LBL-14404
Preprint

Lawrence Berkeley Laboratory
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Submitted to Zeitschrift für Physik A

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August 1982

NUCLEAR SCIENCE DIVISION

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

LBL-14404 c.2

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This work was supported by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

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Abstract

The transition state theory of fission is generalized to allow for trajectories that return from saddle to compound nucleus due to high viscosity at the saddle point. This generalization includes neutron emission from the saddle. Experimental indications seem to support the present approach.

Introduction

The standard Bohr Wheeler (BW) theory of fission decay [1], identical with the transition state theory for chemical reactions, is subject to serious limitations of both quantal and classical nature. We want to consider here the most crucial approximation of the theory, its possible failure, and a generalization designed to overcome part of the difficulty. The BW theory calculates the flux of the density distribution in phase space across a suitably chosen hypersurface normal to the reaction coordinate. This flux is then identified with the reaction rate. This is both the beauty and the trap of the theory. The flux and the reaction rate can be identified if and only if no phase-space trajectory, after crossing the hypersurface, comes back and crosses it again returning to the reactant's region. In order to eliminate, or at least to alleviate, the problem, the "transition state", or the position

of the hypersurface, is chosen to cut across the saddle point in coordinate space, on the hope that, once the saddle point is negotiated, the system irreversibly rolls down towards the product region. This is certainly an extreme approximation, requiring a substantial decoupling (low viscosity) between collective and internal degrees of freedom near the transition state.

A more general approach to the problem of chemical reaction rates was developed in 1940 by Kramers[2]. A particle moving in a viscous medium in thermal equilibrium is subject to an effective force rapidly fluctuating in time in a highly irregular way (brownian motion). If initially the particle is captured in a potential hole, the diffusive force acting on the collective degree of freedom can shuttle the particle over the potential barrier (Q). The reaction rate is the result of the competition between diffusive force and driving force along the path from the initial to the transition state. The essential difficulties arising from the mathematical complexity of the solution of the diffusion equation for a nonstationary process can be overcome if one considers a substantially high barrier. Under this condition a distribution of Boltzmann type is soon established near the initial state, and the resulting quasistationary diffusion can then be dealt with the one-dimensional case where the potentials in the initial configuration and in the transition state are approximated by harmonic oscillator type [2]. In this one-dimensional model the crucial parameters that control the coupling are the viscosity of the medium (η) and the frequency of the harmonic potentials (ω and ω'). The diffusion over the barrier is characterized by three different regimes according to whether the characteristic frequency for viscosity (η) is coupled or not to the characteristic frequency for the internal degrees of freedom in the initial state (ω) and in the transition state (ω').

i) Intermediate viscosity ($\eta \gg \omega$ and $\eta \ll \omega'$). Under these conditions the strong coupling in the initial configuration leads to a Maxwell-Boltzman

distribution for the generalized momentum of the fission coordinate. The probability current across the barrier is directed by the tail of this distribution with no further resistance felt by the system in the transition state. In this limit the transition method holds and gives for the reaction velocity the equation $r = \omega \exp(-Q/T)$.

ii) Low viscosity ($\eta \ll \omega$ and $\eta \ll \omega'$). Due to the small coupling in the intial state the delivery of particle to the transition state is small and the reaction velocity drops rapidly below the transition method value [3] [$r = \eta(Q/T) \exp(-Q/t)$].

iii) Large viscosity ($\eta \gg \omega$ and $\eta \gg \omega'$). The reaction rate can be no longer identified with the flow in the direction (initial configuration) $\rightarrow$ (transition state) $\rightarrow$ (reaction product's region). The net flow through the transition state, as a result of the strong coupling in this region ($\eta \gg \omega$), becomes now smaller than the transition method value [$r = \omega(2\pi\omega'/\eta) \exp(-Q/T)$].

