Relativistic Two-Photon Emission: Lifetime of the 2^1S_0 State of Heliumlike Kr^{34+}

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The two-photon ($2E1$) decay $2^1S_0 \rightarrow 1^1S_0$ in the heliumlike ion Kr^{34+} has been observed and the lifetime measured. Departures are measured for the first time from the predictions of the nonrelativistic theory, and are sensitive to relativistic effects on the transition-matrix elements. The measured lifetime is $\tau(\text{measured}) = 34.08(0.34) \times 10^{-12}$ s compared to $\tau(\text{nonrelativistic}) = 31.94(0.01) \times 10^{-12}$ s.

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Radiative decay of excited atomic and nuclear states by the simultaneous emission of two electric dipole photons ($2E1$ decay) is a relatively rare process that is usually observed only when decay by single-photon emission is rigorously forbidden (e.g., for $0 \rightarrow 0$ transitions) or highly inhibited by the angular momentum and parity selection rules as in the case of the $2S_{1/2} \rightarrow 1S_{1/2}$ transition in hydrogenic ions. To date a number of observations and measurements have been reported in atoms[1] and nuclei.[2] The best tests of the theory have been made on hydrogenic ions in the $2S_{1/2}$ state. The most accurate calculation of the nonrelativistic rate for these ions is due to Klarsfeld[3] which yields for the transition probability per unit time $A_{\text{nonrelativistic}} = 8.229\,43Z^6$ s^{-1}. Measurements have been made[1] on hydrogenic ions through $Z = 18$ and are all in good agreement with this calculation.

Recently, Parpia and Johnson[4] and Goldman and Drake[5] have made calculations of the $2E1$ decay rate using a relativistic formulation. Their work indicates that deviations $\geq 1\%$ from the nonrelativistic rate should exist for $Z \geq 20$. In this paper we report a measurement of the $2E1$ transition $2^1S_0 \rightarrow 1^1S_0$ in the heliumlike ion Kr^{34+}. This measurement shows for the first time depeartures from the predictions of the nonrelativistic theory. The result represents the first test of the relativistic theory of the $2E1$ process and indeed is the first test of the relativistic contribution to the dipole matrix element.

The beam-foil time-of-flight method is employed in these measurements. The apparatus consists of two Si(Li) detectors located on opposite sides of a box through which the Kr^{34+} beam passes (see Fig. 1). The positions of the exciting foil (200 μg/cm^2 carbon) and one of the detectors are fixed thoroughout the experiment. The second detector-collimator assembly is moved parallel to the ion beam by means of a precision translator with a positional accuracy ~ 1 μm. This accuracy is important since the decay length of the 2^1S_0 state is about 2 mm. The raw data consist of the ratio of the number of counts in the $2E1$ spectrum of the movable detector to the number of $2E1$ counts in the fixed detector as a function of the separation of the two detectors. Thus, in this experiment, the fixed detector is used to normalize the counting rate in the movable detector. This differs from the conventional time-of-flight method where the integrated beam current in a Faraday cup is used for normalization and where the foil-detector separation is varied. The method used here has two important advantages over the conventional method: (1) Normalization is directly to the number of ions in the 2^1S_0 state. (2) Changes in the shape and state of exciting foil during the experiment are of no consequence.

A first attempt to observe the 2^1S_0 decay was made with the 35A-MeV krypton beam obtained from the GANIL accelerator (Caen, France). This experiment succeeded in observing the decay and in obtaining a

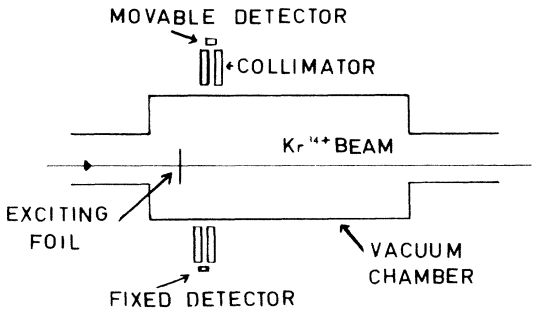

FIG. 1. Schematic of the apparatus.

preliminary value for the lifetime. However, this beam contained a relatively large ratio of $Kr^{35+}/Kr^{34+} \approx 0.25$. This is important since the decay $2S_{1/2} \rightarrow 1S_{1/2}$ in Kr^{35+} is primarily by $2E1$ with a spectrum that is essentially indistinguishable from the 2^1S_0 spectrum under study here. This contamination precludes an accurate lifetime measurement. To surmount this problem an experiment was performed with the 18.9A-MeV krypton beam from the UNILAC accelerator (Gesellschaft für Schwerionenforschung, Darmstadt).

