

## Nuclear Superfluidity and Statistical Effects in Nuclear Fission\*

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The Bardeen, Cooper, Schrieffer formalism is applied to a calculation of the mean-square projection  $K_0^2$  of the angular momentum along the symmetry axis of an excited deformed nucleus. The results are compared with empirical values obtained from analysis of recent data on fission-fragment angular distributions. The comparison corroborates qualitatively the validity of this application of the BCS formalism. Quantitative optimization of the fit to experiment yields the result that the energy gap for a nucleus deformed to the fission-barrier shape is about twice as large as the same quantity at the stable shape. Implications of this result for odd-even effects in nuclear fission are discussed.

### I. INTRODUCTION

THE BCS model of superconductivity<sup>1</sup> has been applied with remarkable success in the description of properties of nuclear ground states and low excited states.<sup>2-6</sup> It has also been suggested<sup>7</sup> that this description may suffice to explain certain data concerned with nuclei at higher excitations, although the basis of evidence in favor of applicability to pertinent statistical properties is at present much less substantial than that on which the ground-state applications are made.

This paper reports an analysis of improved measurements<sup>8,9</sup> of the anisotropy of fission fragments from the neutron-induced fission of Pu<sup>239</sup>, based on the BCS description of the dependence of pairing effects on excitation. The special relevance of such data to pairing in deformed nuclei has already been discussed elsewhere,<sup>7</sup> and seems to outweigh the minor disadvantage that they provide information not on nuclei near their normal ground-state shape, but stretched instead to their saddle-point shape during the fission process.

We first discuss those features of the BCS analysis relevant to superfluid nuclei (Sec. II) and develop their application to those aspects of nuclear structure which determine the fission anisotropy (Sec. III). Then the analysis of the experimental data in terms of parameters characteristic of the nuclear structure is described

(Sec. IV). In Sec. V the comparison of theory and experiment is made and the conclusions are drawn that the superfluid description of this data is clearly superior to an independent particle description, and that it is adequate to describe the data at its present level of accuracy. Finally, in Sec. VI inferences are drawn concerning the properties of nuclei at their saddle shape and their implications for other features of the fission process are noted. The results are summarized in Sec. VII.

### II. THE BCS ANALYSES FOR SUPERFLUID NUCLEI

The statistical analysis of a Fermi system described by a pairing Hamiltonian was first treated by Bardeen, Cooper, and Schrieffer<sup>1</sup> in connection with the superconductivity of the electrons in some metals. By requiring that the free energy of the system be minimal at any given temperature,  $T$ , they obtained a modified single-particle spectrum described by the replacement

$$\epsilon_j \rightarrow E_j = (\epsilon_j^2 + \Delta^2)^{1/2}, \quad (1)$$

where  $\epsilon_j$  is the excitation energy of the  $j$ th particle (or hole) in the absence of the interaction, and  $E_j$  the corresponding excitation energy of the  $j$ th excitation when the interaction is included. The new excitations are sometimes called "quasiparticles" because of the fact that their description corresponds essentially to the description of independent Fermi particles whose spectrum corresponds to Eq. (1) above. This feature is perhaps more transparent when the problem is handled by the alternative, but equivalent, method of Valatin<sup>10</sup> and Bogoliubov.<sup>11</sup>

Besides the modification of the spectrum, BCS derives the equation for the dependence of the gap parameter  $\Delta$  on temperature.

$$\frac{1}{G} = \int_0^{\hbar\omega} \frac{d\epsilon}{E} \tanh\left(\frac{E}{2T}\right). \quad (2)$$

Here  $G$  is the pairing matrix element (assumed constant

<sup>10</sup> J. G. Valatin, *Nuovo Cimento* **7**, 843 (1958).

<sup>11</sup> N. N. Bogoliubov, *Zh. Eksperim. i Teor. Fiz.*, **34**, 58 (1958) [translation: *Soviet Physics—JETP* **7**, 41 (1958)]; *Nuovo Cimento* **7**, 794 (1958). N. N. Bogoliubov, V. Tolmachev, and D. Shirkov, *A New Method in the Theory of Superconductivity* [English translation (Consultants Bureau, Inc., New York, 1959)].

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<sup>1</sup> J. Bardeen, L. N. Cooper, and J. Schrieffer, *Phys. Rev.* **108**, 1175 (1957), referred to in this paper as BCS.

<sup>2</sup> S. T. Belyaev, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* **31**, No. 11 (1959).

<sup>3</sup> D. R. Bes and Z. Szymanski, *Nucl. Phys.* **28**, 42 (1961). V. Soloviev, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Skrifter* **1**, No. 11 (1961). T. Voros, V. Soloviev, and T. Sikolos, *Joint Inst. for Nuclear Research, Dubna, USSR Lab. of Theoretical Physics, Report JINR-E-932*, 1962 (unpublished), and references cited therein.

<sup>4</sup> J. Griffin and M. Rich, *Phys. Rev.* **118**, 850 (1960).

<sup>5</sup> S. G. Nilsson and O. Prior, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* **32**, No. 16 (1960).

<sup>6</sup> C. Gallagher and V. Soloviev, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* (1962).

<sup>7</sup> J. J. Griffin, in *Proceedings of International Conference on Nuclear Structure, Kingston*, edited by D. A. Bromley and E. W. Vogt (University of Toronto Press, Toronto, Canada, 1960), p. 843.

<sup>8</sup> J. E. Simmons (private communication).

<sup>9</sup> R. L. Henkel, R. B. Perkins, and J. E. Simmons (to be published).

between states in a range  $\pm\hbar\omega$  about the Fermi energy),  $g$  is the density (assumed constant) of unperturbed degenerate pairs of single-particle levels (neutrons or protons) near the Fermi sea, and  $E$  is given by Eq. (1). Solution of Eq. (2) provides

(a) the ground-state value of the gap parameter,  $\Delta_0$ , when  $T=0$ ;

(b) the value of the critical temperature  $T_c$  at which Eq. (2) is satisfied by  $\Delta=0$ , and above which the energy is no longer minimized by a modification of the form (1), so that the system reverts to the ordinary description obtained by neglecting the pairing interaction;

(c) the dependence of the gap parameter on temperature when  $0 < T < T_c$ .

