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EVALUATED NEUTRON REACTION DATA FOR URANIUM 233

**R. J. Howerton
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EVALUATED NEUTRON REACTION DATA FOR URANIUM 238

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MS. date: March 21, 1973

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EVALUATED NEUTRON REACTION DATA FOR URANIUM 238

Abstract

Describes methods used to obtain
evaluated neutron interaction and photon
production data for ^{238}U .

I. Introduction

This report describes the procedures used to obtain evaluated neutron interaction and photon production data for ^{238}U for the neutron energy range from 10 μeV to 20 MeV. The evaluation is designated as MAT 4187 in the DNA evaluated nuclear data collection obtainable from the Radiation Shielding Information Center, Oak Ridge National Laboratory. In this energy range a number of nonelastic reactions are energetically possible in addition to elastic scattering. Table 1 lists these reactions and their thresholds.

This evaluation neglects all of the charged-particle-producing reactions, both because there are essentially no measurements and because the Coulomb barrier suppression leads to very small cross sections. The reactions that are included in the evaluated data set, in addition to total and elastic cross sections, are total nonelastic, (n, n') , $(n, 2n)$, $(n, 3n)$, $(n, 4n)$, (n, γ) , $(n, \text{fission})$, and $(n, \text{X}\gamma)$.

The present report is divided into the following sections:

- II. Neutron cross sections
 - III. Neutron angular distributions
 - IV. Neutron energy distributions
 - V. Gamma-ray production cross sections
 - VI. Gamma-ray production angular distributions
 - VII. Gamma-ray production energy distributions
 - VIII. Integral tests of the evaluation
- Each section includes a discussion of errors.

Table 1. Neutron interaction and photon production reactions and thresholds for ^{238}U .

Reaction	Threshold (MeV)
n, n'	0.0452
$n, 2n$	6.17
$n, 3n$	11.32
$n, 4n$	17.89
n, p	3.19
n, np	7.69
n, d	5.46
n, nd	11.41
n, t	5.13
n, nt	9.99
$n, \text{fission}$	0.45
n, α	exoergic ($Q \approx 9 \text{ MeV}$)
n, γ	exoergic ($Q = 4.80 \text{ MeV}$)

¹Work performed under the auspices of the Defense Nuclear Agency and the U.S. Atomic Energy Commission.

II. Neutron Cross Sections

A. Total Cross Section

Total cross section measurements¹⁻²³ exist over the entire energy range of interest. In the energy region below 1 keV, ENDF/B-III MAT 1158 data were used, with temperature broadening to 300°K and deletion of some data points based on a 10% thinning criterion. No resonance parameter data are included explicitly in the evaluation. Individual total cross section data points have quoted errors ranging from about 1% to 10%. The evaluated total cross section curve has an estimated accuracy of 5% to 10%.

B. Nonelastic Cross Section

Nonelastic cross section measurements²⁴⁻³³ span the energy range from 0.15 MeV to 18.5 MeV, and they include measurements made by one of us (M.H.M., Ref. 30). For neutron energies greater than about 4 MeV, the nonelastic cross section is quite constant with energy. The quoted errors are about 10% to 20% at the lower energies and 5% to 10% at the higher energies. The evaluated nonelastic cross section curve has an accuracy of 15% at the lower energies and 10% at the higher energies.

C. Elastic Cross Section

Experimental measurements of the elastic cross section^{14, 18, 34} have been made between 0.3 MeV and 4 MeV and at 7 and 14.1 MeV. At higher and lower energies, the elastic cross section was determined from systematics and from the differences between measured values for the total and nonelastic cross sections. At energies below 1 keV, the elastic cross

section was based on ENDF/B MAT 1158 data, with the data temperature-broadened to 300°K and thinned using a 10% thinning criterion. The thermal neutron elastic scattering cross section is 8 barns (b), as determined by the difference between the total cross section and the capture cross section. Taking into account the errors attributed to the total cross sections and the nonelastic cross sections, we estimate accuracies for the elastic cross sections of about 15%.

D. Partial Nonelastic Cross Sections

These include the (n, γ) , $(n, \text{fission})$, (n, n') , $(n, 2n)$, $(n, 2n)$, $(n, 3n)$, and $(n, 4n)$ reactions.

