Angular Momentum Effects in the Gamma-Ray De-Excitation of Fission Fragments*

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The energy dissipated as γ rays during the de-excitation of fission fragments (7.2±0.8 MeV for U^{235} thermal fission) is somewhat higher than has been estimated theoretically (4.9 MeV). It has been suggested that this discrepancy arises from the angular momentum of the fission fragments. We have made a quantitative evaluation of this possibility, taking into account the angular momentum dependence of the level density, the nonexistence of levels of a given angular momentum below some minimum energy (yrast energy), and the competition between neutron and γ-ray emission. The initial angular momentum distribution is that derived from measurements of isomers produced in fission. Initial excitation energies are based on the known de-excitation properties of fission fragments. All other parameters were derived from sources having no direct connection with the fission process. The calculations for an average pair of fragments (Sr^{96} and Xe^{140}) indicate that 7.1 MeV should appear as γ rays and that the average photon energy is 0.9 MeV. The calculated average neutron energy and number of neutrons are also in agreement with experiment.

I. INTRODUCTION

THE last stage in the de-excitation of a fission fragment to its ground state is normally the emission of γ rays. The total energy dissipated in this manner is 7–8 MeV from the two fragments formed in the thermal-neutron induced fission of U^{235} and 8–9 MeV from the spontaneous fission of Cf^{252}. The average number of γ rays is about 8 for the first case and 10 for the second, corresponding to an average photon energy of about 1 MeV.[1–3]

Attempts to account quantitatively for the energy dissipated in γ rays have given estimates that are lower than these values. For instance, for the spontaneous fission of Cf^{252}, Leachman and Kazek[4] have estimated 4.0 MeV in γ rays and for the thermal-neutron-induced fission of U^{235} Terrell[5] has estimated 4.9 MeV. It has been suggested by a number of people that this discrepancy arises because the theoretical estimates do not take into account the angular momentum of the fragments.[1–3,6] For nuclei with excitation energies somewhat in excess of the neutron binding energy, neutron emission may not be inevitable, as assumed in the theories, but instead may be strongly inhibited with respect to γ-ray emission if the neutrons must carry away a large amount of orbital angular momentum. Thus at a given excitation energy there will be more γ-ray emission and less neutron emission from high-spin fragments than from low-spin fragments.

That this angular momentum effect is important in nuclear reactions has been demonstrated experimentally by Grover and Nagle,[7] who investigated systems in which only a level of spin 0 is available as the product of neutron emission, and found that the neutron-unstable nuclei with spins larger than $\frac{7}{2}$ prefer to decay by γ ray instead of neutron emission, even though the neutrons could be emitted with several hundred keV of kinetic energy. That this angular momentum effect is also important for particle-emission products well above the ground state is strongly suggested by the experimental results of Mollenauer[8] and of Alexander and Simonoff,[9] who found that the energy dissipated in γ rays is roughly proportional to the average angular momentum calculated for the compound nuclei expected to have been prepared in the systems they studied. Grover and Gilat[10] have found, in a theoretical investigation, that for a nucleus of a given angular momentum, γ-ray emission competes favorably with neutron emission if the excitation energy is less than the sum of the neutron binding energy and the energy of the lowest level of that angular momentum (yrast energy). Gordon and Aras have shown in a preliminary way that such an effect can account for the energy that appears as γ rays in fission.[11]

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[1] E. K. Hyde, *The Nuclear Properties of the Heavy Elements* (Prentice-Hall, Inc., Englewood Cliffs, N. J., 1964), Vol. III, pp. 279–286.
[2] S. A. E. Johansson and P. Kleinheinz, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), pp. 805–826.
[3] H. Maier-Leibniz, P. Armbruster, and H. J. Specht, *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 113.
[4] R. B. Leachman and C. S. Kazek, Jr., Phys. Rev. **105**, 1511 (1957).
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[10] J. R. Grover and J. Gilat, Phys. Rev. **157**, 802 (1967).
[11] G. E. Gordon and N. K. Aras, *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 73.

