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Proton Orbital $\frac{1}{2}^-$ [521] and the Stability of Superheavy Elements[(a)]

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We present experimental evidence for the identification of the deformed proton orbital $\frac{1}{2}^-$[521] in ^{251}Es and ^{247}Bk. The implications of this finding with respect to the stability of superheavy elements in the vicinity of $Z \approx 114$ are discussed.

In this Letter, we report on the experimental observation of the proton orbital $\frac{1}{2}^-$[521] and discuss the stability of superheavy elements in the light of this observation.

The proton orbital $\frac{1}{2}^-$[521] has been tentatively assigned in ^{249}Bk by Hoff.[1] In this nucleus, however, the observed decoupling parameter for the rotational band built on this orbital is ~ 0.0. This is in sharp disagreement with the value of ~ 1.0 expected[2] for the pure single-particle state. We believe that we have found this orbital in ^{251}Es and ^{247}Bk, where it is much purer than it is in ^{249}Bk. In ^{251}Es and ^{247}Bk, both the decoupling parameter and the signature observed in reaction spectroscopic studies are in good qualitative agreement with the values expected for the pure single-particle state.

The level structure of ^{251}Es has recently been investigated[3] by measuring the γ-ray and conversion-electron spectra arising from the electron-capture decay of ^{251}Fm (5.3 h). On the basis of the derived multipolarities and logft values the following proton single-particle assignments were made: $\frac{3}{2}^-$[521], 0 keV; $\frac{7}{2}^+$[633], 8.3 keV; $\frac{7}{2}^-$[514], 461.4 keV; and $\frac{9}{2}^+$[624], 777.9 keV. The ^{251}Es ground-state band is not populated directly in the electron-capture process. As $\Omega = \frac{9}{2}^-$ for the ground state of ^{251}Fm, rotational bands having $\Omega \leq \frac{5}{2}$ are not expected to be populated in ^{251}Es. The level at 461.4 keV is also populated by the favored α transition of ^{255}Md (27 min), which confirms the single-particle nature of this state. We have recently studied the reaction ^{250}Cf$(\alpha,t)^{251}$Es with 28.0-MeV α particles from the Argonne National Laboratory tandem Van de Graaff accelerator. The spectrum of outgoing tritons produced in this reaction was measured with an Enge split-pole magnetic spectrograph and is shown in Fig. 1. The three prominent peaks at 411, 452, and 548 keV are associated with an orbital or orbitals not populated in the electron capture and therefore have $\Omega \leq \frac{5}{2}$. Based on single-particle–model calculations,[2] the only logical assignment in this energy range is the proton orbital $\frac{1}{2}^-$[521]. This orbital is calculated to have large values of C_j^2 for $j = \frac{1}{2}$, $\frac{5}{2}$, $\frac{7}{2}$, and $\frac{9}{2}$ and a decoupling parameter of ~ 1.0. Assigning the 411-, 452-, and 548-keV levels as the $\frac{1}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$ members of the $\frac{1}{2}^-$[521] rotational band gives a rotational constant of 6.8 ± 0.3 keV and a decoupling parameter of $+1.0 \pm 0.1$ for this band, in excellent agreement with the expected values for the single-particle state $\frac{1}{2}^-$[521].

We have also studied the levels of ^{247}Bk as observed in ^{247}Cf electron capture and in the reaction ^{246}Cm$(\alpha,t)^{247}$Bk. In the electron-capture study, the following proton single-particle states were assigned[4]: $\frac{3}{2}^-$[521], 0 keV; $\frac{7}{2}^+$[633], 40.8 keV; $\frac{5}{2}^+$[642], 334.9 keV; and $\frac{5}{2}^-$[523], 447.8 keV. The spectrum of tritons produced in the reaction ^{246}Cm$(\alpha,t)^{247}$Bk is shown in Fig. 2. We assign the levels at 704, 743, 815, and 828 keV as the $\frac{1}{2}$, $\frac{5}{2}$, $\frac{7}{2}$, and $\frac{9}{2}$ members of the $\frac{1}{2}^-$[521] rotational band.