(n, γ) Reaction

Measurements of the (n, γ) cross section³⁵⁻⁶² have been made from thermal energies to 7.6 MeV and at 14.5 MeV. At energies below 1 keV, the (n, γ) cross section was based on ENDF/B MAT 1158 data, with the data temperature-broadened to 300°K and thinned using a 20% thinning criterion. Since the magnitude of the (n, γ) cross section above 7.6 MeV is less than 10 mb, any reasonable high-energy extrapolation is adequate for the evaluation. The estimated uncertainty in the (n, γ) cross section is 5% or less at thermal energies, about 25% in the epithermal and keV range, and perhaps 15% in the MeV range up to 14 MeV.

$(n, \text{fission})$ Reaction

Measurements of the $(n, \text{fission})$ cross section⁶³⁻⁸⁴ span the energy region from threshold to 20 MeV. These measure-

ments have an accuracy, relative to the ^{235}U fission cross section, of about 5%. Since the ^{235}U fission cross section has estimated uncertainties of about 5% at low and moderate energies and 10% at 14 MeV, the overall uncertainties in the ^{238}U fission cross sections are slightly larger than those for ^{235}U .

(n, n') Reaction

Since ^{238}U is an even-even nucleus, it has a relatively small number of low-lying levels. For neutron energies up to 4 MeV the evaluation includes (n, n') excitations to several discrete lines plus an (n, n') continuum contribution, with the results based on experimental data.^{14, 24, 26, 34, 85-90} Above 4 MeV only the continuum component is considered, and the evaluated cross sections are based on nuclear systematics. The (n, n') evaluated cross sections have accuracies of about 25% below 4 MeV and 40% above 4 MeV.

(n, 2n) Reaction

Uranium-238 is the only high-Z isotope for which the (n, 2n) excitation function has been accurately measured.^{86, 91-96} The evaluated (n, 2n) cross section was

based on experimental data from 6.4 MeV to 16 MeV, and nuclear systematics were used to extend the evaluation to 20 MeV. The experimental (n, 2n) data have quoted errors of 5% to 10%. The accuracy of the (n, 2n) evaluation is 10% over most of the energy range, with somewhat larger uncertainties near threshold and above 16 MeV.

(n, 3n) Reaction

The (n, 3n) cross section has been measured in the energy range from 12.6 MeV to 18.8 MeV.^{86, 87, 89, 92, 93} These measurements were combined with nuclear systematics to span the range of evaluated energies. The quoted experimental errors range from about 25% to 50%, and the evaluation has an accuracy of about 50%.

(n, 4n) Reaction

Two (n, 4n) data points exist,⁸⁹ at 17.8 MeV and 18.8 MeV. These were combined with nuclear systematics to complete the evaluation. The data points have quoted errors of 40% and 100%, and the evaluation for the (n, 4n) cross section has an accuracy that is comparable with these experimental accuracies.

III. Neutron Angular Distributions

A. Elastic Scattering Angular Distributions

Experimental measurements of elastic scattering angular distributions have been made from 0.075 MeV to 4 MeV and at 7 and 14.1 MeV.^{14, 18, 34, 89} These were combined with optical model calculations to cover the range of energies needed for the evaluation. The total

elastic cross section has an accuracy of about 15%, and the individual differential cross section data points have relative uncertainties of about 15%.

B. Nonelastic Scattering Angular Distribution

A number of measurements have been made on (n, n') angular distributions.^{14, 97-100}

However, except for the lowest (n, n') level (0.045 MeV), these indicate that the (n, n') scattering is essentially isotropic. Hence the lowest (n, n') level was entered with anisotropic angular distributions, but all other inelastic processes were assumed to be isotropic. This

assumption of isotropy leads to no more than 10% to 15% errors in the worst cases for $(n, 2n)$, $(n, 3n)$, and $(n, 4n)$ reactions, but high energy (n, n') reactions may have larger errors than this since they tend to exhibit a forward peaking.

IV. Neutron Energy Distributions

The formalism that was used to specify the nonelastic neutron spectra is described individually for each reaction. Although much of the following discussion is couched in terms of the evaporation model, the energy distributions are presented exclusively as tables. Since not all distributions can be represented in the evaporation formalism but all can be represented by tables, a tabular representation preserves consistency. A minor but pertinent point is the choosing of the low energy cut-off if nuclear temperatures are presented. The tabular representation makes the intended use of the data explicit.

