# Statistical-Model Analysis of Isomeric Ratios in  $(\alpha, xn)$  Compound Nuclear Reactions<sup>\*</sup>

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Cross-section ratios of isomeric pairs produced in the compound nuclear reactions <sup>41</sup>K( $\alpha, n$ ) <sup>44</sup>Sc, <sup>55</sup>Mn( $\alpha, n$ ) <sup>58</sup>Co, <sup>93</sup>Nb(a,n)<sup>98</sup>Tc, <sup>93</sup>Nb(a,2n)<sup>95</sup>Tc, <sup>53</sup>Nb(a,3n)<sup>94</sup>Tc, and <sup>136</sup>Ba(a,3n)<sup>137</sup>Ce are analyzed in terms of th statistical model. In this context Vandenbosch and Huizenga have proposed a formalism for calculating isomer formation from compound nuclei of relatively low excitation energy and spin. It is assumed that the density of levels of spin J in residual nuclei is proportional to  $(2J+1) \exp[-J(\dot{J}+1)/2\sigma^2]$  and that in the case of a product formed by neutron evaporation the emission of charged particles is not of importance. Here  $\sigma$  is the spin cutoff parameter and is related to the effective moment of inertia  $\sigma$ . In this paper the extension of such a model to compound nuclei of much higher excitation energy and spin is considered. Typically by this method our experimental isomeric ratios imply a value of *6* much smaller than that of a rigid sphere,  $s_R$ , even for nuclei excited to well above nucleon binding energies. The introduction of two additional factors in the formalism leads to more reasonable values of *\$/\$R.* First, we invoke a principle of limiting spin. That is, the level density of a residual nucleus is not described by the expression above for all  $J$ ; rather, above some critical  $J$  value, determined by a Fermi-gas model, the level density is taken to be zero. Secondly, we find charged-particle emission, particularly of  $\alpha$  particles, to be of importance in many cases. With the inclusion of these factors experimental isomeric ratios are consistent with an  $\frac{s}{g_R}$  value of unity when the excitation energy *Ef* is greater than about 10 MeV. Below 10 MeV, *\$/\$R* has essentially the same dependence on *Ef* for all of the reactions analyzed.

#### **I. INTRODUCTION**

IN the preceding paper<sup>1</sup> experimental values of<br>isomeric cross-section ratios are given for the reac-N the preceding paper<sup>1</sup> experimental values of tions  ${}^{41}\text{K}(\alpha,n){}^{44}\text{Sc},\ {}^{55}\text{Mn}(\alpha,n){}^{58}\text{Co},\ {}^{93}\text{Nb}(\alpha,n){}^{96}\text{Tc},\ {}^{93}\text{Nb}$  $(\alpha,2n)$ <sup>95</sup>Tc, <sup>93</sup>Nb( $\alpha,3n)$ <sup>94</sup>Tc, and <sup>136</sup>Ba( $\alpha,3n)$ <sup>137</sup>Ce. In each case recoil ranges and excitation functions were used to define the approximate region of bombarding energy in which the compound system has received all of the momentum of the incident  $\alpha$  particle and hence the region of applicability of a compound-nuclear model.

Since a pair of nuclear isomers in general differs only slightly in energy but considerably in spin, the yield ratio of an isomeric pair should be a function of the angular momentum involved in the reaction. Vandenbosch and Huizenga<sup>2</sup> (referred to hereafter as VH) have suggested a formalism based on the statistical model from which isomeric ratios can be calculated for reactions in which the excitation energy of the compound nucleus is relatively low. In several of the reactions analyzed in the present work, compound nuclei of much higher excitation energy and spin than those considered by VH have been generated. It is the purpose of this paper to extend the VH formalism to such systems.

We shall show that it is necessary to take into account the effect of the evaporation of particles other than the kind leading to the particular isomeric pair under consideration. We refer below to this effect as "competition." Further, we find it necessary to invoke a principle of limiting spin, i.e., that residual nuclei cannot have arbitrarily high spins. The effective moments of inertia extracted from the analysis then become reasonable in context of theory. $3-6$ 

Other have undertaken an analysis of systems basically similar to ours but have neglected competition and the principle of limiting  $spin.^{2,7-18}$  Their results

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<sup>6</sup> A. C. Douglas and N. MacDonald, Nucl. Phys. 13, 382 (1959);<br>
N. MacDonald and A. C. Douglas, Nucl. Phys. 24, 614 (1961).<br>
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<sup>\*</sup> Supported in part by the U. S. Atomic Energy Commission under Contract AT(30-1)-1930. This is AEC Report NYO-1930-42. f Present address: Department of Chemistry, Columbia Uni-

versity, New York, New York.<br><sup>1</sup> T. Matsuo, J. M. Matuszek, Jr., N. D. Dudey, and T. T.<br>Sugihara, preceding paper, Phys. Rev. 139, B886 (1965).<br><sup>2</sup> R. Vandenbosch and J. R. Huizenga, Phys. Rev. 1**20**, 1313

<sup>(1960).</sup> 

<sup>3</sup> T. Ericson, Advan. Phys. 9, 425 (1960). 4 D. W. Lang, Nucl. Phys. 42, 353 (1963).

invariably result in effective moments of inertia  $\mathfrak g$ which are 30% to 70% of that of a rigid sphere,  $g_R$ , even at average excitation energies well above the neutron-binding energy. Theoretical models<sup>3-6</sup> appear to require  $\mathfrak{I} \approx \mathfrak{I}_R$  at such energies.

The experimental data analyzed are only those of the preceding paper<sup>1</sup>; however, the formalism should be generally applicable to the many other experimental results which have been reported. We discuss first the general method by which the calculations were performed and describe the choice of functional form and parameters in the nuclear level density.

## **II. CALCULATIONS**

#### **A. General Description of the Model**

In the VH formalism, the compound nuclei of a given excitation energy are formed with a variety of spins. The evaporation of a particle leads to residua] nuclei also with a variety of spins. In the simplest case in which further particle emission is not energetically possible, further de-excitation occurs by  $\gamma$ -ray emission, again changing the spin distribution. Ultimately the  $\gamma$ -ray cascade leads to one or the other isomer. The process is described in terms of the statistical model.

