The Two-Photon Decay of 1s2s $^1S_0$ States in Heavy He-Like Atomic Systems

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Abstract. In He-like systems the decay of the 1s2s $^1S_0$ excited state to the $1s^2$ $^1S_0$ ground state is not allowed. This excited state can only decay to the ground state via the emission of two photons. The spectral shape of the emitted continuum is determined by the complete structure of the atomic system as all bound and continuum P states contribute to the 2E1 decay. For very heavy atomic systems the ^3P states ALSO have to be included and the normalized spectral shape changes with atomic number according to the relative strengthS of both, the electron-electron interaction and of the relativistic effects. A brief survey on the variation of the spectral shape of the two-photon continuum with atomic number is given and compared to experiments ranging from He-like Ni to He-like Au with special emphasis on the heavy relativistic system. The data compare well with fully relativistic calculations.

INTRODUCTION

Traditionally ATOMIC structure is probed by RADIATIVE transitions giving access to binding energies of the involved states. Atomic binding energies are calculated using matrix elements where the Hamiltonian is multiplied from both sides BY the same wave function. Here, variational methods WHICH MINIMIZE the binding energies PROVIDE ACCURATE RESULTS. Lifetime measurements are more sensitive TO the exact shape of the density distributions of the ELECTRONS as the overlap of the wavefunctions weighted by the interaction determines the transition rates. This is correspondingly true for line shapes, in particular for LINE widths. For inner-shell transitions in heavy atomic systems the transition rates are pretty large and HENCE, for fast allowed transitions, lifetimes or line widths are difficult to measure. There, only for metastable states or for "forbidden transitions" CAN lifetimes or linewidths be determined experimentally giving direct access to the structure of the wave functions.

In He-like systems a direct transition from the "metastable" 1s2s $^1S_0$ state to the $1s^2$ $^1S_0$ ground state is absolutely forbidden by selection rules. This state can only decay by the emission of two photons to the ground state, i.e. by the 2E1 decay. To some EXTENT, the same is true for the excited 2s $^2S_{1/2}$ state in H-like ions; however, there a direct M1 decay to the 1s $^2S_{1/2}$ ground state COMPETES with the 2E1 decay. TWO-photon decay was treated theoretically by Göppert-Mayer [1] MORE than 70 years ago. The 2E1 decay rate is determined by a summation over all two-photon transitions possible via all intermediate P states of the atomic system obeying the RULE THAT the sum energy of the two photons corresponds to the total binding energy difference BETWEEN the initial and final state. The summation includes all relevant bound and continuum states. The differential transition probability $W_{2\gamma}$ has the form [1]:

$$dW_{2\gamma} / d\omega_1 = \{\omega_1 \cdot \omega_2 / (2\pi \cdot c)^2\} \cdot |\mathbf{M_{2\gamma}}|^2 \cdot d\Omega_1 \cdot d\Omega_2$$

where ω_j is the energy and $\mathbf{d}\Omega_j$ the solid angle for the j*th* photon and the transition energy ω_0 satisfies ENERGY conservation $\omega_0 = \omega_1 + \omega_2$. The second order matrix element $\mathbf{M_{2\gamma}}$ is given by the expression:

$$\mathbf{M_{2\gamma}} = \boldsymbol{\varepsilon_1} \cdot \boldsymbol{\varepsilon_2} \, \Sigma \, \{ < 1\ {}^1S_0 \,||\, R_{E1}(\omega_2) \,||\, n > < n \,||\, R_{E1}(\omega_1) \,||\, 2\ {}^1S_0 > / \, (E_n - E_{2'So} + \omega_1)$$
$$+ < 1\ {}^1S_0 \,||\, R_{E1}(\omega_1) \,||\, n > < n \,||\, R_{E1}(\omega_2) \,||\, 2\ {}^1S_0 > / \, (E_n - E_{2'So} + \omega_2) \}$$

Here, $\boldsymbol{\varepsilon_j}$ is the polarization vector for the j*th* photon and $R_{E1}(\omega_j)$ is the electric dipole operator. It has to be mentioned that the summation RUNS over all intermediate states n independent OF whether they are empty or occupied [2].

Integrating finally over all possible photon energies yields the total transition rates that are SENSITIVE to the structure of the complete atomic system. Measurements of lifetimes for the 1s2s ${}^1S_0$ and the 2s ${}^2S_{1/2}$ levels in He- and H-like ions, respectively, and their Z dependences give therefore important information on the total structure of heavy few-electron ions; for an overview see e.g. Refs. [3, 4]. The 2E1 transition rates increase strongly with Z – in first approximation with the sixth power of the atomic number, Z^6 – due to the increase in total transition energy with Z. For our cases of interest, heavy He-like ions, the possible different groundstate transitions and their rate dependences are indicated in the level diagram shown in Fig.1 for a heavy L-shell excited ion. An overview OF the rate dependences is given e.g. in Ref. [5] and in the literature cited there.

