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Two-photon decay in strong central fields observed for the case of He-like gold

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Abstract

The photon energy differential shape of the second order matrix element for the two-photon (2E1) decay of the $1s2s^1S_0$ level in He-like gold has been measured. The results are in agreement with a recent fully relativistic calculation. The corresponding 2E1 matrix element deviates from those in lighter He-like systems due to the strong central field in a heavy two-electron ion. © 1999 Published by Elsevier Science B.V. All rights reserved.

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The elementary process of decay with two quantum jumps was first analyzed by Göppert-Mayer in 1931 [1]. In this process two correlated photons are emitted simultaneously during the interaction of an atom with the photon field. Two photons (ω_1, ω_2) with a sum-energy, $\omega_0 = \omega_1 + \omega_2$, corresponding to the difference in energy of the initial and final (atomic) states involved have to be created out of the fluctuations of the surrounding vacuum. A summation over all possible virtual intermediate states (bound and continuum) determines the shape of the continuous two-photon emission spectrum. Hence, a determination of the spectral distribution of the photons $A\,\omega_i\,d\omega_i$ is sensitive to the total structure of

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the atom in contrast to conventional spectroscopy which probes the energies or lifetimes of only a few levels. Studies of the two-photon spectral distribution go far beyond measurement of the lifetimes of a two-photon emitting states which only test the transition probability integrated over the photon distribution. For this reason, a lot of effort has been invested recently in the determination of the exact spectral distribution for two-photon decay in H- and He-like medium heavy ions (Z approximately 18–36) [2,3], as well as for single inner-shell vacancy states in heavy atoms [4]. Moreover, in contrast to studies in medium-Z ions, where from early on lifetime measurements were used to determine the two-photon decay rates [5], lifetime measurements using standard beam-foil spectroscopy are not feasible for very heavy He-like ions because the relevant lifetimes are too short. Thus, measurement of the spectral distribution provides one of the few means for studying two-photon decay at very high-Z where one can probe the influence of a strong central field on the total structure of an ion. Moreover, in this regime, the negative energy continuum, which appears directly in the sum over virtual intermediate states in the second order matrix element may give a non-negligible contribution. To explore these issues we have measured the photon energy distribution from decay of the $1s2s^1S_0$ level in He-like gold ($Z = 79$, A $= 197$) having a total X-ray transition energy $\omega_0 = 68.3$ keV. This isotope is ideal due to its high-Z, simple nuclear shape, and non-zero nuclear spin ($I = 3/2$). The nuclear spin causes hyperfine quenching of the 2^3P_0 state and this eliminates potential contributions from the E1M1 two-photon decay branch from this state [6]. Here, we report the first measurement of two-photon decay in very high-Z two-electron ions. Our results for the energy provide a probe of the influence of a very strong central field on the total structure (i.e. the dependence on a complete set of wavefunctions and energies) of a high-Z atom.

Two-photon decay is a second order elementary process so the electron couples twice to the photon field. Therefore, this process should exhibit the influence of the strong field in an enhanced way. In H-like systems the $2s^2S_{1/2}$ state decays via a 2E1 transition to the $1s^2S_{1/2}$ ground state. For H-like ions the spectral distribution of the 2E1 transitions

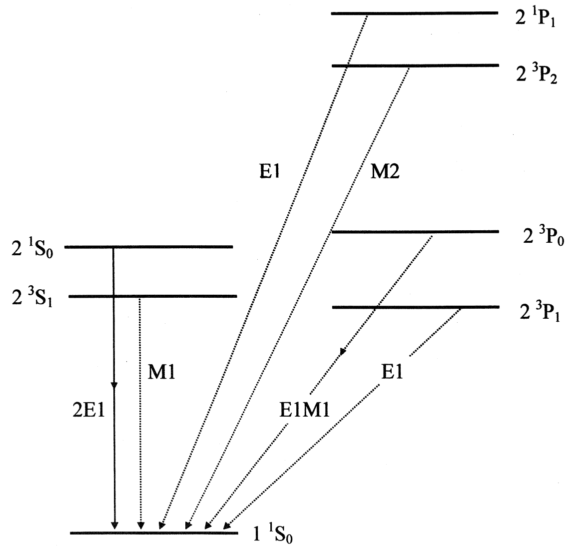

Fig. 1. Level diagram and decay modes for He-like Au^{77+} ions. (The total transition energy ω_0 between the $1s2s^1S_0$ state and the ground state is 68.3 keV.)

