# Selective excitations in actinide nuclei via alpha pickup

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The  $\alpha$ -cluster pickup reaction  $(d, {}^{6}\text{Li})$  has been studied at  $E_d = 55$  MeV on targets of  ${}^{232}\text{Th}$  and  ${}^{238}\text{U}$ . Members of the ground-state rotational bands in  ${}^{228}\text{Ra}$  and  ${}^{234}\text{Th}$  are excited and absolute reduced  $\alpha$  widths obtained from finite-range distorted-wave analysis are in good agreement with values deduced from  $\alpha$  decay. In addition three excited groups of states are very strongly populated in both nuclei with spectroscopic strength per group comparable with those of the respective ground state bands. These groups are apparently excited rotational bands with band heads at  $E_x = 700 \pm 40$ ,  $1070 \pm 60$ , and  $1390 \pm 60$  keV in  ${}^{228}\text{Ra}$  and  $E_x = 810 \pm 30$ ,  $1150 \pm 40$ , and  $1470 \pm 40$  keV in  ${}^{234}\text{Th}$ . The selective and strong excitation in this particular multi-nucleon transfer reaction of several excited bands is not predicted by existing theoretical models. An attempt has been made to describe the systematics of excited 0<sup>+</sup> states in the actinide region with the interacting boson model. Excitation energies are reasonably well described but intruder states are present and transfer strengths are not reproduced properly. The observation of strong  $\alpha$ -cluster pickup to excited rotational bands suggests coherent contributions from both neutron and proton pair excitations which can lead to strong four-body correlations and/or to new types of collective excitations which favor quartet structure. It is found that about 25% of the nuclear charge (matter) at  $r \simeq 10.6$  fm must be associated with  $\alpha$  particles. This high  $\alpha$ -clustering probability indicates  $\alpha$ -particle condensation in low-density nuclear matter.

NUCLEAR REACTIONS <sup>232</sup>Th, <sup>238</sup>U(d, <sup>6</sup>Li), E = 54.8 MeV; measured  $\sigma(\theta)$ ; DWBA analysis; <sup>228</sup>Ra, <sup>234</sup>Th deduced levels,  $S_{\alpha}$ ,  $\gamma_{\alpha}^{2}$  (10.5 fm),  $\alpha$ -clustering probabili-ties.

#### I. INTRODUCTION

Calculations<sup>1</sup> with simple shell-model configurations explain relative  $\alpha$ -spectroscopic effects in heavy nuclei very well, but absolute reduced  $\alpha$  widths deduced from  $\alpha$  decay (and  $\alpha$  transfer) are usually underpredicted by several orders of magnitude unless extensive configuration mixing is introduced.<sup>2</sup> Such mixing and the associated strong  $\alpha$ -particle clustering in the surface of heavy nuclei results from the nature of the interactions between nucleons which is respondible for two-proton two-neutron (and other) correlations.<sup>2, 3</sup>

Experimental absolute reduced  $\alpha$  widths for heavy nuclei have been deduced almost entirely from  $\alpha$ -decay data. Only in recent years have direct  $\alpha$ -transfer reactions such as  $(d, {}^{6}\text{Li})$  and  $({}^{16}\text{O}, {}^{12}\text{C})$  been performed<sup>4-12</sup> for nuclei with A> 100. Absolute reduced  $\alpha$  widths can be extracted from reaction analyses<sup>6-8, 11</sup> when the transfer proceeds predominantly by a direct one-step mechanism. Direct  $\alpha$  transfer extends the study of  $\alpha$ -like correlations to the region of stable nuclei. Moreover, spectroscopic information becomes available for excited states, whereas  $\alpha$ decay data are often limited to low excited states due to the strong energy dependence of the  $\alpha$ -particle penetrability. Multiparticle multihole and other more complicated excitations can hence be studied via  $\alpha$  transfer.

The  $(d, {}^{6}\text{Li})$  reaction is a particularly useful reaction since  $0^{+} \rightarrow 0^{+}$  transitions display characteristic diffractive angular distributions even for the heaviest nuclei. Although cross sections decrease strongly with target mass,<sup>4</sup> problems due to target contaminants are minimal because of the increasingly positive Q values.

Low excited  $0^+$  states in the actinide region are currently the subject of experimental and theoretical studies.<sup>13-29</sup> Such states have been observed<sup>13</sup> via (p,t) with typically 15% of the ground state cross section. No such transitions are usually

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observed<sup>15,16</sup> in (t,p) except for certain 0<sup>+</sup> states<sup>17,19</sup> in <sup>246</sup>Pm and <sup>248</sup>Cm which appear to have the characteristics of pairing vibration states due to a subshell closure at N = 152. Several of the excited 0<sup>+</sup> states are characterized by small  $\alpha$ decay hindrance factors<sup>30-32</sup> and large E0 matrix elements.<sup>33,34</sup> The observation of relatively strong two-neutron pickup is incompatible with the assumption of  $\beta$  vibrations as theoretical description of these states. This conclusion lead to the introduction of other models,  $^{21,29}$  particularly the concept of pairing isomers  $^{21,22,24-26}$  where independent pairing correlations for prolate and oblate orbitals are assumed. The observation in the present experiment of intense  $\alpha$ -pickup transitions to excited states in <sup>228</sup>Ra and <sup>234</sup>Th provides new insight into the intrinsic structures of these states and thus facilitates tests of theoretical models.

The experimental procedures and results are presented in Secs. II and III, and the analysis of the data with the distorted-wave Born approximation (DWBA) is described in Sec. IV. It is followed by discussions of reduced widths (Sec. V), the systematics in the actinide nuclei of excited  $0^+$ states in the framework of the interacting boson approximation (IBA) (Sec. VI), and  $\alpha$ -clustering probabilities (Sec. VII). A summary is presented in Sec. VIII.

