Estimation of the time scale of last chance alpha emission using an "atomic clock"

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(Received 4 October 1993)

The probability of filling a K-vacancy, created on the incoming part of the collision, before α particle emission is used to time the decay of Yb compound nuclei. These nuclei were produced in the fusion reaction 250 MeV 60 Ni + 100 Mo. In general the nuclear decays are too fast to be timed by this clock, however, α -particle emitting compound nucleus states have lifetimes sufficiently long for this technique to work when both the α -particle has low energy and the compound nucleus spin is large. This supports the existence of last, or near last, chance α -particle emission in the deexcitation of high spin compound nuclei.

PACS number(s): 24.60.Dr, 25.70.Gh

Thirty years ago Gugelot [1] pointed out that one can calibrate the decay width (Γ_c) of a compound nucleus in terms of the K x-ray width (Γ_x) for atomic vacancies. All one has to do is prepare an atom with an excited nucleus and a K vacancy and then measure the branching ratio $\Gamma_{\mathbf{z}}/\Gamma_{\mathbf{c}}$. In practice, there are several problems in applying this idea. The K -vacancy production probabilities P_K are low (of the order of 1%) for typical low energy fusion reactions and, in contrast to atomic collisions below the Coulomb barrier, the x rays of interest must be detected in a large background resulting from internal conversion x rays and γ rays emitted by excited nuclear reaction products. Another important issue is photon production from the filling of vacancies in a charged particle exit channel during the time after emission but before the emitted particle is well outside the K -shell Bohr radius (molecular orbital emission). Due to these problems, this old idea has met with success only recently $[2-4]$.

This technique is best suited to the study of the lifetimes of last or near last chance decays of compound systems in the bottom portion (high charge) of the periodic table. Only under these conditions does the K -vacancy lifetime approach the compound nuclear lifetime. Two interesting topics for which this technique is quite promising are: (1) the study of slow fission components and (2) slow "yrast" α -particle emission. The former involves actinide nuclei for which the K -vacancy lifetime is less than 10^{-17} s, and the latter, heavy rare earth nuclei with K vacancy lifetimes between 10^{-16} and 10^{-17} s. The recent paper by Molitoris et al. [4] addresses the first of these topics and this work deals with the second.

Statistical model codes predict that the emission of α particles competes with γ -ray emission at very low excitation energies when the nuclei are trapped near the yrast line at high to moderate spins [5,6]. Grover and Gilat first discussed this process in work published 25 years ago [5]. These early calculations, as well as more recent ones [7], predict that these "stretched" and "last chance" α particles would be emitted from states with lifetimes similar to the K-vacancy lifetime and therefore present an ideal application of the atomic clock method. The predicted α -particle energies are low for these "last chance" particle emissions which, despite the angular momentum removed by the decay, tend to populate moderate to high spin states of the residue. The present work demonstrates for the first time and in a direct way that these long lived α -particle emitting states do exist; however, the mean lifetime of these states appears to be shorter than predicted by the statistical model.

The Holifield Heavy Ion Facility at Oak Ridge National Laboratory was used to produce a beam of 250 MeV $\rm{^{60}Ni}$, which impinged upon a 711 μ g/cm² self-supporting 100 Mo target, enriched to 97.27% in mass 100. This reaction produces $^{160}\mathrm{Yb}$ compound nuclei with a mean excitation energy of 59 MeV and angular momenta up to \approx 57 \hbar . The K-vacancy production probability for a scattered trajectory with no time delay is estimated to be 2% from scaling relations based on semiclassical calculations in the first Born approximation [8]. Additional calculations by Andersen et al. $[8]$ (for monopole ionization) suggest, that for the system studied here, that the fusion trajectory (incoming part of the scattered trajectory) yields roughly 1/3 of the ionization of the full trajectory. The cited scattering calculations reproduce a large volume of both light and heavy-ion scattering data [9]. The half trajectory calculations agree with both ^{40}Ar induced fusion reactions at 4.5 MeV/A [10] and measurements of K-vacancy production in α decay [8,11] to better than a factor of two. Considering the above, we will use an estimate of $P_K \approx 0.7\%$; to which we estimate an uncertainty of a factor of two. On the other hand, there is a rather small uncertainty in the the lifetime of Yb K vacancies: $\tau_x = 20 \times 10^{-18}$ s [12], a value corroborated by the intrinsic line widths $[13]$. In the present experiment, one would expect this time to increase an insignificant amount due to multiple ionization. The x rays were detected in a planar Ge detector at 130° to the beam while α particles were detected with the Dwarf Ball system [14] which covers almost 4π sr in solid angle. The Spin Spectrometer [15] was used to determine the γ -ray fold (k_{γ}) which is used for spin selection. Three coaxial Ge detectors with Compton suppressors replaced three elements of the Spin Spectrometer. These were used to determine the overall γ -ray spectrum.

Figure 1 shows x-ray spectra with three different requirements on k_{γ} and the center of mass energy of the detected α particles, E_{α} . The upper histogram is generated with the selection of a low energy α particle (the lower half of the E_{α} distribution, E_{α} < 17.5 MeV) and the upper third of the k_{γ} distribution. The middle histogram is constructed with the requirement that both k_{γ} and E_{α} be large, upper third and half of the respective distributions, while the lower histogram utilizes the data with intermediate values of k_{γ} and low values of E_{α} . (The data with the lowest values of k_{γ} are not used as they are contaminated with reactions on carbon impurities in the target.) The three spectra have similar statistics and therefore are displayed with vertical offsets. The x-ray peaks from converted transitions in Er residues are the prominent features in all these spectra. What is also apparent are weak Yb x-ray peaks in the upper spectrum. The only Yb x-ray peak with clear statistical significance is K_{α_1} ; however, excess counts also occur at the energies corresponding to K_{α_2} and K_{β_1} energies. The arrows indicate the location and expected relative intensities of the x rays. Within the statistical uncertainty, no evidence for these peaks is found in the other two spectra or in the spectra generated by gating on protons (not shown). This observation is in agreement with our expectation that the slowest emissions involve α particles of lower than average energy and which populate residue states at rather high spin.

