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THE TWO-PHOTON DECAY OF 1s2s $^1S_0$-LIKE STATES IN HEAVY ATOMIC SYSTEMS

PAUL H. MOKLER[a] and ROBERT W. DUNFORD[b]

[a] *GSI, Planckstrasse 1, D-64291 Darmstadt, Germany*

[b] *ARGONNE National Laboratory, Argonne, IL 60439, U.S.A.*

Dedicated to Professor Kseno Ilakovac on the occasion of his 70th birthday

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In He-like systems the direct decay of the 1s2s $^1S_0$ state to the 1s^2 $^1S_0$ ground state is “forbidden”. The transition 1s2s $^1S_0 \rightarrow$ 1s^2 $^1S_0$ by two photons sensitively probes the structure of the complete atomic system. In particular, the shape of the two-photon spectrum is sensitive to it and also reveals for heavy atomic numbers details of relativistic effects in strong central fields. A brief survey on this field of research is given with special emphasis on high nuclear charge Z.

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1. *Atomic structure investigations*

Atomic structure is traditionally probed by measuring photonic transitions. In particular, transition energies give access to the binding energies of the involved states. Theoretically, binding energies are determined by matrix elements where the Hamiltonian is multiplied twice with the same wave function. Best results can be gained by variational principles minimizing the binding energy. Lifetime measurements are more sensitive to details in the structure of the wave functions as the overlap of the two involved wave functions weighted by the interaction determines the transition rates. In this connection, the exact shape of a transition line, in particular its width, has also to be considered. Due to the high transition rates for inner shell transitions in heavy atomic systems, lifetimes or line widths are difficult to measure for the fast, allowed transitions. Only for metastable states or for so-called forbidden transitions are the rates small enough to get experimental access to lifetimes and possibly also to line widths, and hence access to the structure of the wave functions.

A very special case is the two-photon decay of the 2s $^2S_{1/2}$ state in hydrogen-like systems as well as of the 1s2s $^1S_0$ state in helium-like atomic systems. In the He-like case, the 1s2s $^1S_0$ state can only decay via two-photon emission to the $1s^2$ $^1S_0$ ground state; a direct one-photon transition is absolutely forbidden for that case. Theoretically, two-photon decay was originally treated by Göppert-Mayer [1] seventy years ago. This decay rate is determined by a summation over all possible transitions via intermediate P states of the atomic system with the boundary condition that the sum energies of the two photons corresponds to the total binding energy difference of the initial and final states. In this connection, it is important to note that the summation has to run over all intermediate states independent of the fact whether these states are empty or occupied [2]. The summation includes all bound and continuum states. Integrating over all possible photon energies yields the transition rates that are already sensitive to the structure of the complete atomic system. For an overview of lifetime measurements for heavy He-like ions see Refs. [3] and [4]. The two-photon transition rates increase with Z^6, the sixth power of the atomic number. In Fig. 1, a level diagram with the different possible transitions and their rate dependences on Z is shown for heavy He-like ions. For an overview on the rate dependencies on Z for the inner-shell transitions in He-like systems see, e.g., Mokler et al. [5].

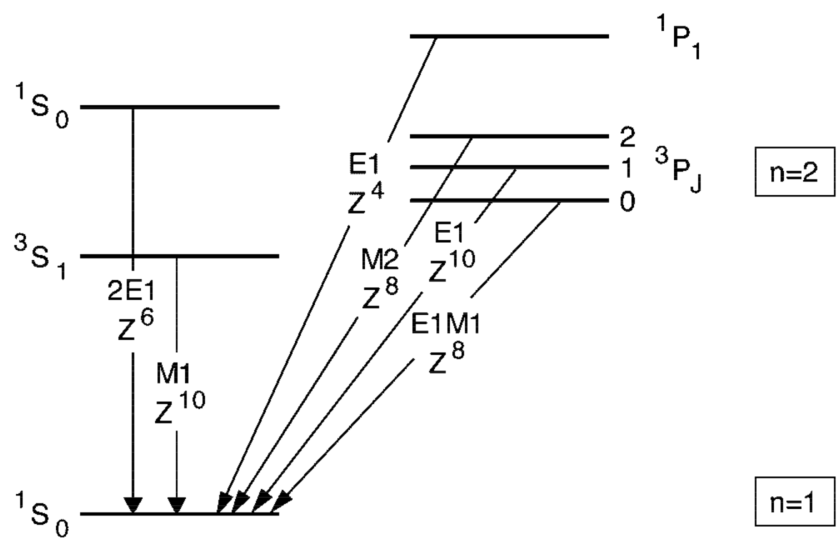

Fig. 1. Level diagram for heavy He-*like ions, possible transitions and their rate dependence on Z are indicated.*