has been calculated for both the nonrelativistic [7] and the relativistic cases [8–10]. However, due to relativistic effects, the direct M1 spin-flip transition gets so fast for the heaviest systems that the 2E1 decay branch is negligible [9,11]. Hence, for the heaviest systems He-like ions are the simplest ones where two-photon decay can be studied experimentally. For illustration a level diagram for He-like gold ions is sketched in Fig. 1. In He-like ions the $1s2s^1S_0$ state can – according to the selection rules – only decay by two-photon emission to the $1s^2s^1S_0$ ground state. Moreover, He-like ions are the simplest ones where electron correlation can be studied. On the other side, with increasing atomic number Z the correlation strength decreases as $1/Z$ so that the heaviest He-like ions behave like H-like ions. Measuring two-photon decay in very heavy He-like ions provides information on both electron-electron correlations in He-like ions and on the strong field effect in an almost H-like system.

According to Goldman and Drake [10], the two-photon spectral distribution, which is symmetric with respect to the two photons, can be described by the energy differential emission rate

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

The matrix element M_{fi} which must also be symmetric about the midpoint (at $\omega_0/2$) is given by a sum over all intermediate states n

$$M_{if} = \sum \left[\frac{\langle f|D_1|n\rangle \langle n|D_2|i\rangle}{\omega_{ni} + \omega_1} + \frac{\langle f|D_2|n\rangle \langle n|D_1|i\rangle}{\omega_{ni} + \omega_2} \right] \quad (2)$$

where D_i is the photon field operator. In the non-relativistic He-like case [7,12], the sum in M_{if} comprises all the intermediate ^1P levels whereas in the relativistic systems the ^3P states have to be included. Fully relativistic calculations have been done only recently by Derevianko and Johnson [13].

The influence of the increasing strength of the central field in very heavy systems is elucidated in Fig. 2. This representation takes advantage of the factorization given in Eq. (1), that was first exploited by Bannett and Freund [14]. Normalizing to the total transition energy ω_0 ($f_1 = \omega_1/\omega_0$ and $f_2 = \omega_2/\omega_0 = 1 - f_1$) we have the following factorization for the distribution of one of the two photons:

$$A(f_1)\,df_1 \;\propto\; f_1(1-f_1)|M_{fi}|^2 df_1 \quad (3)$$

The factor $f_1(1-f_1)$ is responsible for the overall shape of the two-photon spectrum, and is represented by the shaded area in Fig. 2 (normalized in the center to a maximum value of one). Multiplying this parabola with the square of the corresponding energy differential matrix element results in the true shape of the two-photon energy distribution. The curves shown in Fig. 2 are the two-photon distributions according to the fully relativistic calculations of Derevianko and Johnson [13] divided by the factor $f_1(1-f_1)$. Hence, they represent the square of the energy differential matrix element for He and for He-like Ni^{26+} and Au^{77+} ions (also normalized to one at the center). The effects in Fig. 2 are partially explained by the interplay of electron-electron interactions and relativistic effects. At low Z the energy splittings within n = 2 are dominated by electron-electron interactions which decrease in relative importance as Z is increased. Going from He to Ni^{26+}, the energy denominators in the two-photon matrix element, Eq. (2), decrease relative to those for states