## **II. EXPERIMENTAL PROCEDURES**

A 54.8 MeV deuteron beam from the Texas A & M cyclotron was used together with an Enge split-pole magnetic spectrometer ( $\Delta \Omega = 2.1 \text{ msr}$ ,  $\Delta \theta = 3^{\circ}$ ) to identify <sup>6</sup>Li particles from the (d, <sup>6</sup>Li) reaction on <sup>232</sup>Th and <sup>238</sup>U targets. The targets consisted of natural thorium and uranium of about 200  $\mu g/cm^2$  thickness on carbon backings. The target thicknesses were determined from forwardangle elastic scattering data and from the energy loss of 5.5 MeV  $\alpha$  particles. The reaction products were detected and identified in the focal plane of the spectrograph with a position-sensitive proportional counter backed by a silicon surface-barrier detector (5  $cm \times 1$  cm) partially biased to just stop <sup>6</sup>Li particles. Energy loss, total energy, and time-of-flight signals were used for particle identification. Elastic deuteron scattering and  ${}^{12}C(d, {}^{6}Li){}^{8}Be$  were used to calibrate the energy scale of the detector. The energy analyzed beam of >1.5  $\mu$ A on target made it necessary to use a water-cooled Faraday cup which introduced an uncertainty in the integrated current of  $\pm 10\%$ . The low cross sections required average exposures of 25 mC per spectrum. The energy resolution due to target thickness and position resolution was 80 keV full width at half maximum

(FWHM). The detection efficiency was  $75 \pm 10 \%$ due to the limited detector height. The combined uncertainties of target thickness and other effects yield absolute cross sections with estimated uncertainties of  $\pm 40\%$ . The measured spectra are essentially free of background despite a very high rate of incident  $\alpha$  particles and deuterons.

#### **III. EXPERIMENTAL RESULTS**

Energy spectra obtained from <sup>232</sup>Th(d, <sup>6</sup>Li)<sup>228</sup>Ra and <sup>238</sup>U(d, <sup>6</sup>Li)<sup>234</sup>Th at  $\theta_{1ab} = 6^{\circ}$  are displayed in Fig. 1. The spectra cover a range of about 1.6 MeV in excitation energy. Both spectra are characterized by transitions to four distinct groups of states, probably rotational bands. The estimated



FIG. 1. Energy spectra for <sup>6</sup>Li particles from the  $(d, {}^{6}Li)$  reaction. Band heads for various groups of levels are marked by arrows. Level schemes for the residual nuclei and neighboring isobars are from Refs. 43 and 44. Tentative spin assignments (Ref. 31) are given in parentheses and extrapolated energy levels are indicated by dashed lines.

| $\frac{232}{d\sigma} \operatorname{Th}(d, {}^{6}\operatorname{Li})^{228}\operatorname{Ra}$ |                                      |  |       |                                   |    | $^{238}$ U(d, <sup>6</sup> Li) <sup>234</sup> Th |  |       |                                   |  |  |
|--|--------------------------------------|--|-------|-----------------------------------|----|--|--|-------|-----------------------------------|--|--|
| J  | E <sub>x</sub> <sup>a</sup><br>(keV) | $\frac{d\Omega}{d\Omega}$ (6°) <sup>2</sup><br>(nb/sr) | Sα    | $\gamma_{\alpha}^{2}(s)^{c}$ (eV) | J  | E <sub>x</sub> <sup>a</sup><br>(keV)             | $\frac{d\Omega}{d\Omega}$ (6°) <sup>3</sup><br>(nb/sr) | Sα    | $\gamma_{\alpha}^{2}(s)^{c}$ (eV) |  |  |
| 0*   | 0                                    | $137 \pm 40$   | 0.019 | 59                                | 0* | 0  | $92 \pm 24$  | 0.017 | 44                                |  |  |
| 2*   | 64 <sup>d</sup>                      | $287 \pm 58$   | 0.042 | 120                               | 2* | 50   | $151 \pm 31$   | 0.027 | 68                                |  |  |
| 4*   | 205 <sup>d</sup>                     | $108 \pm 36$   | 0.012 | 29                                | 4* | 160  | $115 \pm 27$   | 0.013 | 26                                |  |  |
| 6*   | 409 <sup>đ</sup>                     | $59 \pm 26$  | 0.008 | 14                                | 6* | (331)  | $44 \pm 17$  | 0.004 | 6                                 |  |  |
|  |                                      |  |       |                                   | 8* | (555)  | $69 \pm 21$  | 0.007 | 7                                 |  |  |
| G1   | 0                                    | $530 \pm 76$   | 100°  | 100 °                             | G1 | 9  | $359 \pm 48$   | 100 ° | 100 <sup>e</sup>                  |  |  |
| G2   | $700 \pm 40$                         | $799 \pm 97$   | 158   | 139                               | G2 | $810 \pm 30$                                     | $553 \pm 59$   | 125   | 108                               |  |  |
| G3   | $1070 \pm 60$                        | $282 \pm 58$   | 57    | 47                                | G3 | $1150 \pm 40$                                    | $342 \pm 46$   | 83    | 67                                |  |  |
| G4   | $1390 \pm 60$                        | $223 \pm 51$   | 47    | 36                                | G4 | $1470\ \pm 40$                                   | $309~\pm~44$   | 82    | 63                                |  |  |

TABLE I. Excitation energies, cross sections, spectroscopic factors, and reduced widths for ground state rotational bands and groups of excited states.

<sup>a</sup>Excitation energies from this work are given with uncertainties. The energies for the groups of levels represent band head energies.

<sup>b</sup>Relative uncertainties of c.m. cross sections are indicated. Absolute uncertainties are estimated as  $\pm 40\%$ . <sup>c</sup>Channel radius  $s=1.7A_{core}^{-1/3}$  fm.

<sup>d</sup>From Ref. 31.

<sup>e</sup>Relative values normalized to group G1.

excitation energies of the respective band heads labeled G1, G2, etc., are indicated. The 0<sup>+</sup> and 2<sup>+</sup> members of the g.s. rotational bands are not fully resolved, but individual cross sections for these and the other members can be extracted reliably using standard peak fitting procedures.<sup>35</sup> The cross sections for the states with  $J^{T} \leq 4^{+}$  are a factor of about 2 greater than for higher spin states. The 2<sup>+</sup> state is the most intense. No attempts have been made to resolve the excited groups of levels. The excitation energies and cross sections for the ground state rotational bands and for the three respective excited groups are listed in Table I. The level schemes included in Fig. 1 will be discussed later.