A. $(n, \text{fission})$ Energy Distributions and $\bar{\nu}(E)$

Measurements of the neutron energy spectrum from fission have been made at only two energies.¹⁰¹ However, fission spectra can be obtained at all energies by using the formalism given in papers by Howerton¹⁰² and by Howerton and Doyas.¹⁰³ The first of these papers¹⁰² gives a method of predicting the value of $\bar{\nu}(E)$, where $\bar{\nu}(E)$ is the average number of neutrons emitted per fission at neutron energy E , and the second paper¹⁰³ gives the fission temperature as a function of $\bar{\nu}$.

Thus a combination of these two calculations gives the fission energy spectrum for all incident neutron energies. The slope of the energy dependence of $\bar{\nu}$ changes as the thresholds corresponding to (n, n') fission, $(n, 2n)$ fission, etc., are reached. Hence in the evaluation, the fission neutron energy spectra are quoted at energies that correspond to thresholds and to midpoints between thresholds.

Many measurements have been made of the value of $\bar{\nu}(E)$ that corresponds to prompt neutron emission.^{77,92,104-124}

These measurements are implicitly incorporated into the formalism of Howerton.¹⁰² In particular, values for $\bar{\nu}(E)$ that were predicted on the basis of the Howerton formalism are in good agreement with experimental values¹¹¹ that were measured subsequent to the predictions. The experimental data extend up to 14.8 MeV, and systematics was used to extend these results to 20 MeV.

The neutron energy spectrum from fission is known to an accuracy of about 5% over the secondary neutron energy range from 0.5 MeV to 5 MeV. At lower and higher energies, it is known to about 10%. The accuracy of $\bar{\nu}$ is estimated to about 2%.

B. (n, n') Energy Distributions

For neutron energies below 4 MeV, (n, n') energy distributions were obtained from experimental measurements.^{14, 34, 97-99} For neutron energies above 4 MeV, the (n, n') energy distributions were taken from a model that includes both a low energy temperature component and a high energy tail. The high-energy tail, which includes about 15% of the inelastically-scattered neutrons at 4 MeV and essentially 100% of the neutrons at energies of 15 MeV and above, consists of direct-interaction and pre-equilibrium neutrons, chosen in appropriate fractions. At high energies, if an (n, n') reaction does not result in a final state neutron n' that carries away most of the energy, then the residual nucleus will be left in a highly-excited state, and it will undergo fission or emit another neutron which takes it out of the (n, n') category. Hence the high energy component of the (n, n') reaction yields high energy neutrons almost exclusively. The energy spectrum has an accuracy of about 50% at lower energies and 100% at higher energies.

C. $(n, 2n)$ Energy Distributions

No measurements exist for $(n, 2n)$ energy spectra in ^{238}U . Hence a temperature model was used to provide these spectra. The spectrum was assumed to be the same for both the first and second neutrons, and it was chosen so that energy is conserved on the average, taking into account the energy that appears in the form of gamma rays. Even with the constraint imposed by the conservation of energy, errors can be large for certain portions of the energy spectrum and at certain incident neutron energies, ranging from perhaps 30% in favorable cases to orders of magnitude in unfavorable cases.

D. $(n, 3n)$ and $(n, 4n)$ Energy Distributions

The formalism used for $(n, 2n)$ energy spectra was carried over directly to $(n, 3n)$ and $(n, 4n)$ energy spectra in the absence of data. Energy conservation again served as a constraint, but the errors can be even larger than for $(n, 2n)$ reactions, since more complex decay modes are involved.

V. Gamma Ray Production Cross Sections

At neutron energies of 1.09 MeV and above, comprehensive $(n, X\gamma)$ measurements exist¹²⁵ which encompass all of the gamma-ray-producing reactions. For neutron energies below 1.09 MeV, gamma rays are produced by capture, fission, and (n, n') processes. In the evaluation, the capture, fission, and (n, n') gamma-producing reactions were explicitly included over the energy ranges from their various thresholds up to 1.09 MeV.