The extension of such studies to high excitation energies, where the regime of large viscosity seems to be more likely, offers the stimulating possibility of clarifying the role of viscosity and its dependence on the temperature in the dynamics of the nucleus from the compound state to the saddle point. While the general philosophy of our approach treads in diffusion model's footsteps, the formal apparatus, as will appear clearly in the next section, is somewhat different. We assume high viscosity in the general saddle point neighborhood. As a result, the flux from the compound nucleus is trapped in the saddle region, and the associated randomization leads to a backflow towards the compound nucleus. Furthermore, it is interesting to consider the possibility that, while the system is trapped in the neighborhood of the saddle point, it may undergo particle decay, in particular neutron emission. For this case a natural way to handle the problem is the use of the Master Equation.

The model.

Let us consider a compound nucleus A, a saddle point region B, a region C far down the scission valley, and a nucleus D after one neutron emission. The transition probabilities are λ_1 (from A to B), λ_2 (from B to A), λ_3 (from B to C), λ_n (from A to D), $\lambda_{n'}$ (from B to D).

The master equations are:

$$\dot{\varphi}_A = \varphi_B\lambda_2 - \varphi_A(\lambda_1 + \lambda_n)$$

$$\dot{\varphi}_B = \varphi_A\lambda_1 - \varphi_B(\lambda_2 + \lambda_3 + \lambda_{n'})$$

$$\dot{\varphi}_C = \varphi_B\lambda_3$$

$$\dot{\varphi}_D = \varphi_A\lambda_n + \varphi_B\lambda_{n'}$$

where the φs are the time-dependent populations. Two main differences with respect to the standard BW theory are visible: a) there is a backflow from B to A that makes the decay of A nonexponental (notice that by setting $\lambda_2 = 0$ we recover the BW expression); b) neutrons are allowed to be emitted from the saddle region.

The system of differential equations can be solved in a straightforward way, and the exact solutions are

$$\varphi_A = -\frac{\varphi_o}{\Delta}\left[(r_2 + \lambda_1 + \lambda_n)\, e^{r_1 t} - (r_1 + \lambda_1 + \lambda_n)\, e^{r_2 t}\right]$$

$$\varphi_B = -\frac{\varphi_o}{\Delta\lambda_2}(r_1 + \lambda_1 + \lambda_n)(r_2 + \lambda_1 + \lambda_n)\left[e^{r_1 t} - e^{r_2 t}\right]$$

$$\varphi_C = \varphi_o\frac{\lambda_1\lambda_3}{\lambda_\Delta\Delta}\left[r_2 e^{r_1 t} - r_1 e^{r_2 t} + \Delta\right]$$

$$\varphi_D = \frac{\varphi_o}{\lambda_\Delta\Delta}\left\{r_2(e^{r_1 t} - 1)[\lambda_1\lambda_{n'} - \lambda_n(r_2+\lambda_1+\lambda_n)] - r_1(e^{r_2 t} - 1)[\lambda_1\lambda_{n'} - \lambda_n(r_1+\lambda_1+\lambda_n)]\right\}$$

where

$$\Delta = [(\lambda_2 + \lambda_3 + \lambda_{n'} - \lambda_n - \lambda_1)^2 + 4\lambda_1\lambda_2]^{1/2}$$

$$r_{1/2} = (1/2)[-\lambda_{TOT} \pm \Delta]$$

$$\lambda_{TOT} = \lambda_1 + \lambda_2 + \lambda_3 + \lambda_n + \lambda_{n'}$$

$$\lambda_\Delta = \lambda_1(\lambda_3 + \lambda_{n'}) + \lambda_n(\lambda_2 + \lambda_3 + \lambda_{n'})$$

The initial conditions have been chosen so that

$$\varphi_A(t = 0) = \varphi_o \quad , \quad \varphi_B(t = 0) = \varphi_C(t = 0) = \varphi_D(t = 0) = 0 \quad .$$

From the populations at time infinity one can obtain the following expression for Γ_N/Γ_F:

$$\Gamma_N/\Gamma_F = \frac{\varphi_D(\infty)}{\varphi_C(\infty)} = \frac{\lambda_n}{\lambda_1} + \frac{\lambda_1\lambda_{n'} + \lambda_2\lambda_n + \lambda_n\lambda_{n'}}{\lambda_1\lambda_3} \quad . \tag{1}$$