When  $\Delta_0 \ll \hbar\omega$ , the weak coupling limit applies and Eq. (2) yields

$$\Delta_0 = \frac{\hbar\omega}{\sinh 1/gG} \quad (3)$$

$$T_c = \frac{2\gamma}{\pi} \hbar\omega \exp\left(-\frac{1}{gG}\right) \quad (4)$$

in the extreme cases mentioned, together with the ratio

$$2\Delta_0/T_c = 2\pi/\gamma = 3.5278. \quad (5)$$

Here  $\ln\gamma$  is the Euler-Mascheroni<sup>12</sup> constant,

$$\gamma = 1.781072 \dots$$

All of these formulas are relevant to deformed nuclei with appropriate choice of the constants  $g$ ,  $G$ , and  $\hbar\omega$  and appropriate modification for the fact that the neutrons and protons comprise two independent fermion systems rather than the single fermion system described in the case of electrons in metals. For simplicity we assume in this analysis that the constants relevant to neutrons are the same as those for protons, so that the modifications involve merely insertions of  $2x$  factors in various places and the values of the constants correspond to an average of the actual neutron and proton values.

For nuclei one also needs a description parametrized by the excitation energy rather than the temperature. In a finite system, this feature carries implications which will be discussed more fully elsewhere<sup>13</sup>; for the purposes of this discussion it suffices to assume that an adequate average description of the system may be obtained by identifying each temperature with the average excitation energy calculated at that temperature in the minimal BCS ensemble. The excitation energy is just the difference between ensemble average of the Hamiltonian at temperature  $T$  and the same average at zero temperature, and can be written

$$\begin{aligned} E^*(T) &= U(T) - U(0) \\ &= U_0 + U_{\text{qp}} + U_{\text{sc}} - U_0(0), \end{aligned} \quad (6)$$

<sup>12</sup> B. Muhlschlegel, Z. Physik **151**, 613 (1958); **155**, 313 (1959); **156**, 235 (1959).

<sup>13</sup> M. Rich and J. Griffin, Phys. Rev. Letters **11**, 19 (1963).

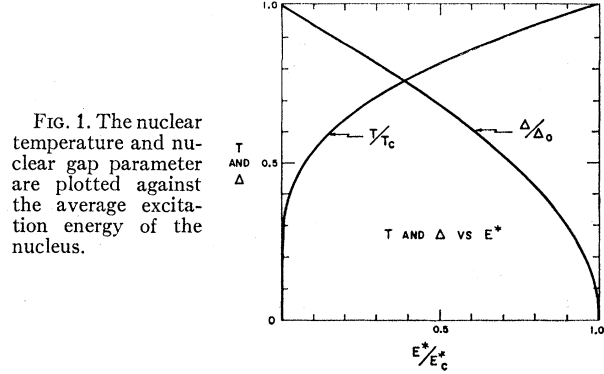


FIG. 1. The nuclear temperature and nuclear gap parameter are plotted against the average excitation energy of the nucleus.

where  $U_0$  is the "ground-state" energy at temperature  $T$ , measured with respect to the ground state of the noninteracting Fermi gas.

$$U_0(T) - U_0(0) = g\Delta^2 - g\Delta^2. \quad (7)$$

The quasiparticle energy  $U_{\text{qp}}$  has the form analogous to a single-particle excitation energy<sup>14</sup>:

$$U_{\text{qp}} = 4g \int_0^\infty d\epsilon f(\epsilon) E = 8gT^2 J(t), \quad (8)$$

where  $t = T/T_c$ .

Finally, the self-consistent energy arises from the minimization procedure and is given by

$$U_{\text{sc}} = -4g\Delta^2 \int_0^\infty \frac{d\epsilon}{E} f(\epsilon) = -4g\Delta^2 L(t). \quad (9)$$

In these expressions  $f(\epsilon)$  is the usual Fermi occupation function for a quasiparticle  $\epsilon$  with energy given by (1).

$$f(\epsilon) = \left\{ 1 + \exp\left[\frac{1}{T}(\epsilon^2 + \Delta^2)^{1/2}\right] \right\}^{-1}. \quad (10)$$

These formulas include already the  $2x$  factor for neutrons and protons; also, temperature is measured throughout in energy units.  $E^*(T)$  is then obtained by numerical evaluation of  $\Delta(t)$ ,  $J(t)$ , and  $L(t)$ ; it is plotted in dimensionless form in Fig. 1, together with the ratio  $\Delta/\Delta_0$ .

### III. THEORETICAL CALCULATION OF FISSION ANGULAR DISTRIBUTION PARAMETER, $K_0^2$

The modifications of the independent-particle results implied by a BCS pairing interaction are especially relevant to data on the angular distribution of fission fragments because of the fact that such angular distributions depend specifically on the quantity  $K_0^2$  which measures the mean square value of the projection of angular momentum on the nuclear symmetry axis. In

<sup>14</sup> Various dimensionless integrals occur throughout this discussion. In each case they depend only on the dimensionless temperature  $t = T/T_c$ . They are defined and, in some cases, tabulated in the Appendix.

the presence of a pairing interaction among the single-particle states (doubly degenerate with projection  $\pm k$  along the symmetry axis), the spectrum of excitations which can contribute to the total projection<sup>15</sup> no longer extends continuously to the ground state; instead, such excitations occur only above minimum energy  $2\Delta$  in even-even nuclei. The result is that, at a given excitation energy, a superfluid nucleus will have a value of  $K_0^2$  significantly less than its noninteracting counterpart. The dependence of  $K_0^2$  upon excitation energy is therefore a good test of the persistence of superfluid effects to finite excitation energies. For this reason the very careful measurements analyzed here are of special value for such a test. In the present section we discuss the relationship between the superfluid model and the quantity  $K_0^2$  derived from these measurements, following essentially a method described earlier.

