta_3=\frac{-\langle 1|\rho^R|-1\rangle+\langle -1|\rho^R|1\rangle}{\langle 1|\rho^R|1\rangle+\langle -1|\rho^R|-1\rangle}. \tag{15}$$

The four Stokes parameters I, η_1, η_2, and η_3 can be evaluated, as functions of the emitted photon angles, by means of our general expression for the matrix elements of the photon-polarization density operator presented by Eq. (5), which is applicable to the steady-state coherent excitation of the initial atomic states in an arbitrary arrangement of static (or quasistatic) electric and magnetic fields. Alternatively, by utilizing the expansion given by Eq. (7), the Stokes parameters may be expressed in terms of the irreducible spherical-tensor components of the photon-polarization density operator. However, the irreducible spherical-tensor components of the photon-polarization density operator can be conveniently related to the irreducible spherical-tensor components of the initial-state atomic density operator only for special high-symmetry situations or for sufficiently weak fields.

III. POLARIZATION OF RADIATIVE EMISSION ALONG THE MAGNETIC-FIELD DIRECTION

Due to the limited line-of-sight access to the anode-cathode region of the applied-B ion diode on the PBFA II and SABRE facilities, the polarized atomic radiative emission has been observed only in the direction of the external magnetic field. In tokamak plasmas, the polarized atomic radiative emission has been measured in a direction perpendicular to the known toroidal magnetic field, and parallel to the poloidal magnetic field of interest. For observation along the magnetic-field direction, the main emphasis has been on the determination of the circular-polarization parameter η_2, which can be defined as follows:

$$\eta_2=(I_+-I_-)/(I_++I_-). \tag{16}$$

Here I_+ and I_- denote the intensities of the right and left circularly polarized radiative emissions. The circular-polarization parameter η_2 may be conveniently expressed, in terms of the diagonal matrix elements of the photon-polarization density operator ρ^R, as follows:

$$\eta_2=\frac{\langle 1|\rho^R|1\rangle-\langle -1|\rho^R|1-\rangle}{\langle 1|\rho^R|1\rangle+\langle -1|\rho^R|-1\rangle}. \tag{17}$$

A. Polarization of radiative emission in the presence of perpendicular (crossed) electric and magnetic fields and coherent excitation processes

For spectroscopic observations in the magnetic-field direction, which will be taken as the direction of the atomic quantization axis, we may set $\theta=0$ in our general expression for the matrix elements of the photon-polarization density operator ρ^R, which is given by Eq. (5). This expression can then be substantially simplified by taking advantage of the relationship $D^{(J)}_{MM'}(\varphi,0,0)=\delta(M,M')$. In addition, we will assume that only a single electromagnetic-multipole contribution (specified by j) is dominant, and we will ignore the interference between different multipole components. Using the orthogonality properties of the final two 3-j symbols, the general expression can then be reduced to the result

$$\begin{aligned}\langle\lambda|\rho^R|\lambda'\rangle=&\sum_{i,i',f}{}' \langle\gamma_i|\rho^A|\gamma_{i'}\rangle\sum_{\Delta_f J_f M_f}\sum_{\Delta'_f J'_f M'_f}\langle\gamma_f|\Delta_f J_f M_f\rangle\\ &\times\langle\gamma_f|\Delta'_f J'_f M'_f\rangle^*\sum_{\Delta_i J_i M_i}\sum_{\Delta'_i J'_i M'_i}\langle\Delta_i J_i M_i|\gamma_i\rangle\\ &\times\langle\Delta'_i J'_i M'_i|\gamma_{i'}\rangle^*\left(\frac{2j+1}{4\pi}\right)A(j)A(j)^*\\ &\times(-1)^{J_f+J'_f-M_f-M'_f-\lambda-\lambda'}(\Delta_f J_f\|Q^{(j)}\|\Delta_i J_i)\\ &\times(\Delta'_f J'_f\|Q^{(j)}\|\Delta'_i J'_i)^*\begin{pmatrix}J_f & j & J_i\\ -M_f & -\lambda & M_i\end{pmatrix}\\ &\times\begin{pmatrix}J'_f & j & J'_i\\ -M'_f & -\lambda' & M'_i\end{pmatrix}.\end{aligned} \tag{18}$$

This simplified expression can be used to evaluate the linear-polarization and circular-polarization parameters for observation along the magnetic-field direction of atomic radiative emission corresponding to a single electromagnetic-multipole component.