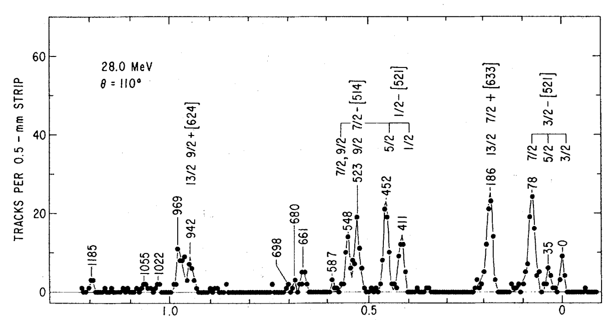

FIG. 1. The triton spectrum from the reaction ^{250}Cf$(\alpha,t)^{251}$Es.

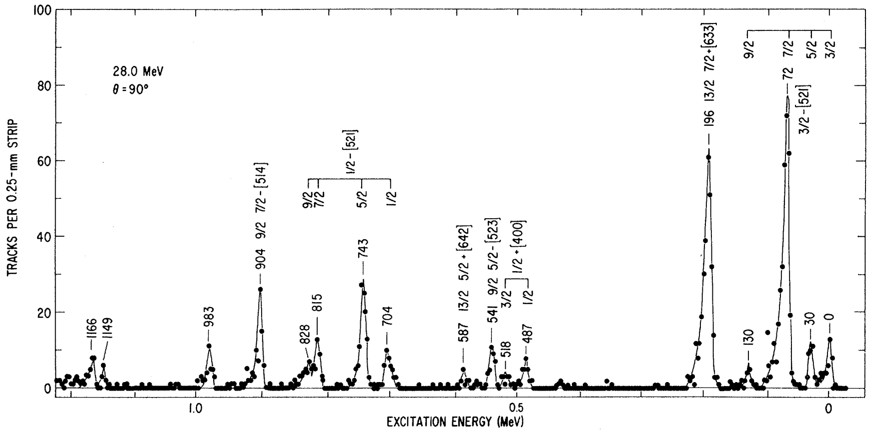

FIG. 2. The triton spectrum produced in the reaction $^{246}Cm(\alpha,t)^{247}Bk$.

These assignments give values of 6.0 ± 0.3 keV for the rotational constant and $+0.7 \pm 0.1$ for the decoupling parameter in this nucleus, in moderately good agreement with the values expected for the pure single-particle state.

A most convincing placement of the $\frac{1}{2}^-[521]$ orbital would come from a spectroscopic investigation of the odd-mass Md isotopes. Presumably, the $\frac{1}{2}^-[521]$ orbital is near the ground state in Md. We already have a hint that the $\frac{1}{2}^-[521]$ and the $\frac{7}{2}^-[514]$ orbitals are quite close in energy in the Lw and Md isotopes. Specifically, we note that the 8.87-MeV favored α group[5] of ^{257}Lw populates a state that is within 30 keV of the ground state in ^{253}Md, as determined from mass systematics.[6] This is evidence for the near degeneracy of the two orbitals, and implies the existence of isomers in Md and Lw isotopes.

The excitation energy of the proton orbital $\frac{1}{2}^-[521]$ in the actinides is of crucial importance in assessing the possible stability of superheavy elements with nuclear charge $Z \approx 114$. In terms of a pure liquid-drop model, nuclides in this mass region ($A \approx 298$) are unstable with respect to fission.[7] Whatever stability there may be with respect to fission arises from the shell effects[8] associated with large gaps in the single-particle spectrum. The stability with respect to fission at $Z = 114$ is partly due to the $f_{7/2}$-$f_{5/2}$ splitting and partly due to the gap in the neutron spectrum at $N = 184$. A decomposition of the $\frac{1}{2}^-[521]$ orbital in terms of spherical orbitals shows[2] that this orbital is $\sim 30\%\ f_{5/2}$. Furthermore, the energy of the orbital $\frac{1}{2}^-[521]$ in the single-particle spectrum is rather sensitive to the position of the $f_{5/2}$ orbital in the spherical potential. The position of the spherical $f_{7/2}$ orbital is largely fixed by the known energy of the deformed state $\frac{3}{2}^-[521]$.

In ^{247}Bk, ^{249}Bk, and ^{251}Es, the orbital $\frac{1}{2}^-[521]$ lies slightly below the orbital $\frac{7}{2}^-[514]$. Calculations[2,9] of single-particle energy-level spacings in ^{249}Bk, using a modified Woods-Saxon oscillator, and folded Yukawa potentials, lead to a prediction of the $\frac{1}{2}^-[521]$ some 300–500 keV above the $\frac{7}{2}^-[514]$ orbital. It is not clear how to manipulate deformation parameters in order to bring the energy of this level into agreement with experiment and maintain good agreement between other single-particle level spacings extracted from the experimental data and the spacings calculated with a conventional Woods-Saxon spin-orbit term. However, Rost[10] has noted that the energy of the $f_{5/2}$ orbital, and accordingly the $\frac{1}{2}^-[521]$ orbital, can be lowered by changing the radius parameter, $r_{s.o.}$, in the spin-orbit interaction. He has suggested that the radius parameter of the spin-orbit term should be taken as $\sim 75\%$ of r_0, the radius parameter used for the central term of the Woods-Saxon potential. We found that this modification has the same effect on our momentum-dependent[11] Woods-Saxon potential.

