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Spectral distribution of the $2S \rightarrow 1S$ two-photon transition in atoms and few-electron ions

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Abstract. The two-photon decay of the $2S$ state to the ground state in dressed atoms and one- or two-electron ions has been studied for several decades. Relativistic calculations have shown an Z-dependence of the spectral shape of this two-photon transition in one- or two-electron ions. We have measured the spectral distribution of the $1s2s\ ^1S_0 \rightarrow 1s^2\ ^1S_0$ two-photon transition in He-like tin at the ESR storage ring using a new approach for such experiments. In this method, relativistic collisions of initially Li-like projectiles with a gaseous target were used to populate exclusively the first excited state, $1s2s$, of He-like tin, which provided a clean two-photon spectrum. The measured two-photon spectral distribution was compared with fully relativistic calculations. The obtained results show very good agreement with the calculations for He-like tin.

Keywords. Two-photon decay; ionization; highly-charged ions.

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1. Introduction

Two-photon decay process was discussed first by Göppert-Mayer [1,2] in the 1930s for hydrogen and later it was described in detail for hydrogen and helium using non-relativistic approach by Breit and Teller [3]. The $2S$ state decays to ground state primarily by the emission of two electric-dipole photons, i.e. $E1E1$ or $2E1$. In this decay process, transition between quantum levels occurs via simultaneous emission of two correlated photons of energies $\hbar\omega_1$ and $\hbar\omega_2$ under the boundary condition that sum of the energies of both the photons equals the total transition energy $\hbar\omega$, i.e. $\hbar\omega_1 + \hbar\omega_2 = \hbar\omega = E_\mathrm{i} - E_\mathrm{f}$. Here E_i and E_f represent the energies of the initial and final states, respectively, of the transition. The differential transition probability (or decay rate) for the simultaneous emission of two-photons is given by [4]

$$A(\omega_1)\mathrm{d}\omega_1 \propto \omega_1\omega_2|M_{fi}|^2\mathrm{d}\omega_1, \tag{1}$$

where ω_1 and ω_2 are the frequencies of the emitted photons. The matrix element $|M_{fi}|$ is defines as

$$|M_{fi}| = \sum \left\{ \frac{\langle f\|D_1\|n\rangle\langle n\|D_2\|i\rangle}{\omega_{ni}+\omega_1} + \frac{\langle f\|D_2\|n\rangle\langle n\|D_1\|i\rangle}{\omega_{ni}+\omega_2} \right\}, \tag{2}$$

where $|i\rangle$, $|n\rangle$, and $|f\rangle$ represent the initial, intermediate and finalstate, respectively. D_i is the photon field operator and ω_i is the frequency of transition i. The frequency difference ω_{ni} in the denominator is defined as $\omega_{ni} = \hbar^{-1}(E_n - E_i)$. The total two-photon decay rate A_T is obtained by integrating eq. (1) over frequencies, i.e.,

$$A_\mathrm{T} = \frac{1}{2}\int_0^{\omega_{if}=\omega} A(\omega_1)\mathrm{d}\omega_1. \tag{3}$$

As both the emitted photons are indistinguishable, the factor $1/2$ in the above equation takes into consideration the double counting of photon ω_1 in the frequency interval $[0, \omega_{if}]$. The total decay rate of the two-photon increase as Z^6 [4–10], Z is the nuclear charge of the atom/ion.

The two-photon transitions are especially strong between those states, where a single photon decay is strictly forbidden by the conservation of angular momentum, e.g. from $J = 0$ to $J = 0$. For $2S \rightarrow 1S$ two-photon transition, the energies of the individual photons form a continuous distribution which has maximum intensity at half of the total transition energy, i.e. $f \equiv \hbar\omega_1/\hbar\omega = 0.50$, and drops to zero at both the end points [8–11]. In contrast to the one-photon decay, which depends on the initial and final states of the transition, the energy distribution of the two-photon continuum is determined by the summation over all intermediate (bound and continuum) states of the atom or ion [8–11]. Hence, the spectral distribution of two-photon transition is sensitive to the entire atomic structure.

