Lyman-α_1 Decay in Hydrogenlike Ions: Interference between the $E1$ and $M2$ Transition Amplitudes

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For the Lyman-α_1 transition ($2p_{3/2} \rightarrow 1s_{1/2}$) in hydrogenlike ions an interference between the leading $E1$ decay channel and the much weaker $M2$ multipole transition gives rise to a remarkable modified angular distribution of the emitted photons from aligned ions. This effect is most pronounced for the heaviest elements but results in a still sizable correction for medium-Z ions. For the particular case of hydrogenlike uranium where the angular distribution of the Lyman-α_1 x rays following radiative electron capture has been measured, the former variance with theoretical findings is removed when this $E1$-$M2$ interference is taken into account.

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Radiative transitions in high-Z heavy ions play a key role for our understanding of the effects of strong Coulomb fields on the electronic structure of atoms and ions [1]. At high-Z, transition rates and energies are strongly affected by relativistic corrections, and even quantum electrodynamical effects show up in a clear way [2]. One of the most prominent examples is the Lyman-α_1 transition ($2p_{3/2} \rightarrow 1s_{1/2}$) in hydrogen or one-electron ions which serves, e.g., as a precise measure for the $1s$ Lamb shift in hydrogenlike ions [3]. In the case of transitions rates, relativistic effects are manifested by the strongly enhanced importance of magnetic transitions; e.g., the $2s_{1/2}$ decay in high-Z one-electron ions is almost entirely governed by $M1$ transitions quite in contrast to the dominant $2E1$ decay at lower Z. In fact, the photon angular distribution of radiative transitions turns out to be more sensitive to magnetic and retardation effects than total decay rates. This was shown for the case of continuum-bound state transitions (radiative electron capture, REC) occurring in collisions of bare uranium ions with light atomic targets [4]. At high-Z and for not too high collision energies, the transition rates for REC and the corresponding cross sections are well described within the dipole approximation [2]. However, this approach fails to describe the associated photon angular distributions which are strongly modified by magnetic and retardation effects [5].

In this Letter we report on an interference between the $E1$ and $M2$ transition amplitudes in the decay of the $2p_{3/2}$ level in aligned hydrogenlike heavy ions which significantly alters the photon angular distribution of the Lyman-α_1 transition ($2p_{3/2} \rightarrow 1s_{1/2}$). Similar effects are well known for γ transitions between nuclear levels where the so-called *multipole mixing ratios,* e.g., for $E2$ and $M1$ transitions, provide detailed information about the nuclear states involved [6]. To the best of our knowledge such effects have not been reported yet for bound-bound transitions in highly charged ions. For L-shell vacancy production following proton impact, however, evidence for multipole mixing has been observed [7]. As emphasized in this Letter, such interferences may have considerable impact also for the interpretation of experimental data. For the particular case of hydrogenlike uranium, where the angular distribution of the Lyman-α_1 x rays following radiative electron capture has been measured, the former disagreement with theoretical findings [8] is removed when taking this interference into account.

Because of parity and angular momentum conservation laws, the Lyman-α_1 ($2p_{3/2} \rightarrow 1s_{1/2}$) decay in hydrogenlike ions can proceed via either $E1$ or $M2$ transitions which reflect different properties of the electron distribution. While the electric dipole component describes the charge oscillations, the magnetic quadrupole reflects the nonspherical part of the electron motion, i.e., its current distribution [1]. However, for all hydrogenlike ions up to the heaviest elements, the magnetic interaction is much weaker than the electric one, although the decay rate for $M2$ transition ($\Gamma_{M2} \propto Z^8$) increases rapidly as a function of the nuclear charge Z when compared to the $E1$ rate ($\Gamma_{E1} \propto Z^4$). But even for hydrogenlike uranium ($Z = 92$) the $E1$ transition rate amounts to $\Gamma_{\rm E1} = 3.92 \times 10^{16}$ s^{-1}, whereas the $M2$ rate $\Gamma_{\rm M2} = 2.82 \times 10^{14}$ s^{-1} contributes less than 1% to the total decay rate. In the past this rather small contribution was one main reason why—till today—the $M2$ component of the radiation field has not been incorporated in computations on the $2p_{3/2}$ decay of hydrogenlike ions or similar $nl \rightarrow 1s$ transitions in the high-Z regime [9,10]. On the other hand, the Lyman-α_1 transition has been studied intensively both by theory and experiments during the last decades because the characteristics of this line, i.e., its polarization and angular distribution, may reveal subtle information on the population mechanisms and, thus, on the dynamical processes of high-Z ions. A number of detailed investigations have been carried out, for instance, for electron-impact excitation [11–13] as well as for radiative electron capture in collisions of fast bare projectiles with light target ions [8,14,15].