The first term to the right is the standard result. The above expression can be obtained without solving the differential equations by summing over the probability tree.

$$P_f = \frac{\lambda_1}{(\lambda_1+\lambda_n)}\left\{\frac{\lambda_3}{(\lambda_2+\lambda_3+\lambda_{n'})} + \frac{\lambda_2}{(\lambda_2+\lambda_3+\lambda_{n'})}\frac{\lambda_1}{(\lambda_1+\lambda_n)}\left\{\frac{\lambda_3}{\lambda_2+\lambda_3+\lambda_{n'}} + \ldots\right\}\right\}$$

$$= (\lambda_1\lambda_3)/(\lambda_1\lambda_3 + \lambda_1\lambda_{n'} + \lambda_2\lambda_n + \lambda_3\lambda_n + \lambda_n\lambda_{n'})$$

from which equation (1) is immediately obtained. The new expression (1) favors neutron decay in two ways: a) by allowing neutron decay from the saddle; b) more importantly, by redirecting part of the flux from the saddle region back to the compound nucleus.

An intermediate and more general situation can be envisaged as follows. For a given viscosity at the saddle, there will be a critical velocity along the fission coordinate, above which the system escapes altogether towards fission and

below which the system gets trapped in the saddle region. The treatment can be modified by splitting λ_1 as follows:

$$\lambda_1 = \frac{1}{2\pi\rho(E)}\left[\int_0^{\varepsilon_0} \rho(E - B_F - \varepsilon)\, d\varepsilon + \int_{\varepsilon_0}^{\infty} \rho(E - B_F - \varepsilon)\, d\varepsilon\right]$$

$$= \lambda_{1S} + \lambda_{1F} \qquad (2)$$

where ρ is the level density, B_F the fission barrier, and ε the kinetic energy along the fission coordinate. The first term to the right corresponds to saddle trapping and the second to complete saddle negotiation. The meaning of the critical velocity introduced in (2) becomes clear if we define $\tau_\eta \approx \eta^{-1}$ the characteristic time necessary to the onset of the equilibration between the fission degree of freedom x and all the other degrees of freedom of the system in the saddle region. The critical value ε_0 is then given from the relation $\varepsilon_0 \propto (1/\tau_\eta)^2 \approx \eta^2$.

For all the phase space trajectories with $\varepsilon > \varepsilon_0$, the system is insensitive to the friction and behaves like a BW system. On the other hand for the trajectories along which the system enters into the saddle region with $\varepsilon < \varepsilon_0$, the equilibration takes place and the associated randomization of motion is responsible for a backflow towards the compound nucleus state.

The situation reminds one of the scaling limit theory applied to the saddle point [4], where a characteristic time is defined after which the driving force decouples the system from the heat bath and the system rolls down towards the scission configuration. Using the expression (2) the general result is now

$$\Gamma_F/\Gamma_N = \frac{\lambda_{1F}(\lambda_2+\lambda_3+\lambda_{n'}) + \lambda_{1S}\lambda_3}{\lambda_n(\lambda_2+\lambda_3+\lambda_{n'}) + \lambda_{1S}\lambda_{n'}} \qquad (3)$$

Again it is reasonable, although not necessary, that, for the systems trapped in the saddle region $\lambda_2 = \lambda_3$. If one disregards the contribution of the neutron decay from the saddle region, one obtains the simple form:

$$\Gamma_F/\Gamma_N = \frac{\lambda_{1F}}{\lambda_n} + \frac{1}{2}\frac{\lambda_{1S}}{\lambda_n} \tag{4}$$

In the equidistant model approximation the level density is given by the expression

$$\rho(E) \propto \exp\left[2(aE)^{1/2}\right]$$

where the preexponential energy factors have been omitted.