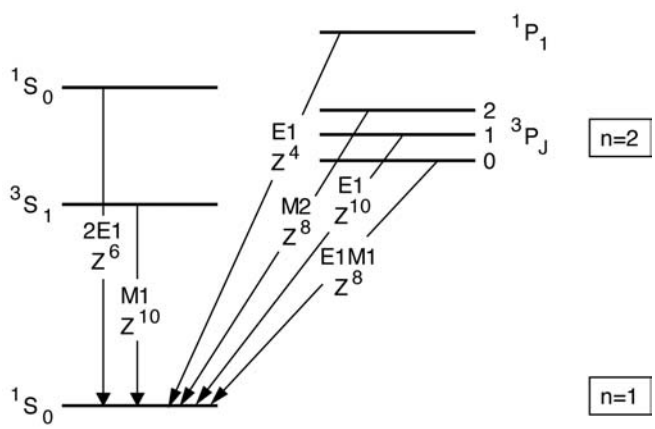


FIGURE 1. Level diagram for heavy He-like ions, possible groundstate transitions and their rate dependences are shown, cf. [5].

THE TWO-PHOTON CONTINUUM

In contrast to lifetime measurements, a determination of the complete two-photon decay spectrum will provide much more detailed information on the complete structure of He-like atomic systems. Here, the shape of a measured two-photon continuum has to be compared with the energy differential transition probability, $dW_{2\gamma} / d\omega_1$, given above. As the two photons are indistinguishable, the spectrum is symmetric IN EACH PHOTON, i.e. it is mirror symmetric around the mid point at half the total transition energy, $\omega_0 / 2$. For convenience in comparing

spectra from different atomic systems, the total transition energy is best normalized to "1" with the fractional energies of the two photons $f_j = \omega_j / \omega_0$ and $f_1 + f_2 = 1$. In this representation the differential transition probability for TWO-photon decay can be approximated by a product of a simple "phase space factor", $f_1 \cdot (1 - f_1)$, and a "structure factor", c.f. Ref. [6]:

$$dW_{2\gamma} / d\omega_1 \propto f_1 \cdot (1 - f_1) \bullet \left| \mathbf{M_{2\gamma}} \right|^2$$

The phase space factor HAS THE shape of a simple parabola and the structure factor is only determined by the square of the energy differential matrix element $\mathbf{M_{2\gamma}}$.

The structure factor contains all the information on the complete atomic system and varies considerably with ATOMIC SPECIES [7-9]. Just CONSIDERING THE WIDTHS (FWHM) of the SPECTRA, THE STRUCTURE FACTOR NORMALLY BROADENS THE DISTRIBUTIONS IN A WAY THAT DEPENDS ON THE SYSTEM. For H-like systems the FWHM of the continuum starts at a value of 0.82 (in relative units of ω_0) for H and decreases WITH INCREASING ATOMIC NUMBER due to relativistic effects. FOR H-like U THE FWHM IS about 0.71, which is also the value for a pure parabola, cf. Ref. [8]. For He the FWHM starts AT 0.75, A NARROWER WIDTH COMPARED TO H. THIS NARROWING IS CAUSED BY THE ELECTRON-ELECTRON INTERACTION. Due to the relative decrease of this interaction with atomic number Z compared to that of the central potential the width for He-like ions widens first towards the width of H-like systems before around the mid-Z region WHERE the relativistic effects turn the tendency back, cf. Ref. [9]. Finally the width for He-like U approaches that of H-like U – an indication that H- and He-like atomic structures approach each other for the heaviest atomic species. However, the widths are only an integral measure OF THE STRUCTURE, more details can be gained by STUDYING the DETAILED shape of the photon continuum.

A recent survey on the two-photon decay in heavy He-like systems is given in Ref. [10] with special emphasis on the change of the spectral shape with atomic number Z. There, the two-photon decay in singly K-shell ionized heavy atomic systems is ALSO included. These quasi He-like atomic systems were investigated in detail by Ilakovac and his group [11 – 13]. Here, we concentrate only on heavy true He-like ions. In Fig.2 the shape of the two-photon distribution and its change with Z is presented for He-like ions. The structure factors for He, Ni^{26+} and Au^{77+} are given along with the form of the parabola according to calculations BY Derevianko and Johnson [9]. The structure factors for He and Au^{77+} are pretty similar whereas that for Ni^{26+} deviates considerably broadening the parabolic shape of the spectrum.