From 1.09 MeV to 20 MeV, the experimental $(n, X\gamma)$ cross sections¹²⁵ were used, with some small modifications as noted below. An $(n, X\gamma)$ measurement at 0.3 MeV also exists,¹²⁵ but the shape of the energy spectrum does not agree well with other nearby measurements¹²⁶ (at 0.3 MeV, the (n, γ) reaction is the only available gamma-producing process for photons with energy greater than 0.3 MeV). Also, and more importantly, the 0.3 MeV

gamma ray spectrum¹²⁵ strongly violates energy conservation. For these reasons the 0.3 MeV data¹²⁵ were omitted from the evaluation.

A. Gamma Ray Production from Neutron Capture

The capture gamma ray spectrum for ^{238}U has been accurately measured only at thermal neutron energies.¹²⁷ Some measurements have also been made in the eV and low keV region.¹²⁶ However, these measurements are made only at resonance energies, which may distort the results, and they have several obvious difficulties: (1) they disagree in spectral shape and in normalization with the very accurate thermal measurement¹²⁷; (2) they extend only down to 1 MeV photon energy, and their integrated spectral intensities suggest that the photon spectrum is very small below 1 MeV—a result that is known to be incorrect; (3) even with a lower energy photon cutoff at 1 MeV, some of the spectra violate energy conservation by as much as 30%. With a reasonable low-energy photon component included, the normalization errors would be as large as 50%. Errors of this magnitude indicate serious deficiencies in the experiment, and these deficiencies could affect both the shape and the normalization of the individual spectra. Thus the data of John and Orphan¹²⁶ were disregarded in the evaluation.

B. Gamma Ray Production and Fission Reactions

The ^{238}U fission spectrum has a threshold at about 0.45 MeV. Since no measurements of the gamma rays from ^{238}U fission exist, the ^{235}U fission gamma ray spectrum¹²⁸ was used for

the evaluation. In the absence of data, the ^{235}U fission gamma ray multiplicity was also used for ^{238}U , and this multiplicity was held constant for all neutron bombarding energies (since the neutron kinetic energy is negligible as compared to the fission energy releases, the constancy of the multiplicity with increasing neutron energy seems reasonable).

C. Gamma Ray Production from (n, n') Reactions

The evaluated neutron data for ^{238}U include several experimentally-measured (n, n') excitation levels, as was discussed in Section IID. The two lowest levels, at 50 and 150 keV, were carried in the evaluation as discrete lines from threshold up to 1.09 MeV. The 150 keV decay was assigned a suitable branching ratio.¹²⁹ Gamma rays from the higher levels were placed in energy bins in a manner that conserves energy. The gamma rays from inelastic scattering are not as energetic as those from capture and fission. In the experimental (n, X γ) data at 1.09 MeV,¹²⁵ the effect of these low-energy (n, n') gammas can be observed as causing the sharp rise in the (n, X γ) cross section below 1 MeV. In fact, the proper shape for the low energy portion of the 1.09 MeV (n, X γ) spectrum¹²⁵ was determined by summing the (n, γ), (n, fission), and (n, n') contributions to the gamma ray spectrum at 1.09 MeV.

D. Gamma Ray Production from (n, X γ) Reactions

Measurements of the (n, X γ) reaction have been carried out at energies extending from 1.09 MeV to 14.8 MeV.¹²⁵ Since the measurements did not extend

down to zero gamma ray energy, it was necessary to make an extrapolation in order to obtain $(n, X\gamma)$ cross sections that reflect the entire gamma ray spectrum. This extrapolation to zero gamma ray energy was made on the basis of overall energy conservation (see Section V:C),

and also by comparing the $^{238}\text{U}(n, X\gamma)$ spectra¹²⁵ to the $^{235}\text{U}(n, X\gamma)$ spectra of Drake,¹³⁰ which extend down to 0.25 MeV photon energy.

The accuracy of the gamma ray production cross section is estimated to be about 30% over the range of energies in the evaluation.

VI. Gamma Ray Production Angular Distributions

There are no data on gamma ray angular distributions in ^{238}U . However, measurements on other heavy elements indicate

that the gamma ray production is essentially isotropic. Hence isotropy was assumed for all gamma-producing reactions.