The transition probabilities λ_{1F}, λ_{1S}, and λ_n then can be calculated in a straightforward way:

$$\begin{aligned}
\lambda_{1S} &= \frac{1}{2\pi\rho(E)}\frac{1}{2a_f}\Big\{\left[2a_f^{1/2}(E-B_F)^{1/2}-1\right]\exp\left[2a_f^{1/2}(E-B_F)^{1/2}\right] \\
&\quad - \left[2a_f^{1/2}(E-B_F-\varepsilon_o)^{1/2}-1\right]\exp\left[2a_f^{1/2}(E-B_F-\varepsilon_o)^{1/2}\right]\Big\} \\
\lambda_{1F} &= \frac{1}{2\pi\rho(E)}\frac{1}{2a_f}\Big\{\left[2a_f^{1/2}(E-B_F-\varepsilon_o)^{1/2}-1\right]\exp\left[2a_f^{1/2}(E-B_F-\varepsilon_o)^{1/2}\right]\Big\} \\
\lambda_n &= \frac{1}{2\pi\rho(E)}\frac{1}{2a_n}\Big\{\left[2a_n^{1/2}(E-B_n)^{1/2}-1\right]\exp\left[2a_n^{1/2}(E-B_n)^{1/2}\right]\Big\}
\end{aligned} \tag{5}$$

where a_f and a_n are the level density parameters appropriate to the saddle point and to equilibrium deformation, respectively. Substitution of (5) into (4) yields

$$\Gamma_F/\Gamma_N = \frac{a_n}{2a_f\left[2a_n^{1/2}(E-B_n)^{1/2}-1\right]}\left\{\begin{aligned}&\left[2a_f^{1/2}(E-B_F-\varepsilon_o)^{1/2}-1\right]\exp\left[2a_f^{1/2}(E-B_F-\varepsilon_o)^{1/2}\right. \\ &\left. - 2a_n^{1/2}(E-B_n)^{1/2}\right] + \left[(2a_f^{1/2}(E-B_F)^{1/2}-1\right] \\ &\exp\left[2a_f^{1/2}(E-B_F)^{1/2} - 2a_n^{1/2}(E-B_n)^{1/2}\right]\end{aligned}\right\} \tag{6}$$

It is interesting to note that the above expression, in the limiting case of no viscosity ($\varepsilon_0 \to 0$), reduces to the standard case, while for large viscosity ($\varepsilon_0 \to E - B_F$), it approaches the limit $(1/2)(\Gamma_F/\Gamma_N)_{\varepsilon_0} = 0$. The latter result is a straightforward consequence of the assumption $\lambda_2 = \lambda_3$ for the systems with $\varepsilon < \varepsilon_0$.

Calculations

The viscosity parameter η is a function both of the intrinsic degrees of freedom of the nucleus, and so of its temperature T, and of the collective mode under consideration. The complete solution of this problem goes beyond the scope of this paper. However, if the effect of the collective motion (i.e. the dependence of the viscosity from the shape of the saddle point) is set aside, the temperature dependence of η can be inferred from qualitative microscopic considerations. The number of quasiparticle collisions per unit time $(1/\tau_\eta)$ allowed by conservation of energy and momentum alone is reduced, because of the blocking effect of the Pauli's principle, by a factor $(T/\varepsilon_F)^2$, with ε_F the Fermi energy. Thus we obtain for ε_0 the following dependence upon the temperature:

$$\varepsilon_0 \propto (1/\tau_\eta)^2 \approx \left[\frac{1}{\tau_0\left(\frac{\varepsilon_F}{T}\right)^2}\right]^2 \propto T^4 \qquad (7)$$

For the compound nucleus ^{180}W, fig. 1 shows the excitation function of Γ_F/Γ_N in the two limits of high viscosity at the saddle point (dashed line) and zero viscosity (full line). Both curves are calculated using the expression (6), where, for simplicity, no effect due to angular momentum has been taken into account. The values of B_F and B_n are from ref. [5]. For the ratio a_f/a_n and for a_n the values 1.11 and A/10 are chosen, and for

ε_0 we use the simple form $\varepsilon_0 = cT^4$, where $T = (E^*/a_f)^{1/2}$ and c is a constant adjusted to fit at low energy the zero viscosity ($\varepsilon_0 = 0$) limit. Note that, for increasing T, Γ_F/Γ_N calculated with $\varepsilon_0 \neq 0$ decrease compared to Γ_F/Γ_N with $\varepsilon_0 = 0$. The general trend agrees with the experimental data [6,7], which show at high excitation energy a decrease of the fission probability compared with the prediction of the standard model. Actually, in this calculation we have overestimated the true result because we have neglected the neutron evaporation from the saddle.

