Anisotropic radiative emission effects on deduced resonant-transfer-excitation cross sections

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The radiation emitted by an ion collisionally excited by an electron beam is anisotropic. We discuss two schemes, one for use with *LS*-coupling cross sections and one for intermediate coupling, whereby theoretical resonant-transfer-excitation cross sections can be modified to take account of this anisotropy when making comparisons with experimental results deduced from observations of radiation at a particular beam angle. We have carried out such calculations for a number of ions initially in an *S* state, viz. F^{6+}, Ca^{q+} ($q=16-19$), and Nb^{q+} ($q=29-31$). In all cases, neglecting hyperfine structure, the allowance for anisotropic radiative emission at right angles to the beam axis was found to result in only a small increase (10–15%) in the cross section over results that assumed isotropy. Allowance for hyperfine structure reduced even this anisotropy by 20% for $^{19}F^{6+}$ ($I=\frac{1}{2}$) and by 80% for ^{93}Nb ($I=\frac{9}{2}$) ions; results for ^{40}Ca ($I=0$) ions remain unchanged.

I. INTRODUCTION

The radiation emitted by an ion collisionally excited by an anisotropic source will be anisotropic in general. This has implications for the extraction of information on collision dynamics from an experiment which only observes the subsequent radiative emission at a particular angle with respect to the beam axis, as in resonant-transfer-excitation (RTE) experiments[1] or in an electron-beam ion-trap[2] (EBIT) experiment. Using the impulse approximation, Brandt[3] showed that resonant-transfer-excitation processes in atom-ion collisions could be calculated from the equivalent free-electron–ion collision process. Consequently, the whole theoretical apparatus for electron-ion collisions could be applied to RTE. In particular, the theory of differential electron scattering, which was formulated by Blatt and Biedenharn[4] and by Percival and Seaton,[5] is the basis for recent calculations on resonant-transfer excitation followed by Auger emission (RTEA) by Badnell[6] and by Bhalla,[7] who also remarked on the differential or anisotropic photon emission problem, which is the subject of this paper.

The theory of polarized electric dipole radiation, which is simply related to the anisotropy, was first developed by Oppenheimer.[8] The first calculations were carried out by Penney,[9] who showed that in general it was necessary to take into account both fine structure and hyperfine structure. The theory was extended by Percival and Seaton[10] and by Fano and Macek.[11] The theory of Percival and Seaton,[10] which was for collisional excitation in the *LS*-coupling approximation, only requires a minor modification for resonant-transfer excitation or dielectronic recombination. The required theory for an intermediate-coupling approximation can then be developed in an analogous fashion. At this stage our theoretical approach is equivalent to that of Inal and Dubau,[12,13] except that we restrict ourselves to electric dipole radiation only. However, we also make allowance for the hyperfine structure, unlike Inal and Dubau,[12,13] and show that its effect is important for $^{19}F^{6+}$ ($I=\frac{1}{2}$) and ^{93}Nb ($I=\frac{9}{2}$) ions. Isotopes with zero nuclear spin are of course unaffected, e.g., ^{40}Ca, $^{58,60}Ni$, and $^{54,56}Fe$, iron being the example chosen by Inal and Dubau.[12,13]

In Sec. II of this paper we develop the theory required to calculate resonant-transfer excitation or dielectronic-recombination cross sections with photon emission at a particular angle. In Sec. III we discuss its application to a number of experiments and in Sec. IV we present the results of our calculations. We then conclude in Sec. V.