TABLE I. Some input data used for the calculations.[a]

Nuclide	B_n[b] (MeV)	B_p	B_α	Level density parameter[d] a (MeV^{-1})	δ[e] (MeV)	$\langle\Gamma_\gamma(E)\rangle^{f}/D(E)$ dipole	$\langle\Gamma_\gamma(E)\rangle^{f}/D(E)$ quadrupole	E (MeV)	$\langle D(E)\rangle^{f}$ (eV)
Xe^{140}	5.663	11.69	1.972	···	···				
Xe^{139}	4.136	11.40	1.457	16.1	0.982	1.9(−3)	5(−6)	7.95	50
Xe^{138}	5.733	11.12	0.768	14.8	0.942	8.0(−4)	2(−6)	8.80	128
Xe^{137}	4.460[c]	10.67[c]	2.278	13.5	0.964	3.3(−4)	8(−7)	7.71	323
Sr^{96}	5.996	14.23	6.587	···	···	···	···	···	···
Sr^{95}	4.122	13.82	6.700	10.2	1.176	3.8(−3)	2.5(−6)	8.37	37
Sr^{94}	7.440[c]	13.39	6.348	10.1	1.176	3.8(−3)	2.5(−6)	9.55	37
Sr^{93}	4.600[c]	12.95	5.637	10.0	1.176	3.8(−3)	2.5(−6)	8.37	37

[a] Notation is as follows: B_n = neutron binding energy; B_p = proton binding energy; $B_\alpha = \alpha$ binding energy; a = level density parameter as defined by Lang; δ = "condensation energy," used in level density and other formulas to take account of evenness or oddness of neutron and proton numbers; $\langle\Gamma_\gamma(E)\rangle$ = mean total dipole or quadrupole radiation width of nucleus at energy E and at a specified spin J = "2"; $D(E)$ = mean level spacing at energy E and spin J = "2."
[b] Unless otherwise indicated, all binding energies are taken from A. G. W. Cameron and R. M. Elkin, Can. J. Phys. **43**, 1288 (1965).
[c] J. H. E. Mattauch, W. Thiele, and A. H. Wapstra, Nucl. Phys. **67**, 32 (1965).
[d] D. W. Lang, Nucl. Phys. **26**, 434 (1961).
[e] A. Gilbert and A. G. W. Cameron, Can. J. Phys. **43**, 1446 (1965). The δ for Sr^{95} was used for all Sr isotopes.
[f] The values of $\langle\Gamma_\gamma\rangle$ and of $\langle D\rangle$ were estimated from resonances seen in slow-neutron spectroscopy on neighboring similar nuclei. For further remarks see Ref. (12).

We present here some results of a calculation of the de-excitation properties of a typical pair of fragments (Sr^{96} and Xe^{140}) that takes into account the angular momentum effects on neutron emission and on the competition between neutron and γ-ray emission. The initial conditions of angular momentum and excitation energy and the various parameters used in the calculation were taken from known experimental data or from theoretical values; no adjustable parameters were used. The results of our calculations of the number of γ rays, energy dissipated as γ rays, average photon energy, average number of neutrons, and average kinetic energy of the neutrons are in good agreement with the experimental values, indicating that the above-mentioned discrepancy can indeed be blamed on the neglect of angular momentum effects. There is thus no reason to expect that the statistical model cannot be successfully applied to evaporative de-excitation of the excited primary fission products, or that any other model is needed to account for the de-excitations.

II. METHOD

The calculations were carried out using the Brookhaven IBM 7094 computer. The calculative procedure used was that described by Grover and Gilat.[12]

The emissions of neutrons, dipole and quadrupole γ rays, protons, and α particles were taken into account. In particular, the de-excitation by cascades of γ-ray emissions was followed all the way to the ground state in each nucleus participating in the chain of successive neutron emissions.

Evaporation Parameters

Many of the input data used in the de-excitation calculations are collected in Table I, together with their sources. The transmission coefficients for $Sr^{95}+n$, $Rb^{95}+p$, $Kr^{92}+\alpha$, $Xe^{139}+n$, $I^{139}+p$, $Te^{136}+\alpha$ were used for all calculations. The optical model and parameters used to obtain the transmission coefficients are those of Bjorklund and Fernbach[13] for neutrons, of Bjorklund, Campbell, and Fernbach[14] for protons, and of Huizenga and Igo[15] for alphas.