Figure 2 displays the quantity $\delta = [(\Gamma_F/\Gamma_N) - \Gamma_F/\Gamma_N)_{BW}]/(\Gamma_F/\Gamma_N)_{BW}$ as a function of the temperature and for different values of c. It is interesting to note the sensitivity of the deviation of our model from the standard BW theory to the variations of c in the low-energy region. The importance of this behavior is obvious; in the low-energy region (i.e. the low viscosity region for our model) Γ_F/Γ_N must converge to the BW limit $(\Gamma_F/\Gamma_N)_{BW}$, and this condition is assured in our model by the phenomenological constant c. The unique determination of c from fit of experimental data in this energy region and the extension of such comparison to higher energies allows for a check of the model.

It is a well-known fact that at high energies the experimental determination of Γ_F/Γ_N is uncertain due to the occurrence of two effects: a) the presence of higher order fission, i.e. the possibility that the nucleus undergoes fission after one or more neutrons have been emitted; b) the increasing contribution to the fission cross section of incomplete fusion reactions, which cause uncertainty in the derivation of the compound nucleus cross section. In the regime of high temperature this is definitely a severe handicap for the comparison of any model with experimental data. The best way to partially overcome it is to perform a direct measure of the prefission

neutron emission $\langle\nu_n\rangle$. Disregarding in first approximation the charged particle evaporation, $\langle\nu_n\rangle$ is given by

$$\langle\nu_n\rangle = \sum_s s\, P_{F,s+1} \prod_{K=1}^{s} P_{n,K}$$

where

$$P_{n,K} = 1/[1 + (\Gamma_F/\Gamma_N)_K] \qquad P_{F,s+1} = 1/[1 + (\Gamma_N/\Gamma_F)_{s+1}]$$

$(\Gamma_F/\Gamma_N)_K$ is the branching ratio for the nucleus of mass $A_K = A_{K-1} - 1$ and excitation energy $E_K^* = E_{K-1}^* - B_{n,K-1} - 2T_{K-1}$. $B_{n,K-1}$ and $2T_{K-1}$ are the binding energy and the kinetic energy of the neutron evaporated from the nucleus A_{K-1}. $(\Gamma_F/\Gamma_N)_K$ is calculated from (6) where for each step B_n is taken from ref. [5]; $a_n = A/10$; $a_f/a_n = 1.02$. For B_F we use the rotating liquid drop barrier times 0.8 as suggested from Gavron [8]. A constant value of $\ell_{crit} = 70$ [8] is given in input for each excitation energy: the angular momentum removed from the neutrons emitted is calculated from ref. [9]. The effect of the backflow from saddle to compound, responsible for the increasing number of neutrons emitted through the inhibition of the fission channel, can be clearly seen in fig. 3 where the quantity $\langle\nu_n\rangle$ is plotted versus excitation energy E^* for the nuclei ^{170}Yb, ^{180}W, ^{186}Os.

This general trend seems consistent with the result of recent accurate measurements of prefission neutron emission [10], which for systems with high fission barrier show substantially larger values of $\langle\nu_n\rangle$ than predicted by the standard model.

We have not performed a comparison with the experimental data because of the strong approximation introduced in the expression (6). This approximation becomes more severe at high temperature where the increasing

role of viscosity implies a longer transition time through the saddle region. This would enhance the possibility of neutrons being emitted directly from the transition region.

This work was supported by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

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Figure Captions

Fig. 1. Branching ratio Γ_F/Γ_N for the compound nucleus ^{180}W versus excitation energy E* for $\varepsilon_0 = 0$ (full line) and $\varepsilon_0 = 0.08\ T^4$ (dashed line).

