Lifetimes of Two-Photon-Emitting States in Heliumlike and Hydrogenlike Nickel

R. W. Dunford, M. Hass, $^{(a)}$ E. Bakke, H. G. Berry, C. J. Liu, and M. L. A. Raphaelian^(b) Physics Division, Argonne National Laboratory, Argonne, Illinois 60439

L. J. Curtis

Department of Physics and Astronomy, University of Toledo, Toledo, Ohio 43606 (Received 16 January 1989)

We report measurements of the lifetimes of the 2¹S₀ state of heliumlike Ni²⁶⁺ and the 2²S_{1/2} state of hydrogenlike $Ni²⁷⁺$, both of which decay predominantly by two-photon emission. Our method differs from previous measurements of the lifetimes of similar states in that we require a coincidence between the two photons. Our lifetime of $217.1(1.8)$ ps for the hydrogenlike decay is in agreement with the theoretical value of 215.45 ps. The heliumlike decay lifetime of $156.1(1.6)$ ps is in fair agreement with the theoretical value of $154.3(0.5)$ ps.

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The $2^2S_{1/2}$ state of hydrogenlike ions decays to the ground state either by the emission of two electric dipole photons $(2E1)$ or by single-photon magnetic dipole $(M1)$ emission.¹ The two-photon branch dominates at low atomic numbers, but the $M1$ branch becomes increasingly important with Z . The calculation of the lifetime of this state is well understood. In the nonrelativisitic limit, the two-photon decay rate^{2,3} is $\omega_{\text{NR}}(2E1)$ $=8.22938Z^{6}$ s⁻¹. The M₁ decay rate (to lowest order)⁴ is $\omega_{\text{NR}}(M1) = 2.496 \times 10^{-6} Z^{10} \text{ s}^{-1}$. Corrections to these rates have been calculated by Johnson, 4 Goldman and Drake,⁵ and Parpia and Johnson.⁶ The most accurate measurement of the lifetime of the $2^2S_{1/2}$ state was carried out in He⁺ by Hinds, Clendenin, and Novick.⁷ The contribution of the $M1$ branch was first observed in Ar^{17+} by Gould and Marrus.⁸ Here, we report the results of a measurement of the lifetime of the $2 \, \frac{2}{5} \, \frac{1}{2}$ state of hydrogenlike Ni^{27+} which is the first to be sensitive to the relativistic corrections to the two-photon decay rate. In addition, our experiment provides the most precise test of the calculation of the $M1$ contribution to the decay rate of this state.

We also report a measurement of the lifetime of the $2^{1}S_0$ state of heliumlike Ni²⁶⁺ which decays only by two-photon emission. The theoretical calculation of the heliumlike two-photon decay is much more difficult because of the need of account for two-electron correlations. Drake has recently reported a calculation³ which takes into account relativistic corrections for the first time. Our measurement in Ni^{26+} is sufficiently precise to be sensitive to these corrections. The only other experiment sensitive to these corrections is a measurement of the lifetime of the $2^{1}S_0$ state of heliumlike Kr³⁴⁺ by Marrus et al.⁹ They quote a lifetime of $34.08(0.34)$ ps, which is 2 standard deviations greater than the theoretical calculations of Drake which gives 33.41(0.14) ps.

Our experiments are the first measurements of the two-photon decay rates in hydrogenlike and heliumlike

ions based on the coincidence technique. Coincidences between the two photons emitted in these decays were first observed in He⁺ by Lipeles, Novick, and Tolk¹⁰ who also verified the $1+\cos^2\theta$ distribution for the angle between the two photons. More recently, the polarization correlation of the two photons emitted in the decay of the $2^2S_{1/2}$ state in deuterium was measured by Perrie et al. as a test of Bell's inequality.¹¹ Two-photon coincidences were also observed in heliumlike and hydrogenike argon by Marrus and Schmieder,¹² but their lifetime measurements were based on yields determined from singles spectra.

A spectrum of the individual photon energies resulting from the $2E1$ decay has the form of a continuous distribution with a broad maximum at half the transition energy which drops to zero at either end. An experiment based on the singles rate, requires distinguishing the two-photon continuum from the background. In a preliminary measurement¹³ of the lifetime of the $2^{1}S_0$ state of heliumlike Ni^{26+} , we used the standard beam-foil time-of-flight technique in which the rate of continuum photons was measured as a function of the foil-detector separation using a Si(Li) detector. In that measurement, the precision was limited by uncertainty in the shape of the background under the two-photon continuum and by uncertainty in the dependence of that shape on the foil-detector separation. In our latest experiments, this problem is eliminated since the requirement of a coincidence with the proper sum energy provides a signature to distinguish the two-photon decay signal from the background. For this reason our experimental error is not limited by considerations of background subtraction.

