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Direct observation of slow fission from the width of K x-ray line

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Abstract. An atomic clock based on the measurement of the intrinsic width of K x-ray lines has been used to obtain evidence of long-lived fission of the highly excited plutonium nuclei produced in the fusion of ^{4}He+^{238}U at E(^{4}He)$_{Lab}$=60 MeV. The mean fission time of the long-lived fission could be obtained from the increase of the intrinsic width of plutonium K x-ray line using quantum energy-time uncertainty principle. The presence of long-lived fission (mean fission time >1×10^{-18} s) has been found and the fluorescence yield per fission event shows that most of the fission events are slow (~10^{-18} s).

1 Introduction

The timescale of the nuclear fission process of highly excited fissile nuclei is a basic characteristic of the underlying fission dynamics. However, an apparently anomalous situation exists regarding the timescale of nuclear fission process. The atomic techniques (K x-ray-fission fragment coincidence and crystal blocking techniques) [1-4] have measured long fission times (~10^{-18} s) for most of the fission events of the highly excited fissile nuclei, whereas nuclear techniques [5-7] have obtained much shorter fission time (~10^{-20} s) for most of the fission events. Long fission times (>10^{-18} s) could be obtained from Langevin fluctuation-dissipation dynamical calculations [8] using a large viscosity parameter inhibiting the fission process. The different measured fission timescales have been attributed to the sensitivity of the nuclear technique to short timescale and that of atomic technique to long timescale and it was argued that the long fission times could provide information about the viscosity [8] of the nuclear medium and might be used as a probe [4] for studying the long-lived superheavy nuclei. It was recently shown [9] that the observed long fission time for the majority of the fissioning events as obtained by the atomic techniques cannot be reconciled with the short fission time obtained by the nuclear techniques for any plausible fission time distribution. So it is very important to obtain evidence for long fission time by more direct means. In earlier K x-ray fission fragment coincidence experiments, very broad K x-ray lines (FWHM ~20 keV) were observed

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because of contributions from different elements with similar Z values and electronic configuration mixing. The long fission times were obtained from the measured K x-ray multiplicity per fission assuming a fission time distribution and estimated probability of K orbital vacancies in the element produced by fusion. Since in ^{4}He+^{238}U fusion reaction, predominantly plutonium is produced by fusion and the effect of electronic configuration mixing should be small, narrow plutonium K x-ray lines should be seen in coincidence with the fission fragments, if there is a significant contribution from the long fission time component. So the intrinsic width of the plutonium K x-ray line should provide direct evidence for a long fission time component from the quantum energy time uncertainty principle, thus opening up a new way of looking at the fissioning nucleus. However, neutron emission from plutonium competes with the fission process and so fission of plutonium with progressively lower excitation energies takes place leading to a distribution of fission time. Statistical model codes predict that on the average about 2 neutrons would be emitted before fission corresponding to an average fission time of ~10^{-20} s, in agreement with the experimental results [7] obtained from ^{243}Am at E_X=60 MeV and the probability of emission of 4 prefission neutrons (corresponding to a long fission time ~10^{-18} s) is ~5%. However, according to Langevin fluctuation-dissipation model, there could be a significant contribution from a long-lived fission component, if the viscosity parameter is large leading to observable plutonium K x-ray yield and we have studied this possibility experimentally.