Fig. 2. The quantity $\delta = [(\Gamma_F/\Gamma_N) - (\Gamma_F/\Gamma_N)_{BW}]/(\Gamma_F/\Gamma_N)_{BW}$ versus temperature for different value of the constant c in the expression $\varepsilon_0 = cT^4$ and for the compound nucleus ^{180}W.

Fig. 3. Average prefission neutron emission $\langle\nu_n\rangle$ versus excitation energy for three compound nuclei (^{170}Yb, ^{180}W, ^{186}Os). The full lines are the result of the calculation with $\varepsilon_0 = 0$, the dashed lines with $\varepsilon_0 = 0.08\ T^4$.

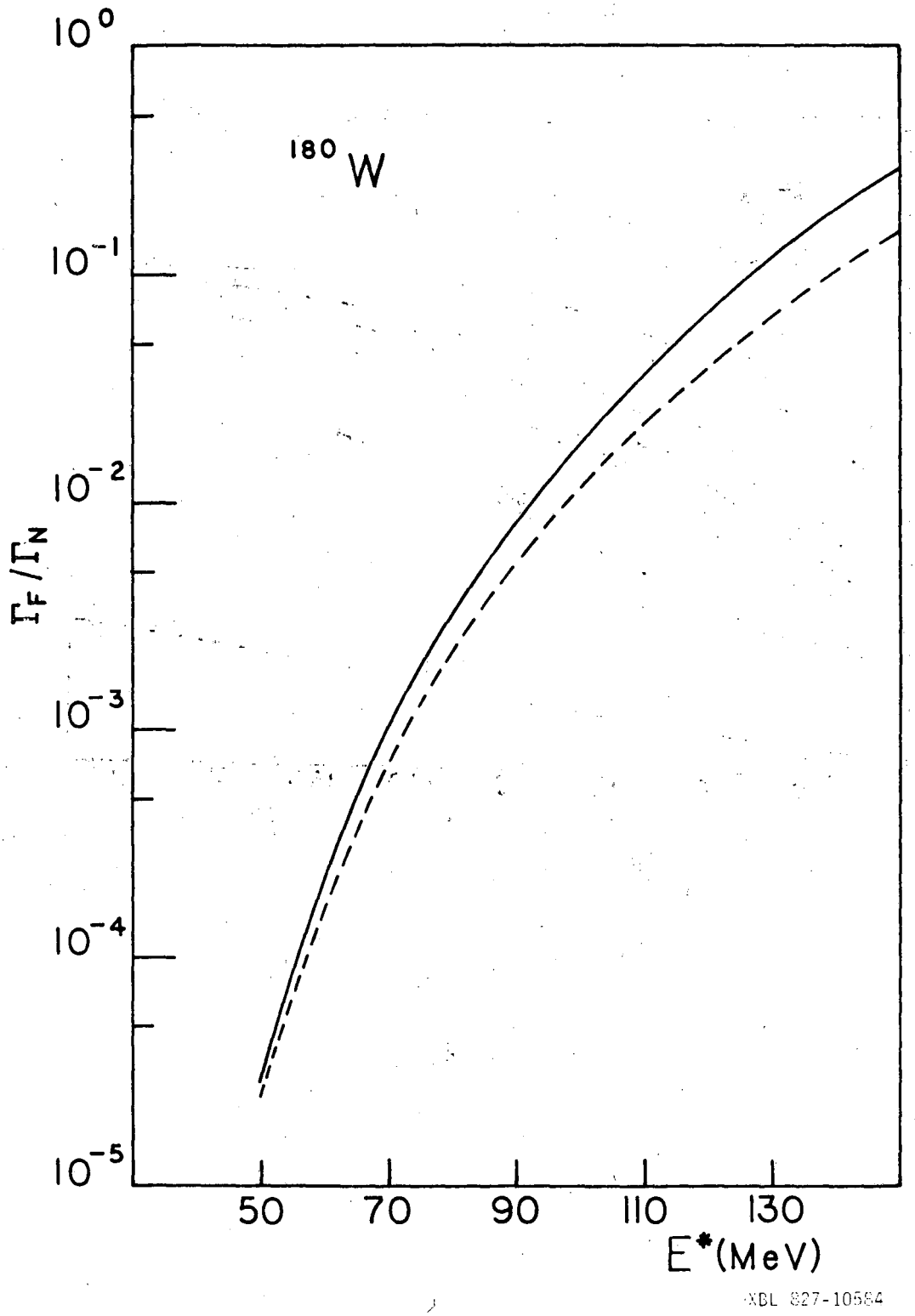
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
Γ_F/Γ_N
^{180}W
50
70
90
110
130
E^*(MeV)
XBL 827-10584

