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RATIO OF Cd^{115} ISOMERS PRODUCED IN HEAVY-ION-INDUCED FISSION

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Abstract

Isomeric cross-section ratios have been measured for the formation of Cd^{115} isomers in C^{12}- and O^{16}-induced fission in Au^{197} and U^{238}. The cross-section ratio (high spin/low spin) varied from 1.60 to 3.43 for the system $Au^{197} + C^{12}$, with the C^{12} ion energy ranging from 73 to 120 MeV. In the same energy range the ratio for the system $U^{238} + C^{12}$ varied from 0.55 to 0.82.

The average intrinsic spin, $\langle \ell_f \rangle$, of the fragment that decays to the Cd^{115} isomer pair was estimated from a simple model. The cross-section ratio showed the same relative increase with $\langle \ell_f \rangle$ for both targets. At the same value for $\langle \ell_f \rangle$ the ratio is much larger for Au^{197} than it is for U^{238}. This difference is related to the fact that in the Au system, the Cd^{115} is produced in a more asymmetric division.

Ratio of Cd^{115} Isomers Produced in Heavy-Ion-Induced Fission*

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It has been reported[1] that the ratio of the yield of Cd^{115m}(I=11/2) to that of Cd^{115g}(I=1/2) is 20 times as great in the fission of gold by carbon ions as in the fission of U^{235} by thermal neutrons. No correction was made in this work for the growing in of Cd^{115} via Ag^{115}, although the authors state that they estimate that approximately 30% of the Cd^{115} comes by this path rather than by direct production. Consequently, the reported ratios reflect variations in the charge distribution in the fission as well as in any angular momentum in heavy-ion-induced fission,[2] and it seemed worth while to study more carefully the Cd^{115} isomer ratio in these reactions. The mode of chemical separation allowed correction of the ratio for production of Cd^{115} isomers via Ag^{115}. Thus, the corrected ratios reflect rather accurately the values for direct production in fission.

Experimental Procedure

The target foil stacks were bombarded in the Berkeley heavy-ion linear accelerator (Hilac) with C^{12} and O^{16} ions accelerated to 10.4±.2 MeV per nucleon. A 0.25-mil Al foil preceded the target stack to ensure that the ions were fully stripped. In a typical gold target stack, four accurately weighed gold foils (approximately 1.18 mg/cm^3 thick) were sandwiched between two 0.7-mil aluminum foils. The sandwich arrangement was necessary to catch

the recoiling fission fragments through a 4π solid angle. Thus, there were four gold foils and eight aluminum foils in each stack. For the uranium targets, uranium tetrafluoride was evaporated to a thickness of 0.3 to 1.5 mg/cm^2 onto an aluminum foil backing 0.2 mil thick. The stack arrangement was as described for the gold targets. The integrated beams varied between 1 and 2 microampere hours (about 3 hours of irradiation time). A water-cooled copper block target holder was used which also served as the Faraday cup. The range-energy curves compiled by Hubbard for heavy ions in metals[3] were used to calculate the median energy of the beam as it passed through each target foil.

Immediately following the termination of bombardment, the target stack was disassembled, and each target foil, plus its two aluminum sandwiching foils, dissolved as a unit with Cd, Sb, and Mo carriers. As rapidly as possible, silver carrier was added to the solution and AgCl precipitated. After centrifugation and washing, the AgCl precipitate was dissolved in NH_4OH and set aside. The cadmium in the supernate was separated from the aluminum by precipitation of $Cd(OH)_2$ with NaOH solution. Addition of Sb and Pd carrier to the solution of the $Cd(OH)_2$ precipitate in excess of 2 M HCl was followed by precipitation of the acid group sulfides with H_2S. Subsequent to centrifugation and decantation, the supernate was diluted with H_2O to precipitate CdS. This precipitate was dissolved in 5 ml of 3 M HCl and reprecipitated by dilution to 40 ml. Following dissolution of the CdS in 10 ml of 3 M HCl, the solution was poured onto a column of Dowex-1 anion-exchange resin in the Cl^- form. The resin bed was washed with 2 M HCl and then with 0.1 M HCl. Finally, the Cd was eluted with 1.5 M H_2SO_4. After addition of Fe, Zr, Te, and La carriers, a hydroxide scavenge was performed using NH_4OH. The supernate was acidified and CdS precipitated. The CdS was filtered and washed

twice each with water and acetone. It was then oven-dried, weighed, and mounted for counting. Cadmium carrier was added to the silver solution (after several hours so as to allow decay of the Ag^{115})and the AgCl reprecipitated. The supernate was scavenged with NH_4OH, and CdS subsequently precipitated as in the latter steps of the above procedure.