II. THEORY

When an initially unpolarized ion is collisionally excited by an unpolarized electron beam, the intensity $I(\theta)$ of the subsequent electric dipole radiative emission at an angle θ with respect to the beam axis, averaged over all polarizations, is given by[10]

$$I(\theta)=\tfrac{1}{3}\bar{I}[1+\beta P_2(\cos\theta)] , \tag{1}$$

where the total intensity integrated over all angles is $4\pi\bar{I}/3$ and

$$P_2(\cos\theta)=\tfrac{1}{2}(3\cos^2\theta-1) . \tag{2}$$

The asymmetry parameter β is related to the polarization fraction P at $\theta=90°$ by

$$\beta=\frac{2P(90°)}{P(90°)-3} \tag{3}$$

and

$$P(90°)=\frac{I_\parallel-I_\perp}{I_\parallel+I_\perp}=\frac{3I_\parallel-\bar{I}}{I_\parallel+\bar{I}} , \tag{4}$$

where $I_\parallel$ and $I_\perp$ are the intensities of emission (at $\theta=90°$) with linear polarization vector respectively parallel and perpendicular to the beam axis. Away from $\theta=90°$,

$$P(\theta)=\frac{P(90°)\sin^2\theta}{1-P(90°)\cos^2\theta} . \tag{5}$$

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From Eqs. (3) and (4) we have

$$\beta=\frac{1}{2}\left[1-\frac{3I_{\parallel}}{\bar{I}}\right] . \tag{6}$$

Percival and Seaton[10] have derived expressions for the intensities $I_{\parallel}$ and $\bar{I}$ in the LS-coupling approximation which allow for fine structure and hyperfine structure. We are interested in similar expressions for use in the intermediate-coupling approximation which also allow for hyperfine structure. We present just the final results in this section and a detailed derivation is given as an appendix to this paper.

Assuming that the excited states are populated directly by collisions (i.e., no cascading) and depopulated by radiative and maybe autoionizing transitions, then the collisional excitation or resonant capture rate coefficient is directly proportional to the radiation intensity. We consider the intermediate-coupling case first. When the ion is initially in a $^1S_0$ state we can take account of the hyperfine structure through angular momentum recoupling (see the Appendix), viz., for nuclear spin I and $J\rightarrow J'$ emission,

$$\frac{I_{\parallel}}{\bar{I}}=\frac{(2J+1)\sum_{F,F'}(2F+1)^2(2F'+1)\begin{Bmatrix} J & F & I \\ F' & J' & 1 \end{Bmatrix}^2 \sum_{M_J,M_I,M_F}\begin{pmatrix} J & I & F \\ M_J & M_I & -M_F \end{pmatrix}^2\begin{pmatrix} F' & 1 & F \\ M_{F'} & 0 & -M_F \end{pmatrix}^2\sigma(JM_J)}{(2I+1)\sum_{M_J}\sigma(JM_J)} , \tag{7}$$

where $\sigma(JM_J)$ is the collision cross section for populating the JM_J state from a $^1S_0$ initial state, ( ) denotes a Wigner $3j$ symbol, and { } a Wigner $6j$ symbol, see e.g., Ref. 14. We can also apply this to a $^2S_{1/2}$ initial state if we neglect the hyperfine recoupling in the initial state. The intermediate-coupling expression in the absence of hyperfine structure is simpler (see the Appendix), viz., for $J\rightarrow J'$ emission,

$$\frac{I_{\parallel}}{\bar{I}}=\frac{\sum_{M_J}(2J+1)\begin{pmatrix} J' & 1 & J \\ M_{J'} & 0 & -M_J \end{pmatrix}\sigma(JM_J)}{\sum_{M_J}\sigma(JM_J)} , \tag{8}$$

where $\sigma(JM_J)$ is the collision cross section for populating the JM_J state from a given initial state. We still require expressions in the LS-coupling approximation for problems which are too big to consider in intermediate coupling. Following Percival and Seaton,[10] when the ion is initially in an S-state fine structure and hyperfine structure can be taken into account through angular momentum recoupling,[10] and we can still work with LS-coupling transition rates, viz., for nuclear spin I and $SL\rightarrow SL'$ emission, see Eq. (9) below.