In contrast to lifetime measurements, a determination of the two-photon decay spectrum will provide much more information on the details of the atomic structure involved. The spectrum is symmetric with respect to both photons, i.e. mirror-symmetric around the mid point at half the total transition energy. Actually, the spectrum can be considered as a product of a phase space factor showing a simple parabola $f_1(1-f_2)$, where f_1 and f_2 are the fractional energies of the two photons, with $f_1+f_2=1$ corresponding to the total transition energy, and a structure factor determined by the transition matrix element and depending only on the details of the structure of the atomic system [6]. This structure factor will vary considerably with the atomic system, H-like, He-like or singly K-ionized atom, and with atomic number Z [7–9]. In particular, for high Z values in He-like systems, the strong central field will overwhelm completely the influence of electron-electron correlation so that relativistic effects will dominate the atomic structure. Here, beyond the summation over all intermediate ^{1}P states, the ^{3}P states have also to be included [9]. In Fig. 2, the reduced matrix elements for the 2E1 decay as function of the normalized transition energy, according to calculations of Derevianko and Johnson [9] are shown. In other words, the phase space factor (see the shaded area in Fig. 2) is already divided out from the energy spectrum for the two-photon distribution.

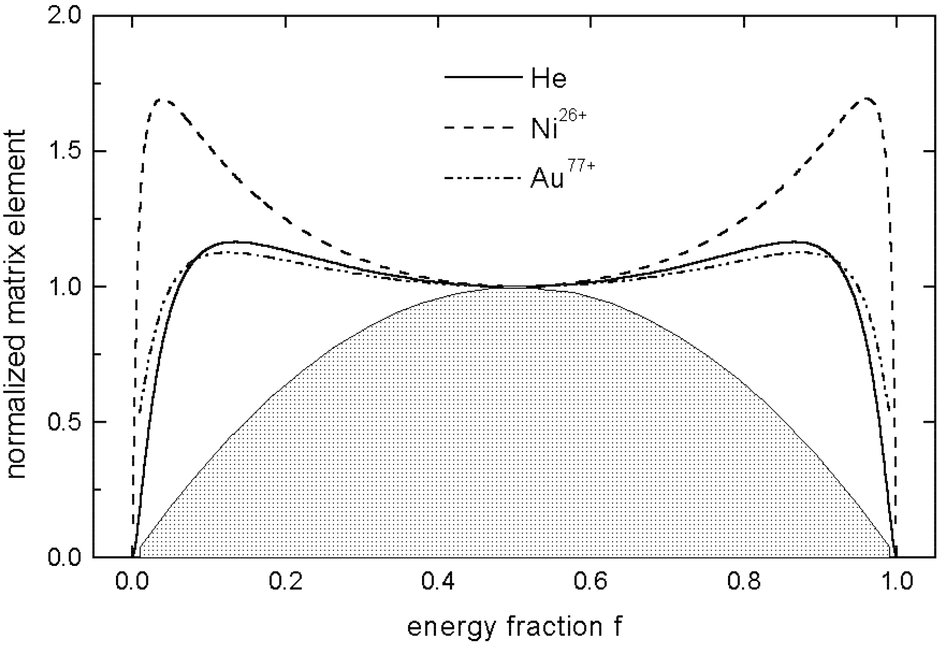

Fig. 2. Reduced matrix element for 2E1 transitions in He-*like ions as a function of the normalized photon energy according to Ref. [9]. The rim of the shaded area represents the parabolic shape of the phase-space factor.*

2. *Two-photon decay branches in heavy atoms*

In atomic systems with one K vacancy, the two-photon decay has to compete with the fast allowed decay modes in singly ionized atoms. Hence, the wings of the characteristic lines that are orders of magnitudes larger heavily disturb the two-photon spectrum. Only a coincident detection of the two photons can discriminate against the tails of the characteristic lines. The first measurement for a heavy atomic system, for Mo, was reported only in 1984, where the K vacancy was produced by photoionization [2]. A much cleaner way to produce the inner-shell vacancy was introduced by Ilakovac and coworkers already two years later [10]. For internal K-vacancy production, they used a decay mode of radioactive nuclei, the electron capture to the nucleus. Detecting the two photons in coincidence, they obtained a clean two-photon spectrum for the transitions 2s → 1s, 3s → 1s, 3d → 1s and 4s/d → 1s in xenon ($Z = 54$) [11], silver ($Z = 47$) and hafnium ($Z = 72$) [12]. All these measurements confirmed quite clearly the existence of the two-photon decay branch also in singly ionized atoms, and demonstrated that for this decay mode all intermediate states have to be included whether they are occupied or empty. Moreover, a drastic difference in shape for the various two-photon decay branches, in particular for 2s → 1s and 3d → 1s transitions showing different phase space factors, was evidently established.