2 Experiment and Data Analysis

A natural uranium-oxide film (≈ 2.5 mg/cm^2) electrodeposited on a ~1micron thick thin aluminum foil was bombarded by a beam of 60 MeV ^{4}He particles (~2 pnA beam current) accelerated by the Variable Energy Cyclotron located in Kolkata, India. A large area solar cell detector covering about 10% of the 4π solid angle was placed at a distance of ~2 cm from the target to detect fission fragments. The solar cell detector is primarily sensitive to heavy fission fragments only. The center of the solar cell was making an angle of 135° with the beam axis. A four-segmented low energy photon spectrometer (LEPS) was placed at a distance of 10.7 cm from the target at a polar angle of 90° with the beam direction for detecting γ-rays and x-rays. Each segment of LEPS subtended a solid angle of about 7 msr at the target position. A coincidence circuit was setup between the LEPS and solar cell detectors. The solar cell detector recorded a fission fragment spectrum of expected shape in coincidence with the γ-ray and x-rays detected by the LEPS detector. In Fig. 1 (top panel), we show random corrected coincidence spectrum. In the coincidence spectrum, we still see the remnants of uranium K x-ray lines produced by the inelastic scattering process because of the imperfect cancellation of the random coincidence events. Our main peaks of interest are plutonium K x-ray lines. However, the presence of fission fragment γ-rays would produce a large background and could complicate interpretations. Fission fragment γ-ray and plutonium K x-ray lines produced in the nuclear fusion reaction ^{4}He+^{238}U could be distinguished by their spectral shapes, because the fission fragment γ-ray peak would be double-humped and broadened due to Doppler effect, whereas plutonium K x-ray line would remain narrow for long-lived fission component. In order to draw an appropriate background curve, we have performed best possible fits of the data points around 103 keV and 98.4 keV by Gaussian functions (magenta color curves) and linear backgrounds (black lines). Typical reduced χ^2 values are ≈1 and FWHM of the Gaussian peaks = 1 keV. There is a narrow (FWHM = 1 keV) peak around 103 keV with >99% confidence level. Broad double-humped peak structures have been seen around 65 keV and 88 keV and they could be best fitted by GEANT3 [10] simulated curves obtained by using actual experimental geometry and linear backgrounds (black lines). A smooth curve (solid red background) has been drawn [Fig. 1 (top panel)] joining these linear backgrounds (black lines) and this red background has been used as the background curve for our analysis. In Fig. 1 (bottom panel), we show red background subtracted coincidence spectrum. We see remnants of uranium K x-ray lines and double-humped broad peaks at 65 keV and 88 keV. Double-humped peak at 65 keV has been fitted by GEANT3 simulation curve assuming fragment speed ≈0.025c corresponding to a very asymmetric mass division of the plutonium nucleus. This peak has been interpreted as $K_{\alpha1}$ x-ray from a heavy element (possibly iridium). Double-humped peaks around 88 keV has been fitted with GEANT3 simulation curve assuming fragment speed ≈0.05c corresponding to approximately symmetric mass split of plutonium nucleus and this peak has been interpreted as a fission fragment γ-ray.

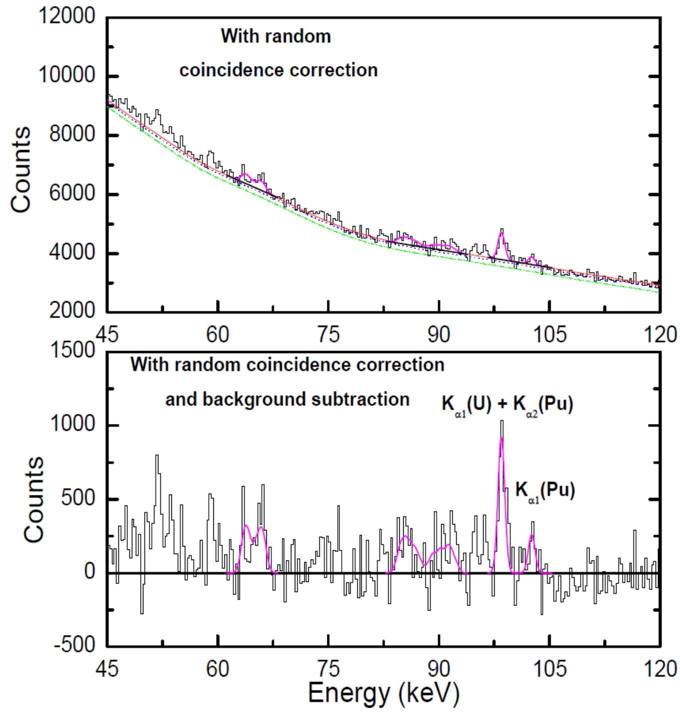

Fig. 1. Random coincidence corrected photon spectrum with solid red, dotted blue and dot-dash green backgrounds in the upper panel. Red background corrected spectrum shown in the bottom panel as discussed in the text.