Another motivation for the coincidence experiment concerns the requirement to distinguish between the hydrogenlike and the heliumlike two-photon decays since both states can be produced in the beam-foil interaction. The continuum spectra of these decays have a nearly

identical energy distribution and it is not possible to separate them experimentally. In the singles measurements of the heliumlike two-photon lifetime, one can determine the hydrogenlike component by observing the single-photon M_1 decay branch, but this method is difficult because the energy resolution of a Si(Li) detector is not sufficient to separate the $M1$ x rays from x rays produced by long-lived excited states which cascade through the $2p$ level. This latter problem also precludes the use of the single-photon $M1$ decay to obtain an accurate measurement of the lifetime of the hydrogenlike $2^2S_{1/2}$ state. In the coincidence experiments, distinguishing the two decays poses no significant problem since the sum peaks from the hydrogenlike and heliumlike decays are separated by about 300 eV and the detector resolution is good enough to distinguish these two transitions.

The data to be presented here were taken using nickel beams from the Argonne Tandem-Linac (ATLAS). Beam energies of 376 and 674 MeV were provided in two separate runs. After acceleration, the ions were stripped in a thick carbon foil (200–500 μ g/cm²) and either the $26+$ or the $28+$ charge state was magnetically selected and directed onto the target. At 376 MeV, 20% of the beam emerging from the foil was in the heliumlike 26+ charge state while the fraction of fully stripped nickel was negligible. At 674 MeV, 46% of the beam was in the 26+ charge state and 7% was in the 28+ charge state. Typical beam currents (electrical) were 9 nA for the Ni^{26+} beam and 1.5 nA for the Ni^{28+} beam. The beam velocity was measured by a time-of-flight velocity analyzer. The uncertainty in the velocity measurement $(\pm 0.3\%)$ is dominated by the correction for energy loss in the stripper foils. 14

A schematic diagram of the apparatus in the target area is shown in Fig. l. A thin carbon target (12 μ g/cm²) was moved relative to three fixed Si(Li) detectors using a precision translation stage which measured the position of the target to better than 25 μ m. The foil-detector distance was varied from 5 to 55 mm. For the higher-energy beams, the hydrogenlike and heliumlike decays have theoretical decay lengths of 10.¹ and 7.2 mm, respectively. At the lower-beam energy, the heliumlike state has a decay length of 5.5 mm. Two of the Si(Li) detectors were collimated so that they observed a region of 5 mm along the beam path and subtended a solid angle of 0.1% of 4π at the beam. The third detector was not highly collimated and accepted photons into a solid angle of 1% of 4π . There was also a lower-resolution silicon x-ray detector attached to the target holder which was used for normalization. The resolution of the Si(Li) detectors was 180 eV at 5.9 keV while the normalization detector had a resolution of 1.8 keV at 14 keV. Typical rates in the Si(Li) detectors at the smallest foil-detector separation used to collect data were 400 Hz for the collimated detectors and 3 kHz for

FIG. 1. Schematic diagram of the apparatus. The foil target is moved relative to three fixed Si(Li) detectors. Two of these detectors are collimated so that the detected photons are localized to a 5-mm region along the beam. A fourth lowerresolution x-ray detector, which moves with the target, is used for normalization.

the uncollimated detector.

Although events were recorded for coincidences between any two of the three Si(Li) detectors, most of the coincidences were between one of the collimated detectors and the large solid-angle detector. The collimators served to localize the events in position along the beam, while the large solid-angle detector provided a good efficiency for detecting the second photon whose direction is only weakly correlated with the first $(1+\cos^2\theta)$ distribution). In Fig. 2 we show sum-energy spectra for coincidence events after subtraction of random coincidences. The widths of the sum peaks are due to a combination of the intrinsic detector resolution and the Doppler broadening associated with the angular acceptance of the detectors. The upper spectrum was taken with fully stripped Ni^{28+} incident on the target, while the lower spectrum was taken with heliumlike Ni^{26+} incident on the target. The peak in the sum-energy spectrum taken with the 26+ beam is almost entirely from the heliumlike two-photon decay. The spectrum taken with the 28+ beam is predominantly from the twophoton decay of hydrogenlike nickel but there is a small contamination from the heliumlike sum-energy peak which lies 300 eV lower in energy. In both lifetime measurements, the relative intensities of the heliumlike and hydrogenlike peaks were determined at each position of the foil by a two-Gaussian fit to the region of interest. In these fits the peak positions and widths were fixed, since these were determined independently in our measurements. The hydrogenlike component in the heliumlike sum peak is negligible; the heliumlike component in the hydrogenlike sum peak is small and well determined and does not contribute significantly to the measurement un-