Four-point angular distributions for transitions to the members of the g.s. rotational band and to the four groups of states in  $^{234}$ Th are displayed



FIG. 2. Experimental angular distributions for transitions to the members of the ground state rotational band in <sup>234</sup>Th and the four unresolved groups of levels indicated in Fig. 1 (filled circles). Cross sections for the corresponding transitions to states in <sup>228</sup>Ra at 6° (open circles) are also indicated. The curves are from finite-range DWBA calculations for <sup>238</sup>U(d, <sup>6</sup>Li)<sup>234</sup>Th described in the text.

in Fig. 2. Cross sections for the corresponding transitions to <sup>228</sup>Ra at  $\theta_{1ab} = 6^{\circ}$  are also indicated. The curves displayed in the figure are from DWBA calculations as described in the next section.

#### **IV. DWBA ANALYSIS**

All data were subjected to a finite-range DWBA analysis utilizing the computer code DWUCK5.<sup>36</sup> The procedures and parameters are identical to those used in a previous comparison<sup>11</sup> between  $\alpha$ decay and  $\alpha$ -cluster pickup [parameter sets P(d), D(<sup>6</sup>Li), GV( $B = A + \alpha$ ), and W(<sup>6</sup>Li =  $d + \alpha$ ) from Table II of Ref. 11]. Macroscopic  $\alpha$ -cluster wave functions with eleven radial nodes bound in Woods-Saxon plus Coulomb potential wells were used to calculate the DWBA form factors. The potential parameters ( $r_0 = 1.35$  fm, a = 0.65 fm) for the  $\alpha$ cluster bound state were chosen in accordance with the known A dependence of nuclear radii as discussed elsewhere.<sup>11</sup> The relative motion wave function between the deuteron and  $\alpha$  particle in the ground state of <sup>6</sup>Li was generated by a Woods-Saxon potential with a hard core. This potential<sup>37</sup> yields the correct  $\alpha + d$  separation energy and reproduces low-energy  ${}^{3}S_{1}$  scattering phase shifts, the rms charge radius of the ground state of <sup>6</sup>Li, as well as absolute  $(d, {}^{6}Li)$  cross sections. Other <sup>6</sup>Li cluster wave functions are discussed in Ref. 11.

The <sup>6</sup>Li optical model parameter set D appears to be most appropriate and predicts correct absolute  $(d, {}^{6}Li)$  cross sections for medium heavy and heavy target nuclei. It is not fully satisfactory, though, as it was derived by extrapolating lower-mass data. Alternate <sup>6</sup>Li parameter sets based on 73, 99, and 154 MeV elastic scattering data have become available recently,<sup>38,39</sup> but were found to reproduce the measured<sup>40</sup> 50 MeV elastic scattering for <sup>208</sup>Pb as well as  $^{208}$ Pb(d, <sup>6</sup>Li)<sup>204</sup>Hg<sub>g.8</sub>. data at  $E_d = 54.8$  MeV (Ref. 9) very poorly. Elastic scattering data obtained at 88 MeV have also just been reported.<sup>41</sup> The need for unique global <sup>6</sup>Li optical parameters still exists.

Spectroscopic factors  $S_{\alpha}$  and reduced widths defined by

$$\gamma_{\alpha,L}^{2}(s) = \frac{\hbar^{2}}{2\mu_{\alpha}s} S_{\alpha} \left| u_{L}^{\mathsf{D}\mathsf{W}}(s) \right|^{2} \tag{1}$$

are determined in the analysis. Here,  $\mu_{\alpha}$  is the reduced  $\alpha$ -particle mass,  $u_L^{DW}(r)$  the one-dimensional radial wave function which is normalized to unity inside a sphere of 20 fm, and s is the channel radius. The latter, taken as  $s = 1.7A^{1/3}$ fm, is chosen near the radius  $r_{\rm reac}$  at which the  $\alpha$  particle is picked up in  $(d, {}^{6}\text{Li})$  because  $\gamma_{\alpha}{}^{2}(r)$ becomes less reliable for  $r < r_{reac}$ . Unlike *R*-matrix theory, the use of Woods-Saxon plus Coulomb potentials makes the choice of channel radius essentially arbitrary. Whereas  $S_{\alpha}$  is very modeldependent, the reduced widths  $\gamma_{\alpha}^{2}(s)$  are quite insensitive to the choice of potential parameters. The  $\alpha$ -clustering probability defined as

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$$A_{\Gamma}^{m(c)}(r) = \frac{\rho_{\alpha,\Gamma}^{m(c)}(r)}{\rho_{\alpha,c}^{m(c)}(r)}$$
(2)

is the ratio of the nuclear matter (or charge) density associated with  $\alpha$  particles in a particular guantum state  $\Gamma$  to the total nuclear matter (or charge) density. The latter is normalized to Aor Z, respectively. The numerator in Eq. (2) is obtained by folding the number density of  $\alpha$  particles in the quantum state  $\Gamma$ .

$$\eta_{\alpha,\Gamma}(r) = \frac{\mu_{\alpha}}{2\pi r \hbar^2} \gamma_{\alpha,\Gamma}^2(r) , \qquad (3)$$

with the matter (or charge) distribution  $\rho_{\alpha}^{m(c)}(r)$  of the free  $\alpha$  particle which is normalized to 4 or 2, respectively (for details see Appendix B of Ref. 11). By summing  $\rho_{\alpha,\Gamma}^{m(c)}(r)$  over the valence  $\alpha$ particles, i.e., those which have the highest available quantum numbers  $\Gamma$ , one can obtain an estimate (lower limit) for the total  $\alpha$ -clustering probability in the surface of nuclei in their ground states. This summation is achieved by adding the reduced width functions  $\gamma_{\alpha,\Gamma}^{2}(r)$  for the observed pickup transitions for ground and excited states. There is no double counting in this procedure for pickup reactions. Experimental charge distributions<sup>42</sup> for the target nucleus and the free  $\alpha$  particle are used in the determination of the (charge)  $\alpha$ -clustering probability  $\sum_{\Gamma} A_{\Gamma}^{c}(r)$  (see Sec. VII).

