Independent isomeric yield ratio of ^{134}I in the photofission of ^{235}U and ^{238}U

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The independent isomeric yield ratio of ^{134}I has been determined radiochemically for the photofission of ^{235}U and ^{238}U with bremsstrahlung with end-point energies ranging from 12 to 30 MeV. The root-mean-square values of the angular momentum of the corresponding fission fragments, J_{rms}, calculated using a statistical deexcitation model show an independence on the compound nucleus angular momentum and excitation energy. This J_{rms} behavior is compared to the results obtained in thermal neutron induced and medium-energy particle induced fission.

[NUCLEAR REACTIONS, FISSION $^{235,238}U(\gamma,F)$, $E_\gamma max=12$, 15, 20, 30 MeV; measured ^{134}I independent isomeric yields; deduced angular momenta.]

A study of the angular momentum of the fission fragments provides information on the scission configuration, leading to a better understanding of the dynamics of the fission process. Measurements of the anisotropy[1] and the number[2] of γ rays, emitted by the fission fragments, have been applied for the determination of the angular momenta of the fission fragments. Further methods, providing information on this subject, are the measurements of ground state band populations in even-even fission products[3] and the determination of isomeric yield ratios. An extensive survey of the results available in the literature, obtained with the latter method is given by Aumann *et al.*[4] These authors found a general increase of the fragment angular momenta in medium-energy-induced fission compared to low-energy-induced fission, attributed to the increase of the excitation energy and angular momentum of the fissioning compound nucleus.

Recently Denschlag *et al.*[5] and Bocquet *et al.*[6] performed experiments at the mass separator Lohengrin of the Institut Laue-Langevin (ILL) Grenoble to investigate the dependence of the fragment angular momentum on their kinetic energy. These experiments showed an increase of the angular momentum with increasing excitation energy of the fragments in most cases. In particular, for the fission fragment corresponding to ^{134}I a strong decrease of the root-mean-square angular momentum (J_{rms}) with the fragment kinetic energy E_K, $\Delta J_{rms}/\Delta E_K=-0.31\pm0.03\hbar$/MeV was found by Denschlag *et al.*[5] in $^{235}U(n_{th},f)$.

We studied the dependence of the isomeric yield ratio of ^{134}I on the bremsstrahlung end-point energy in the range 12–30 MeV for the photofission of ^{235}U and ^{238}U. Using a statistical model analysis the average angular momenta of the fragments corresponding to the measured isomeric ratios are deduced. As the photon absorption in the considered energy range is predominantly $E1$ absorption, the angular momentum, transferred in the reaction is well defined in our experiments.

Samples consisting of 1 g natural uranium [$UO_2(NO_3)_2\cdot6H_20$] or the ^{235}U target-catcherfoil setup, described in Ref. 7, were irradiated with a bremsstrahlung beam, produced in a 0.1 mm thick gold foil by an analyzed electron beam of the Linac of the Nuclear Physics Laboratory. After the irradiation the iodine fraction was separated from the natural uranium samples or the catcherfoils using radiochemical procedures close to the method described by Troutner *et al.* and Wahl.[8] As fission yield monitor we used ^{135}I. Experiments with irradiation times of 10 min and 2 h were performed. For the short irradiation runs the time interval between the end of the irradiation and the chemical separation of the iodine from the tellurium fractions was 2.5 min, for the longer irradiation runs this time interval was 1 hr. Several successive γ spectra were taken using a 50 cm^3 Ortec Ge(Li) detector and a conventional measuring chain. The resolution of the system was 1.8 keV at 1333 keV. The γ spectra were analyzed using the program CAOS.[9] The spectroscopic data of the studied fission products were adopted from Ref. 10. The independent yield of $^{134}I^m$ was deduced directly from the intensity of the 272 keV γ ray in the spectra of the short irradiation runs. The independent yield of $^{134}I^g$ was calculated from the intensity of the 847 and 884 keV γ rays in the spectra of the short and long irradiation runs using the following expression:

$$N=Y_1f_1+Y_2f_2+Y_3f_3 \qquad (1)$$

with

$$f_1=\frac{(e^{-\lambda_2 t_1}-e^{-\lambda_2 t_2})}{\lambda_2(\lambda_2-\lambda_1)}[\lambda_2(1-e^{-\lambda_1\tau})e^{-\lambda_1 t_0}-\lambda_1(1-e^{-\lambda_2\tau})e^{-\lambda_2 t_0}] ,$$

$$f_2=\frac{e^{-\lambda_2 t_0}}{\lambda_2}(1-e^{-\lambda_2\tau})(e^{-\lambda_2 t_1}-e^{-\lambda_2 t_2}) ,$$

$$f_3=\frac{1}{\lambda_2-\lambda_3}\left[\frac{\lambda_3}{\lambda_2}(1-e^{-\lambda_2\tau})(e^{-\lambda_2 t_2}-e^{-\lambda_2 t_1})e^{-\lambda_2 t_0}-\frac{\lambda_2}{\lambda_3}(1-e^{-\lambda_3\tau})(e^{-\lambda_3 t_2}-e^{-\lambda_3 t_1})e^{-\lambda_3 t_0}\right] .$$

In this expression Y_1, Y_2, and Y_3 represent the cumulative yield of ^{134}Te and the independent yields of $^{134}I^g$ and $^{134}I^m$, respectively. The decay constants of ^{134}Te, $^{134}I^g$, and $^{134}I^m$ are λ_1, λ_2, and λ_3. The irradiation time is indicated by τ and the time interval between the end of the irradiation and the chemical separation by t_0. The normalized number of decaying $^{134}I^g$ nuclei during a measurement starting at time t_1 and ending at time t_2 after the chemical separation is represented by N. As Y_3 is deduced from the intensity of the 272 keV γ ray in the short irradiation runs, Y_1 and Y_2 can be solved from a set of equations (1) for the short and long irradiation runs.

The isomeric ratios $\sigma_m/\sigma_g+\sigma_m$ for ^{134}I obtained in our experiments are listed in Table I. The values for the average excitation energy of the ^{235}U and ^{238}U compound nucleus after irradiation with 12-, 15-, 20-, and 30-MeV bremsstrahlung, reported in Refs. 7 and 11 are also included in the table. It is clear that the values for ^{235}U and ^{238}U do not differ significantly and that the isomeric ratios are practically independent of the bremsstrahlung end-point energy.

Using a statistical model analysis for the deexcitation of fission fragments developed by Min and Martinot[12] (computer code MAMI), the average initial angular momenta of the primary fission fragments, leading by the emission of prompt neutrons and γ rays to ^{134}I were deduced from the experimentally determined isomeric ratios. A description of the code MAMI, which takes into account the competition between neutron and gamma emission at each step of the deexcitation path and the feeding of the different discrete levels in the final nucleus, can be found in Ref. 6. The transmission coefficients for neutron emission $T_1(E)$ were taken from the report of Lindner.[13]

TABLE I. Independent isomeric yield ratios of ^{134}I in the photofission of ^{235}U and ^{238}U and deduced root-mean-square angular momenta, J_{rms}.

	^{235}U			^{238}U		
E_e (MeV)	$\langle E_{exc}\rangle$ (MeV)	$\frac{\sigma_m}{\sigma_g+\sigma_m}$	J_{rms} ($\hbar$)	$\langle E_{exc}\rangle$ (MeV)	$\frac{\sigma_m}{\sigma_g+\sigma_m}$	J_{rms} ($\hbar$)
12	9.7	0.51±0.09	8.8±0.8	9.7	0.51±0.05	8.6±0.7
15	11.6	0.54±0.04	9.5±0.6	11.6	0.53±0.04	9.1±0.6
20	13.1	0.53±0.03	9.4±0.6	13.4	0.48±0.04	8.6±0.6
30	14.1	0.49±0.03	8.8±0.6	14.7	0.53±0.04	9.3±0.6

For the probability distribution of the initial spin states of the fragments $P(J_i)$ the commonly used expression

$$P(J_i) \propto (2J_i+1)\exp\left[\frac{-J_i(J_i+1)}{B^2}\right]$$

was adopted with B a parameter similar to a spin cutoff. For each value of the parameter B a corresponding theoretical value for the isomeric yield ratio can be calculated. Using this relationship the B value, corresponding to the measured value of the isomeric yield ratio can, be deduced.