B. Circular polarization of radiative emission in the absence of a perpendicular electric field and a coherent excitation process

In the absence of a perpendicular component of the electric field, axial symmetry may be present and the z components of the total electronic angular momenta can then be treated as conserved quantities. It follows that the final-state magnetic quantum numbers must be equal, i.e., we may set $M_f=M'_f$ in Eq. (18). Coherent excitation of the initial atomic magnetic sublevels, which are designated by M_i, can still occur as a result of a nonparallel directed-excitation process. In the absence of a coherent excitation process, the initial-state atomic density operator ρ^A must be diagonal in the M_i representation, i.e., we can assume that $M_i=M'_i$ in Eq. (18). In the absence of a perpendicular component of the electric field and a coherent excitation of the initial-state magnetic sublevels, the selection rules governing the remaining two 3-j symbols in Eq. (18) can be exploited to obtain the result that the photon-polarization density operator ρ^R can have only diagonal elements, corresponding to $\lambda=\lambda'=\pm1$. Consequently, only the circularly polarized radiative emission will be observable in the direction of the magnetic field. According to Eq. (17), the circularly polarized radiative emission along the magnetic-field direction can be described in terms of the diagonal photon-polarization density-matrix elements

$$\begin{aligned}\langle\lambda|\rho^R|\lambda\rangle=&\sum_{M_i M_f}{}' \langle\gamma_i M_i|\rho^A|\gamma_i M_i\rangle\sum_{\Delta_f J_f}\sum_{\Delta'_f J'_f}\langle\gamma_f M_f|\Delta_f J_f M_f\rangle\\ &\times\langle\gamma_f M_f|\Delta'_f J'_f M_f\rangle^*\sum_{\Delta_i J_i}\sum_{\Delta'_i J'_i}\langle\Delta_i J_i M_i|\gamma_i M_i\rangle\\ &\times\langle\Delta'_i J'_i M_i|\gamma_i M_i\rangle^*\left(\frac{2j+1}{4\pi}\right)A(j)A(j)^*\\ &\times(-1)^{J_f+J'_f-2M_f}(\Delta_f J_f\|Q^{(j)}\|\Delta_i J_i)\\ &\times(\Delta'_f J'_f\|Q^{(j)}\|\Delta'_i J'_i)^*\begin{pmatrix}J_f & j & J_i\\ -M_f & -\lambda & M_i\end{pmatrix}\\ &\times\begin{pmatrix}J'_f & j & J'_i\\ -M_f & -\lambda & M_i\end{pmatrix}.\end{aligned} \tag{19}$$

The indices γ_i and γ_f now represent the quantum numbers for the field-dependent atomic eigenstates, excluding the conserved z components of the total electronic angular momenta. The observation of linearly polarized radiative emission in the magnetic-field direction, which is described in terms of the nondiagonal matrix elements of the photon-polarization density operator ρ^R, could reveal the presence of a perpendicular component of the electric field or a coherent excitation mechanism.

C. Radiative emission in the absence of coherent excitation and the neglect of electric-field and magnetic-field mixing of atomic states

In the conventional description of atomic radiative emission, initial-state atomic coherences and field-induced mixing of initial and final states are ignored. In addition, the initial-state magnetic sublevels are often assumed to be uniformly (statistically) populated. This assumption can be expressed as follows:

$$\begin{aligned}\langle\gamma_i M_i|\rho^A|\gamma_i M_i\rangle&=\rho^A(\Delta_i\Delta_i J_i J_i;0,0)/(2J_i+1)^{1/2}\\ &=N(\Delta_i J_i)/(2J_i+1).\end{aligned} \tag{20}$$