The yrast levels for Sr^{93}, Sr^{94}, Sr^{95}, Xe^{137}, Xe^{138}, and Xe^{139} were estimated in the way described in Ref. 16, where they are also displayed. Since in these calculations the residual interactions (other than the pairing force) are not taken into account, it sometimes happens that the calculated yrast levels for three or more consecutive angular momenta have the same configuration and thus the same energy, as for $J=9/2$, 11/2, 13/2, and 15/2 in Xe^{137} (see Ref. 16). Whenever these situations occurred in the fission-fragment–de-excitation calculation, one or more yrast levels were unable to decay to the ground state by either dipole or quadrupole radiation. We have opted to break these degeneracies quite arbitrarily and to suit our own purposes, but only to the extent necessary to avoid "stranding" nuclei in levels other than the ground state. The energies by which the levels were shifted are much smaller than would usually be expected from the degeneracy-breaking effects of the neglected residual interactions. The effect of these level shifts in the calculated apportionment of excitation energy between neutrons and γ rays is small (see Ref. 10) compared with the over-all effect of simply introducing the yrast levels and keeping track of angular momentum in the neutron and photon emissions. The effect of the shifts on the calculated average numbers of photons is larger, and contributes appreciably to our cited 10% uncertainty in these values (see remarks further on). The yrast level energies used in this work are shown in Figs. 1(a) and (b).

The "yrast temperatures" T_J are an important part of our calculation of level densities, because they de-

[12] J. R. Grover and J. Gilat, Phys. Rev. **157**, 802 (1967).

[13] F. Bjorklund and S. Fernbach, Phys. Rev. **109**, 1295 (1958).
[14] F. Bjorklund, G. Campbell, and S. Fernbach, Helv. Phys. Acta, Suppl. **VI**, 432 (1961).
[15] J. R. Huizenga and G. Igo, Nucl. Phys. **29**, 462 (1962).
[16] J. R. Grover, Phys. Rev. **157**, 832 (1967).

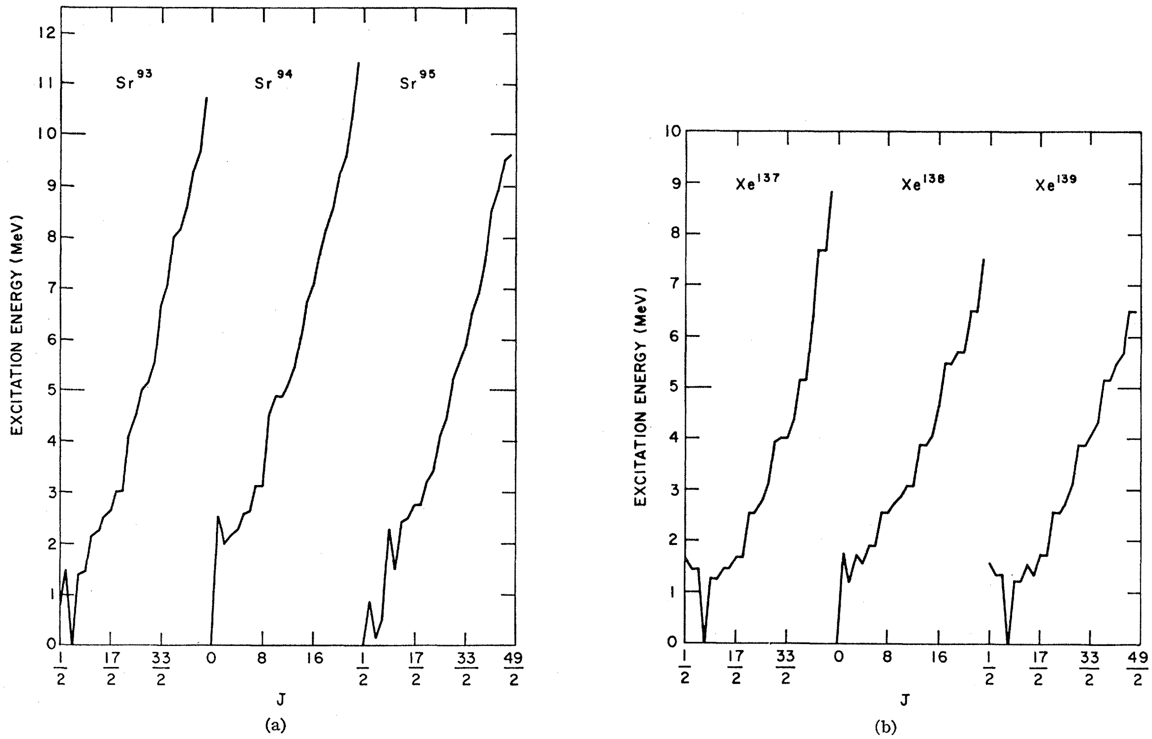

FIG. 1(a) Yrast levels of $Sr^{93,4,5}$ used in the calculations. (b) Yrast levels of $Xe^{137,8,9}$ used in the calculations.