The distribution in spin of the compound nucleus is given<sup>2</sup> by

$$
\sigma_e(E_{e,r}J_e) = \pi \lambda^2 \sum_{S=|I-s|}^{I+s} \sum_{l=|J_e-S|}^{J_e+S} \frac{2J_e+1}{(2s+1)(2I+1)} T_l(\epsilon), \quad (1)
$$

where *Ec* and *Jc* are the excitation energy and spin of the compound nucleus,  $\lambda$  is the de Broglie wavelength of the incoming projectile, *s* is the spin of the projectile, *I* is the spin of the target nucleus, *S* is the entrance channel spin, and  $T_i(\epsilon)$  is the transmission coefficient of the incident particle of channel energy *e* and orbital angular momentum  $l$ . In subsequent discussion it is convenient to use a normalized form of  $\sigma_c (E_c, J_c)$ ,

$$
P_c(E_c, J_c) = \sigma_c(E_c, J_c) / \sum_{J_c=0}^{\infty} \sigma_c(E_c, J_c).
$$
 (2)

The rate  $R_{\nu}d\epsilon_{\nu}$  at which the compound nucleus  $(E_c, J_c)$  emits a particle  $\nu$  to form a residual nucleus of excitation energy  $E_f$  and spin  $J_f$  is given by<sup>19</sup>

$$
R_{\nu}(E_c, J_c, \epsilon_{\nu}, E_f, J_f) d\epsilon_{\nu}
$$
  
= const  $\frac{\Omega(E_f, J_f)}{\Omega(E_c, J_c)} \sum_{s_{\nu} = |J_f - s_{\nu}|}^{J_f + s_{\nu}} \sum_{l_{\nu} = |J_c - S_{\nu}|}^{J_c + S_{\nu}} T_l(\epsilon_{\nu}) d\epsilon_{\nu},$  (3)

where  $T_i(\epsilon)$  is the transmission coefficient for particle *v* with orbital angular momentum *lv* and channel energy  $\epsilon_{\nu}$ . In a Fermi-gas model, the level density  $\Omega(E, J)$  is<sup>3</sup>

$$
\Omega(E,J) = \frac{(2J+1)\omega(E)}{\pi^{1/2}(2\sigma^2)^{3/2}} \exp[-J(J+1)/2\sigma^2] \tag{4}
$$

19 T. D. Thomas, Nucl. Phys. 53, 558 (1964); 53, 577 (1964).

where  $\omega(E)$  is the total density of states. We obtain  $\omega(E)$  from an expression derived for a diffuse nuclear well<sup>20</sup>

$$
\omega(E) = \frac{a^{1/4}}{2\pi E^{3/4}} \exp[2(aE)^{1/2} + (\beta/3)(E/a)^{3/2}].
$$
 (5)

The diffuseness of the well is contained in the quantity  $\beta$ ; its value is usually about unity. The parameter  $\alpha$  is proportional to the spacing of single-particle states at the Fermi level and is given  $by<sup>20</sup>$ 

$$
a = 2\left(\pi/3\right)^{4/3} \mu r_0^2 A \, h^{-2},\tag{6}
$$

where  $\Lambda$  is the mass number,  $\mu$  is the effective nucleon mass, and  $r_0$  is the nuclear radius parameter.

The spin cutoff parameter  $\sigma$  in Eq. (4) is formally related to the effective moment of inertia  $\mathfrak{g}$  by<sup>3</sup>

$$
\sigma^2 = \frac{gT}{\hbar^2},\tag{7}
$$

where  $T$  is the "nuclear temperature" defined by  $1/T = d \ln \omega(E)/dE$ . The nuclear temperature is then approximately

$$
1/T \approx (a/E)^{1/2} - 3/4E.
$$
 (8)

Thus the probability for population of a state  $(E_f, J_f)$  from a state  $(E_c, J_c)$  by emission of a particle  $\nu$  of energy  $\epsilon_{\nu}$  must be determined from emission probabilities which are summed over the spin distribution of the product nucleus and weighted according to the spin distribution of the compound nucleus. We obtain

$$
P_{\nu}(E_c, J_c, \epsilon_{\nu}, E_f, J_f) d\epsilon_{\nu}
$$
  
=  $[N(J_c, \nu)]^{-1} \Omega(E_f, J_f) P_c(E_c, J_c) \sum_{S_{\nu}} \sum_{l_{\nu}} T_l(\epsilon_{\nu}) d\epsilon_{\nu}.$  (9)

Here  $N(J_c, v)$  is a normalizing factor. If the rate  $R_v d\epsilon_v$  $[Eq. (3)]$  is integrated over energy, the total rate to a particular final state of spin  $J_f$  is obtained. If this is now summed over all spins  $J_f$  and the kinds of particles *v* which a compound state can emit, then we account for all of the original compound nuclei. Hence  $N(J_c, \nu)$ is given by

$$
N(J_c,\nu) = \sum_{\nu} \sum_{J_f} \int_{E_f} R_{\nu}(E_c,J_c,\epsilon_{\nu},E_f,J_f) d\epsilon_{\nu}.
$$
 (10)

The calculation is repeated if the residual nucleus of interest requires the evaporation of more than one particle. After the last particle has been emitted, the residual nucleus of excitation energy *Ef* will de-excite by emitting one or more  $\gamma$  rays. The relative probability of decay from a state  $(E_f, J_f)$  to a final state of excitation energy  $E_p$  and spin  $J_p$  by emission of a  $\gamma$  ray of energy  $\epsilon$ <sup>*y*</sup> is assumed to be equal to the product of the density of final levels  $(E_p, J_p)$  and a term related to vector coupling rules. Thus, the total normalized probability of

<sup>20</sup> D. B. Beard and A. McLellan, Phys. Rev. **131,** 2664 (1963).

 $\gamma$  emission leading to  $(E_p, J_p)$  is<sup>21</sup>

$$
P_{\gamma}(E_p, J_p)d\epsilon_{\gamma} = \sum_{J_f}
$$
  

$$
\frac{\epsilon_{\gamma}^{2L+1}\Omega(E_p, J_p)\delta(J_f - J_p - L)P_{\nu}(E_c, J_c, \epsilon_{\nu}, E_f, J_f)}{\sum_{J_p} \int_{E_p} \epsilon_{\gamma}^{2L+1}\Omega(E_p, J_p)d\epsilon_{\gamma}} d\epsilon_{\gamma},
$$
 (11)

where  $\delta(J_f-J_p-L)$  is a delta function which conserves angular momentum, and *L* is the multipolarity of the electromagnetic transition.