In 1973, Freund [12] suggested that two-photon transition between inner-shall vacancy in initially neutral atom could be measurable despite its very feeble decay rates compared to the single photon. Afterwards, several theoretical work, both non-relativistic [13,14] and relativistic [15,16], have been reported. Experimentally, $2S \rightarrow 1S$ two-photon decay process has been measured for selective cases in the decay of K-shell vacancy in initially neutral atom using the photon–photon coincidence technique [14,17–22]. In these measurements, the K-shell vacancy is produced by irradiating the targets by photons [14,17,18] or by using radioactive isotopes that preferably decay by nuclear electron capture [19–22]. All these measurements confirmed the existence of the two-photon decay branch in singly ionized atoms. Further detailed information about two-photon decay from singly-ionized atom can be obtained from Mokler and Dunford [23] and Ilakovac *et al* [24].

Several lifetime measurements were performed for the $2S_{1/2}$ state in H-like ions and the $2\,^1S_0$ state in He-like ions for atomic number $Z < 50$ [23]. In spite of the high precision, the measured lifetime could only test the $2E1$ decay probability summed over all continuum photon energies. Furthermore, the measurements become very difficult for the high-Z systems in which the lifetimes of the $2S$ state are very short. On the other hand, measurements of the $2E1$ energy distribution, which is sensitive to the entire atomic structure, are possible for all elements throughout the periodic table. Moreover, the energy distribution of the $2E1$ transition provides detailed information about the influence of relativistic corrections to the two-photon transition.

For H-like ions, the $2E1$ decay is the dominant channel from the $2S_{1/2}$ state for light atoms, whereas for heavy atoms ($Z > 50$) the $2S_{1/2}$ state decays almost exclusively via a magnetic dipole $M1$ – spin-flip transition [8]. Non-relativistic theoretical calculations predict that the spectral distribution of the $2E1$ transition from the $2S_{1/2}$ state of H-like ions is independent of Z, whereas the relativistic calculations show monotonous decrease of the full-width at half-maximum (FWHM) of the spectral distribution of $2S_{1/2} \rightarrow 1S_{1/2}$ two-photon transition as nuclear charge Z is increased.

Helium-like ions are the most suitable candidates for the two-photon decay investigations in high-Z domain. For the He-like ions, relativistic computations [9,10] show an increase of the FWHM of the energy distribution up to around $Z = 20$ and a decrease if Z is further increased. This variation in the energy distribution of the emitted photons is caused by relativistic and electron–electron interaction effects on the wave functions and energies. Therefore, a measurement of the spectral distribution of the two-photon decay along the isoelectronic sequence of He-like ions probes uniquely our understanding of the interplay between electron–electron correlation and relativistic effects on the structure of the simplest multielectron atomic systems. Experimental efforts were made in the past to accurately determine the spectral distribution of $2^1S_0 \rightarrow 1^1S_0$ transition for several He-like ions (Ni [25], Ge [26], Kr [27] and Au [28]). In these experiments, the $2E1$ decay was selected by the simultaneous detection of the two photons by two detectors, i.e. photon–photon coincidence measurements. These experiments are characterized by a very low efficiency, which results from the efficiency product of the two independent photon detectors.

In this report, we present experimental two-photon spectra by the decay of the 2^1S_0 state of He-like tin. The spectra were measured using an alternative method of exclusive K-shell ionization of initially Li-like ions. It is noted that the measured spectra, obtained by the present technique, are the cleanest $2E1$ spectra obtained so far. The results show that the spectral distribution is in good agreement with fully relativistic calculations and clearly distinguishes the continuum two-photon spectral distribution of He-like tin from that of other He-like ions.

2. Experimental details

The experiment was performed using the heavy-ion accelerator facility at GSI, Darmstadt. The He-like tin ions with an initial energy of 300 MeV/u, delivered from the UNILAC/SIS complex, were accumulated in the ESR storage ring. Typically, 10^8 ions were stored and electron-cooled in the ring. The momentum spread of the ion beam was