Both the original CdS and the milked CdS samples were counted in an end-window proportional counter at appropriate intervals over a 5-month period to allow accurate determination of the half-lives. The decay curves agreed well with the literature values of 53 hours (Cd^{115g}) and 43 days (Cd^{115m}). The cadmium activity in the silver samples allowed correction for the contribution to the Cd^{115} isomers via Ag^{115g}. Unfortunately, there is a short-lived Ag^{115m} for which it was not possible to obtain a direct radiochemical correction. The decay scheme suggested by Coryell et al.[4] can be used to provide an estimate of the upper limit of the possible error involved by neglecting the Ag^{115m} formation. If we assume that all the Ag^{115g} comes from decay of Ag^{115m}, the amount of Cd^{115g} in the milked sample allows us to calculate that the measured ratio of isomer yield would be not more than 3% too large. Since the ratio differences are much larger than this, it is obvious that neglect of the correction for Ag^{115m} is not important to our conclusions. Previously, Porile called attention to the much lower probability for Ag^{115} production than for Cd^{115} in fission of neutron-deficient compound nuclei,[5] which is the situation for heavy-ion-induced fission.

The counting data were corrected for background and resolved into the two components. The cadmium activities in the silver fraction, the bombardment time, and the time of separation of the cadmium from the silver were used to calculate the amount of Cd^{115} produced via Ag^{115}. The cadmium fraction activities were extrapolated back to time at end of bombardment,

corrected for the Ag^{115} decay, and the σ_m/σ_g calculated, assuming that the ratio of the counting efficiencies f_g/f_m was 0.9.[6] It was also necessary to subtract the In^{115m} activity from the Cd^{115g} activity. The correction for In^{115m} was made by observing the growth of the In^{115m}, and was in agreement with the estimate made from conversion coefficients.

Results

The values for the σ_m/σ_g ratio for the different systems studied are given in Table I. A number of other runs on these systems gave similar results.

In addition, several bombardments were performed in which a gold target foil was placed between several layers of 0.25-mil Al foil. Within experimental error there was no variation in the σ_m/σ_g ratio with thickness in aluminum. This means that, within the limitations of these experiments, no difference in angular distribution was observed between isomers.

Discussion

Among the important factors that determine the isomeric ratio are:[7] (a) the distribution in angular momenta J_f of the primary fragment, (b) the number and types of steps in the de-excitation of the primary fragment to the Cd^{115} isomer pair, (c) the angular momentum carried away at each step, (d) the probability of forming states of different spins during each step of the cascade, and (e) the spins of the isomeric states.

A variety of primary fragments, of widely distributed masses and excitation energies, can decay to Cd^{115}. This makes a quantitative treatment of the problem almost impossible. We will instead attempt to discuss the isomer ratio in a qualitative way.

Huizenga and Vandenbosch[7,8] in their quantitative analysis have shown the neutron and especially γ emission do not change the average spin of the nuclei appreciably. The cascade, however, spreads out the distribution of the

spin states. We limit ourselves to estimation of the average value $\langle \ell_f \rangle$ of the primary fragment and compare this with the spins of the Cd^{115} isomer pair, which are 1/2 and 11/2. It is assumed that the excited nucleus just prior to the γ de-excitation chooses to feed the metastable or ground state depending on which transition has the smaller spin change.[7]

Some of the factors determining $\langle \ell_f \rangle$ are the spin, ℓ_n, of the fissioning nucleus and the fraction of ℓ_n carried off as angular momentum by the primary fragment.