The last  $\gamma$  ray to be emitted is assumed to lead the excited nucleus to one of the two isomeric states depending upon which transition has the smaller spin change. The number of  $\gamma$  rays in the cascade is calculated from the expression $21,22$ 

$$
N(E_f) = (aE_f)^{1/2}/(L+1). \tag{12}
$$

## B. Modifications of the Model

Calculations cannot readily be done according to the equations in the previous section. The integrations over energy in Eq. (3) and (10) cannot be carried out in closed form.

Vandenbosch and Huizenga<sup>2</sup> made the simplifying assumption that evaporated particles were emitted with a kinetic energy corresponding to the mean value as given by evaporation theory. We adopt the same practice. The kinetic-energy spectrum was generated for each kind of evaporating particle from evaporation theory, which uses an energy-dependent Coulomb barrier corrected for penetration.<sup>23</sup> The mean kinetic energy  $\bar{\epsilon}_v$  was obtained by averaging over only that part of the spectrum which would lead to the reaction of interest. When an  $(\alpha, n)$  calculation, for example, was being carried out for compound nuclei which have excitation energy sufficient for multiple-particle evaporation, events leading to  $(\alpha, 2n)$ ,  $(\alpha, n\rho)$ ,  $(\alpha, n\alpha)$ , etc. were excluded. The quantitative criterion used was that if the excitation energy  $E_f$  of the residual nucleus after neutron emission was greater than  $B_i+V_i+2$  MeV, particle *i* was emitted, and the low-energy part of the spectrum leading to such *Ef* values was excluded in the averaging process. Here  $B_i$  and  $V_i$  are the separation energy and Coulomb barrier of particle *i.* The quantity 2 MeV is added to take into account the possible competition between  $\gamma$ -ray de-excitation and particle emission.<sup>24</sup> The calculations are not sensitive to its choice between 0.5 and 3 MeV.

A relatively small error appears to be introduced in the distribution of  $J_f$  by using average evaporation

energies.<sup>11,25</sup> However, this assumption could lead to considerable errors in the calculation of emission widths,

We now rewrite Eqs. (9) and (10), dropping *Ec* and  $E_f$  from the arguments since all compound nuclei at a particular bombarding energy have the same excitation energy and  $\bar{\epsilon}$  implies an average excitation energy  $E_f$ . Thus we obtain as a working equation

$$
P_{\nu}^{(1)}(J_f) = \sum_{J_c} \frac{\Omega_{\nu}(\bar{E}_f, J_f) P_c(J_c, E_c) \sum_{S_{\nu}} \sum_{l_{\nu}} T_l(\tilde{\epsilon}_{\nu})}{\sum_{J_c} \sum_{J_f} \sum_{J_f} \Omega_{\nu}(\bar{E}_f, J_f) \sum_{S_{\nu}} \sum_{l_{\nu}} T_l(\tilde{\epsilon}_{\nu})}.
$$
 (13)

Equation (13) is used to calculate the total probability for the compound nucleus to decay by particle emission to a final state of average excitation energy  $\bar{E}_f$  and spin  $J_f$ . For multiple-particle evaporation processes, the distribution  $P_{\nu}^{(1)}(J_f)$  obtained after the emission of the first particle is used in place of  $P_c(J_c,E_c)$  to generate another distribution  $P_{\nu}^{(2)}(J_f)$  for the second evaporation, This is continued for as many steps as necessary.

After the last particle-evaporation step, the distribution  $P_{\nu}^{(n)}(J_f)$  is used with Eq. (11) to calculate the distribution  $P_{\gamma}(\bar{E}_p, J_p)$  following  $\gamma$ -ray emission, as described by Huizenga and Vandenbosch.<sup>26</sup> The calculation is repeated for each of  $N(\bar{E}_f)$  – 1  $\gamma$  rays, and the last  $\gamma$  ray is assumed to populate the isomeric state requiring the smallest spin change. Residual nuclei of spin equidistant between those of the product isomers are divided equally between the two isomeric states.

Several workers<sup>13,18,25,27</sup> have recently commented on the question of multipolarity of the  $\gamma$  rays in the cascade. Quadrupole and perhaps higher order  $\gamma$  rays probably originate from the high-spin end of the distribution; these  $\gamma$  rays lead to states which decay ultimately to the isomer of higher spin in any case. This suggests that the part of the distribution which may be divided between the two isomers is likely to de-excite by dipole radiation. Hence we assume only dipole radiation in the cascade.

#### C. Choices of Parameters

Numerical results depend on the values chosen for the various parameters in the level density  $\Omega(E, J)$ . We discuss these next.

# 1. Spin-dependent Part of  $\Omega(E, J)$ .

In Eq. (4)  $\sigma^2$  is the mean-square deviation of the projection of the nuclear angular momentum on a fixed axis and is given by<sup>3</sup>

$$
\sigma^2 = k \langle m^2 \rangle \tag{14}
$$

<sup>&</sup>lt;sup>21</sup> V. M. Strutinsky, L. V. Groshev, and M. K. Akimova, Nucl.<br>
Phys. 16, 657 (1960).<br>
<sup>22</sup> Equation (12) was derived for the case  $n = 0$  in the expression<br>  $\omega(E) = \text{const.} E^x \exp(2(E))^{1/2}$ . For  $n = -\frac{3}{4}$ ,  $N(E_t)$  changes on

<sup>&</sup>lt;sup>24</sup> J. R. Grover, Phys. Rev. 127, 2142 (1962).

<sup>25</sup> N. D. Dudey, Ph.D. dissertation, Clark University, 1964 (unpublished).