#### V. SPECTROSCOPIC FACTORS AND REDUCED WIDTHS

The spectra of Fig. 1 and those at other angles clearly show the presence of selective excitations of groups of levels. The low spin members of the ground state rotational bands  $(0^+ \text{ to } 8^+)$  are populated as well as three groups each of excited levels. The latter appear to be excited rotational bands (0<sup>+</sup> to  $\geq$  4<sup>+</sup>), and the energies deduced for the respective band heads, as marked in Fig. 1, are given in Table I.

The spectra are accompanied by the known level schemes<sup>43,44</sup> for <sup>228</sup>Ra and <sup>234</sup>Th as well as those for the respective neighboring proton-rich isobars <sup>228</sup>Th and <sup>234</sup>U. Isobars appear to have similar level schemes (see below). The latter are included for comparison with the data as more experimental information is available on them. Excited rotational bands based on  $0^+$  states (quasi  $\beta$  bands) in the isobar <sup>234</sup>U are indeed close to the groups G2 and G3. Tentative assignments<sup>31</sup> of  $0^+$ 

for states in <sup>228</sup>Ra coincide closely with the groups G2 and G3, and another  $0^+$  state in the isobar <sup>228</sup>Th is also close in excitation energy. These facts together with the systematics of excited  $0^+$  states discussed later suggest that the four groups of states in both <sup>228</sup>Ra and <sup>234</sup>Th are rotational bands. Negative parity states (1<sup>-</sup>, 3<sup>-</sup>, and 5<sup>-</sup>; quasi  $\gamma$  bands) are weakly excited if at all.

The calculated angular distributions for the transitions to the members of the g.s. rotational band in  $^{234}$ Th of Fig. 2 agree well with the limited data available. The data have large relative uncertainties due to statistics and the unfold-ing procedure<sup>35</sup> for the 0<sup>+</sup> and 2<sup>+</sup> states as well as the absolute uncertainties mentioned earlier.

No attempts were made to resolve the excited groups of levels in <sup>228</sup>Ra and <sup>234</sup>Th. The calculated angular distributions for the four unresolved groups are obtained assuming L = 2. This appears to be the dominant component as in the ground state band.

Reduced widths  $\gamma_{\alpha}^{2}(s)$  and the model-dependent spectroscopic factors  $S_{\alpha}$  are included in Table I. The cross sections are largest for the 2<sup>+</sup> members of the g.s. rotational band and fall off markedly for the 6<sup>+</sup> and 8<sup>+</sup> states. Reduced widths and spectroscopic factors fall off even more strongly as the reaction mechanism favors transfer with angular momentum L > 8. The observed behavior is very similar to that seen in <sup>166</sup>Er(d, <sup>6</sup>Li)<sup>162</sup>Dy at  $E_d = 35$  MeV (see Fig. 1 of Ref. 8) and <sup>154</sup>Sm(d, <sup>6</sup>Li)<sup>150</sup>Nd at  $E_d = 33$  MeV.<sup>12</sup> It supports the boson-model assumption that only s-

and *d*-bosons are transferred in  $\alpha$  transfer.<sup>45</sup> Table II shows the comparison between the reduced widths, partial decay constants, branching ratios, half-lives, and reduced hindrance factors deduced from reaction and decay data. The inverse reduced hindrance factors RHF<sup>-1</sup> represent relative reduced widths. The widths  $\gamma_{\alpha}^{2}(s)$  (and RHF) are determined in the reaction while  $\lambda_{\alpha}$  and  $I_{\alpha}$  are obtained using calculated penetrabilities. The situation is reversed for decay where  $\lambda_{total}$ and  $I_{\alpha}$  provide the experimental input. The close agreement between both the absolute and the relative values is guite remarkable considering uncertainties in the reaction analysis and the calculation of penetrabilities. Similar agreement has been observed previously<sup>11</sup> in a study of  $\alpha$ transfer between ground states of  $\alpha$  decaying nuclei. It indicates a predominant direct  $\alpha$ -transfer mechanism. Penetrabilities calculated from spherical cluster wave functions have to be replaced by penetrability matrices<sup>46</sup> if the deformation is explicitly included. Also, inelastic excitations in the entrance and exit channels should be included in the reaction analysis. The above result suggests that deformation effects are relatively weak and/or that they affect both analyses similarly. The channel radius used in the comparison of  $\gamma_{\alpha}^{2}(s)$  and RHF is different from the inner classical turning point which is commonly used in the analysis of  $\alpha$ -decay data. However, the reduced hindrance factors are essentially independent of this choice. The values of RHF of Table II are also in excellent agreement with

TABLE II. Comparison of reduced widths, decay constants, branching ratios, half-lives, and reduced hindrance factors for reaction and  $\alpha$ -decay data.