In Fig. 3, we have plotted the extracted single-particle level spacings obtained from the measured ^{247}Bk, ^{249}Bk, and ^{251}Es excitation energies using a density-dependent delta interaction[12] to calculate the pairing-force matrix elements. We also display two calculated single-particle spectra obtained with the momentum-dependent Woods-Saxon potential. The first calculated spectrum was obtained with $r_{s.o.} = r_0 = 1.25$ fm and the second with $r_{s.o.} = 0.75 r_0 = 0.94$ fm. We note that the levels calculated with $r_{s.o.} = 1.25$ fm differ

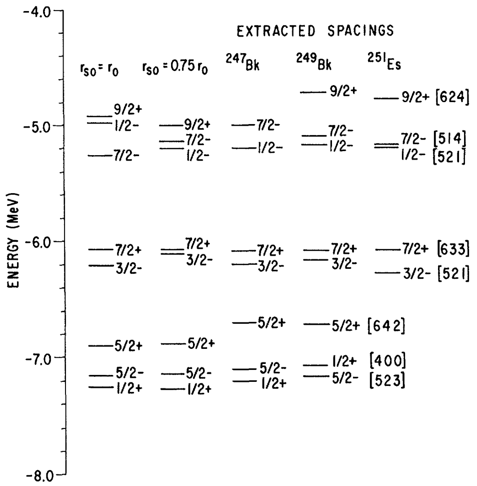

FIG. 3. Comparisons of proton level spacings extracted from experimental data on ^{247}Bk, ^{249}Bk, and ^{251}Es with single-particle level spacings calculated with a momentum-dependent Woods-Saxon potential. The deformation parameters $\nu_2=0.25$, $\nu_4=0.01$, $\nu_6=0.01$ correspond to modified oscillator deformation parameters of $\epsilon_2=0.24$, $\epsilon_4=0.01$, $\epsilon_6=0.01$.

from those shown previously[2] for ^{249}Bk. This change arises from a slight (5%) decrease in the spin-orbit parameter which is needed throughout the actinides to optimize the agreement with extracted level spacings obtained with the density-dependent pairing-force matrix elements. If one uses a constant G pairing force, the magnitude of the spin-orbit force is 5% larger, and the difficulties with the energy of the $\frac{1}{2}^-[521]$ orbit are ~100 keV greater. As can be seen in Fig. 3, the choice of $r_{s.o.}=0.75r_0$ does lower the energy of the $\frac{1}{2}^-[521]$ orbital, in agreement with the experimental data. This calculated spectrum is also in rather good agreement with the extracted level spacings for all of the other single-particle levels assigned in these nuclei. With the modified spin-orbit term, the decoupling parameter calculated for the $\frac{1}{2}^-[521]$ orbital is +1.1 and the decomposition in terms of spherical orbitals (C_j^2) is essentially unchanged from our previous[2] calculation. We note that the $I=\frac{9}{2}$ member of the $\frac{1}{2}^-[521]$ band is expected to be populated weakly relative to the $I=\frac{7}{2}$ member of the band in the (α, t) reaction, although $C_{7/2}^2 \approx C_{9/2}^2$. The distorted-wave Born approximation code DWUCK[13] gives almost an order-of-magnitude reduction here.

We have calculated the proton energy levels for the spherical nucleus $^{298}_{114}X$ using both $r_{s.o.}=r_0$ and

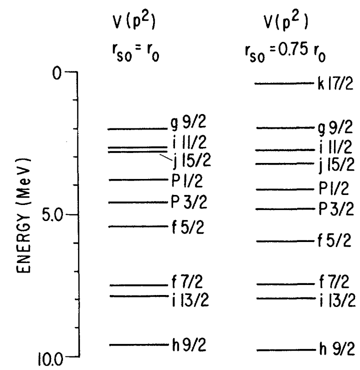

FIG. 4. Calculated proton single-particle spectra for $Z=114$, $A=298$.