In calculating the quantity  $K_0^2$  we must recognize that the intrinsic spectrum of a deformed nucleus is not a complete description of the spectrum, but omits low-lying collective rotational states associated with the degeneracy of the deformed Hartree-Fock solution under rotations.<sup>16</sup> Therefore, although a good description of the average angular momentum along the symmetry axis may be obtained by considering the intrinsic spectrum alone, the moment of inertia  $\mathfrak{F}_1$  perpendicular to this axis must be calculated by appeal to some semiclassical argument such as the "cranking model"<sup>17</sup> or the method of Migdal.<sup>18</sup>

Thus, in the Boltzmann factor,

$$\exp\left(-\frac{K^2}{2K_0^2}\right) = \exp\left\{-\frac{\hbar^2 K^2}{2T} \left[ \frac{1}{\mathfrak{F}_{11}} - \frac{1}{\mathfrak{F}_1} \right]\right\} \quad (11)$$

the quantity  $2T\mathfrak{F}_{11}$  can be evaluated directly from the average of  $K^2$  over the intrinsic spectrum:

$$\frac{T\mathfrak{F}_{11}}{\hbar^2} = \langle K^2 \rangle = 4g \int_{-\infty}^{\infty} d\epsilon k_\epsilon^2 f(\epsilon) [1 - f(\epsilon)], \quad (12)$$

$$= 4g \langle k^2 \rangle_{av} T I(t), \quad (13)$$

$$= \mathfrak{F}_{11}^{rig} T \frac{I(t)}{\hbar^2}. \quad (14)$$

Here  $k_\epsilon^2$  is the square of  $k$  for the quasiparticle excitation with energy  $(\epsilon^2 + \Delta^2)^{1/2}$  and  $f(1-f)$  is the probability that one and only one of the degenerate pair  $\pm k_\epsilon$  is excited. We assume in evaluating  $\langle K^2 \rangle$  that  $k_\epsilon^2$  may be replaced by some average value  $\langle k^2 \rangle_{av}$  and removed from the integral.

The evaluation of  $\mathfrak{F}_1 T$ , on the other hand, must pro-

<sup>15</sup> This statement is true apart from possible collective excitations which may fall in the region of the energy gap. Such states are not considered in this analysis because their contribution to  $K_0^2$  should be small.

<sup>16</sup> D. J. Thouless, Nucl. Phys. **31**, 211 (1962).

<sup>17</sup> D. R. Inglis, Phys. Rev. **96**, 1059 (1954); **97**, 701 (1955).

<sup>18</sup> A. B. Migdal, Nucl. Phys. **13**, 655 (1959).

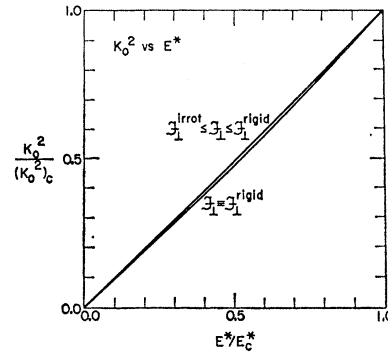


FIG. 2. The calculated parameter  $K_0^2$  is plotted versus excitation energy for the two cases given by Eqs. (19a), (19b).

ceed through a calculation of  $\mathfrak{F}_1$ . Migdal's analysis shows that

$$\mathfrak{F}_1^{irrot} < \mathfrak{F}_1 < \mathfrak{F}_1^{rig} \quad (15)$$

and also that

$$\mathfrak{F}_1 \rightarrow \mathfrak{F}_1^{rig} \quad (16)$$

as the deformation becomes large.

For  $\text{Pu}^{240}$ , with  $x = (Z^2/A)/(Z^2/A)_{\text{critical}}$  approximately equal to 0.74, liquid drop calculations<sup>19</sup> yield for  $R_{\text{max}}/R_0$  at the saddle shape the value 1.71. The irrotational moment for a spheroid of this shape is

$$\mathfrak{F}_1^{irrot} = 0.75 \mathfrak{F}_1^{rig}, \quad (17)$$

which already limits the range of  $\mathfrak{F}_1$  so seriously as to suggest that the approximation  $\mathfrak{F}_1 \equiv \mathfrak{F}_1^{rig}$  should be fairly good at all temperatures. At the opposite allowable extreme, however, is the assumption that  $\mathfrak{F}_1$  is given by  $\mathfrak{F}_1^{irrot} = 0.75 \mathfrak{F}_1^{rig}$  at  $T=0$  and increases to  $\mathfrak{F}_1^{rig}$  at  $T=T_c$  in accordance with the algebraic dependence on  $\Delta$  given by Belyaev<sup>2</sup>:

$$\mathfrak{F}_1 = \mathfrak{F}_1^{rig} \left[ 1 + 0.241 \frac{\Delta(T)^2}{\Delta_0^2} \right]^{-3/2} = \mathfrak{F}_1^{rig} f(t). \quad (18)$$

These two extremes yield

$$\frac{K_0^2}{(K_0^2)_c} = \frac{3.05tI(t)}{[4.05 - I(t)/f(t)]}, \quad (19a), (19b)$$

where  $t = T/T_c$ ,  $(K_0^2)_c$  is the value of  $K_0^2$  at  $T=T_c$ , and  $f(t)$  is (a) identically equal to 1 when  $\mathfrak{F}_1 = \mathfrak{F}_1^{rig}$  is assumed, or (b) defined by (18) when  $\mathfrak{F}_1$  varies in the manner assumed. We have also inserted the relationship  $\mathfrak{F}_1^{irrot}/\mathfrak{F}_1^{rig} = 4.05$  appropriate to the calculated<sup>19</sup> shape of  $\text{Pu}^{240}$  at the saddle point. The functions (19a), (19b) are plotted in Fig. 2.