In Fig. 2, we show background subtracted true coincidence spectrum in the energy range from 96.5 keV to 107 keV with statistical error bars on the data points. In order to decide whether the data points are compatible with a fission fragment γ-ray, a GEANT3 simulation [10] of a sharp 101 keV γ-ray line from a fission fragment has been performed with the actual experimental setup. The speed of the γ-ray emitting fission fragment is taken as ≈0.04c (corresponding to approximately symmetric mass split of plutonium nucleus) so that the simulated high and low energy humps would fall around 103 keV and 98.4 keV peaks to obtain the best fit. An additional yield at 98.4 keV as the remnant of uranium $K_{\alpha1}$ x-ray has been added to the GEANT3 simulation. The yields of the remnant of uranium $K_{\alpha1}$ x-ray and fission fragment γ-ray could be adjusted to obtain the best fit (dotted blue curve) of the data points with a reduced χ^2=1.2. However, for this fit, GEANT3 simulated dotted broad hump become statistically very poorly defined (ratio of hump area to statistical error bar ≈ 2). On the other hand, if the data points are fitted by two isolated Gaussian peaks (FWHM=1 keV) corresponding to plutonium K x-ray lines and an additional yield at 98.4 keV is added as the contribution from the remnant of uranium K x-ray, then we not only get an excellent fit (solid red curve) with the reduced χ^2=1.0, but more importantly plutonium $K_{\alpha 1}$ Gaussian peak at 103 keV become statistically significant with >99% confidence level (ratio of peak area to error bar ≈ 4). So, clearly, the data points shown in Fig. 2 could be better described by two isolated Gaussian peaks (FWHM = 1 keV) centered around 103 keV and 98.4 keV with a high statistical confidence level (>99%).

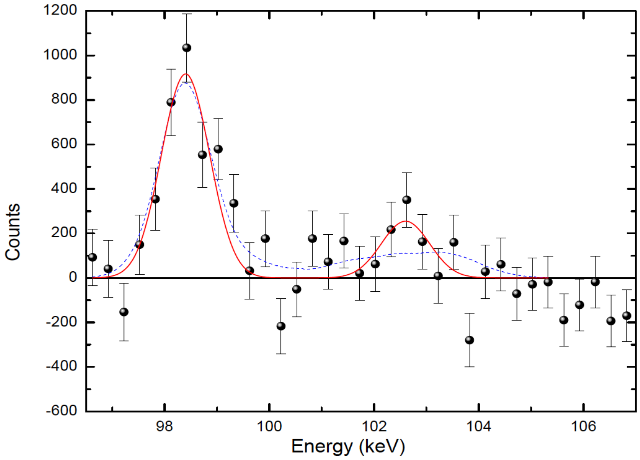

Fig. 2. Random coincidence corrected photon spectrum with statistical error bars. Red curve is the Gaussian fitting of the data points with two isolated peaks. Dotted blue curve is GEANT3 simulation of a sharp 101 keV fission fragment γ-ray along with a 98.4 keV line as discussed in the text.

Another method to distinguish between the fission fragment γ-ray and plutonium K x-ray is to determine photon multiplicities per fission at the relevant photon energies by gating on different regions of the fission fragment energy spectrum. In Fig. 3, we show photon multiplicity per fission versus fission fragment kinetic energy (without target thickness correction) for 103 keV, 88 keV and 65 keV photon energies as obtained by gating on different regions of the fission fragment kinetic energy bins. The number of fission fragments in each bin is the same. The horizontal error bar on each data point indicates the fission fragment kinetic energy region over which the integration has been done. In the case of fitting the 88 keV γ-ray humps, the required speed (≈0.05c) of the relevant fission fragment implies approximately symmetric fission. Correspondingly, we see almost all the photon multiplicity in the central fission fragment kinetic energy bin [Fig. 3(c)]. For fitting 65 keV K x-ray humps the required speed (≈0.025c) of the relevant fragment implies asymmetric mass splitting of the plutonium nucleus and correspondingly the photon multiplicity in the central fragment kinetic energy bin drops [Fig. 3(b)]. If 103 keV photons are from fission fragments moving with a speed of ≈0.04c as obtained from the spectral shape and relatively large yield, then qualitatively speaking, corresponding photon multiplicity should all be in the central kinetic energy bin as observed for the multiplicity plot of 88 keV fission fragment γ-ray. The qualitative observation of about equal photon multiplicity in each bin does not support the interpretation in terms of a fission fragment γ-ray and is compatible with the interpretation of plutonium $K_{\alpha 1}$ x-ray line. The centroid of the plutonium $K_{\alpha 1}$ x-ray line as obtained by taking weighted average over the Gaussian peak region comes at (102.8±0.5) keV and the corresponding plutonium $K_{\alpha 2}$ line would be at 98.6 keV overlapping with the uranium $K_{\alpha 1}$ line. The standard plutonium $K_{\alpha 1}$ and $K_{\alpha 2}$ x-ray energies are 103.7 and 99.5 keV respectively. Our atomic calculations indicate that the lower energy of the plutonium K x-ray lines could be because of the larger size of the deformed rotating fissioning nucleus and ionization of the outer orbitals of the plutonium atom in the fusion process. The width of the plutonium K x-ray lines would provide information about the presence of a long-lived fission component.