Fig. 1

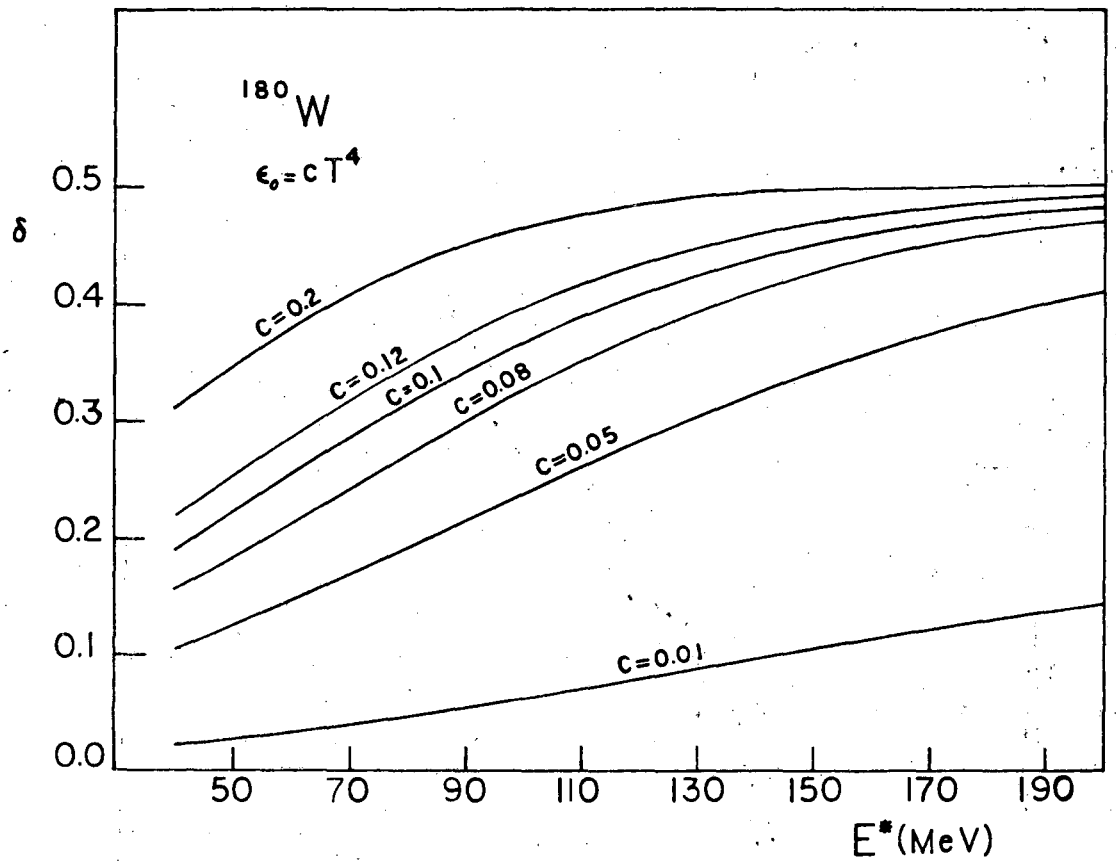
^{180}W
$\epsilon_0 = cT^4$
δ
0.5
0.4
0.3
0.2
0.1
0.0
C=0.2
C=0.12
C=0.1
C=0.08
C=0.05
C=0.01
50
70
90
110
130
150
170
190
E*(MeV)