The total excitation energy of two complementary fragments with mass M_1, M_2, and charge Z_1, Z_2, $E^*_{\text{total}}(M_1,Z_1,M_2,Z_2)$, is given by the difference between the energy release in the considered fission event $Q(M_1,Z_1,M_2,Z_2)$ and the total kinetic energy of the fragments $E_K(M_1,Z_1,M_2,Z_2)$ based on the conservation law of energy. For the calculation of the Q values, we used the mass formula of Garvey *et al.*[14] Following Aumann *et al.*[4] two extreme cases were considered to calculate $E^*_{\text{total}}(M_1,Z_1,M_2,Z_2)$: the total kinetic energy of the fragments for the mass split M_1/M_2 is independent on the charge division Z_1/Z_2 and the total excitation energy of the fragments for the mass split M_1/M_2 is the same for all charge divisions. The information on the total kinetic energy release in the photofission of ^{235}U and ^{238}U was adopted from our previous work.[15,16] The calculated total excitation energy E^*_{total} was divided among the two complementary fragments proportional to the average number of emitted neutrons. The neutron emission curve was derived from the performed energy correlation measurements[15,16] and the determined post neutron mass distribution[7–11] as described in Ref. 17. Owing to the lack of information on the dependence of the variance of the fragment excitation energy on the fragment mass an estimation for this parameter was deduced from the behavior of the dispersion of the number of emitted neutrons as a function of the fragment mass, measured by Signarbieux *et al.*[18] for ^{252}Cf spontaneous fission. The broadening of the fragment excitation energy distribution, due to the use of a continuous bremsstrahlung spectrum was also taken into account.

The primary parentage of ^{134}I, i.e., the relative contribution of the different higher mass iodine isotopes, leading to ^{134}I by neutron and gamma emission, was deduced from our mass and charge distribution studies in photofission[7,11,15,16] and the neutron multiplicities calculated by the MAMI code. Based on the similarity of the values for the width of the post-neutron charge distribution obtained in $^{235}U(n_{\text{th}},f)$ and the photofission of ^{238}U with 20 MeV bremsstrahlung,[19] the value 0.35 for the width parameter σ^2 of the preneutron charge distribution was adopted from Clerc *et al.*[20] The procedure for the calculation of the primary parentage is supported by the good agreement between the average number of emitted neutrons determined experimentally and the output of the program MAMI.

In Fig. 1 the calculated dependence of the isomeric ratio of ^{134}I on the cutoff parameter B is shown for the photofission of ^{238}U with 20 MeV brems-

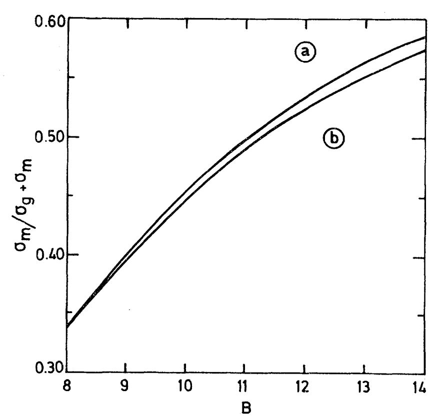

FIG. 1. Calculated isomeric ratio $\sigma_m/\sigma_g+\sigma_m$ of ^{134}I for the photofission of ^{238}U with 20 MeV bremsstrahlung, assuming the total excitation energy of the two complementary fragments (curve a) or the total kinetic energy release (curve b) independent on the charge division for a given mass split.