Here $\sigma(SLM_SM_L)$ is the collision cross section for populating the SLM_SM_L state from a given initial S state. Again, the expression is a little simpler in the absence of hyperfine structure, viz., for $SL\rightarrow SL'$ emission,

$$\frac{I_{\parallel}}{\bar{I}}=\frac{(2L+1)\sum_{\substack{J,J' \\ M_S,M_L,M_J}}(2J'+1)(2J+1)^2\begin{pmatrix} S & L & J \\ M_S & M_L & -M_J \end{pmatrix}^2\begin{pmatrix} J' & 1 & J \\ M_{J'} & 0 & -M_J \end{pmatrix}^2\begin{Bmatrix} L & J & S \\ J' & L' & 1 \end{Bmatrix}^2\sigma(SLM_SM_L)}{\sum_{M_S,M_L}\sigma(SLM_SM_L)} . \tag{10}$$

The above expressions (7)–(10) are applicable to both above- and below-threshold excitation.

We now specifically consider resonant-transfer excitation followed by x-ray stabilization (RTEX) in the isolated resonance approximation. This is the analogous process in ion-atom collisions to dielectronic recombination (DR) in electron-ion collisions. The theory applies equally to RTEX and DR, which only differ through the convolution of the energy-averaged DR cross sections. The quantum numbers for the excited state now refer to the whole recombining ion. We consider LS coupling first. If the ion is initially in an S state and the beam axis is coincident with the z axis then $M_L=0$, $M_J=M_S$, and the asymmetry parameter β is purely algebraic. The same is true for a $^1S_0$ initial state in intermediate coupling since $\sigma(J\frac{1}{2})=\sigma(J-\frac{1}{2})$. When the ion is initially in a $^2S_{1/2}$ state we have $M_J=0,\pm1$ and we can test out the sensitivity of β to the capture cross sections by varying the ratio $\sigma(J1)$ to $\sigma(J0)$.

We end this section by recalling that Eq. (1) allows for the anisotropy of emission averaged over the polarization, which is linear, since we have cylindrical symmetry. We may obtain the anisotropy for a particular linear polarization by again considering the classical source of Percival and Seaton.[10] This results in an additional term for the right-hand side of (1) of the form

$$-\tfrac{1}{2}\bar{I}\beta\sin^2\theta\cos2\phi , \tag{11}$$

where ϕ is the angle between the plane of the experiment, as defined by the incident electron beam and the direction of emission, and the desired linear polarization vector.

$$\frac{I_{\parallel}}{\bar{I}}=\frac{(2L+1)\sum_{J,J'}(2J+1)^2(2J'+1)\begin{Bmatrix} L & J & S \\ J' & L' & 1 \end{Bmatrix}^2\left\{\sum_{F,F'}(2F+1)^2(2F'+1)\begin{Bmatrix} J & F & I \\ F' & J' & 1 \end{Bmatrix}^2\sum_{M_J,M_I,M_F}\begin{pmatrix} J & I & F \\ M_J & M_I & -M_F \end{pmatrix}^2\begin{pmatrix} F' & 1 & F \\ M_{F'} & 0 & -M_F \end{pmatrix}^2\sum_{M_S,M_L}\begin{pmatrix} S & L & J \\ M_S & M_L & -M_J \end{pmatrix}^2\sigma(SLM_SM_L)\right\}}{(2I+1)\sum_{M_S,M_L}\sigma(SLM_SM_L)} \quad (9)$$

Thus, $I_{\parallel}$ and $I_{\perp}$ are given by the modified form of (1) with $\phi=0°$ and $\phi=90°$, respectively ($\theta=90°$, still).

III. APPLICATION TO EXPERIMENT

The intensity of induced light emission in a collision experiment is directly proportional to the collision cross section. In RTEX, for an ion in an initial state i, recombining into an intermediate state j and radiating to a final state k, we have

$$4\pi\frac{d\sigma_x(i;j;k)}{d\Omega}=\sigma_x(i;j;k)[1+\beta(j\rightarrow k)P_2(\cos\theta)] \ . \quad (12)$$