To discuss the $E1$-$M2$ interference effects let us consider a hydrogenlike ion in the $2p_{3/2}$ level whose creation and decay occur as two independent steps, well separated in time (see, e.g., [8]). In the first step the excited level is just populated, for instance, by electron capture or excitation which may occur in ion-atom and ion-electron collisions. Because of the directionality of the collision, the population of magnetic sublevels is likely to deviate from a statistical distribution. In such cases the levels are aligned, thereby the pairs of atomic sublevels with the same magnetic quantum number (but with opposite signs) will be necessarily equally populated. Here we assume that neither the ions nor the target atoms are polarized in ion-atom collisions.

The alignment of an atomic level is commonly described in terms of one or several parameters $\mathcal{A}_k$ which are related to the population cross sections $\sigma(\mu_n)$ of the various sublevels μ_n. In the case of the $2p_{3/2}$ level only the alignment parameter $\mathcal{A}_2$ is nonzero (apart from $\mathcal{A}_0$), and it can be expressed as [16,17]

$$\mathcal{A}_2 = \frac{\sigma(\frac{3}{2}, \pm\frac{3}{2}) - \sigma(\frac{3}{2}, \pm\frac{1}{2})}{\sigma(\frac{3}{2}, \pm\frac{3}{2}) + \sigma(\frac{3}{2}, \pm\frac{1}{2})}, \tag{1}$$

where $\sigma(3/2, \mu_n)$ describes the population of substate μ_n of the $2p_{3/2}$ level.

In the second step, this $2p_{3/2}$ level then decays into the ground state via the emission of a photon. But although this photon emission occurs independently of the particular creation of the level, it may exhibit an anisotropic emission pattern if the level has been aligned. The angular distribution of the photons in the emitter frame is related to the alignment parameter $\mathcal{A}_2$ by [11,15]

$$W(\theta) = A_0 + A_2 P_2(\cos\theta) \propto 1 + \beta_{20}(1 - \tfrac{3}{2}\sin^2\theta), \tag{2}$$

where θ is the angle between the direction of the deexcitation photon and the beam direction while $P_2(\cos\theta)$ denotes the second-order Legendre polynomial. As seen from expression (2), the angular distribution is completely determined by the so-called *anisotropy* coefficient $\beta_{20} = \alpha\mathcal{A}_2$, while the coefficient α depends only on the total angular momenta of the initial and final ionic states, respectively. For the case of the $2p_{3/2} \to 1s_{1/2}$ transition $\alpha = 1/2$.

Expression (2) is well known from the literature (see, e.g., [18]). It includes the contribution from the electric-dipole ($E1$) transition, whereas the—weak—magnetic-quadrupole component ($M2$) is neglected. This $M2$ branch, however, can also be taken into account. For instance, by using the density matrix theory the angular distribution of radiation in its *general form* (i.e., including all allowed multipoles) was obtained by Fano and Racah [19] (see also [11]). Then, the consistent treatment of both decay modes in the Lyman-α_1 transition finally leads to the result that both the alignment and the anisotropy parameters have to be replaced by two corresponding *effective* parameters: $\mathcal{A}_2 \to \mathcal{A}_2^{(\mathrm{eff})}$ and $\beta_2 \to \beta_2^{(\mathrm{eff})}$. Note, the overall shape (2) of the angular distribution is preserved since it depends only on the quantum numbers of the initial and the final ionic states, respectively. The two effective parameters $\mathcal{A}_2^{(\mathrm{eff})}$ and $\beta_2^{(\mathrm{eff})}$ can be expressed as products of the original parameters with a *structure* function

$$\begin{aligned} \mathcal{A}_2^{(\mathrm{eff})} &= \mathcal{A}_2 \cdot f(E1, M2); \\ \beta_{20}^{(\mathrm{eff})} &= \beta_{20} \cdot f(E1, M2). \end{aligned} \tag{3}$$

The alignment parameter (1) depends only on the population mechanism of the excited level, i.e., on collisional parameters such as the projectile velocity or the charge of the target. In contrast, the *structure* function $f(E1, M2)$ is independent of the creation process and merely reflects the electronic structure of the ion. By applying the density matrix theory, this function $f(E1, M2)$ can be expressed for the $2p_{3/2} \to 1s_{1/2}$ transition as

$$f(E1, M2) = \left[\frac{\langle||E1||\rangle^2 - \langle||M2||\rangle^2 + 2\sqrt{3}\langle||E1||\rangle\langle||M2||\rangle^*}{\langle||E1||\rangle^2 + \langle||M2||\rangle^2}\right] \propto \left[1 + 2\sqrt{3}\,\frac{\langle||M2||\rangle}{\langle||E1||\rangle}\right], \tag{4}$$

where $\langle||E1||\rangle = \langle 2p_{3/2}||\alpha\mathbf{A}^{(\mathbf{e})}(L = 1)||1s_{1/2}\rangle$ and $\langle||M2||\rangle = \langle 2p_{3/2}||\alpha\mathbf{A}^{(\mathbf{m})}(L = 2)||1s_{1/2}\rangle$ are the reduced matrix elements for the electric (magnetic) bound-bound multipole transitions of rank L [15,20].