The summations over the magnetic quantum numbers M_i and M_f in Eq. (19) can now be performed. The total intensity of the right or left circularly polarized radiative emission in the magnetic-field direction can then be reduced to the result

$$\langle\lambda|\rho^R|\lambda\rangle=\left(\frac{1}{4\pi}\right)\left[\frac{N(\Delta_i J_i)}{2J_i+1}\right]|A(j)|^2|(\Delta_f J_f\|Q^{(j)}\|\Delta_i J_i)|^2, \tag{21}$$

which is independent of the photon helicity λ. With these approximations, the relationship between the irreducible spherical-tensor components of the photon-polarization and atomic-system density operators may be simply expressed as follows:

$$\rho^R(j,j;0,0)=\rho^A(\Delta_i\Delta_i J_i J_i;0,0)=\left(\frac{1}{4\pi}\right)\times\left(\frac{1}{2j+1}\right)^{1/2}|A(j)|^2|(\Delta_f J_f\|Q^{(j)}\|\Delta_i J_i)|^2, \tag{22}$$

and the irreducible spherical-tensor components $\rho^R(j,j;J,M)$ of the photon-polarization density operator ρ^R corresponding to $J\neq 0$ must vanish. For anisotropic initial-state atomic-level population distributions, represented by an atomic-system density operator ρ^A with nonvanishing irreducible spherical-tensor components $\rho^A(\Delta_i\Delta_i J_i J_i;K,N)$ corresponding to $K\neq 0$, the photon-polarization density operator ρ^R can have nonvanishing irreducible spherical-tensor components with $J\neq 0$. This is in accord with the conservation of total angular momentum in the field-free spontaneous radiative-emission process.

D. Electric-dipole transitions

Nearly all atomic systems are customarily assumed to undergo spontaneous radiative decay predominantly by electric-dipole transitions, and interference of different electromagnetic multipole components is usually ignored. For electric-dipole transitions in the absence of electric and magnetic fields, the matrix elements of the electromagnetic interaction are given by the standard approximation

$$\langle\Delta_f J_f M_f,\vec{k}\lambda|V|\Delta_i J_i M_i,0\rangle=-i(2\pi\hbar\omega)^{1/2}\times\langle\Delta_f J_f M_f|\vec{D}_\lambda|\Delta_i J_i M_i\rangle, \tag{23}$$

where $\vec{D}_\lambda$ denotes the component of the many-electron electric-dipole-moment operator corresponding to photon helicity λ. The familiar (lowest-order perturbation theory) expression for the spontaneous electric-dipole emission rate may be evaluated using the Fermi golden-rule formula, which contains the factor $2\pi/\hbar^2$ and the density-of-final-states (per unit frequency) factor $\omega^2/(2\pi c)^3$. The diagonal matrix elements of the photon-polarization density operator describing spontaneous electric-dipole emission are given by

$$\langle\lambda|\rho^R|\lambda\rangle=\left(\frac{\omega^3}{2\pi\hbar c^3}\right)\left[\frac{N(\Delta_i J_i)}{2J_i+1}\right]|(\Delta_f J_f\|Q^{(1)}\|\Delta_i J_i)|^2. \tag{24}$$

The conventional spontaneous radiative-emission rate (or Einstein A coefficient) is defined in terms of the summation over final-state magnetic sublevels M_f and photon polarizations λ, together with the integration over the photon emission angles. The result thereby obtained can be expressed in the familiar form

$$A_r(\Delta_i J_i\rightarrow\Delta_f J_f)=\left(\frac{4}{3}\right)\left(\frac{\omega^3}{\hbar c^3}\right)\left(\frac{1}{2J_i+1}\right)|(\Delta_f J_f\|Q^{(1)}\|\Delta_i J_i)|^2. \tag{25}$$

Equations (24) and (25) have been recovered by assuming a uniform (statistical) distribution of the initial-state magnetic-sublevel populations and ignoring field-induced mixing of the unperturbed initial and final atomic eigenstates.