VII. Gamma Ray Energy Distributions

A. Gamma Energy Distributions from Neutron Capture

The capture gamma ray energy spectrum has been reliably measured only at thermal neutron energies,¹²⁷ as was discussed in Section V:A. If the thermal neutron capture gamma ray spectrum is used at higher neutron energies, then in order to conserve energy we must either increase the multiplicity or else harden the spectrum. When the thermal spectrum is plotted on a linear scale (instead of the usual semi-logarithmic representation), it can be seen that the high energy tip of the spectrum is vanishingly small. Thus it is not feasible to conserve energy by extending the miniscule high energy tip of the spectrum. It is necessary to modify other parts of the spectrum as well. In the absence of data, we chose the simplest way; we changed the multiplicity. The multiplicity and the high energy cutoff could both be modified, of course, but such complexity is unwarranted

without more complete experimental data.

B. Gamma Energy Distributions from Fission Reactions

As discussed in Section V:C, the ^{235}U thermal fission gamma ray spectrum was also used for ^{238}U , with the multiplicity held constant at the ^{235}U value for all incident neutron energies.

C. Gamma Energy Distributions from (n, n') Reactions

Gamma rays from the 50 and 150 keV inelastic levels were treated as line spectra. Gamma rays from the higher (n, n') levels were placed in broad energy bins in a manner that conserved energy for the total neutron scattering reaction.

D. Gamma Energy Distributions from $(n, X\gamma)$ Reactions

Experimental measurements of the $(n, X\gamma)$ cross section included measurements of the gamma ray spectra.¹²⁵ The

experimental spectra extended down to 0.5 MeV photon energy. Extension to zero photon energy was made by comparing to the data of Drake¹³⁰ and by requiring overall energy conservation. At 1.09 MeV, detailed calculations of gamma ray contributions from (n, γ), (n, fission), and (n, n') reactions were summed to give the proper shape for the low energy part of the (n, X γ) spectrum.

A small high-energy tail was also added to each spectrum to allow for the (experimentally unobserved) fission gamma ray contribution to the overall (n, X γ) spectrum.

The overall accuracy of gamma ray spectra is about 30% in the middle portions of the spectra, with possible errors being much larger at very low and very high photon energies.

VIII. Integral Tests of the Evaluation

The only realistic estimate of the quality of evaluated nuclear data comes from calculating parameters of appropriate integral experiments and comparing the results with the corresponding measurements of those parameters.¹³¹ Unfortunately, an unequivocal certification of the validity of the complete evaluation is never possible since there is an intimate coupling of the effects of uncertainties in the quantities being tested. One is left then with the situation that the results of integral data testing can be used to indicate that an evaluation is poor, but they can not be used to certify that an evaluation is good. Furthermore, there exist no integral experiments sufficiently precise to test the evaluation over all incident particle energy regimes.

Among integral experiments, one must also differentiate between those which test an evaluated library and those which can be used to test a specific material. Those integral experiments which are made up of combinations of materials, each in a substantial fraction of the whole, can test only the general quality of an entire evaluated library, since no one

material can dominate. This is especially true for critical assemblies. On the other hand, there are relatively few one-material integral neutron experiments, possible exceptions being poor-geometry-broomstick-target experiments (which

Table 2. K_{eff} calculations for 11 simple spherical critical assemblies in which the reflectors were uranium and the cores were ²³³U, ²³⁵U or ²³⁹Pu.

Reflector thickness (cm)	Core material	k_{eff}^a calc	Δk_{eff} calc
2.30	²³³ U	0.999	± 0.003
5.31	²³³ U	1.004	± 0.003
19.91	²³³ U	0.998	± 0.003
1.76	²³⁵ U	1.306	± 0.003
4.47	²³⁵ U	1.010	± 0.003
9.96	²³⁵ U	1.004	± 0.003
18.10	²³⁵ U	1.002	± 0.003
1.93	²³⁹ Pu	0.995	± 0.003
4.13	²³⁹ Pu	1.004	± 0.003
6.74	²³⁹ Pu	1.002	± 0.003
19.60	²³⁹ Pu	1.002	± 0.003

$$k_{\text{eff}}^a = \frac{\sum_{i=1}^{11} k_{\text{eff}i}}{11} = \frac{11.026}{11} = 1.0021$$

test primarily the minima in the total cross section) and the LLL pulsed spheres.