We note that, in the absence of pairing,  $I(t) = f(t) = 1$ , and Eq. (19) reduces to the usual independent particle result that  $K_0^2$  is simply proportional to  $T$ . Even in the presence of pairing, this identity applies at the critical temperature ( $t=1$ ), so that the continuous transition

<sup>19</sup> Stanley Cohen and W. J. Swiatecki, University of California, Lawrence Radiation Laboratory Report UCRL-10450, 1962 (unpublished).

to the independent particle behavior for  $T \geq T_c$  is assured.

Equation (19) provides a theoretical description of the dependence of  $K_0^2$  on temperature [or on excitation energy, through Eq. (6)] which can be compared with suitable experimental data. In the process of comparison, two parameters can be adjusted to optimize the fit:  $(K_0^2)_e$  and  $T_c$  (or  $E_c^*$ ). This comparison is discussed in Sec. V.

#### IV. ANALYSIS OF DATA

The experimental data of Simmons<sup>8</sup> and Henkel and Simmons<sup>9</sup> were combined to correct for the 5% Pu<sup>240</sup> present in the target sample. The resulting Pu<sup>240</sup> anisotropies (Table I) were analyzed by means of the ANG-

TABLE I. Angular distributions of fission fragments from Pu<sup>239</sup>+n (Ref. 8), corrected for 5% Pu<sup>240</sup> by the measurements of Ref. 9. The Pu<sup>239</sup> data used here lacked certain small experimental corrections (see Ref. 8) which are inconsequential for purposes of the present analysis.

$E_n$	$W(67.5^\circ)$	$W(45^\circ)$	$W(22.5^\circ)$	$W(10^\circ)$
	$W(90^\circ)$	$W(90^\circ)$	$W(90^\circ)$	$W(90^\circ)$
1.00	1.018±0.011	1.039±0.010	1.076±0.011	1.070±0.010
1.50	1.017±0.011	1.048±0.011	1.088±0.011	1.097±0.011
1.75	1.026±0.010	1.058±0.012	1.091±0.012	1.108±0.012
2.00	1.025±0.019	1.066±0.016	1.079±0.012	1.114±0.012
2.25	1.026±0.008	1.056±0.007	1.101±0.007	1.111±0.008
2.50	1.024±0.007	1.056±0.007	1.107±0.007	1.116±0.007
2.75	1.017±0.005	1.049±0.005	1.100±0.006	1.114±0.005
3.00	1.021±0.005	1.054±0.005	1.089±0.005	1.110±0.005
3.25	1.018±0.009	1.051±0.009	1.098±0.009	1.111±0.009
3.50	1.018±0.006	1.058±0.006	1.089±0.006	1.103±0.005
4.00	1.026±0.011	1.066±0.011	1.093±0.011	1.119±0.013
4.50	1.007±0.014	1.060±0.013	1.103±0.015	1.119±0.016
5.00	1.025±0.013	1.055±0.012	1.089±0.012	1.106±0.013

code<sup>20</sup> which searches the value of  $\beta = 1/2K_0^2$  which best fits the data according to the formula<sup>21-24</sup>

$$W(\vartheta) = \sum_I \sum_M \sum_K G(I, M) v(\beta; I, K) |D_{MK}^I(\vartheta)|^2, \quad (20)$$

where

$$G(I, M) = \sum_L \sum_i \sum_{M_0} (2L+1) \times T_L(E_n) |C_{M_0 M}^{i L I}|^2 |C_{M_0, M-M_0, M}^{I_0 S j}|^2 \times [(2S+1)(2I_0+1) \sum_L (2L+1) T_L(E_n)]^{-1}, \quad (21)$$

<sup>20</sup> L. Blumberg (private communication). See also Ref. 24.

<sup>21</sup> A. Bohr, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1956* (United Nations, Geneva, 1956), Vol. 2.

<sup>22</sup> I. Halpern and V. M. Strutinskii, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15.

<sup>23</sup> J. Griffin, *Phys. Rev.* **116**, 107 (1959).

<sup>24</sup> L. Blumberg, thesis, Columbia University, 1962 (unpublished).

and

$$v(\beta; I, K) = e^{-\beta K^2} \left[ \sum_{K=-I}^I e^{-\beta K^2} \right]^{-1}. \quad (22)$$

For the neutron-induced fission of Pu<sup>239</sup>,  $I_0 = \frac{1}{2}$  and  $S = \frac{1}{2}$  in the above formulas. The penetration coefficients  $T_L(E_n)$  for neutrons were taken from optical-model calculations of Blumberg<sup>24</sup> based on a Woods-Saxon potential characterized by the following parameters:  $r_0 = 1.3A^{1/3}$  F,  $d = 0.5$  F; and  $V_0 = 44$  MeV to 43 MeV,  $W_0 = 3.3$  MeV to 3.6 MeV for  $E_n = 1.0$  MeV to 5.0 MeV, respectively.<sup>25</sup>

By searching  $\beta$  so as to minimize squared differences between the theoretical expression (20) and the observations of Table I one obtains at each neutron energy the value of  $\beta$  which gives the best fit, and also an estimate of the variance of  $\beta$  about this best value. In Table II,

TABLE II. Results of analysis of the data of Table I.

$E_n$	$E^* - E_f$	$\frac{1}{2\beta} = K_0^2$	$\pm \sigma(K_0^2)$	$\chi^2$
1.00	2.60	16.03	1.521	1.57
1.50	3.10	17.78	1.399	0.15
1.75	3.35	17.86	1.387	1.44
2.00	3.60	19.76	1.525	3.18
2.25	3.85	19.33	0.893	1.87
2.50	4.10	19.35	1.315	1.76
2.75	4.35	21.65	0.723	1.75
3.00	4.60	23.87	0.784	2.62
3.25	4.85	24.30	1.396	0.25
3.50	5.10	27.20	0.999	1.83
4.00	5.60	27.43	2.361	3.09
4.50	6.10	28.62	2.593	0.46
5.00	6.60	35.26	2.938	0.78

these results are tabulated (in terms of  $K_0^2 = 1/2\beta$ ) against the excitation energy in excess of the fission threshold. Table II is then the raw material for a comparison of the empirical dependence of  $K_0^2$  on  $E^* = E_f$  with theory.