scribe the behavior of the nuclear level density in a region of energy and angular momentum in which the conventional formulas are inapplicable.[17] Much of the γ-ray cascade takes place in this region. It is assumed that the level spacings between the lowest-energy levels at given spin J, beginning with the yrast level E_J, decrease with increasing excitation energy E in proportion to $\exp[-E/T_J]$. This relationship gives the definition of T_J. It is easily shown that $T_J \approx (E-E_J)/\ln N_J(E)$

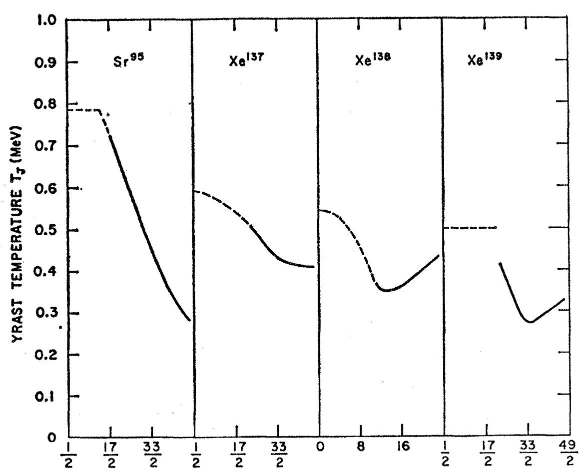

FIG. 2. Yrast temperatures used in the calculations.

[17] T. Ericson, *Advances in Physics*, edited by N. F. Mott (Taylor & Francis, Ltd., London, 1960), Vol. 9, p. 425.

where N_J is the total number of levels of spin J at and below energy E. The level density resulting from this prescription near the yrast levels is made to join smoothly at higher energies with the level density calculated by Lang's prescription (see Table I). The yrast temperatures used in this work are shown in Fig. 2. The procedure for the calculation of level densities is given in Ref. 12, the relevant parameters used here being listed in Table I. The dipole transition rates were estimated from slow-neutron capture data and the quadrupole transition rates were obtained by assuming that the reduced rate for dipole emission is 10^4 times the reduced rate for quadrupole emission. The factor 10^4 is approximately the ratio of single particle rates.

Computational Approximations

Unfortunately the limits on computer speed and memory size restrict our program to the use of only one table of level densities, while the properties of two nuclei are simultaneously important to the calculation at each stage of nuclear de-excitation: (i) the nucleus to which neutron emission proceeds, and (ii) the neutron-emitting species, for which γ-ray de-excitations are also calculated. We chose to calculate the table of level densities to be appropriate for the neutron-emission products, and then to approximate the level densities in the nucleus for which γ-ray de-excitation is calculated. This was done by adjustment of the level density table upwards or downwards in energy as appropriate by the condensation energy δ (except that no level was allowed

TABLE II. Comparison of calculated and experimental quantities (in MeV) for the de-excitation of Sr^{96} and Xe^{140}.

	Sr^{96}	Xe^{140}	Total	Expt	Xe^{140} $\mathcal{J}=\infty$
Total γ-ray energy	3.97	3.11	7.08	7.2 ± 0.8[a]	2.00
Number of γ rays	4.18	3.99	8.2	7.4 ± 0.8[a]	1.73
Average energy per photon	0.95	0.78	0.87	0.97 ± 0.15	1.16
Average number of neutrons	1.38	1.12	2.50	2.43[b]	1.29
Average kinetic energy of neutrons	1.13	0.97	1.06	1.20[c]	1.15

[a] F. C. Maienschein, R. W. Peele, W. Zobel, and T. A. Love, in *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), p. 366.
[b] Reference 21.
[c] Reference 23.

to assume a negative energy). We thus calculate the divisions of energy into neutrons and photons accurately, and accept inaccuracies in the calculated photon spectra, which are not serious for the purpose of this paper. An account of the calculative errors in the γ-ray spectra does not seem appropriate for this work but will be incorporated into a paper now in preparation, that deals with the γ-ray spectra *per se*. However, we estimate that the average number of photons emitted (Fig. 4) at any given excitation energy is accurate to about $\pm10\%$. On the other hand the gross variations, i.e., the existence and positions of the maxima, and minima, are representative of real features. The grand average photon energy reported in Table II should be more accurate than any given point in Fig. 4 since the errors tend to compensate when an average is taken over a wide spread of energy.