Using the impulse approximation,[3] the integrated partial RTEX cross section $\sigma_x(i;j;k)$ may be written in terms of energy-averaged DR cross sections $\bar{\sigma}_d(i;j;k)$; thus

$$\sigma_x(i;j;k)=J(Q)\bar{\sigma}_d(i;j;k)\frac{\Delta E_c}{2I_H}\left[\frac{MI_H}{E}\right]^{1/2} \ . \quad (13)$$

$J(Q)$ is the Compton profile of the target gas with Q given by

$$Q=\frac{1}{2I_H}\left[E_c+E_t-\frac{Em}{M}\right]\left[\frac{MI_H}{E}\right]^{1/2} , \quad (14)$$

E is the projectile-ion energy in the laboratory frame, E_c is the $j\rightarrow i$ Auger energy, and E_t is the binding energy of the target electron, both in the rest frame of the projectile. M is the ionic mass, m is the electron mass, and I_H is the ionization potential energy of hydrogen. The energy-averaged DR cross section is given by

$$\bar{\sigma}_d(i;j;k)=\frac{(2\pi a_0 I_H)^2}{E_c\Delta E_c}\frac{\omega(j)}{2\omega(i)} \times\frac{\tau_0 A_r(j\rightarrow k)\sum_l A_a(j\rightarrow i,E_c l)}{\sum_h\left[A_r(j\rightarrow h)+\sum_l A_a(j\rightarrow h,E_c l)\right]} , \quad (15)$$

where ΔE_c is the energy bin width, $\omega(j)$ is the statistical weight of the $(N+1)$-electron doubly excited state, $\omega(i)$ is the statistical weight of the N-electron initial state, and $(2\pi a_0)^2\tau_0=2.674\,11\times10^{-32}$ cm^2 sec, where a_0 and τ_0 denote, respectively, the Bohr radius and time. A_a and A_r denote autoionization rates and radiative rates, respectively. The total RTEX cross section is obtained by summing Eq. (12) over all possible states j and over all final states k that are stable against autoionization.

The left-hand side of Eq. (12) is the form of the quantity determined by an RTEX experiment[1] and, in principle, an EBIT experiment, although in this case the Compton profile of Eqs. (13) and (14) is replaced by the experimental electron-ion velocity distribution. However, the first EBIT experiments, both excitation[15] of Ba^{46+} and DR (Ref. 2) of Ni^{26+}, measured in effect the collision-induced radiation relative to radiative recombination (RR). The anisotropy of dipole RR is again of the form of Eq. (1) and this accounts for most, if not all, of the anisotropy in the collision-induced radiation.[2,15] Equation (12) assumes that the Si (Li) detector used in RTEX experiments is not

polarization sensitive. An example where this is not true is for the Ge crystal spectrometer used in the EBIT measurement[16] of relative excitation cross sections for Fe^{24+}. However, as we saw with the example in Eq. (11), this does not require the calculation of any new collision quantities.

Finally, we note that if the primary interest of the experiment is the determination of the collision quantity σ rather than radiative anisotropy, then observation at the "magic angle" of $\theta=\cos^{-1}\sqrt{1/3}$ or a measurement of the polarization fraction at $\theta=90°$ obviates the need for a theoretical calculation of β.

IV. RESULTS

We now apply the theory of Sec. II to our previously calculated RTEX cross sections for F^{6+}, Ca^{q+} ($q=16-19$), and Nb^{q+} ($q=29-31$) ions (Refs. 17, 18, and 19, respectively) to compensate for the fact that the experimental observations (Refs. 17, 20, and 1, respectively) were made at $\theta=90°$ to the beam axis. Integrated cross sections have also been computed for a number of these ions by Hahn and co-workers.[21,22]

We first consider K-shell transitions in F^{6+}. We use Eq. (10) with our LS-coupling results[17] to take account of fine-structure effects on the polarization, our intermediate coupling results[17] differing by less than 2% from LS coupling. At $\theta=90°$ the peak RTEX cross section given by Eq. (12) is increased by 15% over the integrated cross section. Allowance for the hyperfine structure of $^{19}F^{6+}$ ($I=\frac{1}{2}$), Eq. (9), reduces this anisotropy by 20%, i.e., to a 12% increase.