E. Directed excitation processes

In the anisotropic excitation of the atomic system, by an electron beam or a laser source, a nonuniform distribution of initial-state magnetic-sublevel population densities can be established. Transition rates between pairs of individual magnetic sublevels can be expressed, in terms of the usual M-averaged Einstein A coefficients, as follows:

$$A_r(\gamma_i\rightarrow\gamma_f)=\left|\begin{pmatrix} J_f & j & J_i \\ -M_f & m & M_i\end{pmatrix}\right|^2\times(2J_i+1)A_r(\Delta_i J_i\rightarrow\Delta_f J_f), \tag{26}$$

which is valid for any single-multipole radiative emission process. In order to evaluate the matrix elements of the photon-polarization density operator, the initial-state atomic-level population densities $N(\gamma_i)=N(\Delta_i J_i M_i)$ must be determined from an M-resolved collisional-radiative model. A steady-state radiative-cascade model may be adequate for sufficiently low particle densities, for which the effects of collisional deexcitation processes can be neglected. If the total rate due to electron and photon excitations is assumed to be balanced only by the rates for spontaneous radiative transitions, we obtain the steady-state equations

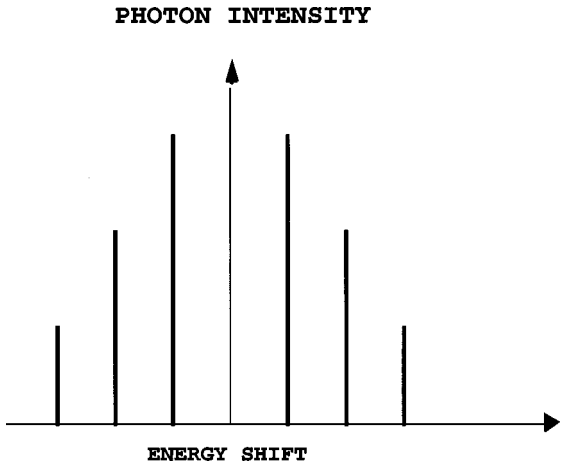

FIG. 2. A weak-field Zeeman pattern describing a $^3P_2\rightarrow{}^3P_1$ magnetic-dipole emission. The symmetrically-shifted right- and left-circularly polarized emissions are observed in the magnetic-field direction, in the absence of electric fields, and for uniformly populated magnetic sublevels.

$$N(g)W_{ex}(g\rightarrow\gamma_i)+\sum_{\gamma_j>\gamma_i}N(\gamma_j)A_r(\gamma_j\rightarrow\gamma_i)=N(\gamma_i)A_r(\gamma_i), \tag{27}$$

where $W_{ex}(g\rightarrow\gamma_i)$ denotes the total excitation rate from the ground state g and $A_r(\gamma_i)$ represents the sum of the radiative-decay rates. The number of important higher excited levels $\gamma_j>\gamma_i$ is expected to grow rapidly with increasing electron or photon energy, especially in an M-resolved description. This model has been used in an analysis of x-ray emission from EBIT [58]. Radiative emission following inner-shell-electron ionization and dielectronic-recombination radiation can be treated by extending the conventional bound-state radiative-cascade model to include autoionizing states [59]. In an M-resolved collisional-radiative model, it may be necessary to take into account collisional and radiative transitions involving a large set of bound excited and autoionizing states. Fujimoto and Kawachi [60] have employed a collisional-radiative model based on the density-matrix approach for axially symmetric excitation in the absence of electric and magnetic fields. The most general time-independent kinetic description would be based on the master equations for all elements of the initial-state atomic-density operator, allowing for a general set of steady-state excitation processes in an arbitrary arrangement of static (or quasistatic) electric and magnetic fields.