Ilakovac and coworkers measured coincidences for back-to-back emission of the two photons, i.e. for 180° emission. Also, Ilakovac observed the resonance effect in 3d → 1s two-photon decay in silver and hafnium [13], which is seen as an increase of

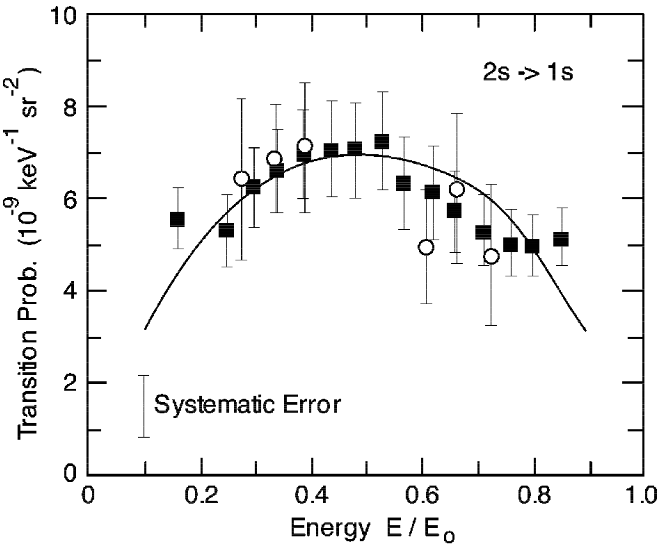

Fig. 3. The two-photon spectra for singly K*-ionized* Ag *atoms as a function of the normalized photon energy; measurements from Refs. [12] and [14] (open circles and full squares, respectively), and theory (full line) according to Ref. [15].*

the differential probabilities for lower energies of one photon ($x_1 < 0.5$) and higher energies of the other photon ($x_2 > 0.5$), in comparison to the case of equal sharing of energy ($x_1 = x_2 = 0.5$). In a recent thesis performed within the group of the authors of this paper, Schäffer [14] reported also on measurements of the two-photon decay branches in Ag induced by nuclear electron capture, confirming completely and with improved statistics the original findings of Ilakovac and his group. In Fig. 3, the 2E1 transition probability per K-vacancy decay in silver atoms is plotted as a function of the normalized transition energy; measurements of Ilakovac et al. [12] and Schäffer [14] are compared to theory [15].

Moreover, the angular dependence of the two-photon emission was probed experimentally by Schäffer and coworkers [14]. Whereas for ns → 1s transitions a $(1 + \cos^2\theta)$ angular distribution is predicted, we expect for nd → 1s transitions a $(1 + [1/13]\cos^2\theta)$ distribution [15–17]. Beyond the 180° emission, Schäffer also studied 90° emission in order to confirm additionally the difference not only in the shape of these two-photon transitions, but also in their angular distribution. As can be read from the Table 1, there is a reasonable agreement between the experimental results [14] and theoretical values from Tong et al. [15].

TABLE 1. Intensity ratio $I(180°)/I(90°)$ for the two-photon decay branches in Ag*; experimental values from Ref. [14], theory according to Ref. [15].*

Transition	Experiment	Theory
2s → 1s	1.90 ± 0.27	2.00
3s → 1s	1.49 ± 0.34	2.00
3d → 1s	1.22 ± 0.25	1.08

3. *Heavy* He-*like systems*

From theoretical point of view, ionic two-electron systems, i.e. He-like ions, are a lot cleaner than atoms as the screening from other atomic electrons has not to be considered. Heavy He-like ions with Z ranging from about 20 to 90 can be produced by stripping after acceleration and have correspondingly a large velocity, typically in the region of 1 to 50% the velocity of light. The excited 1s2s $^1S_0$ state is normally produced by atomic collisions in an exciter foil using the so-called beam foil spectroscopy technique [18]. For heavy He-like systems, the lifetimes of the 1s2s $^1S_0$ states vary from the ns region to the 10 fs region (for $Z = 20 - 90$, respectively) [5]. This means, the excited states in the fast He-like ions decay downstream of the foil according to their velocity and lifetime. For the heaviest ions the two-photon decay can already be considered as practically prompt in the foil.