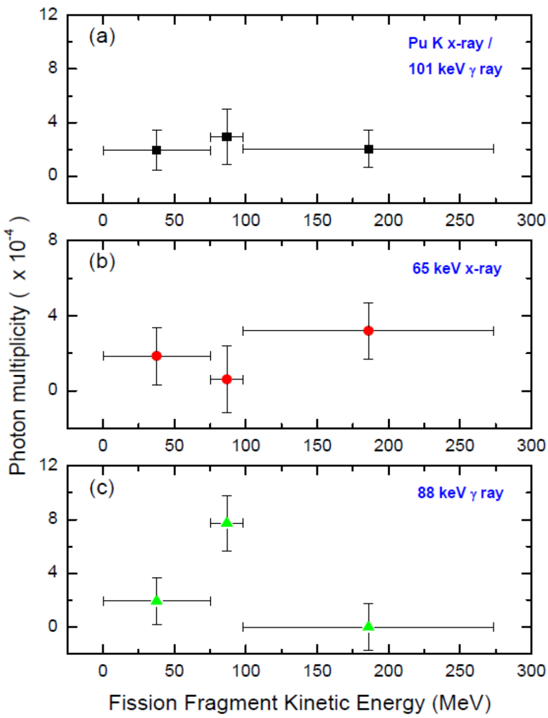

Fig. 3. Photon multiplicities at 103 keV, 88 keV and 65 keV as discussed in the text.

Gaussian fitting of the data points around 103 keV (from 101.7 keV to 105.3 keV) gives FWHM of the peak = (1±0.3) keV and a double Gaussian fit of the data points around 98.4 keV gives FWHM of the plutonium $K_{\alpha 2}$ x-ray line = (1±0.4) keV. Combining the Gaussian fit analyses, we obtain that the FWHM of the plutonium ion's K x-ray line is = (1±0.24) keV. The energy resolution (FWHM) of the LEPS detector as obtained from the singles uranium K x-ray lines is = (1.00± 0.01) keV and the intrinsic width of plutonium K x-ray line is =0.1 keV. So the increase of the intrinsic width of K x-ray line for a fissioning plutonium nucleus is = (0±0.24) keV. This result indicates the presence of long-lived fission for the highly excited plutonium nucleus and the mean fission time of the long-lived component could be estimated as [11] $> \frac{\hbar}{2.35\times0.24\ keV}$ $=1\times10^{-18}$ s. Here $\hbar = \frac{h}{2\pi}$ and h is Planck's constant. Hence the fission time should at least be on the order of 10^{-18} s. Since the fast fission events would produce very broad (>10 keV) spectral shape of low intensity, they would be cut out by the background curve. So this method is sensitive to long fission time component only and we could not determine the percentage of slow fission events by this method. In order to determine the percentage of slow fission event, we obtained K x-ray fluorescence yield per fission (σ_K/σ_f), where σ_K and σ_f are the cross-sections of K x-ray and fission respectively and the probability of K orbital vacancy (P_K) of plutonium produced in the fusion reaction. σ_f for this reaction has been taken as = (2±0.1) barn from a previous measurement [12]. σ_K has been obtained from the plutonium K x-ray yield in the coincidence spectrum. This yield has been corrected by the coincidence efficiency and normalized with respect to the singles uranium K x-ray yield. The absolute cross-section of uranium K x-ray produced by $^4He+^{238}U$ at $E(^4He)_{lab}$=60 MeV has been obtained from the measured and calculated K x-ray cross-sections [13] from different heavy elements by ^4He projectile at different energies. Using ref. [14], K orbital ionization probability of uranium atom due to the collision with a 60 MeV ^4He has been estimated as $P_U = 5.3\times10^{-4}$ with ~25% uncertainty. In the case of a fusion reaction, K orbital ionization probability should be halved [4] as only the incoming trajectory would contribute to the ionization. These uranium K orbital vacancies would be transferred to plutonium and a significant contribution would come due to shake-off ionization. We have used sudden approximation method of ref [15] and obtained shake-off ionization probability of K orbital of plutonium $P_{shake\text{-}off} \geq 4.5\times10^{-4}$. Hence total value of $P_K=(P_U/2+ P_{shake\text{-}off}) \geq 7.2\times10^{-4}$. Assuming an extreme bimodal fission time distribution with a very long fission time ($\tau_f \rightarrow \infty$) and a very short fission time ($\tau_f \rightarrow 0$), the percentage of long fission time component is given by $f_L = \frac{\sigma_K}{P_K \sigma_f} \times 100\%$. Considering the lower limit for P_K ($P_K=7.2\times10^{-4}$), we obtain f_L=78%. For any other fission time distribution, the percentage of slow fission components would increase. However P_K has two components P_{el} and $P_{shake\text{-}off}$. Although P_{el} has been obtained from experimental data with ~25% uncertainty, $P_{shake\text{-}off}$ has been calculated assuming sudden approximation. This method of calculating shake-off ionization has been very successful [15] in the case of β-particle emission and should be applicable for fusion also. Considering our uncertainties in estimating value of P_K, we conclude qualitatively that most of the fission events are slow.