With the calculative procedure that we used, it is necessary to choose the energy mesh size that will give the most accurate results possible. Unfortunately our calculations could not be simultaneously optimized for both the neutron-γ-ray energy apportionment, and the γ-ray spectrum (see Ref. 12). Each calculation was therefore performed twice; once with an energy mesh size of 0.1 MeV (or 0.2 MeV for the largest excitation energies considered) to obtain accurate energy apportionment results, and again with an energy mesh of 0.333 MeV (for Xe^{140}) and of 0.5 MeV (for Sr^{96}) to improve the accuracy of the calculated γ-ray spectra.

Fission Parameters

To perform the calculation it is necessary to know the distribution of initial excitation energies and angular momenta of the fission fragments. Unfortunately, this information is not available at present, and we must resort to plausible estimates instead.

Assuming that the energy and angular momentum are uncorrelated, we treat their joint distribution as a product of an angular momentum distribution and an energy distribution. For angular momentum we have taken the functional form used by Vandenbosch and Warhanek[18] and by Sarantites, Gordon and Coryell[19] for their analysis of isomer yields from fission

$$N(J)\propto(2J+1)\exp[-J(J+1)/2b^2],$$

where $N(J)$ is the probability of forming a fragment with a particular angular momentum J. The parameter b is taken to be 6 on the basis of the isomer ratio results. The most probable angular momentum is about $5.5\hbar$, somewhat lower than is predicted theoretically by Nix and Swiatecki[20] for fission of At^{213}. We assume that the distribution of excitation energies is Gaussian about a mean value and we must therefore know this mean and a width parameter. The mean energies were taken to be 13.0 MeV for Sr^{96} and 10.1 MeV for Xe^{140} and were determined in the following way. The average number of neutrons per fragment was taken from Terrell's work (1.12 for the light fragments and 1.31 for the heavy).[21] This quantity was multiplied by an average neutron binding energy (6.12 MeV for the light fragment and 4.44 for the heavy) estimated from Seeger's masses,[22] and by the average center-of-mass neutron energy (1.20 MeV) given by Terrell.[23] The sum of these two products gave the portion of the excitation energy dissipated in neutron emission. The energy emitted as γ rays (7.5 MeV) was divided between the two fragments in proportion to the average neutron-binding energies, since the higher this energy the higher the excitation energy at which γ-ray emission will compete with neutron emission. We may perhaps be accused of assuming what we want to prove by using this method of determining the initial excitation energy. However, our goal is not to perform a theoretical calculation of the initial excitation energy, which depends in some way on the fission process itself, but rather to show that once we are given the correct initial excitation energy we can calculate the correct division into neutron emission and γ-ray emission.

Since total energy is conserved, the dispersion in total excitation energy must be equal to the dispersion in total kinetic energy. We have used the value $\sigma_{tot}=9.3$ MeV, determined by Thomas, Gibson, Safford, and Miller[24] for the 96–140 mass split in the thermal-neutron-induced fission of U^{235}. (The quantity σ is the usual width parameter of a normalized Gaussian distribution). To determine from this width the appropriate parameters for the excitation-energy distributions we need to make some assumption about the correlation between the excitation energies of the two fragments.

[18] H. Warhanek and R. Vandenbosch, J. Inorg. Nucl. Chem. **26**, 669 (1964).
[19] D. G. Sarantites, G. E. Gordon, and C. D. Coryell, Phys. Rev. **138**, B353 (1965).
[20] J. R. Nix and W. J. Swiatecki, Nucl. Phys. **71**, 1 (1965).
[21] J. Terrell, Phys. Rev. **127**, 880 (1962).
[22] P. A. Seeger, Nucl. Phys. **25**, 1 (1961).
[23] J. Terrell, *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 3.
[24] T. D. Thomas, W. M. Gibson, A. J. Safford, and G. L. Miller (to be published).

We have assumed that there is perfect positive correlation—that is, as one excitation energy increases, the other increases in proportion. In this case the width parameters σ_L and σ_H for the light and heavy fragment distributions are given as

$$\sigma_Q = \langle E_Q \rangle / \langle E_{tot} \rangle \sigma_{tot},$$

where the index Q stands for either the light or heavy fragment and $\langle E_Q \rangle$ and $\langle E_{tot} \rangle$ are the single fragment and total average excitation energies, respectively. (The assumption of perfect positive correlation is not in agreement with the theoretical calculations of Nix and Swiatecki,[20] who predict a negative correlation. The effect of this assumption on the results presented here is not great).