This fraction is a function of the scission shape of the nucleus. Recently Cohen and Swiatecki have, as an approximation to the exact liquid-drop calculations, developed formulae for calculating the total energy of two uniformly charged collinear spheroids.[9] This energy for varying ratios C/A of the major to minor axis is characterized by a specific minimum for a given value of the fissionability parameter X [where $X = (Z^2/A)/50.13$]. It has been experimentally demonstrated[10] that this ratio predicts fairly well the fragment kinetic energy release and thus the scission shape. The fragments are emitted along the symmetry axis, which in our case is the C axis.[11] At high angular momentum the spin axis is along the principal axis, perpendicular to the C axis.[11] If the moment of inertia is assumed to be that of a rigid body, the intrinsic spin, $\langle \ell_f \rangle$, of the spheroid with mass m is given by

$$\langle \ell_f \rangle = \ell_n (1 + K^2) R^{5/3} / (1 + K^2)\,(R^{5/3} + 1) + \frac{5K^2}{A_n} R\,(R^{1/3} + 1)^2 . \qquad (1)$$

Here $C/A = K$, $m_1/m_2 = R$, and $A_n = m_1 + m_2$ is the mass of the fissioning nucleus. We have assumed C/A to be the same for both fragments. Values for C/A were taken from Cohen and Swiatecki.[9] Further, we assumed first-chance fission to be the most probable at the excitation energies we are dealing with; thus $A_n = A_{cn}$, the mass of the compound nucleus. The choice of A_{cn}

was straightforward when Au was used as a target. In this case the fission occurs only after complete fusion (CF) of the target nucleus and the projectile.[12] However, when U^{238} is the target, only a part of the fission (85% with C^{12} and 75% with O^{16}) occurs from a CF reaction.[13]

The remaining fraction (i.e., 15% with C^{12}, 25% with O^{16}) comes from an incomplete fusion (ICF) reaction, in which approximately four nucleons are transferred to the target nucleus before fission.[13] We have assumed this ratio to be the same over the entire range of the bombarding energy.

The ratio R represents the primary ratio between the fragment masses before neutron evaporation. Average values for R were obtained from the average number of neutrons emitted, as estimated according to excitation energies and neutron binding energies involved; 4 MeV was chosen as the average kinetic energy of the neutrons.[13]

A most difficult problem was connected with the choice of ℓ_n. For a first-chance fission, $\ell_n = \ell_{cn}$, the spin of the compound nucleus. Average values $\langle \ell_{cn} \rangle$, have been calculated by Thomas for a square potential nuclear well with $r_0 = 1.5 \times 10^{-13}$ cm.[14] This model predicts fairly well the total interaction cross section in heavy-ion-induced reaction of U^{238}.[15] As stated above, the CF reaction contributes between 75 and 85% to this cross section. The ICF reactions occur at the expense of the complete amalgamation which produces states of high angular momentum.[13] Therefore, the $\langle \ell_{cn} \rangle$ values calculated by Thomas[14] are too high. Based on the measured cross sections for complete fission, average values $\langle \ell_{CF} \rangle$ for this process can be calculated by assuming the same ratio between the CF and ICF reactions over the energy range considered. For the ICF reactions, the average spin of the fissioning nucleus may be of the same order as $\langle \ell_{CF} \rangle$, since the transferred nucleons have maximum impact parameters. Consequently we assume $\langle \ell_n \rangle \approx \langle \ell_{CF} \rangle$.

For Au^{197} we can assume the cross section for the CF reaction to be the same as was found for U^{238}. The transfer of four nucleons to Au^{197} does not deposit enough excitation energy in the nucleus for it to undergo fission. Values for $\langle \ell_n \rangle$, calculated according to a procedure described in Ref. (15), are given in Table I together with the $\langle \ell_f \rangle$ values estimated from Eq. (1) for the fragments that decay to Cd^{115} isomers.