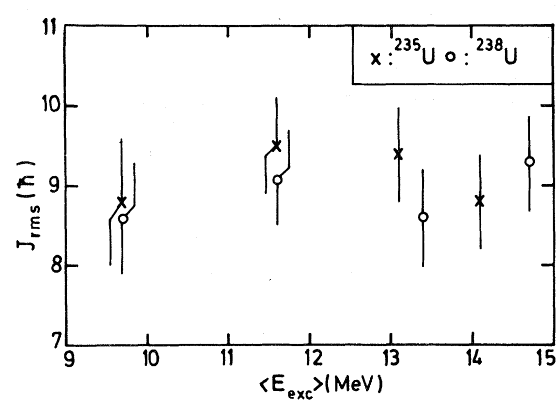

FIG. 2. The root-mean-square values J_{rms} deduced from the isomeric ratio of ^{134}I for the photofission of ^{235}U and ^{238}U as a function of the average excitation energy of the compound nucleus.

strahlung. In the case of curve *a*, the total excitation energy of the two complementary fragments is assumed to be independent on the charge division for a given mass split. For curve *b* the total excitation energy is calculated by assuming the total kinetic energy release independent on the charge division. From this figure it is apparent that the value for *B* and the corresponding value for J_{rms}, deduced from the experimental value of the isomeric ratio, is not very sensitive to the considered assumption.

The root-mean-square values J_{rms} deduced from the isomeric ratio of ^{134}I for the photofission of ^{235}U and ^{238}U are also given in Table I and depicted graphically in Fig 2. The indicated uncertainties on J_{rms} are based on the experimental errors on the isomeric ratios.

From an examination of our results, one can conclude that the average angular momenta of the fragments, leading to ^{134}I by neutron and γ emission, are almost independent on the compound nucleus excitation energy in the considered energy range. This was also observed in our previous work[7,11] for the angular momenta of the fragments corresponding to ^{131}Te but in the case of ^{126}Sb, ^{128}Sb, and ^{132}I a slight increase with the bremsstrahlung end-point energy was found. Diksic and Yaffe[21] determined the isomeric ratios of different iodine and tellurium isotopes in the fission of ^{238}U with protons of energy 30–85 MeV (compound nucleus excitation energy 35–90 MeV) and deduced also corresponding J_{rms} values. According to these authors, the average angular momenta for ^{131}Te and ^{134}I show also almost no increase with the bombarding energy in contradiction to the J_{rms} behavior observed in the other cases.

From Fig. 2 it is clear that the J_{rms} values for fragments leading to ^{134}I for the photofission of ^{235}U and ^{238}U are the same within the experimental uncertainties, although the spin and parity of the compound nucleus ^{235}U is $\frac{5}{2}^+$, $\frac{7}{2}^+$, and $\frac{9}{2}^+$ and spin and parity of the compound nucleus ^{238}U is 1^-. This independence of the fragment angular momentum on the spin of the fissioning nucleus was also observed in thermal neutron induced fission.[22] However, the J_{rms} value, $6.1 \pm 0.6\hbar$, deduced by Denschlag *et al.*[5] from the isomeric ratio of ^{134}I in ^{235}U(n_{th},f) (compound nucleus excitation energy 6.5 MeV) at the average kinetic energy of the fragments, is significantly lower than the values obtained in our photofission experiments.

According to calculations of Dietrich and Zielinska-Pfabé,[23] the dependence of the average fragment angular momentum on the fragment mass has a sawtooth form, resulting from the influence of the shell structure on the bending mode. These calculations predict in the mass region 134–136 values for the average angular momentum ranging from 3.5 to $5\hbar$ for the fissioning nucleus ^{234}U and from 4 to $6\hbar$ for the fissioning nucleus ^{236}U, depending on the intrinsic temperature of the system. The average angular momenta, deduced from our measurements of the isomeric ratio of ^{134}I for the photofission of ^{235}U and ^{238}U, which are about $1\hbar$ lower than the J_{rms} values, summarized in Table I, are significantly higher than these predictions. Our results support the conclusions of Bocquet *et al.*[6] that the angular momentum of the fragments is not so strongly correlated to their deformation.

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