| System                                      | J <b>"</b> | Reaction<br>$\gamma_{\alpha}^{2}(s)^{a,b}$<br>(eV) | $\frac{\lambda_{\alpha}}{(\sec^{-1})}$   | Ι <sub>α</sub><br>(%) | RHF <sup>a,b</sup> | $\alpha \text{ decay} \\ \gamma_{\alpha}^{2}(s)^{b} \\ (eV)$ | λ <sub>α</sub> <sup>c</sup><br>(sec <sup>-1</sup> )                        | I <sub>α</sub> <sup>c</sup><br>(%) | RHF <sup>b</sup> |
|---|------------|--|--|-----------------------|--------------------|--|--|------------------------------------|------------------|
| $^{232}$ Th = $^{228}$ Ra + $\alpha$        | 0+         | 59   | $9.5 \times 10^{-19}$  | 75                    | 1.0                | 73   | $1.2 	imes 10^{-18}$   | 77                                 | 1.00             |
|   | $2^+$      | 120  | $3.2	imes10^{-19}$   | 25                    | 0.5                | 133  | $3.6 	imes 10^{-19}$   | 23                                 | 0.55             |
|   | 4*         | 29   | $1.2	imes10^{-21}$   | 0.10                  | 2.0                | ~73  | $\sim 3.1 	imes 10^{-21}$  | $\sim 0.20$                        | ~1.0             |
|   | 6 <b>*</b> | 14   | $\frac{9.3 \times 10^{-25}}{1.3 \times 10^{-18}}$<br>T ~ 1.7 × 10 <sup>+10</sup> , d | 7 × 10 <sup>-5</sup>  |                    |  | $\overline{1.6 \times 10^{-18}}$   |                                    |                  |
| $^{238}\text{U} = ^{234}\text{Th} + \alpha$ | 0*         | 44   | $2.6 \times 10^{-18}$  | 73                    | 1.0                | 65   | $1_{\alpha} \sim 1.4 \times 10^{-18}$<br>$3.8 \times 10^{-18}$             | 77                                 | 1.00             |
|   | $2^+$      | 68   | $9.6 \times 10^{-19}$  | 27                    | 0.7                | 82   | $1.1 \times 10^{-18}$  | 23                                 | 0.80             |
|   | 4*         | 26   | $1.3 	imes 10^{-20}$   | 0.36                  | 1.7                | $\sim 23$  | $\sim 1.1 \times 10^{-20}$   | ~0.23                              | ~2.8             |
|   | 6*         | 6  | $1.5 	imes 10^{-23}$   | $4 \times 10^{-4}$    | 7.0                |  |  |                                    |                  |
|   | 8*         | 7  | $8.2 	imes 10^{-27}$   | $2 \times 10^{-7}$    | 6.8                |  |  |                                    |                  |
|   |            |  | $\frac{3.6 \times 10^{-18}}{T_{\alpha} \approx 6.2 \times 10^{+9} y^{\rm d}}$        |                       |                    |  | $\overline{4.9 \times 10^{-18}}$ $T_{\alpha} \approx 4.4 \times 10^{+9} y$ |                                    |                  |

<sup>a</sup>This work.

<sup>b</sup>Channel radius  $s=1.7A_{core}^{1/3}$  fm.

<sup>c</sup>Half-lives and branching ratios from Refs. 43, 44, and 47.

<sup>d</sup>Uncertainties  $\leq$  a factor of 2.

values predicted from their known systematic A dependences<sup>47</sup> except for the decay value RHF (4<sup>+</sup>) of <sup>228</sup>Ra which, based on systematics, is probably a factor 2 to 3 too small. The known minimum<sup>47</sup> of RHF (2<sup>+</sup>) near A = 224 leading to  $\gamma_{\alpha}^{-2}(2^+) > \gamma_{\alpha}^{-2}(0^+)$  for this region is directly reflected in the measured (d, <sup>6</sup>Li) cross sections, as they are approximately proportional to the reduced widths in the nuclear surface.

The excited groups of levels, presumably rotational bands, are populated with cross sections partly exceeding those of the respective ground state bands. The relative spectroscopic factors and reduced widths of Table I are estimated from the comparison with the calculated angular distributions of Fig. 2. The groups G2 are the most intense  $[\gamma_{\alpha}^{2}(G2) > \gamma_{\alpha}^{2}(G1)]$  and the combined reduced widths for all transitions are about 3.5 times those of the corresponding g.s. bands. Such transitions to excited groups of levels are in qualitative accord with the observed strong  $\alpha$ -decay branches to excited  $0^+$  states in actinide nuclei. At least 12 such branching ratios have been measured<sup>30, 31</sup> for nuclei from <sup>222</sup>Ra to <sup>240</sup>Pu including two for <sup>234</sup>U. These excited states are indicated in Fig. 4. The (reduced) hindrance factors are quite small, typically about 7. However, the relative strengths of  $RHF^{-1} \simeq 15\%$  are still considerably less than those observed in  $\alpha$  pickup. The transfer data of course suffer from the limited energy resolution and the lack of detailed angular distributions which would establish L = 0 characteristics. Since  $(d, {}^{6}Li)$  cross sections are nearly proportional to the reduced widths, the presence of significant  $\alpha$ -spectroscopic strength for these excited states, independent of their exact nature, is clearly evident. No such selective excitations are predicted by existing theoretical models.

The fact that in all but one case only one excited  $0^+$  state is observed in  $\alpha$  decay is likely due to the penetrabilities which decrease the branching ratios for higher  $0^+$  states by additional orders of magnitude and make them difficult to observe. The difference for the relative reduced widths, ~15% in  $\alpha$  decay,  $\geq 100\%$  in  $\alpha$  transfer, is more difficult to explain. It may be due to a dependence of the calculated penetrabilities or cross sections on nuclear size and shape parameters (smaller radius). Such an explanation might be in accord with the known strong E0 transitions<sup>33, 34</sup> and (p, t) angular distributions,<sup>13</sup> which are slightly different for the ground and excited  $0^+$  states.

### VI. SYSTEMATICS OF EXCITED 0<sup>+</sup> STATES IN ACTINIDE NUCLEI

The excited  $0^+$  states in the actinide region are often characterized as  $\beta$ -vibrational states. However, the

observation<sup>13</sup> of significant (p, t) strength over a wide range of nuclei contradicts such a description since transitions to  $\beta$  bands are not favored. A new type of collectivity has therefore been suggested.<sup>13</sup> The cross sections of the twelve measured (p, t) transitions from <sup>224</sup>Ra to <sup>246</sup>Cm are typically 15% of the ground states. Only one excited  $0^+$  state is usually seen, but two are seen for three of the targets. Strong L = 0pickup transitions have also been observed<sup>14</sup> in neighboring odd-A nuclei. In contrast, (t,p)transitions<sup>15, 16</sup> to these same states are generally very weak. They are populated with at most a few percent of the ground states. Only in the heavier actinide nuclei, <sup>248</sup>Cm and <sup>246</sup>Pu, are excited  $0^+$  states seen in (t,p) (Refs. 17,19) with about 35% of the ground state cross section. These states, together with a  $0^+$  state in <sup>250</sup>Cf which is observed<sup>18</sup> as E0 transition in a conversion electron spectrum, are identified as neutron pairing vibration states due to an apparent subshell closure at neutron number  $N = 152.^{17-19}$ 

The other excited  $0^+$  states have several additional unusual features. As remarked earlier, the observed  $\alpha$ -decay branching ratios of  $10^{-6}$  to  $10^{-4}$ are relatively large. This yields small reduced hindrance factors and large relative reduced widths in the range 10 to 20%. Furthermore, these states exhibit strong electromagnetic E0 transitions to the respective ground states while E2 transitions between members of the excited and ground state rotational bands are weak.