We see that $\langle \ell_f \rangle$ in all cases have values between the spins 11/2 and 1/2 of the isomer pair. The ratios observed are therefore, to a first approximation, reasonable. In particular this crude model explains qualitatively the low ratios: the largest fraction of the high spin of the fissioning nucleus is carried off as orbital angular momentum of the fragments. We further observe an expected increase in the ratio with increasing $\langle \ell_f \rangle$. The calculations also give a higher value for $\langle \ell_f \rangle$ with Au as target than for U at the same bombarding energy; this partly explains the higher isomer ratio for this system. There are, however, large differences between the ratios for the different targets at the same value for $\langle \ell_f \rangle$. A possible explanation for this discrepancy is that the $\langle \ell_n \rangle$ values are incorrect. The level width for fission is expected to increase with spin. This might have the effect with Au as target (where the fission threshold is fairly high) of increasing the average value of the spin of the fissioning nuclei compared with the $\langle \ell_{CF} \rangle$ values. There must, however, be large changes in order to change $\langle \ell_f \rangle$ appreciably.

Other factors, therefore, are contributing. Because of the spheroidal shape of the fragments at scission, the repulsive Coulomb forces may introduce angular momenta. This would alter the $\langle \ell_f \rangle$ values estimated above. The fission mode producing Cd^{115} therefore can have a strong influence on the isomer ratio.

For the U + C system, the Cd^{115} is formed by symmetric fission, since A = 118 is approximately the center of the mass-yield curve. However, for Au + C and Au + 0, Cd^{115} is formed by more asymmetric fission, as A = 98 is the center of the mass-yield curve.[16]

In the system U + 100-MeV protons, the Cd^{115} is produced via symmetric fission and the ratio is 0.3,[17] similar to that for U + C. In the fission of Bi by 450-MeV protons, the Cd^{115} ratio was found to have a value of 3.5. A = 115 lies on the heavy side of the bismuth mass-yield curve, similar to the systems Au + heavy ion. Consequently, there seems to be evidence that the asymmetry of the fission fragments strongly influences the relative yields of the isomeric states. Porile has suggested that an increase in the fission asymmetry, resulting in an increase in the deformation energy, might result in higher angular momentum for the fission fragments.[5]

A few other isomer ratios have been determined for independent fission yields. Talat-Erben et al. found that for Te^{131} formed in the thermal-neutron fission of U^{235} the ratio of high-spin to low-spin isomer was 4.8.[18] Haller and Anderson measured the isomer ratio for Br^{80} in the 70- to 160-MeV proton fission of uranium, and again the high-spin isomer was favored.[19] Croal and Glendenin report that, in the thermal-neutron fission of Pu^{239}, the low-spin isomers of Pd^{111} and Rh^{106} are favored.[20] In all but the Rh^{106} case, these ratios are in agreement with the suggestion that asymmetric fission imparts more angular momentum to the fragments, resulting in increased yield of the high-spin isomer, whereas symmetric fission, having lower angular momentum, favors the low spin state.

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Table I. Experimental ratios σ_m/σ_g and estimated values $\langle \ell_n \rangle$ and $\langle \ell_f \rangle$ for various systems

Bombardment No.	Ion	Target	Ion energy (lab) (MeV)	σ_m/σ_g	$\langle \ell_n \rangle$	$\langle \ell_f \rangle$
1	O^{16}	Au^{197}	72	1.75	9.1	0.9
			97	2.80	26	2.4
			128	3.72	40.0	3.7
			154	4.23	49.4	4.6
2	O^{16}	Au^{197}	97	3.14	26	2.4
			128	4.00	40.0	3.7
			154	4.25	49.4	4.6
3	C^{12}	Au^{197}	73	1.60	20.7	2.0
			90	2.64	29.4	2.8
			103	3.18	35	3.2
			120	3.43	40.5	3.7
4	C^{12}	Au^{197}	90	2.80	29.4	2.8
			103	3.08	35	3.2
			120	3.44	40.5	3.7
5	C^{12}	U^{238}	74	0.55	18.4	1.4
			95	0.77	30.4	2.3
			123	0.82	39.6	3.0
6	C^{12}	U^{238}	120	0.73	39.6	3.0
Ref. 1	O^{16}	Au^{197}	85	2.0[a]		
			102	2.3[a]		
Ref. 1	N^{14}	Au^{197}	89	2.1[a]		
Ref. 1	C^{12}	Au^{197}	64	1.6[a]		
			78	1.8[a]		

[a] Uncorrected for Ag^{115} decay

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