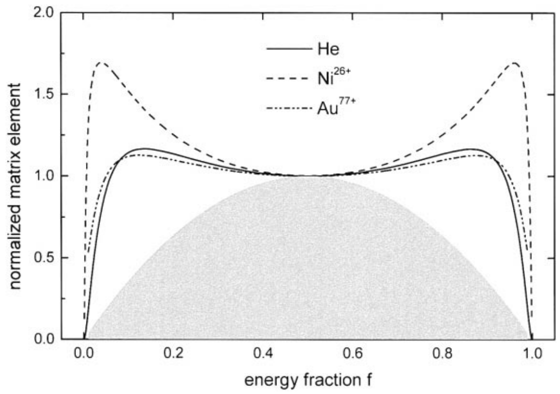

Fig. 2. Components of the spectral distribution for two-photon decay of the $1s2s\,^1S_0$ level in He-like ions: The shaded area represents the distribution $f_1(1-f_1)$ that has to be multiplied by the differential matrix element factor (top curves) to get the final two-photon spectrum. The photon energy f_1 is given as a fraction of the transition energy. Heights at the center are normalized to one. Matrix element factors are given for helium as well as for He-like Ni^{26+} and Au^{77+}.

with $n \neq 2$. This enhances the contributions at either endpoint and broadens the distribution. Going from Ni^{26+} to Au^{77+} relativistic corrections (due to the strong fields) start to dominate and the energy denominators for states within $n = 2$ begin to increase (as Z^4) relative to those of the $n \neq 2$ states which increase as Z^2. This causes a decrease of the contributions at near the endpoints and a corresponding narrowing of the distribution [13]. The goal of the present work is to extract the experimental shape of the energy differential matrix element in He-like Au ions and to compare it to the fully relativistic theory.

He-like Au^{77+} ions were provided by the heavy ion accelerator facility SIS (GSI-Darmstadt) at an energy of 106 MeV/u (44% of the velocity of light). After penetrating a 100-mg/cm^2 thick Al target $1s2s\,^1S_0$ projectile states were excited and decayed directly behind the target (within a few μm). Photons were detected with two X-ray detectors A and B placed at 60 degrees to the beam direction in the laboratory frame, (a sketch of the experimental set-up is shown in Fig. 3). In the emitter system the detection angles transform to 90 degrees with respect to the beam axis giving an angle θ of 180 degrees between the directions of the two photons which corresponds to the maximum intensity as a function of opening angle. Detector A was a 500-mm^2 single

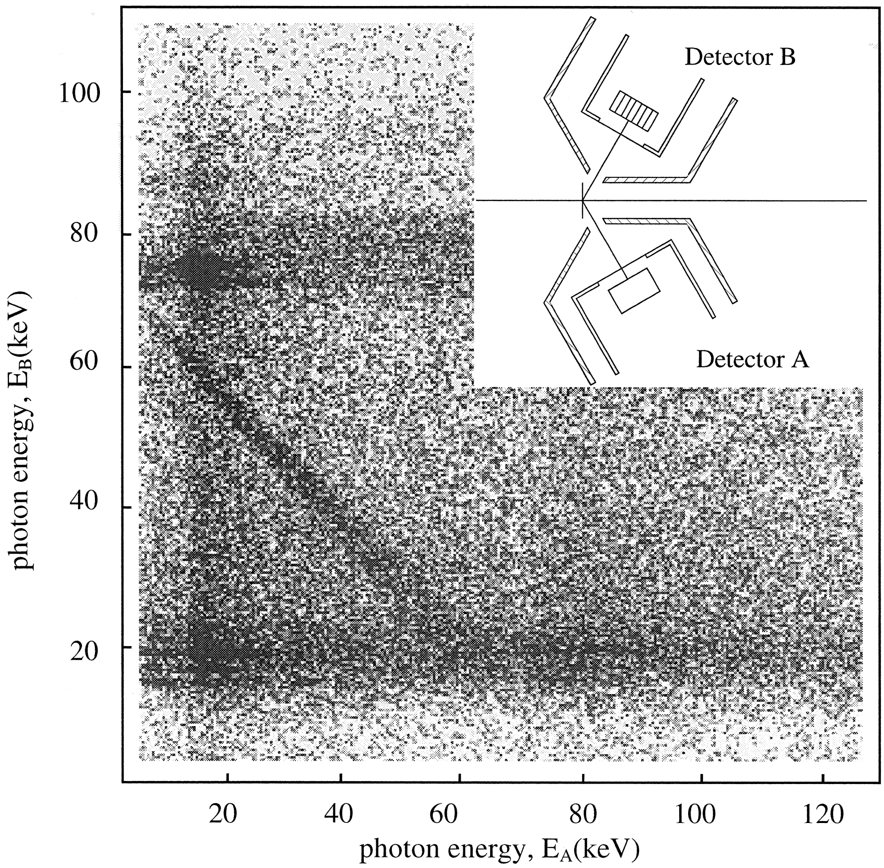

Fig. 3. Scatter plot for two-photon events for Au^{77+} excited in an Al target at 106 MeV/u. The two X-rays are measured in coincidence by detector A and segment 6 of detector B. The diagonal ridge represents the two-photon events. The detector arrangement is given in the inset.