*<sup>™</sup>* J. R. Huizenga and R. Vandenbosch, Phys. Rev. **120,** 1305

<sup>(1960).</sup>  27 H. Warhanek and R. Vandenbosch, J. Inorg. Nucl. Chem. 26, 669 (1964).

where *k* is the number of excited nucleons and *m* is the projection of the angular momentum of a single nucleon on a fixed axis. The largest value  $J_0$  for which a Gaussian form of the level density is expected to hold occurs when all nucleons contributing to the spin have a maximum projection  $\langle m^2 \rangle^{1/2}$  on the fixed axis. Then

$$
J_0 = k \langle m^2 \rangle^{1/2} \approx (\sigma^2 \hbar^2 / \pi) (6a/3)^{1/2} \tag{15}
$$

for a Fermi gas.<sup>3</sup> The maximum spin *Jm* which the nucleons can have is *km;* all contributing angularmomentum vectors are parallel. If the probability  $\rho(m)$ that a nucleon will have a particular *m* value is independent of *m,* then

$$
\langle m^2 \rangle = \int_0^{m_0} \rho(m) m^2 dm / \int_0^{m_0} \rho(m) dm = m_0^2 / 3, \quad (16)
$$

where *mo* is the maximum value of *m. 2S* This implies that

$$
J_m/J_0 = 3^{1/2}.\tag{17}
$$

The preceding analysis indicates that for spins above *Jo* the number of levels is expected to be less than that predicted by Eq. (4) and that above  $J_m$  the number of levels should be zero. Grover<sup>24</sup> has also estimated the maximum value of angular momentum for which levels exist at a particular excitation energy. It is of interest to note that his value is approximately  $3^{1/2}J_0$ .

Calculations have been made based on three assumptions regarding  $\Omega(E, J)$ .

Method 1: For 
$$
J \leq J_0
$$
,  $\Omega(E, J)$  given by Eq. (4); for  $J > J_0$ ,  $\Omega(E, J) = 0$ .

*Method 2:* For  $J \leq J_m$ ,  $\Omega(E, J)$  given by Eq. (4); for  $J>J_m$ ,  $\Omega(E,J) = 0$ .

*Method 3:* For all *J*,  $\Omega(E, J)$  given by Eq. (4).

Method 1 underestimates the number of available levels, while methods 2 and 3 overestimate the number.

In the VH formalism, method 3 was used; for systems involving only relatively low  $J$  this may cause no difficulty. In the cases we have studied involving multiple-neutron evaporation, it is only the final product nucleus in which  $J_0$  or  $J_m$  is less than the maximum  $J$  value in the spin distribution.

#### *2. The Moment of Inertia*

One of our fundamental motivations for studying isomeric ratios was to obtain information about the parameter  $\sigma$ , which through Eq. (7) is related to the effective nuclear moment of inertia  $\mathcal{I}$ . Generally  $\sigma$  is left as a free parameter in the calculation. The value of *6* so obtained is then compared to the rigid-body value  $g_R$  which is calculated for a rigid sphere of radius  $R = 1.2 A^{1/3}$  F.

In order to determine  $\vartheta$  from  $\sigma$ , it is necessary to know

the nuclear temperature. For all excitation energies above about 10 MeV we have calculated the nuclear temperature from Eq. (8). It is expected on theoretical grounds that the nuclear temperature is constant for excitation energies below about 10 MeV.4,29,30 In order to identify these temperatures explicitly we denote them by  $T_0$ . Their values have been taken from experiment when possible. The data of Bramblett and Bonner<sup>31</sup> give  $T_0 = 0.57$  MeV for the <sup>93</sup>Nb( $\alpha, xn$ ) reactions; from Sherr and Brady<sup>32</sup> we have taken  $T_0$  as 1.20 MeV for the  $55Mn(\alpha,n)$  reaction. By extrapolation we assume that  $T_0$  is 0.5 MeV for the  $^{136}Ba(\alpha,3n)$  reaction and 1.5 MeV for the  $^{41}K(\alpha,n)$  reaction.

It is theoretically expected<sup>3,4</sup> that  $\beta$  should be less than  $\mathfrak{I}_R$  at low excitation energies and with increasing energy approach the rigid-body value as an upper limit. Therefore, we have assumed that  $\mathfrak{s} = \mathfrak{s}_R$  for all product nuclei whose excitation energies are greater than 10 MeV, and allowed  $\mathcal{I}/\mathcal{I}_R$  to be an adjustable parameter for all nuclei whose excitation energy is less than 10 MeV. In general this means that the last step in a particle-evaporation chain and the  $\gamma$ -ray cascade occur at a constant nuclear temperature and with a value of  $\frac{g}{g_R}$  less than 1. The ratio  $\frac{g}{g_R}$  for each particular case is then adjusted to give agreement with the observed isomeric ratio. A value of  $\frac{g}{g_R}$  obtained in this way is an average value; it represents the average moment of inertia of those residual nuclei whose average excitation energy is  $\overline{E}_f$ . Furthermore, it averages over the entire  $\gamma$ -ray cascade.

## 3. Parameters in  $\omega(E)$

The parameter *a* in the state-density expression  $[Eq. (5)]$  is involved in many parts of the calculation. The average energy of evaporated particles, the number of cascade  $\gamma$  rays, the nuclear temperature, the value of  $J_0$  and of  $\omega(E)$  all depend on the choice of a. We have used the value  $a = \overline{A}/10.7$  MeV<sup>-1</sup>, as suggested by Beard and McLellan.<sup>20</sup> This corresponds  $\lceil \overline{Eq}$ . (6) to  $r_0$ = 1.35 F. One of us has examined in detail elsewhere<sup>25</sup> how a calculated isomeric ratio is affected by a change in *a.* To obtain the same isomeric ratio, one must use a moment of inertia  $\beta$  about 10% smaller for  $A/13.5$ than for *A/10.7.* The former *a* value corresponds to  $r_0 = 1.2$  F.

In a few calculations involving <sup>137</sup>Ce a pairing-energy term was included to compensate for the odd-even character of residual nuclides in evaporation steps. The energy available for evaporation was reduced by 2.4 MeV if the residual nucleus was even-even, 1.2 MeV if it was *odd-A,* and zero if it was odd-odd. In general a moment of inertia smaller by about  $10\%$  was required

<sup>&</sup>lt;sup>28</sup> We are indebted to T.D. Thomas for suggesting this approach.

<sup>&</sup>lt;sup>29</sup> L. E. H. Trainer and W. R. Dixon, Can. J. Phys. 34, 229 (1956).<br>
<sup>30</sup> T. Ericson, Nucl. Phys. 11, 481 (1959).<br>
<sup>31</sup> R. L. Bramblett and F. W. Bonner, Nucl. Phys. **20**, 395

<sup>(1960).</sup>  32 R. Sherr and F. P. Brady, Phys. Rev. 124, 1298 (1961).



to fit experimental isomeric ratios when the pairingenergy correction was included. In the calculations reported in this paper this correction has not been included.