Fig. 2

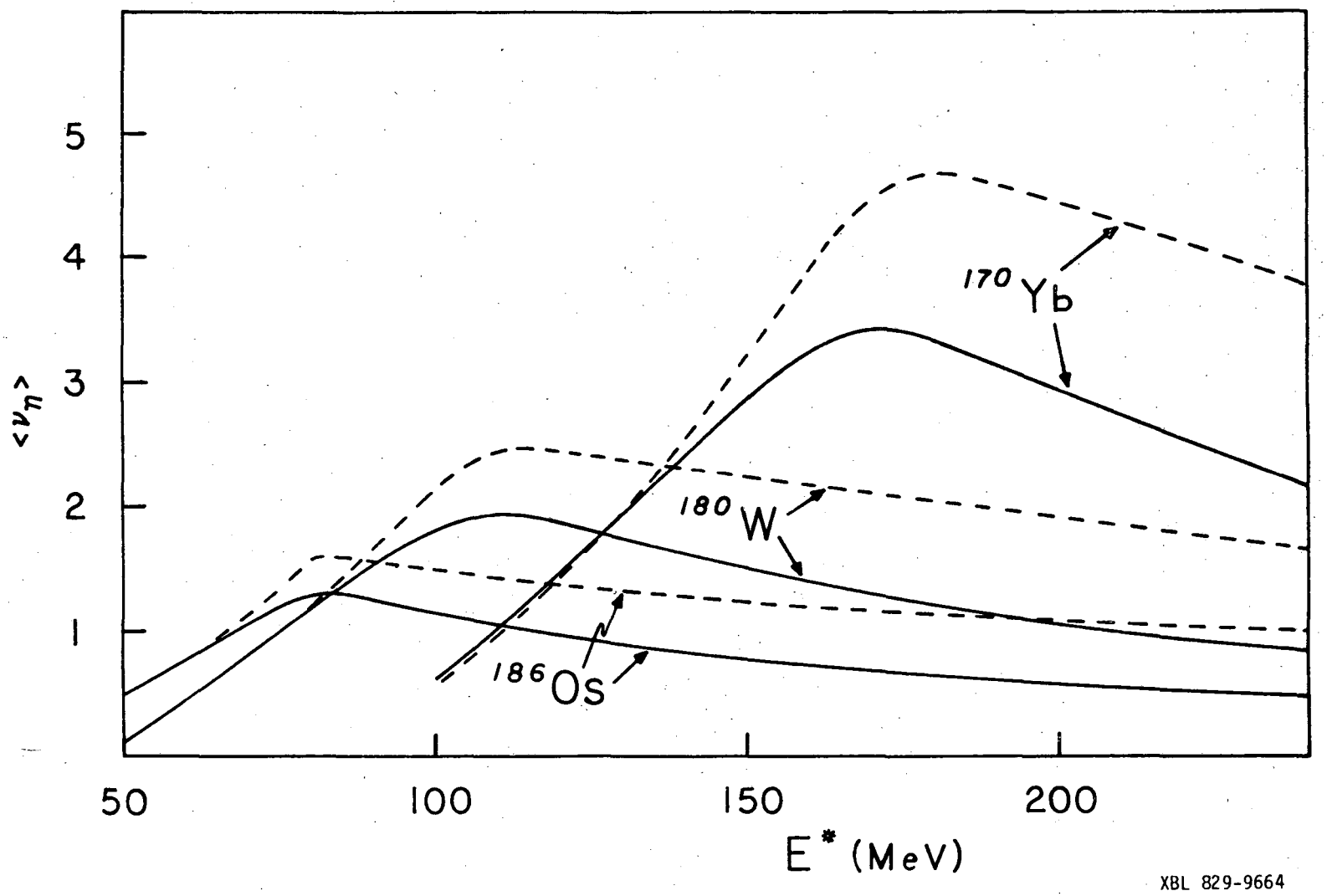
5
4
3
2
1
$\langle \nu_n \rangle$
50
100
150
200
E^* (MeV)
^{170}Yb
^{180}W
^{186}Os
XBL 829-9664

Fig. 3

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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