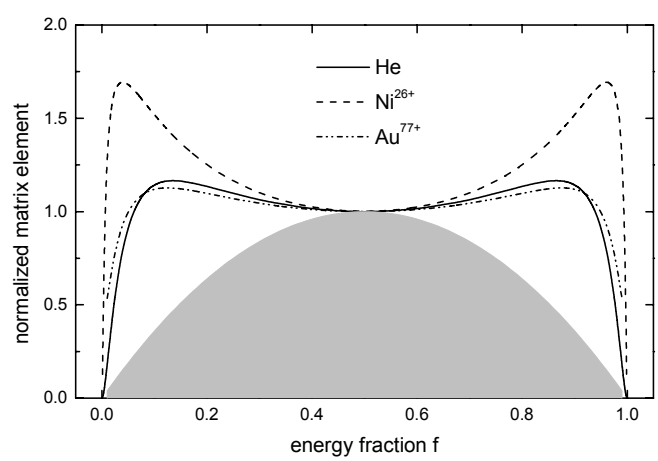
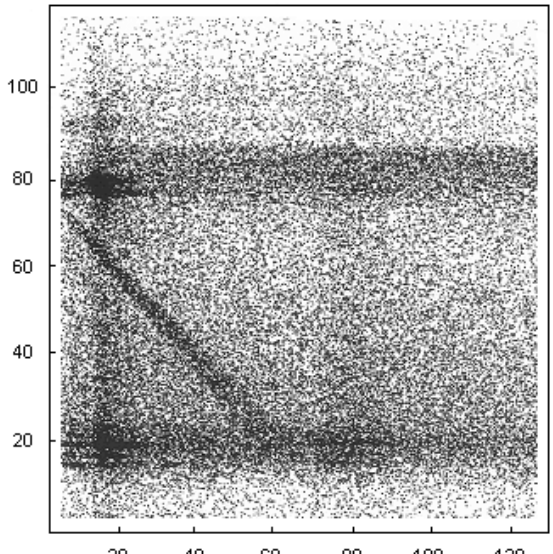


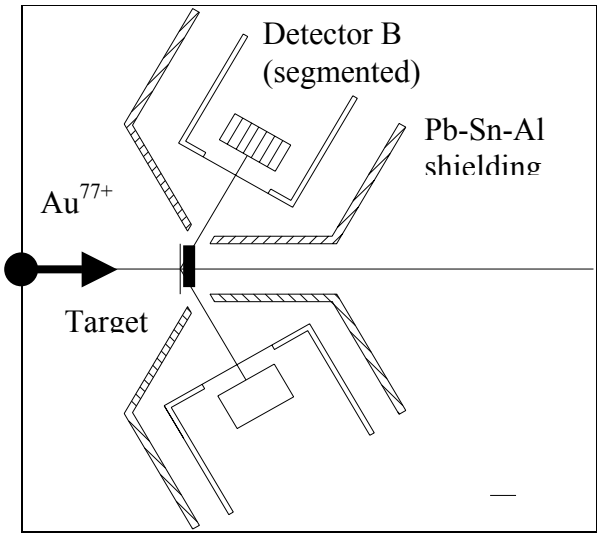
FIGURE 2. The shape factors for the 2E1 decay continuum in He-like ions. The structural factor for He, Ni^{26+} and Au^{77+} are given as normalized matrix elements (squared) along with the parabolic shape of the phase-space factor (rim of the shaded area) as a function of the fractional photon energy, acc. to [9].

EXPERIMENTAL TECHNIQUES

In order to produce He-like heavy ions the acceleration-stripping technique is NORMALLY used, where high velocities are needed FOR HEAVY IONS. To EFFICIENTLY produce He-like species of Ni and Au typical energies around 10 and 100 MeV/u are needed (compare the binding energies for the K electrons of about 9 and 80 keV), respectively. Then, the He-like fast ions are magnetically selected before they are excited in a second "exciter foil" with a certain probability to the 1s2s $^1S_0$ state. The lifetime of this state is 154 ps in Ni^{26+} [14] and only 0.32 ps in Au^{77+}[15]. The radiative decay of these excited states IS detected behind the foil by at least two solid-state x-ray detectors (Si(Li) and Ge(i) detectors, respectively) looking face-to-face perpendicular to the downstream ion beam. The two photons of the decay are preferentially emitted under 180^0 and are detected there in coincidence.