In the dipole approximation, $\langle||M2||\rangle \approx 0$ is taken to be negligible and, thus, $f(E1, M2) \equiv 1$. As seen from Eq. (4), the main correction to this approximation arises from the term which is proportional to the ratio of the transition amplitudes $\langle||M2||\rangle/\langle||E1||\rangle$. For high-$Z$ ions this ratio is of the order $\sim$0.1, leading to a 1% contribution of the $M2$ component to the total decay rate. Note that the $E1$-$M2$ interference term does not contribute to the transition probabilities (and, hence, the lifetimes) because the angular distribution [Eq. (2)] has to be integrated over all photon directions. From the properties of the Legendre polynomials, $\int P_2(\cos\theta)\, d\Omega = 0$, it is seen that the integral of the second term in Eq. (2), which contains the $E1$-$M2$ contribution, vanishes. For H-like uranium, this dimensionless function is as large as 1.28 due to the $E1$-$M2$ interference. Since this function basically depends on the ratio $\langle||M2||\rangle/\langle||E1||\rangle$ of the reduced matrix elements, a non-negligible effect of a few percent remains even for medium-Z ions. We finally note that this structure function scales approximately as $f(E1, M2) \propto Z^{2.24}$ for high-Z ions, while it is $f(E1, M2) \propto Z^{2.03}$ at lower values of Z. Obviously, this is different from a Z^4 scaling as one might expect at a first glance from the corresponding decay rates.

In the following, we focus on REC in relativistic collisions of bare high-Z ions (e.g., U^{92+}) with low-Z target atoms [8,14,15]. In this process, the REC photon must carry away the excess energy and momentum when the electron is captured in any of the ionic bound states. Several theoretical [14,15] and experimental [8] studies were performed in the past to explore the capture into the ground and into the excited states. From a measurement of the anisotropic emission of the subsequent Lyman-α_1 photons, a rather significant alignment was deduced as confirmed by relativistic theory. However, when the theoretical and the observed angular distributions were compared in detail, a remarkable variance was found which could be attributed neither to additional cascade feeding processes (for the excited $2p_{3/2}$ level) nor to further corrections to the electron capture process. This deviation was surprising also in the sense that REC is otherwise one of the best studied processes for bare and few-electron high-Z ions in relativistic collisions for which an excellent agreement between theory and experiment is typically found [4].

As an example, the observed Ly-α_1 angular distribution (full squares) measured for 309 MeV/u $U^{92+} \rightarrow N_2$ collisions is given in Fig. 1 as a function of the laboratory observation angle θ_{lab}. The experimental anisotropy coefficient was determined by normalizing the intensity of the investigated Ly-α_1 transition to that of the Ly-α_2 (+**M1**) radiation. Since the latter is isotropic in the projectile frame and energetically close to the Ly-α_1 line, this method allows us to strongly reduce the influence of possible systematic uncertainties. For the particular case displayed in Fig. 1, an effective anisotropy parameter β_{20}^{eff} of -0.23 ± 0.02 was deduced from a least-squares adjustment of Eq. (2) to the experimental Ly-α_1/Ly-α_2 intensity ratios by considering the correct relativistic angle and solid angle transformation (see, e.g., [8]). Following Eqs. (1) and (2), this means that REC favors the population of the magnetic substates $\mu_n = \pm 1/2$ [8].

For our present study we also evaluated the theoretical alignment parameters for the process of REC occurring in collisions of bare uranium with light gaseous targets. The actual computations were all carried out in the framework of Dirac's theory. The calculation of the alignment parameter $\mathcal{A}_2$ in Eq. (1) requires the evaluation of bound-free matrix elements for Dirac 4-spinors and was carried out already before [4,15]. Apart from the *impulse approximation* (see, e.g., [4]), no further approximation was made for the first step of the electron capture. As a result, the theoretical alignment parameters are the same as reported previously [15]. However, in order to compare with the experiments they need to be multiplied with the structure function $f(E1, M2)$.