#### *4. Transmission Coefficients*

In our calculations we have assumed neutrons, protons, and  $\alpha$  particles to be emitted in each evaporation step whenever energetically possible. Transmission coefficients were taken from Huizenga and Igo<sup>33</sup> *(a* particles); Auerbach<sup>34</sup> (protons); Feshbach, Shapiro, and Weisskopf<sup>35</sup> (neutrons and protons); and Moldauer<sup>36</sup> (neutrons). Optical-model coefficients were used when possible. For neutrons and protons, several sets of coefficients are available; the results of our calculations were not sensitive to their choice.

#### **D. Overall Calculation**

The computation of isomeric ratios was carried out on IBM 7094 computers at Massachusetts Institute of Technology and Columbia University. The general approach was to calculate the isomeric ratio for an arbitrarily chosen value of  $\frac{\mathcal{J}}{\mathcal{J}_R}$  in the residual nucleus. The calculation was then repeated for another choice of  $3/\mathfrak{g}_R$ . The program was not designed to search for the best value of  $\frac{s}{g_R}$  to fit experimental isomeric ratios.

The calculations were performed for the six  $(\alpha, xn)$ reactions of the previous paper at the bombarding energies listed in Table I. The energy range was restricted to that for which compound-nucleus formation seemed assured.

 $\overline{\phantom{m}}$ 

FIG. 1. Isomeric cross-section ratios (high-spin product to low-spin product) as a function of bombarding energy in the reactions (a) <sup>41</sup>K( $\alpha$ ,*n*)<sup>24</sup>Sc, (b) <sup>55</sup>Mn( $\alpha$ ,*n*)<sup>58</sup>Co, and (c) <sup>93</sup>Nb- $(\alpha,n)^{96}$ Tc as calculated by several methods. In method 1 (solid line) the maximum spin of a residual nucleus is taken to be  $J_0$  [Eq. (15)] and charged-particle competition is in-cluded. Methods 2 (dot-dash line) and 3 (dotted line) are similar to 1 except that in 2, the limiting spin is  $3^{1/2}$ . and in 3, no limit is set. Method 4 (short-dashed line) assumes the same limiting spin as in 1 but chargedparticle competition is neglected. Method 5 (long dashes) assumes *a*=4 in all evaporation steps. Experimental points are from Ref. 1. In (b) and (c), methods 2 and 3 gave essentially the same results. For clarity only one line is shown.

## **E. Spins of Isomers**

The spins of the isomers of the various residual nuclei are given in the preceding paper.<sup>1</sup> In the case of  $94g$ Tc the assignment is uncertain; we have taken it to be 6 + in our calculations.

## **III. RESULTS AND DISCUSSION**

#### **A. Results**

The general results of the calculations are summarized in Figs. 1 and 2. In both figures, the points represent experimental data from the previous paper

TABLE I. Effective moment of inertia  $\mathcal{I}$  of the final product nucleus, in units of the rigid-body moment *3R,* as deduced from statistical-model analysis of isomeric ratios by calculational method 1 (see text). The average excitation energies  $\bar{E}_f$  of the final product nucleus are those before  $\gamma$ -ray emission.

Reaction	$E_a$ (MeV)	$\mathfrak{g}/\mathfrak{g}_R$	$\bar{E}_f$ (MeV)
${}^{41}$ K $(\alpha,n)$ <sup>44</sup> Sc	10	0.58	3.9
	14	0.60	6.7
	18	0.90	10.0
	22	1.00	9.8
$55Mn(\alpha,n)$ <sup>58</sup> Co	10	0.50	5.7
	14	0.60	8.0
	18	0.90	10.1
${}^{33}\mathrm{Nb}(\alpha,n){}^{96}\mathrm{TC}$	14	0.50	5,5
	16	0.60	7.3
	18	0.80	9.2
	22	0.90	9.8
${}^{93}\text{Nb}(\alpha,2n){}^{95}\text{Tc}$	22	0.45	3.3
	26	0.50	6.5
	30	0.60	9.8
	34	0.65	10.0
$93Nb(\alpha,3n)94Tc$	34	0.40	2.3
	38	0.80	5.3
	42	0.90	8.6
$^{136}\text{Ba}(\alpha,3n)^{137}\text{Ce}$	34	0.40	3.4
	38	0.60	6.6
	42	0.90	9.8

<sup>33</sup> J. R. Huizenga and G. J. Igo, Argonne National Laboratory Report ANL-6373, 1961 (unpublished). 34 E. H. Auerbach, Brookhaven National Laboratory, ABACUS-2

program.<br><sup>35</sup> H. Feshbach, M. Shapiro, and V. P. Weisskopf, U. S. Atomic<br>Energy Commission Report NYO-3077, 1953 (unpublished).<br><sup>36</sup> P. A. Moldauer, Argonne National Laboratory Report<br>ANL-6323, 1961 (unpublished).

**2 6 34 34 38** 

**(MeV)** 



FIG. 2. Isomeric cross-section ratios (high-spin product to low-spin product) as a function of bombarding energy in the reactions (a) <sup>93</sup>Nb( $\alpha$ ,2*n*)–<br><sup>95</sup>Tc, (b) <sup>93</sup>Nb( $\alpha$ ,3*n*)<sup>93</sup>Tc, and (c) <sup>136</sup>Ba( $\alpha$ ,3*n*)<sup>137</sup>Ce as calculated by several methods. See caption of Fig. 1.

and the various lines have been obtained by calculation. Lines labeled 1, 2, and 3 were obtained by methods 1, 2, and 3. Line 4 was obtained as in method 1 except that charged-particle competition has been neglected. Line 5 results from the assumption that  $\sigma = 4$  in all evaporation steps (including charged-particle emission); no limiting spin was set.

 $\tilde{k}$ 

In Table I are listed the values of  $\frac{g}{g_R}$  which were assumed for residual nuclei when their average excitation energy  $\bar{E}_f$  was less than 10 MeV. As described earlier, we required  $\mathcal{I} = \mathcal{I}_R$  when  $\bar{E}_f > 10$  MeV. The particular  $\frac{s}{s_R}$  values in Table I provide a reasonable fit to experiment for method 1. The same value of  $\mathcal{I}/\mathcal{I}_R$  was used in methods 2, 3, and 4 to illustrate the dependence of a calculated isomeric ratio on the maximum spin allowed in a residual nucleus and on the evaporation of particles other than those leading to the residual nucleus of interest. In method 5 the value of  $\mathcal{I}/\mathcal{I}_R$  is fixed by the value assumed for  $\sigma$ . In general  $\mathcal{I}/\mathcal{I}_R$  is much less than 1 in all evaporation steps when  $\sigma$  is taken to be 4.