F. Spectral patterns due to the circularly polarized radiative emissions

The weak-field Zeeman-effect spectral patterns (corresponding to uniformly populated initial-state magnetic sublevels in the absence of electric fields) are characterized by equal intensities of symmetrically shifted left- and right-circularly polarized radiative emissions in the magnetic-field direction. The photon-energy shift is the difference between the atomic-energy-level shifts produced by the magnetic-field perturbation $H^A(B)=-\vec{M}\cdot\vec{B}=\mu_B g_e\vec{J}\cdot\vec{B}$, where g_e denotes the electron gyromagnetic ratio and μ_B is the Bohr magneton. Using first-order perturbation theory, the photon-nergy shift may be expressed in the linear-Zeeman-effect form [61,62]

$$\begin{aligned}\Delta\hbar\omega(B)&=\langle\Delta_f J_f M_f|H^A(B)|\Delta_f J_f M_f\rangle\\&\quad-\langle\Delta_i J_i M_i|H^A(B)|\Delta_i J_i M_i\rangle\\&=[M_f g(\Delta_f J_f)-M_i g(\Delta_i J_i)]\mu_B B,\end{aligned} \tag{28}$$

where $g(\Delta_i J_i)$ and $g(\Delta_f J_f)$ denote the gyromagnetic ratios or g factors for initial and final states, respectively. The weak-field linear-Zeeman-effect spectral pattern for $^3P_2\rightarrow{}^3P_1$ magnetic-dipole transitions is illustrated in Fig. 2. Due to Doppler broadening and other broadening mechanisms, one usually observes two blended spectral features, corresponding to the unresolved components for each of the two circular polarizations. These patterns can be conveniently analyzed in terms of the intensity-weighted average of the shifts for the unresolved right- or left-circularly polarized components [7,8].

The more complex Stark-Zeeman spectral patterns may be describable in terms of additional Stark shifts of the Zeeman-effect patterns. Since the electric-dipole interaction $H^A(E)=-\vec{D}\cdot\vec{E}$ has nonvanishing matrix elements only between unperturbed (field-free) atomic eigenstates with different parities, the Stark shifts must be evaluated at least in second-order perturbation theory. Ignoring the diamagnetic (quadratic-Zeeman-effect) contribution, the additional photon-energy shift can be expressed in the resolvent-operator form

$$\begin{aligned}\Delta\hbar\omega(E)&=\left\langle\Delta_f J_f M_f\left|\vec{D}\cdot\vec{E}\frac{1}{E(0)-H^A(0)}\vec{D}\cdot\vec{E}\right|\Delta_f J_f M_f\right\rangle\\&\quad-\left\langle\Delta_i J_i M_i\left|\vec{D}\cdot\vec{E}\frac{1}{E(0)-H^A(0)}\vec{D}\cdot\vec{E}\right|\Delta_i J_i M_i\right\rangle.\end{aligned} \tag{29}$$

If the magnetic field is sufficiently strong to uncouple the orbital and spin angular momenta, the uncoupled magnetic-sublevel (Paschen-Back-effect) representation may be more appropriate and the diamagnetic contribution should be included. An extension of the average-shift analysis could be useful for perpendicular (crossed) electric field and magnetic fields. Our general density-matrix description should be applied starting with initial and final eigenstate expansions that have been obtained from a diagonalization of the field-dependent atomic Hamiltonian matrix.

IV. SUMMARY AND CONCLUSIONS

A density-matrix description has been developed for polarized atomic radiative emission under general steady-state excitation conditions in arbitrary arrangements of static (or quasistatic) electric and magnetic fields. Our main result is a general expression for the matrix elements of the photon-polarization density operator determining the total intensity, angular distribution, and polarization of the atomic radiative emission. We deduce that the observation of linearly polarized radiative emission in the direction of the magnetic field could reveal the presence of a perpendicular electric field or a coherent excitation process. To apply our formalism, it is necessary to obtain the field-induced energy-level splittings and eigenstate transformations by perturbation theory or direct diagonalization of the total field-dependent atomic Hamiltonian matrix in a field-free angular-momentum basis set. It is also necessary to determine the steady-state, field-dependent initial-state atomic-level populations and static coherences, taking into account directed collisional and radiative excitation processes. In a future investigation, we will treat the time evolution of the emitted-photon and atomic-system density operators produced by the action of collisional and radiative relaxation processes in the presence of electric and magnetic fields. It will also be necessary to provide a self-consistent description of the spectral-line shapes

associated with the individual, possibly overlapping, Stark-Zeeman components [63].

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