In Fig. 2, we compare the experimental results (solid points) ([8]) for the effective anisotropy parameters β_{20}^{eff} with the corresponding theoretical findings (full line) as obtained from Eqs. (3) and (4). The dashed line, obtained assuming $f(E1, M2) \equiv 1$, i.e., neglecting the $E1$-$M2$ interference term, represents the theoretical treatment of the anisotropy parameter β_{20} as calculated by Eichler

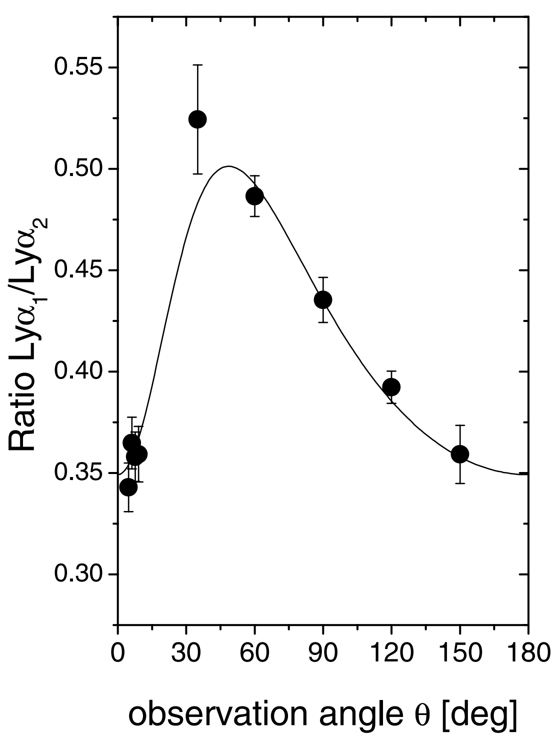

FIG. 1. Experimental Ly-α_1/Ly-α_2 intensity ratio (solid circles) measured for 309 MeV/u $U^{92+} \rightarrow N_2$ collisions (laboratory frame) [5]. The solid line depicts the result of the least-squares adjustment of Eq. (2) to the experimental data, considering the correct relativistic angle and solid angle transformation.

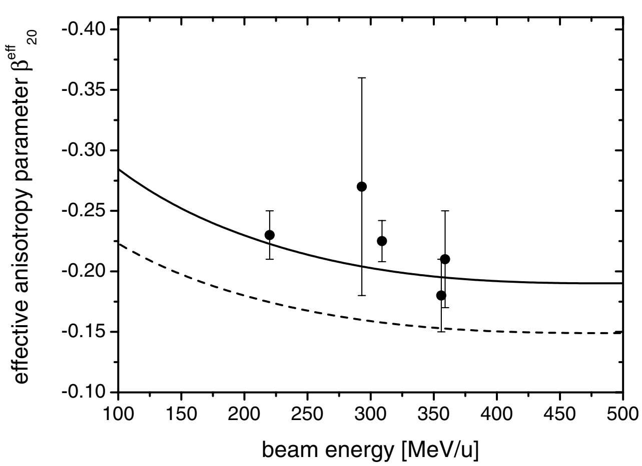

FIG. 2. The experimentally determined effective anisotropy parameters β_{20}^{eff} (solid points) for the Lyman-α_1 radiation of U^{91+} produced in $U^{92+} \rightarrow N_2$ collisions as a function of collision energy. The dashed line represents the theoretical predictions for $f(E1, M2) \equiv 1$, i.e., when the interference term is neglected. The solid line shows the theoretical β_{20}^{eff} parameter as defined by Eqs. (3) and (4).

et al. [15]. As seen from the figure, the former disagreement of the theoretical results from the experimental values [8] is removed when the interference term is taken into account.

In conclusion, an interference between the leading $E1$ decay channel and the—weak—$M2$ branch was studied for the case of the Lyman-α_1 transition in aligned hydrogenlike ions. This interference is found to affect considerably the angular distribution of the emitted photons. Similarly, it also affects the linear polarization of the Lyman-α_1 radiation, a topic which will be discussed in a forthcoming publication [21]. For the particular case of the Lyman-α_1 transition in the hydrogenlike uranium following electron capture, the former deviation between the experimental and theoretical findings for the alignment of the excited ion state [8] is removed when the interference correction is taken into account. Also, we have to add that one may expect similar sizable corrections for any other atomic transitions in the high-Z regime where beside the leading **E1** term, higher multipole contributions are small but allowed. Here, e.g., doubly excited states in He-like ions such as produced by dielectronic recombination must be mentioned [9,10]. More general, the study of decay rates and transition matrix elements of atomic transitions are of great importance to test and advance our basic knowledge about the physics of strong Coulomb fields as they are present at high-Z. However, at high-Z, most of these transitions exhibit such fast decay rates that lifetime measurements are excluded. Because of the sensitivity of the effective alignment parameter on the reduced matrix elements of the multipole transitions involved, the latter can be addressed by measuring precisely the associated photon angular distributions. It represents therefore an experimental tool to study the decay properties of atomic states in the realm of high-Z ions.

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