Ge(i) X-ray detector, whereas detector B was segmented with 7 independent segments (3.5 mm by 25 mm stripes perpendicular to beam and photon directions). Due to the detector segmentation the photon energies could be Doppler corrected individually for the different observation angles. In the emitter system the energy of the photon registered by detector A corresponds to the total transition energy ω_0 of 68.286 keV diminished by the emission energy of the other photon. The two photons were recorded event by event using standard fast-slow coincidence electronics. Details of the experimental set-up and the data acquisition system can be found elsewhere [15,16].

Fig. 3 gives a scatter plot of events where two photons were detected, one in detector A and the other one in segment 6 of detector B. The axes are labeled by the energies of the corresponding photons. The diagonal ridge contains events with a constant sum energy equal to the total transition energy ($\omega_0 = E_A + E_B$), i.e. the ridge corresponds to the desired two-photon decay spectrum. From these raw two-dimensional spectra, background and random events were subtracted and the true events were analyzed according to the procedure used in the earlier Kr and Ni experiments [2,3]. By appropriate cuts and projections, energy distributions were generated for two-photon events seen by the various segments of detector B. These single lab-frame spectra were then transformed individually to the emitter system and summed up resulting in the X-ray continuum displayed at the top of Fig. 4.

The continuum spectrum is a convolution of the true photon distribution with all the electronic and

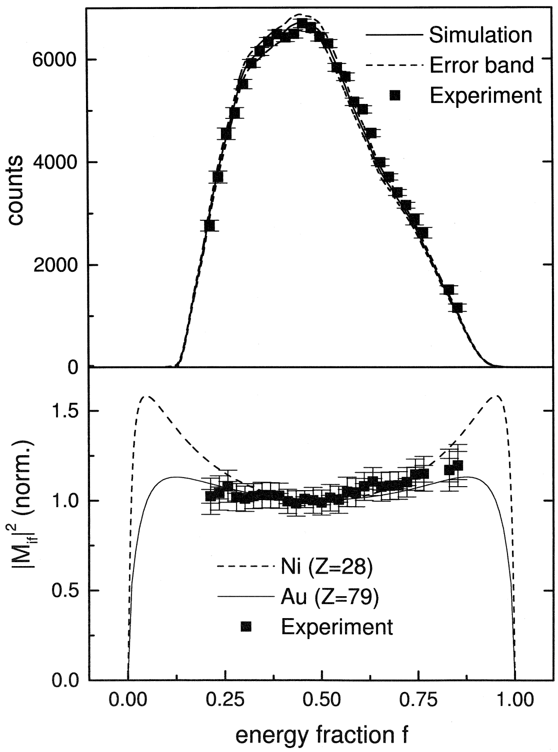

Fig. 4. The X-ray continuum for the two-photon distribution of He-like Au^{77+} ions (top part). The measured data points are compared with a Monte Carlo simulation (full curve; the dashed curves gives the error band). The square of the energy dependent matrix element (bottom part) extracted from the experiment is compared with the fully relativistic theory for Au^{77+} and Ni^{26+} (full and dashed lines, respectively [13]).

detector efficiencies. In order to compare this with the theoretical prediction, we performed a Monte Carlo simulation. This simulation is based on the theoretical two-photon distribution [13], the angular correlation of the two photons $(1+\cos^2\theta)$ [1], and all the relevant experimental factors like detector geometry and efficiencies, beam parameters and Doppler corrections for the X-ray energies. The program is based on the simulation program described in Ref. [2]. The result of the simulation is shown together with the experimental spectrum at the top of Fig. 4. The error band of the simulation is indicated in the graph as well (see dashed curves). An overall good agreement is found between experiment and simulation based on the fully relativistic calculation of Derevianko and Johnson [13]. A χ^2 test for the experimental data clearly favors the theoretical results for Au ($\chi^2 = 3.84$) over those for Ni ($\chi^2 = 7.35$). However, it is apparent that a further extension of the experimentally accessible energy range to lower and higher energies is needed in future experiments. Currently, the systematic errors in the Monte Carlo simulation limit the accuracy of the comparison.