The above comments are made to put the problem of "evaluating an evaluation" into proper perspective, not to indulge in "hand-wringing." Integral testing of evaluations is a valid activity, but it should be carried out with the understanding that the results are not definitive.

To test the present evaluation of ^{238}U neutron reaction data, k_{eff} values were calculated for 11 spherical assemblies in which reflectors of ^{238}U (natural uranium) reflected cores of ^{235}U , ^{233}U and ^{239}Pu . The results are presented in Table 2.

The uncertainty in k_{eff} resulting from uncertainties in the calculational models of the critical assemblies is usually about 0.7%. An analogue Monte Carlo neutronics code (TART) performed the k_{eff} calculations. Reaction cross sections are represented by 175 histograms on a standard preselected energy grid. Angular distributions of secondary neutrons are represented, at the incident energies supplied by the evaluator, by 32 intervals of equal probability in the cosine of the emission angle. Linear interpolation in energy at these cosine values is carried out by the neutronics code to obtain angular distributions at arbitrary energies. Energy distributions for secondary nonelastic neutrons are represented by 32 equally-probable increments in secondary energy, except for the fission process where 256 equally probable increments are used. Interpolations are as with angular distributions. The number of groups used for the cross sections, and the number of intervals used for angular and energy distributions, are adequate in the sense that increasing the

number of increments does not affect the k_{eff} calculations. The Monte Carlo statistics for the calculation are the values listed in the column of Table 2 headed Δk_{eff} . These represent a 67% confidence limit; when they are combined with the mass and geometry uncertainties in the critical assembly specifications, the result is that any calculated k_{eff} value between 0.99 and 1.01 represents the "just critical" configuration as well as would a value of 1.0000.

There is no reason to assume that the specifications of the critical assembly masses or geometrical configurations contain a built-in bias. Since neither the evaluations for the core materials nor those for ^{235}U have been adjusted to force agreement with critical assembly calculations, deficiencies in assembly specifications or in reflector material evaluations can be averaged out by considering the 11 k_{eff} calculations of Table 2 as a whole (the evaluations used for the core materials are those in the LLL evaluated library). This leaves the ^{238}U evaluated data as the only common factor among the various calculations. The $\overline{k_{\text{eff}}}$ value of 1.0024 presented at the end of Table 2 then tends to provide a measure of the validity of the ^{238}U evaluation for fast and moderately fast neutronics systems.

In addition to the calculated k_{eff} values, time spectra of neutrons from a 14 MeV pulsed sphere¹³² were also calculated for detectors placed at 30° and 120° with respect to the incident deuteron beam. The results are displayed in Figs. 1 and 2. The agreement with experiment is generally within calculational statistics except at the first minimum and for late arrival times, where disagreements

Fig. 1. A comparison of the experimental neutron spectrum from a 14-MeV pulsed sphere of ^{238}U vs that calculated using evaluated nuclear data (MAT 4187). The detector was at 30° with respect to the incident deuteron beam.

between calculation and experiment are as much as a factor of two for the latest arrival times.

The apparent disagreement in the first minimum is not real. This calculation includes no correction for collimator inscattering. In other calculations the contribution of this correction amounts to the apparent discrepancy.

The question remains whether the disagreements between calculation and experiment are acceptable. To provide a basis for judging the acceptability of this evaluation, we present corresponding pulsed sphere calculations using ENDF/B-III (MAT 1158) cross sections in Figs. 3 and 4. Comparison with Figs. 1 and 2 clearly shows the improved agreement.

Fig. 2. A comparison of the experimental neutron spectrum from a 14-MeV pulsed sphere of ^{238}U vs that calculated using evaluated nuclear data (MAT 4187). The detector was at 120° with respect to the incident deuteron beam.

Fig. 3. A comparison of the experimental neutron time spectrum from a 14-MeV pulsed sphere of ^{238}U vs that calculated using ENDF/B-III evaluated nuclear data (MAT 1158). The detector was at 30° with respect to the incident deuteron beam.

Fig. 4. A comparison of the experimental neutron time spectrum from a 14-MeV pulsed sphere of ^{238}U vs that calculated using ENDF/B-III evaluated nuclear data (MAT 1150). The detector was at 120° with respect to the incident deuteron beam.

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