The presence of intrinsic excitations with  $K^{r} = 0^{+}$  and  $2^{+}$  below the threshold for two-quasiparticle excitations *in addition* to what was then believed to be  $\beta$ - and  $\gamma$ -vibrational bands, was recognized earlier.<sup>20</sup> This was noted for the region near A = 236 where the additional excitations decay predominantly to lower-lying intrinsic excitations rather than the ground state band, suggesting similar structures for the excited bands.

Excited  $0^+$  states are known to be strongly excited in two-nucleon transfer near major shells or subshell closures in deformed nuclei where the pairing gap is smaller than the gap in single particle or Nilsson orbitals, respectively. Such transitions are also observed in regions of rapid shape transitions, but none of these possibilities apply to the behavior observed in the actinide nuclei. Shape isomers with considerably larger deformation which are observed at higher excitation energies<sup>48</sup> are also ruled out.

Various theoretical models have been developed<sup>21-29</sup> to explain the observed phenomena, most notably the concept of pairing isomers.<sup>21, 22, 24-26</sup> This represents a new and stable type of collectivity whereby the inhomogeneity of prolate and oblate (downsloping and upsloping) Nilsson orbits near the Fermi surface with independent pairing correlations leads to two classes of collective states. The oblate collective state is below the Fermi surface and can therefore only be observed in two-nucleon pickup. However, there appears to exist disagreement<sup>17, 22, 24, 25</sup> as to whether or not the systematics of the 0<sup>+</sup> states can indeed be described by this concept. It also seems unclear whether the excited states are indeed collective in nature as originally suggested<sup>13</sup> or represent shell-model excitations.<sup>22</sup> The selective and strong excitation of three groups of excited states in  $\alpha$ particle pickup is not predicted by the model.

In order to provide additional insight into the properties of the states, we have attempted to describe the states phenomenologically by adjusting the parameters of the interacting boson model Hamiltonian<sup>49</sup>

$$H = \epsilon \left( n_{d_{\tau}} + n_{d_{\tau}} \right) - \kappa Q_{\tau}^{(2)} Q_{\nu}^{(2)} .$$
(4)

Here, the quadrupole operators  $Q_{\mathbf{r}}^{(2)}$  and  $Q_{\nu}^{(2)}$ are expressed as sums of seniority nonconserving and seniority conserving components with relative strengths  $\chi_r$  and  $\chi_{\nu}$ , respectively. No distinction was made between the parameters for the neutron and proton bosons. Also, a projection procedure<sup>50</sup> was used to project out the states which are symmetric in the neutron and proton degrees of freedom<sup>51</sup> (maximum F spin). The parameters  $\epsilon$ ,  $\kappa$ , and  $(\chi_{\nu} + \chi_{\pi})$  were obtained<sup>52</sup> for nuclei from <sup>222</sup> Ra to <sup>242</sup>Pu by fitting known positive parity levels.<sup>32,43,44,47,53</sup> The results are displayed in Fig. 3. All parameters follow welldefined trends except for the quantity  $(\chi_{r} + \chi_{r})$  for <sup>224</sup>Ra and <sup>228</sup>Th. In order to extract averaged global parameters it became necessary to factorize the strength  $\kappa$  of the quadrupole-quadrupole interaction as a function of the number of proton and neutron bosons, respectively, or express it as a function of the total number of bosons outside the <sup>203</sup>Pb core,  $N_{T} = N_{T} + N_{\nu} = \frac{1}{2}(A - 208)$ . Both approaches were performed, but the latter was found to give a slightly more systematic behavior provided  $\kappa$  is redefined as interaction  $\tilde{\kappa}$ between all bosons and the operators  $Q_{\mathbf{r}}^{(2)}$  and  $Q_{\boldsymbol{\mu}}^{(2)}$ in Eq. (4) are replaced by  $Q^{(2)}$ . This is equivalent to the simple substitution in Eq. (4) of the expression  $\kappa(N_{\tau}, N_{\nu}) = [N_{\tau}(N_{\tau} - 1)/2N_{\tau}N_{\nu}]\tilde{\kappa}(N_{\tau})$ . It seems to imply that in this region of nuclei the quadrupole-quadrupole interaction acts between all bosons and not just between the proton and the neutron bosons. The curves included in Fig. 3 represent the averaged parameters.

Figure 4 displays the experimental excitation energies of the ground state rotational bands up to the  $6^+$  state together with all known excited  $0^+$ 



FIG. 3. Parameters  $\epsilon$ ,  $\kappa$ ,  $\tilde{\kappa}$ , and  $(\chi_r + \chi_\nu)$  in the simplified Hamiltonian of the interacting boson model [Eq. (4)]. The data points are from a search using known positive parity levels. The curves represent the average global parameters used to calculate the excitation energies of Fig. 4.

states as function of  $N_T = \frac{1}{2}(A - 208)$  for Z > 88. Most of the data are from a compilation<sup>53</sup> of quasiground, quasi- $\beta$ , and quasi- $\gamma$  bands, but additional 0<sup>+</sup> states are from other references. <sup>13, 17-19, 30, 31</sup> The states observed in the present experiment are included with error bars. The 0<sup>+</sup> states labeled A show a smooth dependence on  $N_T$  with a mini-



FIG. 4. Excitation energies of the ground state rotational bands  $(0^{+}, 2^{+}, 4^{+}, 6^{+})$  and of excited  $0^{+}$  states in the actinide nuclei as function of the total number of bosons outside the <sup>208</sup>Pb core,  $N_T = \frac{1}{2}(A-208)$ . Excited  $0^{+}$  states with small  $\alpha$ -decay hindrance factors or large (p, t) transition strength are marked by short horizontal lines on the left or right, respectively. The solid lines are predictions from the interacting boson model as described in the text.

mum near  $N_T = 11$  (A = 230). Another set of 0<sup>+</sup> states in the heavier actinides labeled *B* intersects set *A* at mass number A = 236 and becomes energetically lower with a minimum near  $N_T = 16$  (A = 240). The two rotational bands in <sup>238</sup>U with band heads at 926 and 993 keV are attributed to sets *B* and *A*, respectively. Three additional 0<sup>+</sup> states labeled *C* are described as neutron pairing vibration states.<sup>17-19</sup> The apparent subshell closure at neutron number N = 152 is not visible in the systematics of the ground state rotational bands, however.