The experimental X-ray continuum shows primarily the parabolic shape of the $f_1(1-f_1)$ factor from Eq. (3) which can be extracted very easily. To de-convolute the spectral shape we need a response function to transform the experimental data into the underlying spectral distribution and visa-versa. In a first step this response function can be derived by a comparison of the theoretical energy distribution of Ref. [13] with a simulation based on that distribution. Dividing the experimental data by the response function and by the factor $f_1(1-f_1)$ gives the square of the desired matrix element according to Eq. (3). This ''experimental matrix element'' is compared with the direct result of Derevianko and Johnson at the bottom of Fig. 4. For the experimental data points the inner error bars represent the statistical uncertainties of the measurement, whereas the outer error bars refer to the systematic errors mainly introduced by the uncertainties of the simulation. For comparison, the energy differential matrix element for He-like Ni^{26+} is shown by the dashed line. This shape was verified in an earlier experiment [3].

We would like to mention that 2E1 transitions from the excited 2^3S_1 level and E1M1 transitions from the 2^3P_0 level may, in principal, contribute to the observed experimental two-photon continuum, cf. the level diagram in Fig. 1. Two-photon decay of the 2^3S_1 level has a very small branching ratio even at high-Z and E1M1 decay of the 2^3P_0 level is suppressed by hyperfine quenching resulting from the non-zero nuclear spin. Estimates using the relevant branching ratios and excitation cross sections indicate negligible contributions from both of these rare two-photon decay modes [15]. The same is true for possible photon backscattering between the detectors. Contributions from two-photon decay of H-like ions are also completely negligible [15]. Hence, the data points displayed at the bottom of Fig. 4 represent the experimentally determined dependence of the matrix element on photon energy for the 2E1 decay of the $1s2s\,^1S_0$ state in He-like Au^{77+} ions.

In summary, our experiment using He-like Au^{77+} ions provides the first indication of the influence of a strong central field on the matrix element for two-photon transitions. The effects are most pronounced for asymmetric photon energies in the highest Z ions, so improvement of the experiment to provide a more extended photon energy range and carrying out measurements in ions with even higher atomic numbers is urgently needed in order to obtain a deep understanding of the complete structure of very heavy ions.

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References

[1] M. Göppert-Mayer, Ann. Phys. (Leipzig) 9 (1931) 273.
[2] R. Ali, Phys. Rev. A55 (1997) 994.
[3] H.W. Schäffer, Phys. Rev. A59 (1999) 245.
[4] K. Ilakovac, Phys. Rev. A46 (1992) 132.
[5] R. Marrus, R.W. Schmieder, Phys. Rev. A5 (1972) 1160.
[6] G.W.F. Drake, Nucl. Instr. Meth. B9 (1985) 465.
[7] G.W.F. Drake, Phys. Rev. A34 (1986) 2871.
[8] W.R. Johnson, Phys. Rev. Lett. 29 (1972) 1123.
[9] F.A. Parpia, W.R. Johnson, Phys. Rev. A26 (1982) 1142.
[10] S.P. Goldman, G.W.F. Drake, Phys. Rev. A24 (1981) 183.
[11] R. Marrus, P.J. Mohr, Adv. At. Mol. Phys. 14 (1978) 181.
[12] G.W.F. Drake, private communication (1995)
[13] A. Derevianko, W.R. Johnson, Phys. Rev. A56 (1997) 1288.
[14] Y. Bannet, I. Freund, Phys. Rev. A7 (1984) 299.
[15] H. Schäffer, Dissertation University Giessen, unpublished (1998)
[16] H.W. Schäffer, Nucl. Instr. Meth. B146 (1998) 62.