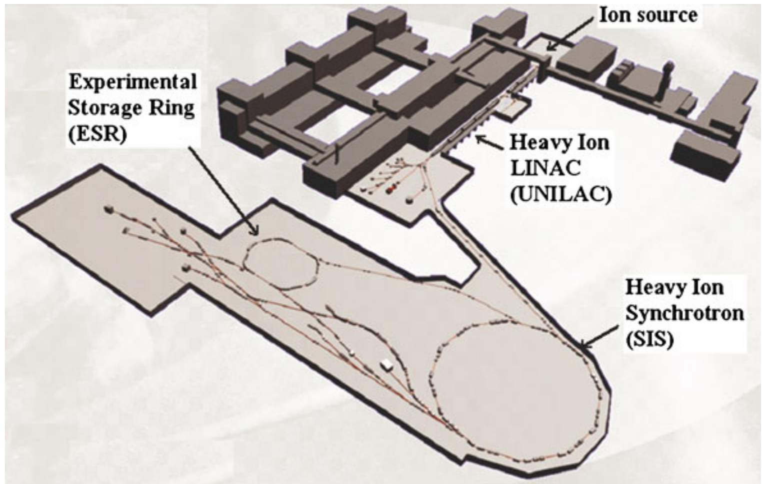

Figure 1. Lay-out of the accelerator facility and experimental areas at GSI, Darmstadt.

close to $\Delta p/p \approx 10^{-5}$. The well-collimated (∼2 mm diameter) ion beam was made to collide with a supersonic gas-jet nitrogen molecular target beam of around 5 mm diameter. The areal density of the target used was about 10^{12} particles/cm^2. After crossing the target region, the projectiles which lost an electron were separated from the primary beam downstream behind the dipole bending magnet of the ring. The up-charged tin (He-like) ions were registered by position-sensitive particle detectors (multiwire proportional

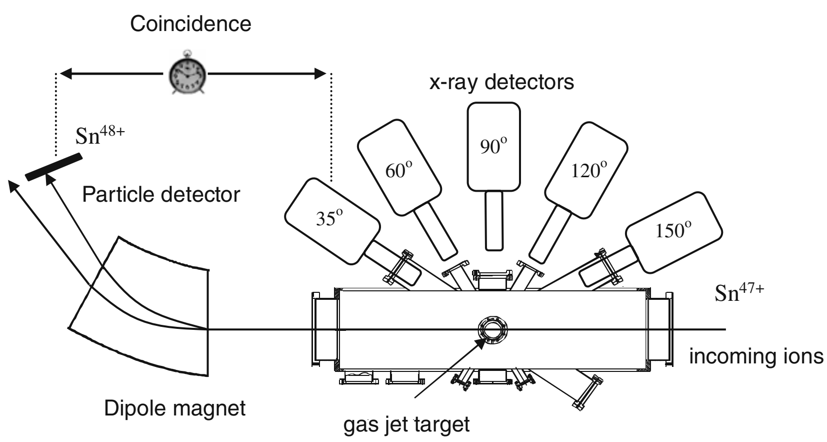

Figure 2. Lay-out of the experimental set-up at the internal jet target. X-ray detectors view the target interaction zone at different observation angles. Photon emission was observed in coincidence with the up-charged ions, detected in the particle counter located behind the dipole magnet.

counters). Schematic of the GSI accelerator facility and the experimental set-up at the target area of the ESR storage ring are depicted in figures 1 and 2, respectively.

The photons emitted from the beam–target interaction region were detected by a planar HPGe detector placed at 35° observation angles with respect to the beam axis. The detector was separated from the ultra-high vacuum of the ring (10^{-11} mbar) by beryllium window (of 100 μm thick). As the source was moving, a 4-mm wide copper–lead slit was mounted in front of the detector to reduce the Doppler broadening. Finally, the X-ray detector was energy- and efficiency-calibrated using thin ^{57}Co and ^{241}Am radiation sources. For details of the experimental set-up, see [29,30].

3. Results and discussions

The X-ray spectra were collected in coincidence with the He-like tin ion (Sn^{48+}). The measured spectra contained a broad continuum, resulting from the $2E1$ decay of the 2^1S_0 state ($1s2s\ ^1S_0 \rightarrow 1s^2\ ^1S_0$), and a monoenergetic line corresponding to the $M1$ decay of the 2^3S_1 state ($1s2s\ ^3S_1 \rightarrow 1s^2\ ^1S_0$). In the obtained spectra, the absence of any additional X-ray lines from the decay of the $1s2p$ state or other higher levels indicate that there is a very selective K-shell ionization of the Li-like tin ions and subsequent exclusive formation of the corresponding $1s2s^{1,3}\ S$ states of He-like tin. Such a high level of selectivity of the K-shell ionization has been reported earlier by our group for relativistic collisions of Li-like U ions [29]. For further analysis of the observed X-rays, the higher-energy half of the spectrum (figure 3a) was selected owing to two reasons: (1) the $2E1$ energy distribution is symmetric because of an equal energy sharing [9,10] and, hence, either of the halves can be utilized to provide the Z-dependent information and (2) the higher-energy half is clean and the detector efficiency is well known in the present energy range.