III. RESULTS

Calculations were made for many fragment-excitation energies between 6 and 22 MeV. The resulting computer outputs were neutron and γ-ray spectra, the probability of emitting a given number of neutrons, and the probability of emitting a proton or an α particle. From the spectra we calculate the total energy emitted as γ rays, the total number of γ rays, the average photon energy, the average neutron energy, and the total number of neutrons. A portion of the outputs is shown in Figs. 3 and 4 where we have plotted the number of γ rays and the total γ-ray energy against excitation of the fragments. We have arbitrarily assumed straight line interpolation between zero yield of γ rays at zero excitation energy and the values at the lowest-excitation energies considered (i.e., the neutron binding energies in Sr^{96} and Xe^{140}).

We see immediately that for the average pair of fission fragments, Sr^{96} at an excitation energy of 13.0 MeV and Xe^{140} at 10.1 MeV, the calculated number of γ rays and the energy dissipated as γ-rays are close to the experimental values. We obtain a more realistic result by averaging the results of Figs. 3 and 4 over the distribution of excitation energies of the two fragments.

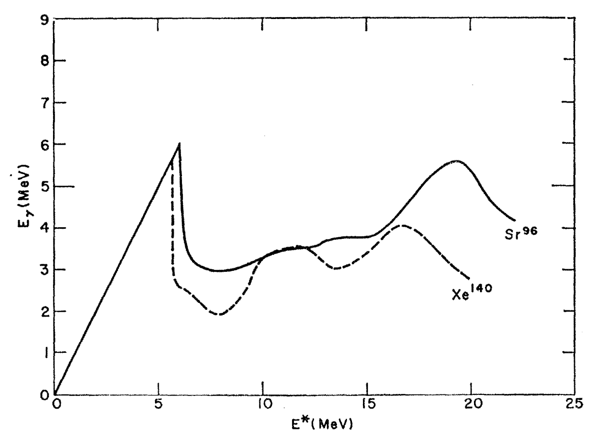

FIG. 3. Calculated mean total energy carried away by γ rays in the de-excitation of Sr^{96} and Xe^{140} excited to energy E^*.

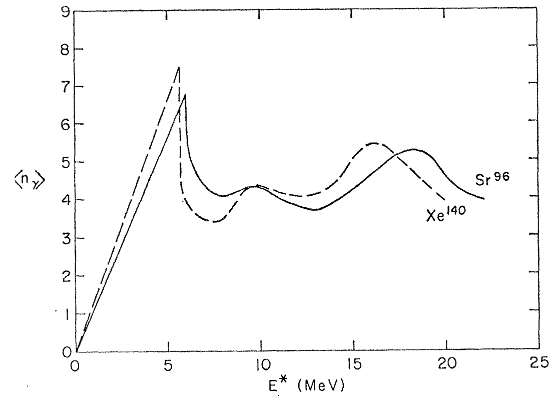

FIG. 4. Average total number of photons emitted from nuclei of Sr^{96} and Xe^{140} excited to energy E^*, including the contributions from the excited products of the neutron-emission cascade.

In order to perform this average we make the assumption described in the previous section, that the distributions are Gaussian, centered at 13.0 MeV with a width parameter σ of 5.2 MeV for the light fragment, and centered at 10.1 MeV with $\sigma = 4.0$ MeV for the heavy fragment. The use of these widths implies a strong positive correlation between the excitation energies of the two fragments. If there is no correlation then the widths should be larger by a factor of about $\sqrt{2}$ in order for the width of the assumed distribution of total excitation energy to agree with that of the distribution of kinetic energies. If, as Nix and Swiatecki[20] predict, the correlation is negative, the widths should be still greater. Inspection of Figs. 3 and 4 indicates, however, that for the regions of excitation energy for which these distributions predict a high probability, the quantities to be averaged do not change rapidly with excitation energy. As a result, the averages we are interested in are not very sensitive to errors in the width. This is not the case for the spectrum of γ rays, for instance, since the high-energy γ rays originate (for the fragment pair Sr^{96}, Xe^{140}) mostly from nuclei with very high initial excitation energies. The probability $P(\nu)$ of emitting a certain number ν of neutrons is very sensitive to assumptions about the correlation of excitation energies.

The results of the averaging are summarized in Table II, together with the experimental results and the results of a calculation done with the nuclear moment of inertia set effectively equal to infinity, all the yrast levels put at zero energy, and all the yrast temperatures made zero. This last calculation demonstrates the effect of not taking into account the angular momentum of the fragments. We see that the more realistic calculation yields numbers that are in good agreement with the experimental results for both γ ray and neutron emission. Furthermore, as the last column of Table II indicates, neglect of angular momentum results in a calculated energy dissipated in γ rays that is only $\frac{2}{3}$ of the correctly calculated quantity.