3 Statistical significance of the results

The ratio of the counts under narrow (FWHM=1 keV) Gaussian peak around 103 keV to its statistical error is ≈4, implying that the narrow peak could be defined with >99% statistical confidence. In order to study the effect of background subtraction on our qualitative results, we have drawn two other background curves (Fig. 1). Dotted blue curve (Fig. 1) has been drawn below the solid red curve, but close to it. The purpose of drawing the dotted blue curve close to the solid red curve is to see the sensitivity of our results to small changes in drawing background. We find that the FWHM of the narrow Gaussian peak around 103 keV remains essentially the same for both the solid red and dotted blue backgrounds. However, for the dotted blue background, there are more counts under all the peaks, implying more random coincidence events. After correcting the area under the peaks by the coincidence efficiency corresponding to the dotted blue background, we qualitatively arrive at the same result that most of the fission events are slow with fission time at least on the order of 10^{-18} s. We have also studied the effect of drawing dot-dash green background curve (Fig. 1) that appears to be very low. In the case of dot-dash green background, there is essentially no negative count in the background subtracted spectrum and as a result, there are large counts under all the peaks. In this case, the data points around 103 keV could be best fitted by a straight line (reduced χ^2 =1.26), whereas the best fit by a Gaussian function gives a reduced χ^2=1.4 with a large Gaussian area and FWHM=4.4 keV. The combination of a large Gaussian area and width imply an unrealistically large value of P_K, if it is interpreted as plutonium $K_{\alpha 1}$ x-ray peak. In this case, the best fit of the data points around 103 keV in the background subtracted spectrum by a GEANT3 simulation of 101 keV fission fragment γ-ray gives a reduced χ^2=2, implying a poor fit. Since a straight line gives the best fit of the data points, it implies that the peak is riding on a large background, because the background curve (dot-dash green background) has been drawn too low. So for any reasonable background curve (such as the solid red or dotted blue background curves), we get a statistically significant narrow (FWHM=1 keV) peak around 103 keV and qualitatively similar results.

4 Conclusions

We have seen plutonium K x-ray lines from the fusion reaction $^4He+^{238}U$ at $E(^4He)_{Lab}$=60 MeV and obtained evidence for slow fission (~10^{-18} s) from the intrinsic width of K x-ray line. The percentage of slow fission events was determined from the measured K x-ray fluorescence per fission and the probability of K orbital ionization of plutonium in the fusion process, assuming a fission time distribution. It was found that most of the fission events are slow. Our results are in agreement with earlier fission time measurements of highly excited ^{238}U nucleus measured by the atomic techniques [1,2]. However, using prefission neutron multiplicity measurement technique, Saxena et al. [7] found that most of the fission events are fast with a mean fission time ~10^{-20} s. The general disagreement in the measurement of fission time by atomic and nuclear techniques might indicate new physics beyond fission dynamics.

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