The response function of the detector was simulated for the $M1$ transition and compared to the measured spectrum. The detector simulations were found to be in good agreement with the measured position and intensity of the K-escape peaks. Furthermore, the theoretical $2E1$ spectral distribution was convoluted with the detector response function for comparison with the measured spectral shape. The intensity of the theoretical two-photon decay continuum was normalized to the measured spectra at the reduced photon energy of 0.50, i.e., for half of the energy of the $1s2s\ ^1S_0 \rightarrow 1s^2\ ^1S_0 : 2E1$ transition (figure 3). To check the sensitivity on Z, the measured spectral shape for tin was compared with relativistic theoretical calculations [9,10] for different He-like ions, convoluted with the simulated detector response function. Figure 3b depicts the reduced χ^2 of the fits between the experimental data for Sn^{48+} and the calculated spectral shapes for different He-like ions using the Minuit minimization code. The results show good agreement with the relativistic calculations only if a proper nuclear charge was chosen, thus confirming the sensitivity on Z. Detailed information of the presented measurement can be found in our recent publication by Trotsenko *et al* [32].

4. Summary and future scope

In summary, we measured the spectral distribution of the two-photon decay of the 2^1S_0 state in He-like tin. The measurement was performed using a new experimental approach

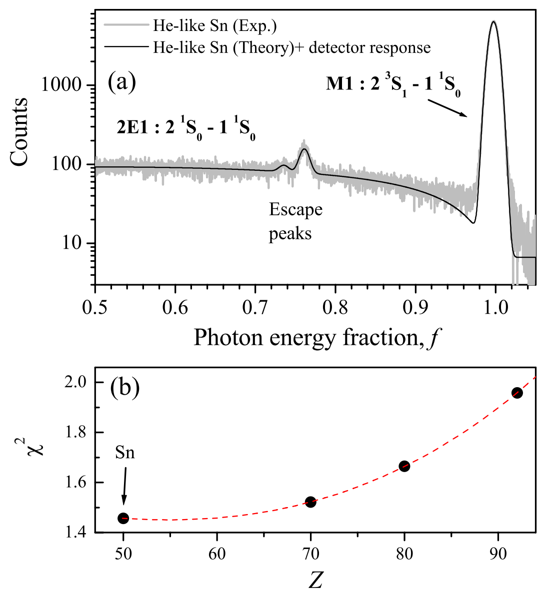

Figure 3. (**a**) Higher-energy part of the projectile X-ray spectra (gray line) for $Sn^{47+} \rightarrow N_2$ collisions measured in coincidence with the Sn^{48+} ions. Black line represents the spectral distribution calculated using the fully relativistic approach for He-like Sn, convoluted with the simulated detector function and Doppler broadening [31,32]. Abscissa is the fraction of the total transition energy carried by either of the photons; (**b**) reduced χ^2 obtained by fitting calculated spectral distribution for different He-like ions to the measured distribution for He-like Sn.

at the ESR storage ring where relativistic collisions of Li-like projectiles with a gaseous target were used to form the desired initial state, which allowed for a measurement of the clean two-photon energy distribution. Our results [32] show the best agreement between theory and experiment for the correct nuclear charge (Sn, $Z = 50$), thus confirming the prediction of relativistic calculations for the Z sensitivity of two-photon spectral distribution.

In the dipole $2E1$ approach for the $2S \rightarrow 1S$ transition, the photon–photon correleation is expected to have $1 + \cos^2\theta$ behaviour, where θ is the opening angle between both the photons, which, therefore, shows symmetry with respect to $\theta = 90°$. Recently, exact relativistic calculations of the photon–photon correlation for two-photon decay from $2S_{1/2}$ state of H-like ions [33] and 2^1S_0 state for He-like ions [34] for few medium and high-Z ions were reported to have an asymmetric shift in the angular correlation function. This asymmetry is expected because of the contribution of higher-order multipoles, which have negligible contribution compared to the $E1E1$ in the spectral distribution spectrum. Further experiments on the photon–photon correlation studies in the two-photon decay of high-Z ions are planned for the future.

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