#### V. COMPARISON WITH THEORY

The experimental values of  $K_0^2$  of Table II were fit to the theoretical Eqs. (19a) and (19b) by choosing values of  $E_c^*$  and searching for corresponding values of  $(K_0^2)_e$  which give the best fit to the data in the  $\chi^2$  sense. Because of the approximately linear relationship between  $K_0^2$  and  $E_c^*$ , both formulas can be fit almost equally well by a range of parameters. Moreover, the values of  $\chi^2$  associated with the two extreme dependences of  $\mathfrak{F}_L$  do not differ significantly. Therefore, the best fitting values of  $(K_0^2)_e$  for both dependences are plotted in Fig. 3 against the corresponding values of  $E_c^*$ . The values of  $\chi^2$  along these curves are such that the low-energy data alone does not exhibit a clear-cut preference for any particular pair of parameters.

<sup>25</sup> R. G. Schrandt, J. R. Beyster, M. Walt, and E. W. Salmi, Los Alamos Scientific Laboratory Report LA-2099, 1959 (unpublished).

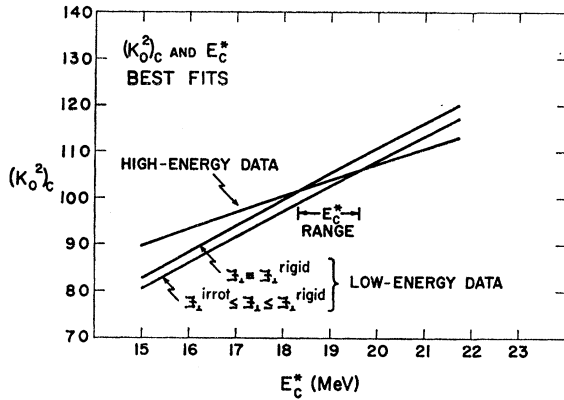


FIG. 3. The loci of best fits of the low energy data of Table IV to Eqs. (19a), (19b) are plotted. A similar curve is plotted for the high-energy data (Table II). Also indicated is the acceptable range of values for the critical energy.

In order to choose the best values of  $E_c^*$  and  $(K_0^2)_c$  from the curves of Fig. 3, we can consider data on  $K_0^2$  taken at higher excitation energies. Vandenbosch *et al.*<sup>26</sup> have measured the fission anisotropy for  $U^{233} + \alpha$ , which is especially favorable in that the percentage of second-chance fission (which is a complicating factor in the analysis) is quite low. For excitation energies above 16 MeV, they deduce values of  $K_0^2$  listed in Table III.

TABLE III. Empirical values of  $K_0^2$  at higher excitation energy. These values were obtained from Fig. 9 of Ref. 26 by subtraction of  $\langle k^2 \rangle_{av} = 10$  from those values.  $\sigma(K_0^2)$  is also taken from the uncertainty indicated in that figure. The author is grateful to Dr. John Huizenga for supplying an enlargement of this figure to facilitate this procedure.

$E^*$ (MeV)	$K_0^2$	$\pm\sigma(K_0^2)$
16	88	10
18	95.5	10.5
20	104.5	8.5
22	110.5	6.5
24	118.0	6.0
26	125.5	5.5
28	134.0	4.0
30	142.0	4.0
32	150.0	4.0
34	157.0	4.0

By demanding that the parabolic dependence of the independent particle model (which should be the correct description for  $E^* \geq E_c^*$ ) provide a good fit to these measurements, we can determine the intersection between the locus of best fits to the low-energy data and to the higher energy data. The result is a unique determination of the critical excitation energy  $E_c^*$  and the corresponding value of  $(K_0^2)_c$ .

In carrying out this combination of high- and low-energy data, one major feature must not be overlooked; viz., the parabolic dependence applicable at high energy should, if extrapolated to low energy, predict  $K_0^2 = 0$ —

<sup>26</sup> R. Vandenbosch, H. Warhanek, and J. R. Huizenga, Phys. Rev. **124**, 846 (1961).

not at  $E^* = 0$ , but at the ground-state energy of the independent particle system, which lies at an excitation energy  $E^* = g\Delta_0^2$  in the paired system. Moreover, since the fissioning nucleus  $Pu^{237}$  is odd in this case, one must have  $K_0^2 = \langle k^2 \rangle_{av}$  at the ground state rather than  $K_0^2 = 0$ , which would apply for an even-even fissioning system such as the  $Pu^{240}$  measured in the low-energy region.

When these features are taken into account, the independent particle model dependence of  $K_0^2$  on the excitation energy  $E^*$  of a paired system is given by

$$K_0^2 = \langle k^2 \rangle_{av} + c(E^* - g\Delta_0^2)^{1/2}. \quad (23)$$

Since by Eqs. (5) through (10),  $E_c^*$  is equal to  $3.1148 g\Delta_0^2$ , the requirement that (20) optimize the fit to  $\alpha$ -induced measurements above  $E^* = 16$  MeV provides an equation for pairs of values  $[(K_0^2)_c', E_c^*]$  (where the prime indicates that  $\langle k^2 \rangle_{av}$  has been subtracted to make this parameter correspond to that for the even-even low-energy data) which are consistent with his data. This relationship is also plotted in Fig. 3. Its intersections with the loci of best fits to the low-energy data specify the range of the parameters consistent with all the data considered and with the range of variations for  $\mathfrak{F}_1$  between the extremes of Eqs. (19a) and (19b), respectively,

$$\begin{aligned} 18.3 \text{ MeV} < E_c^* < 19.6 \\ 101.5 < (K_0^2)_c < 106.1. \end{aligned} \quad (24)$$

One notes that the total  $\chi^2$  for the two sets of data (23 data in all) varies from 16.3 to 15.8 between these extremes. The probabilities that  $\chi^2$  exceeds these values in a distribution with 21 degrees of freedom both lie between 70% and 80%. Thus, there is hardly any statistical preference for either assumption (19a) or (19b) indicated by the data.