Due TO the short lifetimes of the Au ions these ions decay CLOSE TO the exciter foil despite their high velocities. Moreover, in this relativistic velocity regime the Lorentz transformation of the photon emission angle from the fast ion system into the laboratory system leads to a forward tilt of the "90^0" observation angle [16, 17]. the experimental arrangement for the Au^{77+} case is given ON THE RIGHT SIDE OF Fig.3 . In order to be able to compensate for the Doppler effect of the emitted photons AT different observation angles one of the Ge(i) detectors is granularly divided into stripes. For a precise determination of the true two-photon SPECTRUM, emission angels, detector efficiencies, photon transmission losses and electronic coincidence efficiencies have to be determined with high accuracy. Fig.3 gives a contour plot FOR THE AU EXPERIMENT of coincident photons measured with both the detectors. The ridge along the diagonal line describes the wanted two-photon continuum convoluted with all the relevant efficiencies.





Detector A 1 cm

FIGURE 3. Contour plot for the coincident emission of two photons for Au ions (106 MeV/u Au^{77+} excited in Al foil; detector A vs. detector B, photon energies in keV). The experimental arrangement is given at the right side. Along the diagonal ridge in the contour plot the sum energy of both photons is constant and corresponds to the total transition energy of the 2E1 decay [16, 17].

RESULTS

Due to the COMPLICATED efficiency RESPONSE OF THE DETECTION SYSTEM, the measured coincident two-photon spectra cannot be compared directly to theory. Instead of unfolding the measured spectra with all the uncertainties INVOLVED, the theoretical spectral distributions are normally convoluted with all the relevant efficiencies and then compared directly with the experimental ones. This convolution is done best by Monte Carlo simulations starting from the theoretical spectral distribution and then including all the experimental correction factors, cf. Ref. [4]. This method was successfully introduced for the case of the two-photon continuum in He-like Kr^{34+} ions [4]. A clear change in the relative spectral distribution for the two-photon decay compared to the one for atomic He was established. Unfortunately, the systematic errors are still somewhat large. A further improvement in accuracy was achieved for Ni ions by comparing the two-photon decay for H-and He-like species, Ni^{25+} and Ni^{24+}, respectively [6]. Assuming that the spectral distribution for H-like ions can be treated correctly all the experimental efficiencies can be extracted with high accuracy; and then, the spectral distribution for the He-like case can be compared to theory yielding the most precise determination for the two-photon continuum in heavy He-like ions done up to now. However, at this Z range the relativistic effects ON THE SPECTRUM are still too small to be uniquely detected. Relativistic [9] and non-relativistic [18] calculations still deliver here quite similar distributions.

For the heaviest investigated system, He-like Au^{77+} ions, relativistic effects are strong. Hence, a comparison of the experimental spectrum with the simulation is displayed at the top of Fig.4 [17]. Within the error band for the simulation and the experimental error BARS, good agreement results between measurement and fully relativistic theory. For comparison, a corresponding simulation for the non-relativistic case of Ar^{16+} ions is also given (dashed line). At the bottom the residues of the experimental data with respect to the results of the simulations are given. It is evident that the non-relativistic approach is at variance with the experimental findings, only the fully relativistic approach is in agreement.