Next we consider K-shell transitions in Ca^{q+} ($q=16-19$). It is now necessary to take account of fine structure in the integrated RTEX cross section.[18,23] Thus, we make use of Eq. (8), there being no hyperfine structure for ^{40}Ca ions. At $\theta=90°$, both our Ca^{16+} and Ca^{18+} results[18] are increased by about 15%, uniformly with energy. For Ca^{17+} and Ca^{19+} we must additionally consider the relative populations of the $M=0$ and ±1 magnetic sublevels. Assuming that the $M=0$ population is much larger than $M=\pm1$, we obtain a uniform 10% increase in the cross section[18] for Ca^{17+}; this falls to about 7% when $\sigma(JM=0)=\sigma(JM=\pm1)$. The effect on Ca^{19+} is larger and nonuniform with energy. For only $M=0$ populated, the KLL peak of Ca^{19+} is increased by 18% and the KLn ($n>L$) by 12%. This falls to 9% and 6% when $\sigma(JM=0)=\sigma(JM=\pm1)$. Even though there is no hyperfine structure to reduce the anisotropy for ^{40}Ca ions, the observed effect may still be substantially reduced by the presence of any external electric fields in the interaction region, which will then redistribute the population of the magnetic sublevels of the autoionizing states. We have already[18] compared our integrated RTEX cross sections for Ca^{q+} with the experimental results of Tanis *et al.*,[20] which contain relative uncertainties of $\pm10\%$ and an estimated absolute uncertainty of $\pm30\%$. The maximum calculated anisotropy for Ca^{q+} ($q=16-18$) is too small ($<15\%$) and uniform with energy to deduce anything from comparison with experiment, while the Ca^{19+} experiment was dominated by a large background signal.[20]

We now turn to L-shell transitions in Nb^{q+} ($q=29-31$) ions. The RTEX cross sections for Nb^{29+} and Nb^{30+} were only calculated[19] in LS coupling. Using Eq. (10) to take account of fine structure only, the RTEX cross section at $\theta=90°$ is increased by 12–14% over the integrated value. This is reduced substantially, by 80%, to less than 3% on taking into account the hyperfine structure in ^{93}Nb ($I=\frac{9}{2}$) ions via Eq. (9). In fact, the same results are obtained with our intermediate-coupling cross sections for Nb^{31+} on using Eqs. (8) and (7). Again, this anisotropy is too small to show up in comparisons with the experimental results of Bernstein *et al.*[1]

V. CONCLUSION

We have shown how to modify both LS coupling and intermediate-coupling integrated RTEX cross sections to allow for anisotropic radiative emission, including the effect of hyperfine structure. We found that, for ions initially in an S state, the allowance for the anisotropy of radiation emitted at right angles to the beam axis results in no more than a 15% increase, in general. Even this can be substantially reduced by hyperfine structure.

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APPENDIX

Here we give details of the derivation of the intermediate-coupling intensity ratios presented in Sec. II, allowing for hyperfine structure. We equate the collisional rate of population of the state $JIFM_F$ from an initial state i with the rate of depopulation due to radiative and maybe autoionizing transitions; thus

$$N_e N_i v_i \sigma_i(JIFM_F) = N(JIFM_F) \sum_{\substack{F'J' \\ M_{F'}}} A(JIFM_F \rightarrow J'IF'M_F) , \tag{A1}$$

where N_e, N_i, and $N(JIFM_F)$ are the relevant population densities; $v_i\sigma_i(JIFM_F)$ is then the collisional rate coefficient $i \rightarrow JIFM_F$. From Edmonds,[14] Eq. (3.5.6) we have

$$\sum_{\substack{F',J' \\ M_{F'}}} A(JIFM_F \rightarrow J'IF'M_{F'}) = A(JIF) , \tag{A2}$$

i.e., the total rate of depopulation of $JIFM_F$ is independent of M_F.