The krypton beam is prepared in the following way. The 18.9A-MeV krypton beam is prestripped by a thick carbon foil producing a distribution of Kr ions in highly charged states. A dispersing magnet selects only the Kr^{34+} charge state which is then deflected into our beam line. This beam is then excited by a ~ 200-$\mu g/cm^2$ C foil which produces the Kr^{34+} ions in the 2^1S_0 state. This procedure ensures a high ratio of Kr^{34+}/Kr^{35+} ions in the beam. Charge-fraction measurements[6] made with the Kr beam on C foils of various thicknesses show that at this energy $N(Kr^{34+})/N(Kr^{35+}) \geq 90{:}1$. Even with this ratio, there is still a small number of $2S_{1/2}$ counts under the $2E1$ spectrum. These are measured and subtracted out by a procedure described later in this paper.

The lifetime measurement requires an accurate measurement of the decay length (~ 2 mm in this experiment) and the beam velocity. The accuracy of the decay length is ensured by our establishing the position reference relative to a fixed detector rather than to the exciting foil. If we use a precision translator to move the second detector and take care to align the detector translation axis parallel to the beam axis, the error in the decay length from mechanical and alignment errors is $\leq 0.05\%$. The beam velocity is measured by a time-of-flight system installed after the first stripping foil but before the exciting foil. The system consists of two detectors, separated by about 16 m, that detect arrival pulses from the beam bunches. The system yields for the beam energy a value of 18.934A MeV. The correction to the energy resulting from passage through the exciting foil is based on the energy-loss tables by Ziegler[7] for 19A-MeV ^{84}Kr on carbon. The result is $\Delta E = -5.1$ MeV.[7] Hence, the beam velocity after the foil is $\beta = v/c = 0.198\,42(29)$.

The raw data consist of spectra taken at different detector positions. A sample spectrum used in the decay-length measurement is shown in Fig. 2. In evidence is the two-photon spectrum. Analysis of the peak ($M1 + M2$) shows that $n = 2 \rightarrow n = 1$ transitions in Kr^{35+} and Kr^{34+} are also present, and finally there is a fairly uniform background at $E \geq 19$ keV. The electronics was calibrated before and after the run and was found to be stable to ≤ 20 eV. For analysis purposes, the data under the continuous $2E1$ spectrum

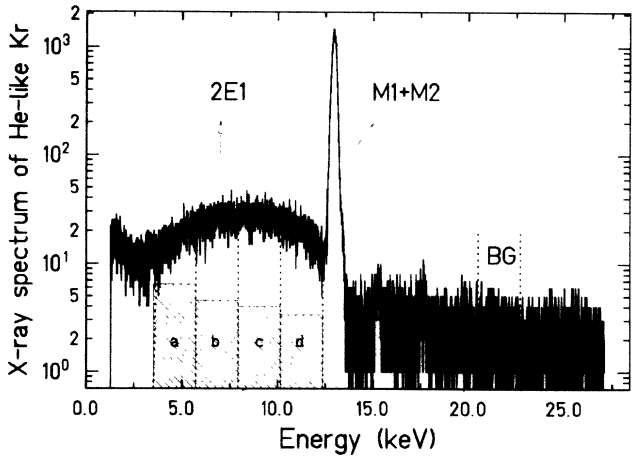

FIG. 2. Sample spectrum taken at near foil-detector separation. The "bins" labeled a,b,c,d were arbitrarily selected for independent analysis of the $2E1$ decay rate. The crosshatched area indicates the background level.

were separated into four equally spaced energy windows from 3.5 to 12.38 keV (Fig. 2). The first step was to subtract from each window counts arising from hydrogenlike ions in the beam and counts from ions in the 2^1S_0 state that arise from cascades from higher states. The number of counts from hydrogenlike ions is found by fitting of the peak at 13.0 keV (Fig. 2). In evidence are the 2^3S_1, $2^3P_2 \rightarrow 1^1S_0$ of Kr^{34+} and a relatively small peak arising from the single-photon $M1$ decay $2S_{1/2} \rightarrow 1S_{1/2}$ of Kr^{35+}. From the known branching ratio[8] $2E1/M1 = 1.96$ for this decay and the measured window and detector efficiency, the number of $2E1$ counts from hydrogenlike ions is unambiguously subtracted from the raw data. The number of counts from ions which have cascaded into the 2^1S_0 state can be unambiguously determined from the small observed peaks at 15.3 keV and higher. These result from $np \rightarrow {}^1S_0$ transitions. From the known branching ratios $(np \rightarrow 2^1S_0)/(np \rightarrow 1^1S_0)$ an unambiguous correction (upper limit) is made to the data for the cascading ions. This correction is small ($< 10^{-4}$ of the decay length).