Excited  $0^+$  states with low  $\alpha$  decay reduced hindrance factors and/or strong (p,t) cross sections are marked in Fig. 4 by short horizontal lines on the left and right hand side of the data points, respectively. With two exceptions, all these states belong to sets A and/or B. Similar intrinsic structures for these sets of excited states are again suggested. The lowest of the excited groups of states (groups G2) observed in the present experiment are also attributed to set A.

The solid curves in Fig. 4 are obtained with the averaged global parameters found above. The 0<sup>+</sup> states labeled A are well reproduced except for the  $0_2^+$  state in <sup>228</sup>Th ( $N_T = 10$ ) which is slightly higher. The states labeled B can be reproduced with slightly different parameters  $\kappa$  and  $\chi_{\mathbf{r}}(\mathbf{r})$ (see Fig. 3) but not simultaneously with the states A. The presence of the states B and other "intruder" states suggests that other types of excitation and/or more than one type of boson may have to be considered. It should be noted that additional states which are not fully symmetric in the neutron and proton degrees of freedom<sup>51</sup> ( $F < F_{max}$ ) are neglected in the present approach.<sup>50</sup> These states are usually predicted at higher excitation energy and should not interfere with the region of interest,  $E_x \le 1.5$  MeV. The parameters of Fig. 3 are not very different from those found in the deformed rare earth nuclei.<sup>54</sup> The wave functions are thus close to the SU(3) limit of the IBA model which identifies the  $0_2^+$  states primarily as  $\beta$ -vibration states, which is at variance with other known properties of these states. An estimate for <sup>230</sup>Th where the experimental excitation energy  $E_*(0_2^+)\approx 650~{\rm keV}$  has a minimum places a more pure SU(3) state (large  $\kappa, \chi_{\pi} \approx \chi_{\nu} \approx -\frac{1}{2}\sqrt{7}$ ) at slightly higher energies ( $E_r = 900$  to 1100 keV).

Similar results are obtained for the negative parity states.<sup>28,31</sup> The observed low excitation energies cannot be reproduced on the basis of quadrupole and octupole vibrations alone. It is worth noting that the negative parity states display a systematics similar to the  $0_2^+$  states. They form two parabolas intersecting at  $N_T = 13$  with minima at  $N_T = 8$  ( $E_x \approx 300$  keV) and  $N_T = 15$  to 16 ( $E_x$   $\approx$  700 keV). Other similarities between the positive and negative parity states with  $K^{\mathbf{T}} = 0^+$  and  $0^-$  suggesting related intrinsic structures were pointed out recently.<sup>29</sup>

Two-nucleon transfer strengths based on the above IBA wave functions were calculated<sup>52</sup> for the experimentally observed<sup>13</sup> (p, t) transitions. Whereas mixing between symmetric  $(F = F_{max})$  and the neglected antisymmetric  $(F \leq F_{max})$  states may affect the calculated strengths, the effect is not expected to be large in analogy to the results<sup>50</sup> for the Nd, Sm, and Gd isotopes. It was found that the relative spectroscopic strength are very small (<1%) for both  $0_2^+/0_1^+$  and  $0_3^+/0_1^+$ . The corresponding (t, p) transitions, however, are predicted to be quite strong for  $0^+_2/0^+_1$  with a maximum of ~13% at A = 230 and decreasing values for smaller and larger A. This is in agreement<sup>45</sup> with theoretical predictions in the extreme SU(3) limit of the IBA model in shells which are less than half filled with bosons, but it is in complete disagreement with the experimental evidence. Similar results are predicted for two-proton transfer for which no data exist. Amplitudes for  $\alpha$ -particle transfer can be expressed as coherent sums of two-neutron and two-proton amplitudes.<sup>55</sup> Pickup transitions to excited  $0^+$  states are therefore predicted to be extremely small, which also disagrees with the observed  $(d, {}^{6}Li)$  transfer strengths. The contradictory situation for twoneutron transfer can be rectified for the heavier actinide nuclei if N = 152 is introduced as a magic neutron number, thus reversing the predictions for (p, t) and (t, p) due to the presence of neutron boson holes. Indeed, a ratio of 15% for  $0^+_2/0^+_1$ is then calculated for  ${}^{240}$ Pu $(p,t)^{238}$ Pu. The systematics of the ground state rotational bands does not support the above assumption, however, and the observation of uniform (p, t) strength over the entire range of actinide nuclei can clearly not be reproduced. It thus appears that a satisfactory and comprehensive description of the excited  $0^+$ states in the actinide nuclei in terms of a *simple* IBA model is not possible despite its success in describing excitation energies. This result is to be contrasted with the excellent agreement<sup>54,56</sup> between experiment and IBA predictions including two-neutron stripping and pickup which is obtained for the samarium isotopes. Reactions involving the transfer of one or two bosons [e.g.,  $(p, t), (t, p), (d, {}^{6}\text{Li})$ ] seem to provide definitive tests of IBA wave functions. It would be highly desirable, for instance, to investigate the presence of two-proton pickup strength to excited states in actinide nuclei which might display characteristics similar to that experimentally observed in (p, t).