The dependences of  $K_0^2$  on  $E^*$  implied by the extreme values (24) of the parameters are presented in Fig. 4, together with the empirical values at low energy (Table I) and the higher energy data (Table II) measured by Vandenbosch *et al.*,<sup>26</sup> corrected to the corresponding even-even value by the subtraction of the odd particle contribution ( $\langle k_p^2 \rangle_{av} = 10$ ).

## VI. IMPLICATIONS OF PRESENT ANALYSIS

### A. Implied Increase in Gap Parameter

To simplify further discussion, we assume that  $E_c^* = 19$  MeV, a value intermediate to the extremes allowed by the present analysis. If the level density parameter has the value  $g = 4.5$ , Eq. (6) implies

$$\Delta_0 = 1.36 \text{ MeV} \quad (25)$$

for an even-even nucleus like  $Pu^{240}$  stretched to its saddle-point shape. This value is somewhat larger than the corresponding values for similar nuclei at their stable ground-state deformation, a result which carries implications discussed below.

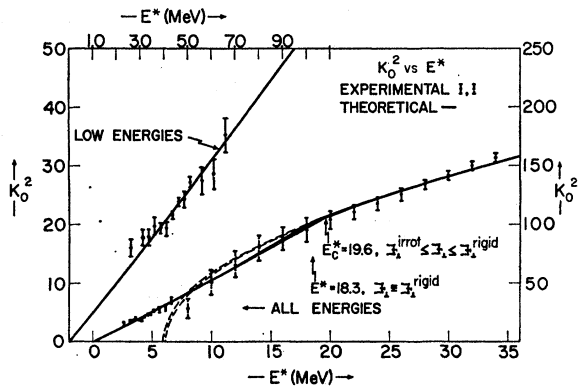


FIG. 4. The comparison between the low energy data (magnified scales) and the theory for the extreme best fitting values [Eq. (24)] is exhibited, together with the same comparison for the high and low energy data taken together. Dotted bars indicate data not utilized in determining best fits.

### B. Odd-Even Differences in the Fission Process

Figure 5 provides a pictorial description of how a difference between the gap parameter for the stable shape  $\Delta_0^s$  and that for the fissioning saddle shape  $\Delta_0^f$  can lead to a retardation of the spontaneous fission half-lives of odd nuclei with respect to even nuclei and to a shift in the corresponding thresholds for energetic fission processes.<sup>27</sup> There the potential surface of the even-even nuclei is traced as a function of deformation. On the same scale is plotted the corresponding potential surface for an adjacent odd- $A$  nucleus which lies higher by an amount approximately equal to the gap parameter  $\Delta$ , which may in general vary with deformation, assuming values  $\Delta_0^s$  and  $\Delta_0^f$  at the stable and saddle-point deformations, respectively. One sees that spontaneous fission of the odd nucleus requires penetration of a potential barrier higher by an energy

$$\Delta V = \Delta_0^f - \Delta_0^s \quad (26)$$

than that of the even-even nucleus. We have assumed here that fluctuations of ground-state energies away from the smooth mass surfaces have been corrected for in the manner proposed by Swiatecki.<sup>28</sup>

One can estimate the magnitude of this effect by using the result of Ref. 29 that for  $Z^2/A = 36.8$ , one millimass unit increase in the potential barrier corresponds to an increase in lifetime by a factor approximately equal to  $10^{4.3}$ . Since the value of  $\Delta_0^f$  implied by our analysis is approximately 700 keV larger than that<sup>29</sup> of stable Pu<sup>240</sup>, one estimates a retardation factor of about  $10^{9.5} \approx 2 \times 10^9$ .

It should be noted that the retardation of odd- $A$

<sup>27</sup> P. Fong, Phys. Rev. **122**, 1545 (1961) also discusses the effect of nuclear pairing on odd-even effects in fission, and makes certain arguments in favor of an enhanced pairing energy at the saddle shape.

<sup>28</sup> W. Swiatecki, Phys. Rev. **100**, 936, 937 (1955); **100**, 936 (1955).

<sup>29</sup> E. K. Hyde, University of California, Lawrence Radiation Laboratory Report UCRL-9036, 1960 (unpublished).

spontaneous fission half-lives has also been attributed<sup>30,31</sup> to the "specialization energy" associated with the constancy of the  $K$ -quantum number as the nucleus deforms toward the saddle point. In particular, Johansen<sup>32</sup> has analyzed this effect quantitatively and concludes that it is of the correct order of magnitude to explain the observed retardation factor ( $\sim 10^9$ ).

It must be said that, although it would be very difficult to improve this calculation, it is based on some questionable simplifying assumptions (e.g., that the nucleus retains a spheroidal shape to the saddle point), and so need not be considered the final word on the question at hand.

Moreover, this explanation of odd-even effects is much less cogent when one attempts to explain with it similar effects evident in energetic fission processes, since these involve the passage through an intermediate region of small deformation and relatively large internal excitation, during which it is hard to believe that the initial value of the projection  $K$  actually retains its identity as a good constant of the motion.

Indeed, if one assumes that  $K$  is a perfect constant of the motion for spontaneous fission, but not constant at all for energetic fission, it is possible to untangle the two effects in question. This is due to the fact that the pairing effect described here would then apply to both situations, whereas the constant- $K$  effect would be in evidence only for spontaneous fission. Then any difference between the even-odd effect in spontaneous fission and in threshold values for energetic fission would be attributed to the constancy of the  $K$ -quantum number, whereas the identical remainder would be attributable to the pairing effect discussed here.