The first detailed study on the spectral shape of the two-photon decay in heavy He-like ions beyond $Z = 20$ was performed by the Argonne group of Dunford and coworkers for Kr ions confirming the theoretical approaches [4]. The measurements confirmed the calculations of Drake [19] based on a non-relativistic approach. A clear change in the relative spectral distribution (normalized to the total transition energy) compared to the one expected for atomic He was established.

In a further investigation of that group with Ni ions, the accuracy in determining the shape of the two-photon spectra, caused by the uncertainties in the efficiencies, was significantly improved by comparing the spectral distributions for the two-photon decays in H-like and He-like Ni ions [6]. At this medium Z value, the measurements are still in accordance with both the non-relativistic calculations of Drake [7] and the full relativistic calculations of Derevianko and Johnson [9] as both these approaches are not really at variance in that case. However, the accuracy of this measurement is so promising that for heavier atomic systems a clear distinction between the different calculations can be expected.

The heaviest He-like system probed so far in detail by the two-photon decay is Au [20]. At the heavy-ion synchrotron facility SIS at GSI-Darmstadt, He-like Au^{77+} ions were produced by stripping at 106 MeV/u and then excited in a thin C foil. The emitted X rays were detected by Ge(i) detectors using the photon-photon coincidence technique. A clear variation to the relative distribution in Ni 26^+ was established and the spectral distribution confirms the fully relativistic calculations of Derevianko and Johnson [9]. Still further improvements of the accuracies towards the ends of the spectral distribution are needed for a final conclusion of the two-photon decay structure in strong central fields.

Recently a new method to efficiently produce the "metastable" 1s2s $^1S_0$ state for heavy ions was introduced by Stöhlker and coworkers [21]. Li-like U ions at 217 MeV/u, coasting in the heavy ion storage ring ESR, collide with nitrogen molecules at the internal gas jet target. At that collision energy, the projectile K-shell is predominantly ionized leaving the 2s electron untouched. Hence, by this specific ionization, the ions ends up predominantly in the excited 1s2s $^3S_1$ and 1s2s $^1S_0$ states of the He-like ions and only pure M1 and 2E1 X-ray transitions are observed, respectively. The spectral distribution for the two-photon decay is clearly observed without the need for measuring coincidences between the two photons.

4. *Future aspects*

It is evident that for heavy systems, i.e. for the real strong-field case, a lot of work still has to be done. For very heavy He-like ions, the field has just been touched, however, the results are promising and there is no obstacle to improving the experimental accuracy for the spectral distribution. In particular, the accuracies towards the ends of the spectra (low and high photon energy sides) can be improved for heavy two-electron systems. This would give enhanced sensitivity to the complete structure of the ion for the strong-field case. For very heavy He-like ions there is also a chance to observe the E1M1 decay of the 1s2p $^3P_0$ state. With the beam foil technique, the "prompt" 2E1 and "delayed" E1M1 decay can be clearly separated in that case. For instance, for He-like Au^{77+} ions, the corresponding transition rates are 7.6×10^9 and 8.1×10^8 s^{-1} from the excited $^1S_0$ and $^3P_0$ states, respectively [14].

The new technique to produce excited He-like ions by selected K ionization of Li-like fast heavy projectiles may lend a quite new and exciting access to the two-photon spectra, and hence, to the complete structure of heavy He-like systems.

On the other side, there are also new techniques to selectively produce singly K-ionized atomic systems by synchrotron radiation. Hence, now all heavy atomic systems are accessible to structure investigations using the two-photon coincidence technique applied by Ilakovac and his coworkers for his radioactive atomic systems. In future, comparing equivalent singly K-ionized and He-like heavy atoms will lead to additional insight into the structure and its changes due to additional electron interactions in strong field conditions.

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DVOFOTONSKI RASPAD STANJA SLIČNIH 1s2s $^1S_0$ U TEŠKIM ATOMSKIM SUSTAVIMA

U sustavima sličnim He je izravan raspad stanja 1s2s $^1S_0$ u osnovno stanje 1s^2 $^1S_0$ "zabranjen". Prijelaz 1s2s $^1S_0 \to$ 1s^2 $^1S_0$ emisijom dvaju fotona je osjetljiva proba strukture cijelog atomskog sustava. Oblik dvofotonskog spektra je posebno osjetljiv i otkriva detalje relativističkih učinaka u jakim središnjim poljima teških atoma. Daje se kratak pregled ovog polja s posebnim naglaskom na sustave visokog Z.