$r_{s.o.}=0.75r_0$. The spectra are displayed in Fig. 4. With $r_{s.o.}=r_0$, we obtain an $f_{7/2}$-$f_{5/2}$ splitting of 2.0 MeV; with $r_{s.o.}=0.75r_0$, this splitting becomes 1.5 MeV, in good agreement with the value of 1.6 MeV obtained by Rost.[10] We note that removing the momentum dependence from our potential gives a spectrum that is very similar to that of Rost. As has been noted previously,[14] the shell corrections calculated for superheavy elements appear to be overly sensitive to small differences in extrapolating the single-particle level schemes. The validity of this observation has been re-inforced by the large reported differences[15-17] in recent calculations of the shell correction for the hypothetical nucleus $^{354}_{126}X$. Our observation of the $\frac{1}{2}^-[521]$ orbital places a meaningful constraint on the magnitude of the proton shell correction at $Z=114$. In the past, level schemes with $f_{7/2}$-$f_{5/2}$ splittings ranging from 1.6 to 2.4 MeV have been advocated.[14] These level schemes give rise to proton shell corrections between -2.5 and -5.0 MeV at $Z=114$. As the fission lifetime depends on the sum of the neutron and proton shell corrections, a change of 1 MeV in either the neutron or proton shell correction in the mass-300 region changes[18] the spontaneous-fission lifetime by a factor of ~100. Thus the range of -2.5 to -5.0 MeV in shell corrections corresponds to an uncertainty of ~10^5 in the fission lifetime. For a momentum-independent potential similar to that of Rost,[10] we find a proton shell correction of -3.1 MeV. Adjustment of the energies of the $h_{9/2}$, $i_{13/2}$, $j_{15/2}$, and $g_{9/2}$ levels to simulate the second spectrum of Fig. 4 reduces this shell correction to -2.5 MeV. Our observation of the $\frac{1}{2}^-[521]$ orbital removes an uncertainty of ~10^4 in the fission lifetimes of nuclides in the mass-300 region.

There remain, however, large uncertainties in the fission lifetimes of superheavy elements because of uncertainties in the neutron single-particle spectrum at $N=184$, particularly in the energies of the highly degenerate orbitals $h_{11/2}$, $i_{13/2}$, and $k_{17/2}$. The observation of the neutron single-particle states $\frac{1}{2}^-[761]$, $\frac{1}{2}^-[750]$, and $\frac{1}{2}^+[880]$ in the mass-250 region would resolve this problem.

In conclusion, we have observed the proton orbital $\frac{1}{2}^-[521]$ in the mass-250 region. The position of this level gives a value of ~ 1.5 MeV for the $f_{7/2}$-$f_{5/2}$ splitting and a proton shell correction of ~ -2.8 MeV at $Z=114$.

[a] Work performed under the auspices of the Division of Physical Research of the U. S. Energy Research and Development Adminstration.

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Observation of Two-Photon Optical Free-Induction Decay in Atomic Sodium Vapor

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We have used the Stark-switching technique to observe two-photon optical free-induction decay in the visible using sodium vapor. Transients were observed with use of both a resonant and a nonresonant intermediate state for the two-photon transition.

Since the first observations[1] of photon echoes many single-photon coherent optical transients have been measured.[2] The extension of the techniques of coherent optical transients to two-photon transitions has recently become of interest. These techniques lead to spectroscopic studies in the time domain which are complementary to those made in the frequency domain. We report in this Letter the first use of the Stark-switching technique[3] to observe two-photon optical free-induction decay (FID). These measurements were made in the visible with use of sodium vapor, and the transients occurred on a nanosecond time scale. Two situations were studied: Simple two-photon FID was observed when the intermediate state for the two-photon transition was nonresonant. However, when the intermediate state was resonant, a different behavior was seen. This latter situation has not previously been studied experimentally and our results presently are not fully understood.

The interest in two-photon coherent effects began when Hartmann first discussed the possibility of observing Raman echoes.[4] These Raman echoes have only recently been observed by Hu, Geschwind, and Jedju.[5] Shoemaker and Brewer have observed a coherent transient phenomenon known as Raman beats.[6] Detailed theoretical analysis of Raman beats has been made[7-9] and the close relationship to two-photon free-induction decay has been discussed.[7] Other experimental work on two-photon transients include the reported observation of two-photon self-induced transparency[10] and measurements of the decay of two-