In Ref. 28 occur analyses of both thresholds and spontaneous lifetimes, which imply an odd-even threshold difference of 1.2 mmu for  $Z^2/A \approx 37$  and an equivalent barrier height difference of 1.53 mmu from spontaneous-fission lifetimes. This would suggest a saddle-point gap parameter larger by 1.2 mmu than that at the stable shape. Our data analysis gives the comparable result of 700 keV = 0.75 mmu.

The difference could easily be resolved by a less ex-

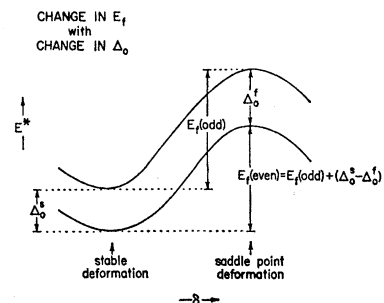


FIG. 5. The effect of a deformation-dependent gap on the differences between fission barriers for odd mass and even-even compound nuclei is illustrated.

<sup>30</sup> J. O. Newton, Progr. Nucl. Phys. **4**, 234 (1955).

<sup>31</sup> J. A. Wheeler, *Niels Bohr and the Development of Physics* (Pergamon Press, Ltd., London, 1955) p. 163.

<sup>32</sup> S. E. Johansson, Nucl. Phys. **12**, 449 (1959).

treme assumption than that used here, viz., assuming a *tendency* for  $K$  to remain constant even in energetic process, but with perfect constancy only for the spontaneous fission case. A quantitative statement of this explanation would, however, add nothing to the evidence at hand, which seems sufficient only for the qualitative conclusion that the dependence of the gap parameter on deformation could easily be as important in determining even-odd effects in fission as the assumed constancy of the  $K$ -quantum number.

### C. Other Implications of the Saddle-Point Gap

The implication of the present analysis that the gap at the saddle shape is even larger than that at the stable deformation suggests several qualitative features that can be expected in the angular distributions of fission fragments. In particular, it implies that, for excitation energies less than about 2.50 MeV above the fission threshold: (a)  $K_0^2$  ought to be quite small in even-even nuclei, and (b) the average  $K_0^2$  in odd- $A$  compound nuclei should be determined only by the mean square projection,  $\langle k_p^2 \rangle_{av}$  associated with a single odd particle.

Unfortunately, the study of neutron-induced fission-fragment angular distributions from even-even compound nuclei is limited by two features: (1) fission thresholds are generally below the neutron binding energy in the heavy elements, and (2) very low-energy neutrons carry only small angular momenta and thus limit the precision with which  $K_0^2$  can be determined in situation (a). One hopes that these difficulties can be obviated by the use of  $(d, pf)$  reactions which are capable of producing fission at excitation energies below the neutron binding energy<sup>33</sup> in reactions involving reasonably large angular momenta.

As regards the odd- $A$  case (b), one must there seek a difference in the angular distributions attributable to the difference between the distribution in  $K$  for only one excited single particle and that for three or more single particles. For small anisotropies this is a very subtle distinction, and probably requires angular distribution data better by about one order of magnitude than that available at present. Such data might be obtainable by means of present-day solid-state counter technology. Alternatively, the  $(d, pf)$  process might also in this case be used effectively to alleviate the lack of angular momenta available in neutron processes at low energy.

## VII. QUANTITATIVE UNCERTAINTIES

Although the qualitative (almost linear) dependence of  $K_0^2$  on energy at low energies is adequately described by the superfluid theory used here, the choice of specific parameters necessary to quantify this theory (and the inferences drawn in the present analysis) is by no means unique. To achieve the specific numerical results

obtained it was necessary to assume values for three parameters: (a) The mean square contribution,  $\langle k_p^2 \rangle_{av}$ , of a single odd neutron to  $K_0^2$ , (b) the ratio of the rigid moments,  $\mathfrak{I}_1^{rig}/\mathfrak{I}_0^{rig}$ , and (c) the density  $g$  of pairs of single particle eigenstates in the deformed potential. In each case the assumption was made *a priori*, without specific reference to optimizing the agreement between theory and experiment.

$\langle k_p^2 \rangle_{av}$  was taken as the average of  $K^2$  over all the single particle eigenstates of the last unfilled major shell with total oscillator quantum number,  $N$ :

$$\langle k_p^2 \rangle_{av} = N^2/6, \quad (27)$$

where  $7 < N < 8$  is expected, which implied  $8 < \langle k_p^2 \rangle_{av} < 11$ . One sees from Fig. 4 that a vertical translation by  $\pm 3$  units in  $K_0^2$  of the curve tracing out the best fits to the high-energy data might change  $E_c^*$  by 2 MeV (or 10%), with a corresponding 5% change in  $\Delta_0$ .

The ratio  $\mathfrak{I}_1^{rig}/\mathfrak{I}_0^{rig}$ , was taken from liquid drop calculations<sup>19</sup> of the shape of the nucleus at the saddle point and exact integrations of the corresponding moments of inertia. To estimate the sensitivity of the results to this parameter, we have performed calculations with the liquid-drop value ( $\mathfrak{I}_1^{rig}/\mathfrak{I}_0^{rig} = 4.05$ ) changed by  $\pm 20\%$ . Such changes result in corresponding changes in the best fitting  $E_c^*$  by +1.4 and -2.0 MeV, in the implied value (25) of  $\Delta_0$  by +4% and -6%, and in  $\chi^2$  at the best fit by -1.4 and +1.7, respectively. These increments can provide estimates of uncertainty if only one makes the reasonable assumption that the liquid drop shape gives moments accurate to 10%.

It should be noted that a (perhaps) different value of this ratio might be obtained from experimental estimates<sup>34</sup> of the nuclear saddle-point shape. However, such an estimate involves a nuclear temperature and can therefore be made in a manner consistent with the present theory only by taking appropriate account of the modified energy-temperature relationship which the superfluid system exhibits. Moreover, the experiments cited appear to be consistent with the liquid-drop results we have utilized, so that one would be forced at the end of such a procedure to consider any difference between its results and the present ones attributable to experimental uncertainty.