## **B. Competition**

In curve 4, competing evaporation paths were neglected (i.e., it was assumed that only neutrons were evaporated) but in all other respects the calculational method was the same as that of method 1. Competition cannot be neglected except at low bombarding energies. The isomeric ratios obtained without competition are generally larger. To obtain a better fit to experiment by method 4, one must use a smaller value of  $\frac{3}{4}$ .

Vandenbosch, Norman, and Haskin<sup>18</sup> have considered competition in the reaction  ${}^{87}Sr(d,2n)$  with 18-MeV deuterons. For this case, they find proton competition to be unimportant. This conclusion is not inconsistent with our results since our systems generally involve higher spin and higher excitation energies.

Higher excitation energies mean that more energy is available for overcoming the Coulomb barrier and hence charged-particle emission becomes more favorable. The increased angular momentum of the compound nucleus may also be involved. This point is examined in the following manner.

**42 34 38 42** 

We define  $F(\nu)$  to be the decay fraction, that is, the fraction of all compound states which de-excite by the emission of particle *v.* A slight modification of our computer program permits us to calculate the partial decay fraction  $F(\nu, J)$ , that is,  $F(\nu)$  for each value of J of the emitting nucleus. In Fig. 3 we have plotted  $F(\nu, J)$  for the excited nuclei  $\rm{^{97}Tc}$  and  $\rm{^{95}Tc}$  as produced in the reaction <sup>93</sup>Nb+42-MeV *a.* The data are normalized such that the sum of the three partial emission fractions for each value of  $J$  is unity. These two figures clearly show that  $F(\nu, J)$  and consequently the normalization constant  $N(J,\nu)$  are functions of J. Thomas has arrived at a similar conclusion.<sup>19</sup> Furthermore, the dependence of  $F(\nu, J)$  on *J* is greater when  $\mathcal{I}$  is smaller. Although the relative magnitude of  $F(\nu, J)$  may be in error because we have used average evaporation energies rather than integrating over the energy spectra, the change as a function of *J* and  $\frac{s}{g_R}$  should be qualitatively correct.

#### **C. The Form of the Level-Density Expression**

Curves 1, 2, and 3 illustrate the consequences of invoking a limiting-spin principle. The difference in calculated isomeric ratios between methods 2 and 3 is relatively small compared to the difference between either of them and method 1. Methods 2 and 3 typically result in much higher ratios for a given value of  $\frac{g}{g_R}$ . To obtain a fit to experiment by these methods, smaller values of *6* are required, as has been observed by others.<sup>37</sup>

As mentioned previously, a sharp cutoff in  $\Omega(E, J)$  at  $J = J_0$  underestimates the number of levels available, while a cutoff at  $J = J_m$  overestimates it. A more correct functional form of  $\Omega(E, J)$  might be a smooth falloff

<sup>37</sup> D. Bodansky, Ann. Rev. Nucl. Sci. 12, 79 (1962).



FIG. 3. Partial decay fraction  $F(\nu, J)$ as a function of the spin  $J$  of the emitting nucleus. The sum of  $F(n, J)$ ,  $F(p, J)$ , and  $F(\alpha, J)$  is unity for each *J* value. In (a) the emitting nucleus is <sup>97</sup>Tc excited to 44 MeV and *\$/\$<sup>R</sup>* of residual nuclei is unity. In (b) the emitting nucleus is <sup>95</sup>Tc excited to 20 MeV and *\$/\$R* of <sup>94</sup>Tc is 0.9. Calculations of  $F(\nu, J)$  have been made under the assumption that the limiting spin is  $J_0 = 9h$  (solid line) and  $3^{1/2}J$ *= 16h* (dashed line). The enhanced contribution of  $\alpha$ -particle emission is clearly evident for large  $J$  values.

from  $J=J_0$  to zero at  $J=J_m$ . The calculated isomeric ratios would then be somewhat larger than the values given as curve 1. To fit the experimental data, one would have to select somewhat smaller values of *\$/\$R*  than those listed in Table I. An approximation to this is provided by choosing a sharp cutoff for a  $J$  value intermediate between  $J_0$  and  $J_m$ . For example, placing the cutoff half way between  $J_0$  and  $\bar{J}_m$  [i.e., at  $J=\frac{1}{2}J_0(1+3^{1/2})$ , we find a reasonable fit to experimental isomeric ratios in the reaction  $^{136}Ba(\alpha,3n)$  for  $\mathcal{A}/\mathcal{A}_R$  values of 0.3, 0.4, and 0.7 for bombarding energies of 34, 38, and 42 MeV, respectively. In the case of  $^{93}Nb(\alpha,n)$  at 14, 18, and 22 MeV,  $\frac{g}{g_R}$  values are 0.4, 0.6, and 0.7, respectively. These values are approximately  $75\%$  of those listed in Table I.

Some analyses<sup>10,14,15</sup> of isomeric ratios have been made under the assumption that the spin cutoff parameter  $\sigma$  was 4 in all steps of particle evaporation chains and in the  $\gamma$ -ray cascade. There appears to be no theoretical justification for such a choice. If we repeat calculation 3 with  $\sigma = 4$  in all evaporation steps, we obtain the isomeric ratios plotted as curve 5. The agreement with experiment is generally poor.

Recent analyses<sup>11,17</sup> of isomeric ratios have invoked a qualitative notion called "spin fractionation" which resembles in part the principles considered here. Consider a compound system in which two reactions such as  $(\alpha, n)$  and  $(\alpha, 2n)$  can occur with comparable cross section. The excitation energy of the compound nucleus must then be such that the  $(\alpha, n)$  residual nuclei are formed with relatively high excitation energy on the average while the average excitation energy of  $(\alpha, 2n)$ residual nuclei is generally much smaller. Since typically there are not many low-lying levels of high spin, this suggests that compound nuclei of high spin may preferentially lead to the  $(\alpha, n)$  reaction and those of low spin to the  $(\alpha, 2n)$  reaction, thus fractionating the compoundnuclear spin distribution.