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### VII. ALPHA-CLUSTERING PROBABILITIES

The  $\alpha$ -clustering probabilities  $A_{r}^{c}(r)$  defined in Eq. (2) of Sec. IV and Appendix B of Ref. 11 are calculated as ratios of the nuclear charge densities associated with  $\alpha$  particles in the quantum state  $\Gamma$  with that for all protons, respectively. Charge densities are used instead of matter densities since experimental charge distributions are well known<sup>42</sup> from electron scattering and other methods. The charge distribution of the free  $\alpha$ particle is also well established.<sup>42</sup> It is needed to derive the charge density  $\rho^{c}_{\alpha,\Gamma}(r)$  at large radii from the density of  $\alpha$ -particle centers,  $\eta_{\alpha,\Gamma}(r)$ . The results of Fig. 5 are obtained for simplicity by using a uniform charge dsitribution of the  $\alpha$ particle with radius  $R = (\frac{5}{3})^{1/2} \langle r^2 \rangle^{1/2}$ , where  $\langle r^2 \rangle^{1/2}$ is the experimental rms charge radius. Figure 5 displays the clustering probability in  $^{238}$ U,  $A^{c}_{\mathbf{g},\mathbf{s}}(\mathbf{r}) = \rho^{c}_{\alpha,\mathbf{g},\mathbf{s}}(\mathbf{r})/\rho^{c}_{\text{nucl}}(\mathbf{r})$ , as calculated from the reduced width function  $\gamma_{\alpha}^{2}(r)$  for the ground state transition  $^{238}U(d, ^{6}Li)^{234}Th$ . The quantum numbers  $\Gamma$  are here those which describe the lpha-cluster wave function of the <sup>238</sup>U ground state relative to <sup>234</sup>Th in its ground state only. The total  $\alpha$ -clustering probability  $\sum_{\Gamma} A_{\Gamma}^{c}(r)$  for all valence  $\alpha$  particles in the ground state of <sup>238</sup>U, also shown, is ob-



FIG. 5. Calculated alpha-clustering probabilities  $A_{g.s.}^{c}(r)$  and  $\Sigma_{\Gamma}A_{\Gamma}^{c}(r)$  (all valence  $\alpha$  particles) for the ground state of <sup>238</sup>U. The rapidly decreasing curve represents the relative nuclear charge density.

tained by adding the reduced width functions  $\gamma_{\alpha}^{2}(r)$ for all observed pickup transitions. Also shown in Fig. 5 is the relative nuclear charge density in <sup>238</sup>U in units of the central density. The graphs show that  $A^{c}_{\mathbf{g},\mathbf{s}_{\star}}(r)$  and  $\sum_{\mathbf{r}} A^{c}_{\mathbf{r}}(r)$  reach maximum values of 1.8% and 20%, respectively, at r = 10.65fm. Here, the corresponding nuclear charge density is 0.18%. Similar results are obtained for <sup>232</sup>Th: the maximum values are 2.5% and 30%, respectively, at r = 10.60 fm, where the relative nuclear charge density is 0.12%. (The total clustering probabilities may be increased further due to more tightly bound  $\alpha$  particles with lower harmonic oscillator quantum numbers which are not included here.) The above  $\alpha$ -clustering probabilities agree with values reported earlier<sup>11</sup> for other  $\alpha$ -decaying nuclei including several rareearth nuclei with  $A \approx 150$ .

The  $\alpha$ -clustering probabilities decrease beyond 11 fm because  $\rho_{\alpha}^{c}(r)$  decreases more rapidly with r than  $\rho_{nucl}^{c}(r)$ . This is due to the dependence of barrier penetration on charge and mass. The maximum  $\alpha$ -clustering probabilities of 20-30% are surprisingly large. They may indicate  $\alpha$ -particle condensation. This phenomenon is predicted<sup>57</sup> for infinite nuclear matter at reduced density where complete  $\alpha$ -particle clustering leads to lower energy per nucleon than uniform nuclear matter. It must be emphasized, though, that the situation in real nuclear matter is not entirely equivalent due to the presence of a surface. The presence of  $\alpha$ condensation effects in the nuclear surface may have consequences for certain atomic effects which depend on high moments of the nuclear charge distribution.

#### VIII. SUMMARY

Excellent agreement between the absolute and relative reduced widths deduced from both  $\alpha$  transfer and  $\alpha$  decay to low-spin members of the ground state rotational bands in <sup>228</sup>Ra and <sup>234</sup>Th has been obtained. It supports the assumption of a direct  $\alpha$ -pickup mechanism for the (d, <sup>6</sup>Li) reaction. Strong transitions are observed to several groups of excited states, presumably rotational bands, in both<sup>228</sup>Ra and <sup>234</sup>Th. The spectroscopic strengths are about equal to those of the ground state bands. This result suggests coherent contributions from both neutron and proton excitations, as two-neutrons pickup strength to excited  $0^+$  states, seen uniformly over the entire range of actinide nuclei,<sup>13</sup> is weaker. The selective excitations observed in the present experiment are seemingly not predicted by existing theoretical models.

Weak branching ratios for excited  $0_2^*$  states observed in  $\alpha$  decay<sup>30, 31</sup> confirm the presence of considerable  $\alpha$  strength for these excited states (small reduced hindrance factor). However, the relative reduced widths are smaller than those inferred from the present experiment. A *smaller* radius for the excited 0<sup>+</sup> states might explain this difference in agreement with conclusions<sup>13</sup> drawn from (p, t) experiments and in accord with the observation of strong E0 transitions.

Existing theoretical models do not seem to provide a comprehensive description of the rather unusual properties of the excited  $0^+$  states in the actinide nuclei. A description in terms of a *simple* IBA model is also inadequate.

The  $\alpha$ -clustering probabilities associated with the valence  $\alpha$  particles at  $r \approx 10.6$  fm in the ground states of both <sup>232</sup>Th and <sup>238</sup>U calculated from the

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reduced widths of the  $(d, {}^{6}\text{Li})$  reaction are found to be about 25%. This large value may indicate  $\alpha$ -condensation effects<sup>57</sup> in low-density nuclear matter as predicted by infinite matter calculations.

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