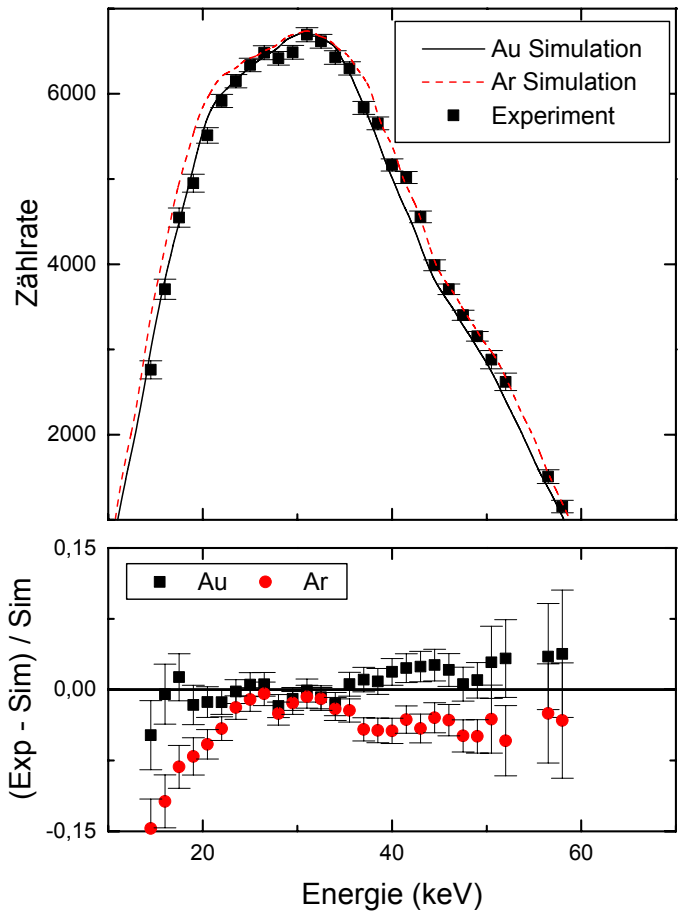


FIGURE 4. The x-ray continuum for the two-photon emission from He-like Au^{77+} ions (top part) [17]. The measured data points are shown along with Monte Carlo simulations one for fully relativistic Au^{77+} ions and one for non-relativistic Ar^{16+} ions (dashed line). At the bottom the residues of the experimental data points compared to the simulations are given.

Taking out the trivial phase space factor from the continuous spectrum one OBTAINS the energy dependent square of the matrix element, see Fig.5. For comparison the normalized square of the theoretical matrix elements are shown for He-like Ni and Au ions along with the experimental data for Au^{77+}. The difference in THE distributions demonstrates quite clearly the relativistic influence, whereby all intermediate ^3P states are included in the calculations [9]. The experimental points evidently favor the fully relativistic Au distribution; a χ^2 test gives values of 3.48 and 7.35 for the Au and Ni predictions, respectively. However, it is also evident, that additional and more precise measurements are needed for a more refined comparison with theory.

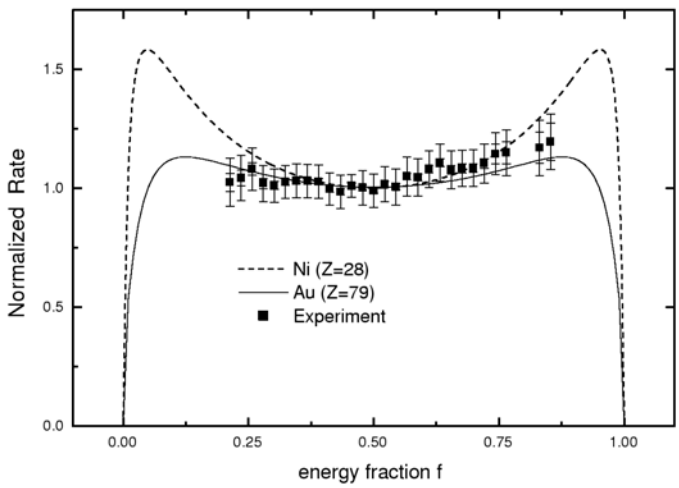


FIGURE 5. The normalized square of the energy dependent matrix element (rate) extracted from the experiment is compared to the fully relativistic theory for Au^{77+} and Ni^{26+} (full and dashed lines, respectively [9]).

OUTLOOK

Both experimentally and theoretically, important steps towards a better understanding of the complete atomic structure of He-like heavy ions were made recently by investigating the two-photon decay continuum associated with the excited 1s2s $^1S_0$ state. Recently, non-relativistic approaches [18] valid up to the MEDIUM Z region (around 30) were complemented by fully relativistic calculations [9] including all intermediate ^3P states. Measurements in the medium Z region STILL compare well with BOTH calculations as the relativistic effects are not yet dominant there [4, 6]. For a true relativistic system, Au^{77+}, the first experiment clearly favors the fully relativistic calculation. Higher accuracies in future experiments are certainly feasible and expected for the next experiments using the described beam foil excitation technique.

A recent experiment demonstrated a new method to produce the 1s2s $^1S_0$ AT MODERATE RELATIVISTIC ION VELOCITIES. IT WAS FOUND THAT selectively IONIZING the K shell of L-like ions leads with HIGH efficiency TO THE 1s2s $^1S_0$ excited state [19]. This new experimental method may give IMPROVED access to this area. Moreover, with the THIRD generation synchrotron radiation facilities new methods are at hand to produce singly K ionized

atoms of any atomic number. These singly K ionized atoms decay A SMALL FRACTION OF THE TIME via the two-photon decay branch and THUS give information on the complete atomic structure of multi-electron systems which finally can be compared to the corresponding true He-like cases. IN THE PAST, MOST OF THE DATA ON INNERSHELL TWO-PHOTON DECAY WERE OBTAINED FROM nuclear unstable species decaying by nuclear K electron capture. THIS METHOD WAS demonstrated by Ilakovac and his group [11 – 13]. Further measurements, both at heavy ion accelerators and at modern synchrotron radiation facilities, will give unprecedented access to the complete atomic structure of He-like or quasi-He-like atomic systems in the strong field domain of the heaviest atomic species.

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