Background counts under the $2E1$ spectrum were treated in two ways. First, a window was established at ~ 22 keV and the counts in this window were assumed to arise from background. It was then assumed that the background in each of the four $2E1$ windows was proportional to the background with proportionality constant K_α different for the windows ($\alpha = a,b,c,d$) but was the same for all detector positions. The value of K_α was then chosen so as to minimize χ^2 when all of the points are fitted by a single exponential to yield a decay length. The second procedure used for fitting to the data was to take the data corrected for hydrogenlike ions and cascades in each of the four windows

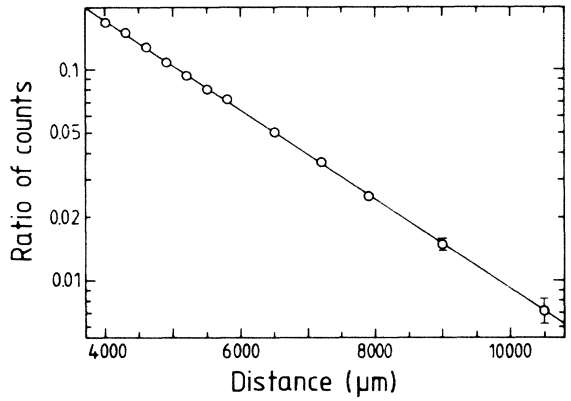

FIG. 3. Sample decay curve obtained for window c (see Fig. 2). The solid line indicates the best fit.

and then do a least-squares fit by a single-exponential plus constant background. The two procedures yield results which differ by less than 1%. A sample decay curve obtained by this fitting procedure is shown in Fig. 3.

Our final experimental result is $A_{\text{expt.}}(2^1S_0) = 2.934(30)\times 10^{10}\ \text{s}^{-1}$, or $\tau(2^1S_0) = 34.08(34)\times 10^{-12}$ s.

The error is mainly the statistical error associated with the fitting procedure. Errors associated with possible systematic effects are shown in Table I.

Our experimental result may be compared with theory in the following way. Drake[9] has recently performed a nonrelativistic variation calculation of $A_{\text{NR}}(2^1S_0)$ for heliumlike Kr^{34+} and has established $A_{\text{NR}}(2^1S_0) = 3.131(1)\times 10^{10}\ \text{s}^{-1}$, which can be compared with our experimental value $A_{\text{expt.}}(2^1S_0) = 2.934(30)\times 10^{10}\ \text{s}^{-1}$. To assess the effect of relativistic corrections we write $A_{\text{NR}}(2^1S_0)$ as a power-series expansion in Z as follows:

$$A_{\text{NR}}(2^1S_0,Z) = (16.458\,86\ \text{s}^{-1})Z^6 + A_5Z^5 + A_4Z^4 + \ldots \quad (1)$$

The leading term is twice the hydrogenic $2S_{1/2} \rightarrow 1S_{1/2}$ rate and is known exactly. The coefficients A_5 and A_4 are unknown. However, the contribution to $A_{\text{NR}}(2^1S_0)$ from terms other than the leading term is the difference between the result of the variational calculation and the leading-order term. The relativistic correction to the leading term has been determined by Parpia and Johnson[4] and by Goldman and Drake.[5] With use of Eq. (4.7) of Goldman and Drake, the decay-rate equation (1) becomes

$$A_{\text{rel}}(2^1S_0) = (16.458\,86\ \text{s}^{-1})Z^6\left[\frac{1+3.9448(Z\alpha)^2-2.040(Z\alpha)^4}{1+4.6019(Z\alpha)^2}\right] + A_5Z^5[1+O(Z\alpha)^2] + A_4Z^4[1+O(Z\alpha)^2] + \ldots \quad (2)$$

We thus obtain for the relativistic decay rate

$$A_{\text{rel}} = 2.981(30)\times 10^{10}\ \text{s}^{-1}.$$

We note the following: (1) Relativistic effects decrease the decay rate, even though they increase the 2^1S-1^1S energy separation. Thus this measurement is primarily sensitive to the relativistic effects on the transition matrix elements. (2) Our measurement is in good agreement with the result using the leading-order correction. However, the error in the theoretical value is due mainly to the uncalculated $O(Z\alpha)^2$ correction to the Z^5 term. Since the theoretical error is of the same order as the experimental error, it will be necessary to calculate the higher-order term in order to confirm the agreement.

TABLE I. Contributions to experimental error (%).

Beam velocity	0.1
Cascade correction	0.01
Geometry and misalignment	0.05
Hydrogenic $2S_{1/2}$ contamination	0.2
Dead time	0.1
Fitting error	0.9

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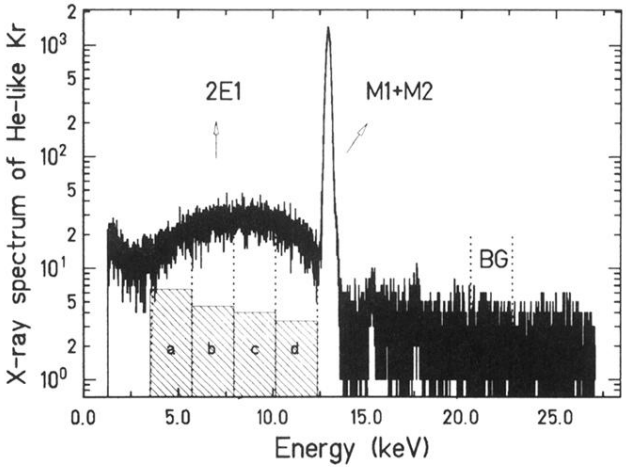

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