Finally, we must consider the assumed value of  $g$  (4.5/MeV), an error in which would produce the same percentage error in the inferred value of  $\Delta_0$ . Our value was defined in the first rough estimate by counting the average density of levels in the Nilsson diagram in the neighborhood of the last filled lead in Pu<sup>240</sup>. This gave a range  $4 < g < 4.8$  for the average of the neutron and proton single-particle level density. The specific value chosen ( $g = 4.5$ ) within this range corresponds rather well with the value for  $A = 240$  indicated by the work of Newton<sup>35</sup> with the empirical correction suggested by

<sup>33</sup> Note added in proof. H. C. Britt, R. H. Stokes, W. R. Gibbs, and J. J. Griffin, Phys. Rev. Letters **11**, 343 (1963) report a realization of this hope.

<sup>34</sup> R. Chaudry, R. Vandenbosch, and J. R. Huizenga, Phys. Rev. **126**, 220 (1962).

<sup>35</sup> T. D. Newton, Can. J. Phys. **34**, 804 (1956).

Ericson.<sup>36</sup> There may be some basis for disagreement about this value, but favoring more likely a smaller value<sup>37</sup> rather than a larger, which would effect an increase in the inferred value of  $\Delta_0$  over that given in Eq. (25).

Over all, a reasonable estimate of the uncertainty in the value of  $\Delta_0$  given in Eq. (25) might be  $\pm 20\%$ .

*Note added in proof.* The direct measurement of Ref. 33 indicates rather better than 20% agreement with the value (25).

### VIII. SUMMARY

The present data and their successful description in terms of the statistical mechanics of a Fermi superfluid support the validity of extending the analogy between nuclei and superfluids to higher excitation energies.<sup>38</sup> Such an extension will imply deviations from ordinary independent particle model results for excitation energies less than  $E_c^* = 3.11 g\Delta_0^2$  in nuclei properly described by the weak-coupling limit (deformed nuclei).

In its detailed results, the present analysis implies that the energy gap at the saddle shape is somewhat larger than that at the stable shape for Pu<sup>240</sup>. This implication suggests a straightforward qualitative explanation of observed even-odd fission threshold differences as well as explaining a good portion of the odd-mass spontaneous fission retardation, previously attributed entirely to the constancy of the quantum number,  $K$ . Although this increase of the gap parameter with deformation runs counter to the trend observed for the heavy elements at their stable deformations, no clear-cut contradiction with theory can be established without more detailed study of the behavior of the parameters which determine the system in the weak-coupling limit.

### ACKNOWLEDGMENTS

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### APPENDIX I

#### A. Dimensionless Integrals

Several dimensionless integrals occur in the discussion of Secs. II and III. These are defined in terms of

<sup>36</sup> T. Ericson, in *Proceedings of the International Conference on Nuclear Structure, Kingston*, edited by D. A. Bromley and E. W. Vogt (University of Toronto Press, Toronto, Canada, 1960).

<sup>37</sup> F. H. Bakke, *Comptes Rendus du Congrès International de Physique Nucléaire aux Basses Energies et Structure des Noyaux, Paris* (Dunod Press, Paris, 1958).

<sup>38</sup> *Note added in proof.* Y. T. Grin, Zh. Eksperim. i Teor. Fiz. 43, 1880 (1962) [translation: Soviet Physics—JETP 16, 1327 (1963)] verifies this point independently, but does not utilize the analysis to obtain new information about the saddle-point gap.

the dimensionless temperature,  $t = T/T_c$ , as follows:

$$J(t) = \int_0^\infty dx \mathcal{E}(x,t) [1 + \exp \mathcal{E}(x,t)]^{-1},$$

$$L(t) = \int_0^\infty dx [\mathcal{E}(x,t) \{1 + \exp \mathcal{E}(x,t)\}]^{-1},$$

$$I(t) = 2 \int_0^\infty dx [\exp - \mathcal{E}(x,t) [1 + \exp - \mathcal{E}(x,t)]^{-2}],$$

where

$$\mathcal{E}(x,t) = [x^2 + x_0^{-2}]^{1/2},$$

and

$$\bar{x}_0 = \frac{\pi d(t)}{\gamma t}.$$

Here

$$d(t) = \frac{\Delta(t)}{\Delta_0}$$

is obtained by solution of the dimensionless gap equation in the weak-coupling limit:

$$0 = \int_0^{\hbar\omega/\Delta_0} dx \left\{ (x^2 + d^2)^{-1/2} \times \tanh \left[ \frac{\pi}{2\gamma t} (x^2 + d^2)^{1/2} \right] - (x^2 + 1)^{-1/2} \right\}.$$

This equation is obtained from Eqs. (2) and (5) by direct substitution.

TABLE IV. Calculated values of the excitation energy and gap parameter for specified values of  $t$ .

$t = T/T_c$	$E^*/E_c^*$	$d(t) = \Delta/\Delta_0$
1.001	1.000	0.000
0.980	0.9354	0.2436
0.960	0.8734	0.3416
0.940	0.8138	0.4148
0.920	0.7568	0.4749
0.900	0.7023	0.5263
0.85	0.5764	0.6303
0.80	0.4653	0.7110
0.75	0.3682	0.7759
0.70	0.2846	0.8288
0.60	0.1551	0.9070
0.50	0.0707	0.9569
0.40	0.0238	0.9850
0.20	0.0002	0.9999
0.00	0.0000	1.0000

For the convenience of the reader, the quantities  $d$ ,  $E^*/E_c$ , and  $K_0^2/(K_0^2)_c$ , which are of direct physical interest, are plotted in Figs. 1 and 2 and tabulated in Table IV.

We note that, although  $L(t)$  diverges at  $t=1$ , the quantity  $d^2 L(t)$ , which is of physical interest [Eq. (9)], remains well defined.