Our calculations take this notion into account quantitatively, not only with respect to the  $(\alpha, 2n)$  reaction competing with the  $(\alpha, n)$  reaction, but with respect to  $(\alpha, \beta)$  and  $(\alpha, \alpha')$  as well. A limiting spin in the residual nucleus also means that emitting nuclei of very high spin will probably decay by other than neutron emission, as indicated in Fig. 3.

## D. Relationship between  $\mathcal{I}$  and  $\overline{E}_f$

In Fig. 4 we have made a crude attempt to relate *6*  and  $\bar{E}_f$  in the various product nuclei. The values of  $3/\mathfrak{g}_R$  shown are from Table I and correspond to method 1. The optimum method with a choice of parameters which give the "best" fit to experiment would lead to  $\frac{s}{s_R}$  values about  $20\%$  smaller than those in Table I. It should be emphasized that  $s/s_R$  is taken to be unity when  $\bar{E}_f > 10$  MeV, as in intermediate nuclei in an evaporation chain.

The values of  $\frac{3}{4}$  obtained from isomeric ratios are model-dependent. Errors in the value of *T,* particularly  $T_0$ , may be as large as  $25\%$ . This will result in equivalent errors in the values of  $\frac{g}{g_R}$ . Not only is it difficult to find agreement between measurements of nuclear temperatures for the same residual nucleus, but the dependence of temperature upon residual energy has not been experimentally established. If the nuclear temperature increases with excitation energy for energies less than 10 MeV, then the curve implied in Fig. 4 would increase less rapidly with increasing *Ef.* 

Considerations of this sort do not alter the general statement that for the six reactions studied,  $\beta$  increases with  $\bar{E}_f$ ; it has a very similar dependence upon energy for several residual nuclei; and it approaches the rigidbody value in the range 10-15 MeV. This trend is in excellent agreement with Lang and LeCouteur<sup>5</sup> who have made a detailed study of pairing-energy effects in excited nuclei. They conclude that for excitation energies near the neutron binding energy,  $\frac{g}{g_R} \approx 0.5$ ; it decreases for lower excitation energies and exhibits a slow trend toward unity at higher energies. In addition Lang<sup>4</sup> has compared the pairing-energy model to a superconductor model. He finds both models predict



FIG. 4. Dependence of *\$/\$R* in final product nucleus on average excitation energy *E<sup>f</sup> .* These *g/gR* values provide a reasonable fit to experimental data by method 1 (see text). The "best" fit by the optimum method would require  $s/s_R$  values about 20% smaller.<br>The points plotted are <sup>44</sup>Sc (open circles), <sup>58</sup>Co (filled circles), <sup>99</sup>Tc (filled tircles), <sup>99</sup>Tc (filled squares), <sup>94</sup>Tc (filled squares), and <sup>137</sup>Ce (o

 $\frac{s}{s}$  1 for nuclei at low energies. However, the superconductor model predicts a larger value of  $\frac{\delta}{\delta R}$  at the neutron binding energy and gives a return to the rigidbody value at much lower energies than the pairing model. The results shown in Fig. 4 are in good agreement with both of these models but unfortunately the accuracy of these calculations does not permit a choice.

## **E. Isomeric Ratios**

Because our model leads to results which agree well with theoretical expectations, we feel that many of the correct features have been incorporated. On this basis we can account at least semiquantitatively for the energy dependence of isomeric ratios. The ratio  $(\sigma_H/\sigma_L)$ of high-spin product to low-spin product in an *(a,xn)*  reaction, and very probably any  $\alpha$ -particle-induced reaction, invariably increases with bombarding energy at first because higher *I* waves are admitted and the average spin  $\langle J_c \rangle$  of the compound nucleus increases. The isomeric ratio in residual nuclei formed by nucleon evaporation processes cannot increase indefinitely, however, even if  $\langle J_e \rangle$  continues to do so, because of the effect of competing reactions. In Fig. 3 it is shown that the relative probability of emitting an alpha particle, as compared to that for nucleon emission, increases rapidly as the spin of the emitting nucleus increases. This effect is most important when the excitation energy is large compared to the Coulomb barrier energy.

Increasing  $\langle J_c \rangle$  does not then lead to correspondingly higher  $\langle J_f \rangle$  in the residual nuclei resulting from nucleon evaporation. The isomeric-ratio curve of  $(\alpha, xn)$  products would be expected to level off. With increased excitation energy, all emitted particles have higher kinetic energy and can carry off more angular momentum, and the Coulomb barrier is less restrictive of the emission of heavier particles; hence even a decrease in the isomeric ratio could occur. In the reaction  $^{93}Nb(\alpha,2n)$  such a change in slope has been observed<sup>1</sup>; this can be accounted for without invoking a direct-interaction process.

These arguments can be applied to both nucleoninduced reactions and heavy-ion reactions. Compound nuclei formed at a particular excitation energy by incident nucleons generally have lower  $\langle J_c \rangle$  than those produced by  $\alpha$  particles. Evaporation of particles other than nucleons will then be less important in nucleoninduced reactions. In these reactions the slow increase in  $\langle J_e \rangle$  with increasing bombarding energy will be reflected by a corresponding increase in the isomeric ratio which continues to higher excitation energies than in the  $\alpha$ -particle-induced case before leveling off. For these reactions the intrinsic spin of the target nucleus may be a much more important factor than in  $\alpha$ -particle-induced reactions.

Heavy-ion reactions should show a different effect. Heavy ions typically deposit much more angular momentum than do  $\alpha$  particles, but the states of very high  $J_c$  probably decay by  $\alpha$ -particle emission or even  $\gamma$ -ray emission if states below  $J_m$  cannot readily be reached by nucleon emission. Thus the high input angular momentum is not necessarily found in the residual nuclei which result from neutron evaporation; these are likely to be formed from states of lower *Jc.*  This is in agreement with the unusually large yield of evaporated  $\alpha$  particles observed in heavy-ion reactions.<sup>38</sup>

## **IV. CONCLUSIONS**

This analysis has shown that two factors not included in the VH formalism are of importance in determining isomeric ratios. First, competing reaction paths should be included, since their dependence upon angular momentum affects spin distributions in residual nuclei. Second, while these calculations support the "Gaussian" form of the level-density expression [i.e., Eq. (4)], a limiting  $J$  value is necessary. With the inclusion of these factors, and the requirement that nuclear temperatures be constant for low excitation energies, the effective moments of inertia are consistent with the rigid-body value for nuclei excited above 10 to 15 MeV. For lower excitation energies the moment of inertia appears to be a function of the excitation energy.