A conceivable source of Yb x-rays, in the α -particle gated spectra, could be from converted transitions of Yb residues formed from the decay of Hf compound nuclei; which would be formed if Ru existed in the target. X-ray Photoelectron Spectroscopy was used to investigate this possibility. No suggestion of Ru was found. The sensitivity of this technique is, conservatively, 0.1 at. %. At this level, assuming the mean cascade averaged conversion coefficients for Yb and Er are the same, no more than 4 of the 90 \pm 30 excess counts in the K_{α_1} could be from Ru contamination. Futhermore, a Ru contamination would not preferentially influence the spectrum generated with the selection of low energy α particles and high values of k_{γ} .

The mean lifetime, τ_c , of the selected compound nuclei can be estimated from the known lifetime of a K vacancy and the fraction of events which emit the CN x ray,

$$
f_{x,\alpha} = \frac{P_{x,\alpha}}{P_K} = \frac{\Gamma_x}{\Gamma_x + \Gamma_c} = \frac{\tau_c}{\tau_x + \tau_c}.
$$
 (1)

In the expression above, $P_{x,\alpha}$ is the x-ray coincidence probability per alpha particle, corrected for the x-ray detection efficiency, the relative intensity of the x-ray branch (K_{α_1}) in this case) and the fluorescence yield. In this work the α -particle singles data were not recorded, so the number of α particles was determined from the beam current and the estimated cross sections.

The extracted lifetime as a function of P_K is shown in Fig. 2. The thick line represents the most likely values and the thin lines give our estimate of the uncertainty of this measurement. If one assumes that $P_K = 0.7\%$, then the best estimate for the mean lifetime of the selected α -particle emitting states is $\approx 6 \times 10^{-18}$ s.

It is also worth noting that this time scale, $\approx 10^{-17}$ s, is more than two orders of magnitude longer than the time required for an α -particle to reach the Bohr orbit radius. This indicates that molecular orbital emission in the decay channel is not probable in this case. This is,

FIG. 1. X-ray spectra gated on high γ fold (k_{γ}) and low E_{α} energy (top histogram, offset by +100), high k_{γ} and high E_{α} (middle histogram), and intermediate k_{γ} and low E_{α} (bottom histogram, offset by -100). The characteristic energies and intensities for K x rays from $_{70}Yb$ and $_{68}Er$ are indicated by the arrows on the top and bottom of the figure, respectively.

FIG. 2. Estimated mean lifetimes of the selected compound nuclei (those which emit a low energy α particle and possess high angular momentum) prior to the emission of the x-ray. The lifetime is given as a function of the K -vacancy production probability. The thick line indicates the most likely values while the region between the thin lines represents the values within the estimated uncertainty of the present measurement. This estimate considers the statistical uncertainty of the yield of CN x-rays and the uncertainty of the total α particle cress section.

of course, a required condition for this "atomic clock" technique.

In principle, an independent method exists to estimate the mean lifetime of the selected α -particle emitting states. That being: to use the relative magnitude of the Er K_{α_1} and Yb K_{α_1} yields. In order to use this method an average conversion coefficient, for the cascades of interest, needs to be deduced. This can be accomplished by folding the efficiency and response corrected photon spectrum from the Ge detectors with calculated conversion coefficients [16]. However, in practice this procedure cannot yield accurate results unless data are available on the multipolarity mixture. Our attempts to use this technique were frustrated by our lack of knowledge of the extent of Ml transitions at low energy.

Statistical model calculations [7] yield an estimate of 40×10^{-18} s for the lifetime of the states consistent with the requirements used to select the data shown in the top histogram in Fig. 1 (lower portion of the E_{α} distribution and upper portion of the residual spin distribution.) Comparison of this value to the experimental results (shown in Fig. 2) suggests one or both of the following: that the K -vacancy production for these fusion reactions is less than 0.7% by at least a factor of 2 (which is unlikely, see discussion above and Refs. $[8-11]$, or that

the mean lifetime of the states is shorter than predicted by the statistical model.

In this work the x-ray atomic clock method was used to provide an estimate of the characteristic emission time of low energy α particles from high spin compound nuclei. The compound nucleus x-ray peak is very small, and therefore the uncertainties are large. Nevertheless, this is positive evidence that "slow" and last, or near last, chance α -particle emission occurs in the deexcitation of high spin compound nuclei. While the present work provides some weak evidence that the emission time is faster than predicted from the statistical model, a definitive conclusion is not possible at this point due to the large statistical uncertainties of the present work and since the absolute probability of creating a K vacancy was not determined.

This work was supported by the Director, Office of High Energy and Nuclear Physics, Nuclear Physics Division of the U.S. Department of Energy under contracts Nos. DE-FG02-87ER40316 and DE-FG02-88ER40406. Oak Ridge National Laboratory is operated by Martin Marietta Energy Systems, Inc. under Contract No. DE-AC05-84OR21400 with the U.S. Department of Energy.

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