The rate of $JIF \rightarrow J'IF'$ emission of photons with electric dipole vector parallel to the z axis is given by[10,14]

$$N_e N_i K_z(JIF \rightarrow J'IF') = \sum_{M_F, M_{F'}} N(JIFM_F)(2F+1) \begin{bmatrix} F' & 1 & F \\ M_{F'} & 0 & -M_F \end{bmatrix}^2 \times A_r(JIF \rightarrow J'IF') , \quad \text{(A3)}$$

which defines the rate coefficient K_z for this process. The rate of $JIF \rightarrow J'IF'$ emission of all photons is given by[10,14]

$$N_e N_i K(JIF \rightarrow J'IF') = \sum_{\substack{M_F, M_{F'} \\ \mu}} N(JIFM_F)(2F+1) \begin{bmatrix} F' & 1 & F \\ M_{F'} & \mu & -M_F \end{bmatrix}^2 \times A_r(JIF \rightarrow J'IF') = \sum_{M_F} N(JIFM_F) A_r(JIF \rightarrow J'IF') , \quad \text{(A4)}$$

which defines the rate coefficient K for this process. Substituting for the population $N(JIFM_F)$ from (A1) into (A3) and (A4) we have

$$K_z(JIF \rightarrow J'IF') = \frac{v_i A_r(JIF \rightarrow J'IF')}{A(JIF)} \sum_{M_F, M_{F'}} (2F+1) \begin{bmatrix} F' & 1 & F \\ M_{F'} & 0 & -M_F \end{bmatrix}^2 \sigma_i(JIFM_F) \quad \text{(A5)}$$

and

$$K(JIF \rightarrow J'IF') = \frac{v_i A_r(JIF \rightarrow J'IF')}{A(JIF)} \sum_{M_F} \sigma_i(JIFM_F) . \quad \text{(A6)}$$

The intensities of emission $I_{\parallel}$ and $\bar{I}$ are directly proportional[10] to the rate coefficients K_z and K. Thus the anisotropy and polarization parameters may be evaluated from (A5) and (A6) for $JIF \rightarrow J'IF'$ emission. We are interested in $J \rightarrow J'$ emission for a given nuclear spin I. We thus make use of[10,14]

$$A_r(JIF \rightarrow J'IF') = (2J+1)(2F'+1) \begin{Bmatrix} J & F & I \\ F' & J' & 1 \end{Bmatrix}^2 A_r(J \rightarrow J') \quad \text{(A7)}$$

and, for an ion initially in an $i = {}^1S_0$ state,

$$\sigma_i(JIFM_F) = \frac{1}{(2I+1)} \sum_{M_I, M_J} (2F+1) \begin{bmatrix} J & I & F \\ M_J & M_I & -M_F \end{bmatrix}^2 \sigma_i(JM_J) \quad \text{(A8)}$$

to sum over F and F' in (A3) and (A4). We then obtain for K_z

$$K_z(J \rightarrow J') = \frac{v_i A_r(J \rightarrow J')}{A(J)(2I+1)} \sum_{F', F} (2J+1)(2F+1)^2(2F'+1) \begin{Bmatrix} J & F & I \\ F' & J' & 1 \end{Bmatrix}^2 \times \sum_{M_J, M_I, M_F} \begin{bmatrix} J & I & F \\ M_J & M_I & -M_F \end{bmatrix}^2 \begin{bmatrix} F' & 1 & F \\ M_{F'} & 0 & -M_F \end{bmatrix}^2 \sigma_i(JM_J) , \quad \text{(A9)}$$

and for K,

$$K(J \rightarrow J') = \frac{v_i A_r(J \rightarrow J')}{A(J)} \sum_{M_J} \sigma_i(JM_J) \quad \text{(A10)}$$

the ratio of which gives equation (7) as required. When there is no hyperfine structure present the intensity ratios may be obtained from (A5) and (A6) on dropping the JI labels and then replacing F with J, in agreement with Eq. (8).

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