The analysis reported here suffers from several assumptions. The most important possibly is the averaging which is necessary to obtain the mean kinetic energy of evaporated particles. Other weaknesses involve the need for a better treatment of the  $\gamma$ -ray cascade step and an accurate value for the nuclear temperature. Nonetheless, the agreement between *\$/\$<sup>R</sup>*

<sup>38</sup>  **A. R. Quinton** and **H. C. Britt, Bull.** Am. Phys. Soc. **6,** 286 **(1961)**; W. **J.** Knox, A. R. Quinton, and C. E. Anderson, Phys. **Rev. 120, 2120 (1960).** 

and  $\bar{E}_f$  and the consistency of  $\frac{s}{g_R}$  as a function of  $\bar{E}_f$  for six reactions on four widely varying nuclei support the validity of this method for interpreting isomeric-ratio data. This agreement suggests the possibility of using experimental isomeric ratios to obtain quantitative information about the region of applicability of the Gaussian form of the level-density expression, the dependence of the nuclear level density upon angular momentum, the dependence of  $\mathfrak s$  upon excitation energy, and the spin distribution in nuclei formed following particle emission.

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# Investigation of the  $T=\frac{3}{2}$  State at 16.97 MeV in Be<sup>9</sup>

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A resonance in the Li<sup>7</sup>+d reaction at a deuteron bombarding energy of  $361 \pm 2$  keV, corresponding to a level in Be<sup>9</sup> at 16.97 MeV, has been investigated. Total cross sections for  $Li^7(d,p_0) Li^8$  were measured by detecting induced Li<sup>8</sup> activity for deuteron energies between 340 and 780 keV. In agreement with earlier work of Woods and Wilkinson, a resonance in this reaction was observed on a background rapidly decreasing with decreasing deuteron energy. This resonance and the surrounding region from 0.1 to 1.1 MeV were also studied with the  $Li^7(d,\gamma)$  Be<sup>9</sup> reaction by measuring the yield of the gamma-ray transitions to the lower states of Be<sup>9</sup> with a NaI crystal. The measured width of the level was less than 600 eV in the laboratory system. The angular distribution of the on-resonance gamma rays to the ground state was isotropic to within 7%. Branching ratios for the gamma rays to several states in Be<sup>9</sup> were measured with a NaI crystal, a scintillation pair spectrometer, and a Nal crystal surrounded by an anticoincidence shield. The relative intensities for the gamma-ray transitions to the ground state and to the states at 1.70, 2.43, 3.04, and 4.74 MeV were found to be 100,  $8.5 \pm 4.3$ ,  $10.6 \pm 5.3$ ,  $\leq 4.5$ , and  $9.6 \pm 4.8$ , respectively. The 15.3-MeV gamma-ray transition indicates the existence of a level in the usual sense at 1.70 MeV in Be<sup>9</sup> . From the gamma-ray measurements below resonance at 300 keV an upper limit of  $1.6 \times 10^{-30}$  cm<sup>2</sup> was found for the direct  $(d, \gamma)$ cross section to low-energy states in Be<sup>9</sup>. For the  $\text{Li}^7(d,n)$  reaction, the yield of neutrons with energies above 10 MeV showed a resonance at the same energy. From neutron time-of-flight measurements relative to coincident gamma rays, neutron groups were seen corresponding to breakup of levels in Be<sup>9</sup> at 2.43 and 4.74 MeV. The  $\dot{\text{Li}}^7(d,d)$   $\text{Li}^7$  and  $\text{Li}^7(d,\alpha)$  He<sup>5</sup> reactions did not show observable resonance effects. From the above results, the reduced widths for neutron and alpha-particle emission are found to be considerably smaller than the corresponding width for protons, thus supporting the hypothesis that the state in Be $9$  at 16.97 MeV has an isobaric spin of  $\frac{3}{2}$ .

## **I. INTRODUCTION**

A VERY narrow resonance in the  $Li^7(d,p)Li^8$  reaction at a deuteron bombarding energy of about 360 keV was first observed by Woods and Wilkinson<sup>1</sup> while studying low-Q stripping reactions; no analysis of the level was presented at that time. Confirming results and additional information on this resonance were obtained by Imhof, Chase, and Fossan.<sup>2</sup> Recent interest in this state has resulted from the observations by Lauritsen, Lynch, and Griffith<sup>3</sup> and by Griffith<sup>4</sup> of a

narrow  $\left( < 5 \text{ keV} \right)$  level in Be<sup>9</sup> at  $14.392 \pm 0.005$  and by Middleton and Pullen<sup>5</sup> of the first excited state of Li<sup>9</sup> at  $2.691 \pm 0.005$  MeV. These latter results, coupled with the early observations by Woods and Wilkinson<sup>1</sup> and by Imhof, Chase, and Fossan<sup>2</sup> have enabled Woods and Wilkinson<sup>6</sup> to make a probable identification of this level in Be<sup>9</sup> at 16.97 MeV as the second  $T=\frac{3}{2}$  state which corresponds to the first excited state of  $\tilde{Li}^9$ . The  $14.392$ -MeV level in Be<sup>9</sup> which is analogous to the ground state of Li<sup>9</sup> would be the first  $T=\frac{3}{2}$  state.

Further analysis of the 16.97-MeV level in Be<sup>9</sup> has been limited by the lack of complete experimental information. If the level is  $T=\frac{3}{2}$ , the neutron, deuteron, and alpha channels would be strongly inhibited because

<sup>1</sup> D. H. Wilkinson, *Proceedings of the International Conference on Nuclear Structure, Kingston, 1960* (North-Holland Publishing

Company, Amsterdam, 1960), p. 42.<br>
<sup>2</sup> W. L. Imhof, L. F. Chase, and D. B. Fossan, Bull. Am. Phys.<br>Soc. 9, 391 (1964).<br>
<sup>2</sup> L. L. L. L. P. L. T. Phys. P. C. Criffith, Pull. Am. Phys. Soc.

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<sup>&</sup>lt;sup>5</sup> R. Middleton and D. J. Pullen, Nucl. Phys. 51, 50 (1964).

<sup>6</sup> J. B. Woods and D. H. Wilkinson, Nucl. Phys. 61, 661 (1965).