FIG. 2. Sum-energy spectra for true coincidences between one of the collimated Si(Li) detectors and the large solid-angle Si(Li) detector at the closest foil-detector distance. (a) Data taken with fully stripped Ni^{28+} incident on the target at 664 MeV. The peak near 8.0 keV is predominantly from twophoton sum coincidences from decay of the hydrogenlike $2²S_{1/2}$ state. The dashed curve shows the contribution from heliumlike two-photon decays. The peak near 9.⁵ keV is due to true coincidences from $n = 3 \rightarrow n = 2 \rightarrow n = 1$ cascades. (b) Data taken with heliumlike Ni^{26+} incident on the target at 664 MeV. The peak near 7.7 keV is predominantly from the twophoton decay of the heliumlike $2^{2}S_0$ state of Ni²⁶⁺.

certainty.

Typical decay curves are shown in Fig. 3. These data have been normalized using the counts from the x-ray detector attached to the target holder and corrected for pileup and cascades. No correction is required for dead time because the data-acquisition electronics were designed so that all four detectors had identical live times. Considerable care was taken to ensure that the errors in the various corrections were less than a few tenths of a percent. The cascade correction was determined assuming that repopulation of the hydrogenlike $2²S_{1/2}$ state or the heliumlike $2¹S₀$ state from high-lying np states have a branch to the ground state giving counts in the singles spectra in the region above 9 keV. The correction is readily obtained using these counts and the known branching ratios¹⁵ $np \rightarrow 2s(np \rightarrow 1s$. The upper curve in Fig. 3 was obtained by fitting the sum-energy peak obtained with the Ni^{28+} beam on target for the hydrogenlike two-photon component. The lower curve is

FIG. 3. Decay curves for two of the four lifetime measurements. These measurements were made with a 58 Ni beam of 664 MeV incident on a $12-\mu g/cm^2$ foil in the target chamber. The data in the upper curve (solid dots) correspond to decays of the hydrogenlike $2^2S_{1/2}$ level in Ni²⁷⁺ and were obtained with a fully stripped Ni^{28+} beam incident on the target. The data in the lower curve (crosses) correspond to decays of the neliumlike $2^{1}S_0$ level in Ni²⁶⁺ and were obtained with a Ni² beam incident on the target. The solid lines are twocomponent fits to the data (see text).

based on fits to the sum-energy peak obtained with the $Ni²⁶⁺$ beam on target. The solid lines are twocomponent fits to the data with the lifetimes and the amplitudes as the only free parameters. The lifetimes and errors obtained from the two-component fits to the data for each of the three detector pairs were combined and then corrected for time dilation to obtain the final result for each lifetime measurement. A number of additional systematic effects were considered but found to be negligible: these included gain shifts, quenching in electric and magnetic fields, contributions from ions with highying spectator electrons, ¹⁶ and backgrounds from coincidences caused by Compton scattering of Lyman- α photons between two of the Si(Li) detectors. It should be emphasized that the limiting error in the present coincidence measurement is determined by the counting statistics in contrast to the singles measurements for which the limiting error involves the uncertainty in the background-subtraction procedure.

Two separate measurements were made for each of the lifetimes. For the heliumlike decay, the measurements were made in two separate runs. The major difference between these two runs was the beam energy, but in addition, one of the three Si(Li) detectors was replaced and new foil targets were used. The two hydrogenlike measurements were both completed during the same experimental run. The conditions for these two measurements were nearly identical. The only differences being the replacement of both the first and

second foil targets and an increased average beam current in the second measurement. For both the hydrogenlike and heliumlike decays, the two lifetime measurements were consistent within errors. We combined the measurements to obtain the quoted average results.

For the heliumlike decay, the combined result is 156.1(1.6) ps which is in fair agreement with the theoretical³ value of 154.3(0.5) ps. This result provides a test of the relativistic corrections which increase the calculated lifetime by 4.3 ps. For the decay of the $2^{2}S_{1/2}$ state in hydrogenlike Ni²⁷⁺ we find a lifetime of $217.1(1.8)$ ps which agrees with the theoretical value⁶ of 215.45 ps. The error in this measurement, expressed in terms of the decay rate, is $\pm 0.38 \times 10^8$ s⁻¹. This is the first measurement of the $2^{2}S_{1/2}$ state lifetime to be sensitive to the relativistic correction^{6} to the two-photon decay rate which is -1.019×10^8 s⁻¹ and it is also the most sensitive test of the $M1$ contributions⁶ to the decay rate which is 7.7×10^8 s⁻¹.

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 (b) Permanent address: The University of Illinois, Chicago,

IL 60680.

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⁽a) Permanent address: The Weizmann Institute of Science, Rehovot, Israel.