

and is fragmented over at least twice as many expected states. About 5% more strength may be present in groups at 8.965, 13.40, and 13.90 MeV, but transitions to levels at 5.125, 6.862, 11.45, and 11.47 MeV are definitely not  $M8$ .

In summary the qualitative, but not quantitative, features of the observed  $M8$  strength are explained by the simple 1p-1h model including both  $T = 1$  and  $T = 2$  states. The missing 78% of the predicted strength may be accounted for in several possible ways. These include a deformed  $^{58}\text{Ni}$  ground state resulting in incomplete closure of the  $f_{7/2}$  shell<sup>15</sup> (e.g., 2p-2h excitations in the  $^{56}\text{Ni}$  core), partial occupancy of the  $g_{9/2}$  orbit producing a blocking effect, and destructive interference<sup>4</sup> between 1p-1h and 3p-3h configurations in the excited  $8^-$  states. There is also the possibility that the strength is fragmented among many weakly excited states arising from mixing between multiparticle-hole configurations.

Hopefully, these measurements and observations will stimulate more realistic model calculations and other experiments in an effort to unravel the physics giving rise to these magnetic excitations. It might soon be possible to study  $(n, p)$  or  $(\pi^-, \pi^0)$  to locate the high-spin  $T = 2$  states in the parent analog  $^{58}\text{Co}$ .

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<sup>1</sup>T. W. Donnelly and G. E. Walker, Ann. Phys. (N.Y.) **60**, 209 (1970).

<sup>2</sup>L. W. Fagg, Rev. Mod. Phys. **47**, 638 (1975).

<sup>3</sup>T. W. Donnelly, J. D. Walecka, I. Sick, and E. B. Hughes, Phys. Rev. Lett. **21**, 1196 (1968).

<sup>4</sup>I. Sick, E. B. Hughes, T. W. Donnelly, J. D. Walecka, and G. E. Walker, Phys. Rev. Lett. **23**, 1117 (1969).

<sup>5</sup>T. W. Donnelly, J. D. Walecka, G. E. Walker, and I. Sick, Phys. Lett. **32B**, 545 (1970).

<sup>6</sup>H. Zarek *et al.*, Phys. Rev. Lett. **38**, 750 (1977).

<sup>7</sup>C. Ngo-Trong, Toshio Suzuki, and D. J. Rowe, to be published.

<sup>8</sup>W. Bertozzi *et al.*, Nucl. Instrum. Methods **141**, 457 (1977).

<sup>9</sup>J. R. Ficenec, W. P. Trower, J. Heisenberg, and I. Sick, Phys. Lett. **32B**, 460 (1970).

<sup>10</sup>T. DeForest, Jr., and J. D. Walecka, Adv. Phys. **15**, 1 (1966).

<sup>11</sup>K. Itoh, M. Oyamada, and Y. Torizuka, Phys. Rev. C **7**, 458 (1973).

<sup>12</sup>H. C. Lee, Chalk River Nuclear Laboratories Report No. AECL-4839 (unpublished).

<sup>13</sup>H. Uberall, *Electron Scattering from Complex Nuclei* (Academic, New York, 1970), Pt. A.

<sup>14</sup>C. W. DeJager, H. DeVries, and C. DeVries, At. Data Nucl. Data **14**, 479 (1974).

<sup>15</sup>G. Oberlechner and J. Richert, Nucl. Phys. **A191**, 577 (1972).

## Evidence against Superheavy Elements in Giant-Halo Inclusions Re-examined with Synchrotron Radiation

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The giant-halo inclusion 19D reported to show the strongest evidence for superheavy elements on the basis of proton-induced x-ray fluorescence has been re-examined with  $\approx 55$  times greater sensitivity by employing synchrotron radiation as the exciting source. It is shown conclusively that at concentration levels of  $\approx 5 \times 10^8$  atoms per inclusion, superheavy elements are not present in GH19D or in numerous other giant-halo inclusions studied.

The detection of characteristic x rays is one of the best methods for identifying the atomic number ( $Z$ ) of a new element. Proton-induced x-ray

emission (PIXE) techniques were employed recently by Gentry *et al.*<sup>1</sup> at the Florida State University (FSU) tandem Van de Graaff accelerator

to obtain evidence for the possible presence of superheavy elements (SHE) in giant-halo (GH) monazite inclusions. Spurred by this report, which if found correct would have important experimental and theoretical consequences, several searches have been undertaken,<sup>2</sup> all with negative results. The many samples searched were chosen because of their similarity to those examined in Ref. 1, but none were giant-halo inclusions. A more definitive experiment<sup>3</sup> examined eleven GH inclusions from the same piece of mica as those reported to show positive evidence for SHE. The inclusions were excited with the intense photon flux at the Stanford Synchrotron Radiation Project (SSRP). No evidence was found for SHE at concentration levels about 10 times smaller than those reported in Ref. 1. Meanwhile, Fox *et al.*<sup>4</sup> had shown that a  $\gamma$  ray excited in the reaction  $^{140}\text{Ce}(p, n\gamma)$  would interfere with the  $L\alpha_1$  peak of  $Z=126$ . This evidence together with the results of our earlier measurements<sup>3</sup> might have ended the matter except for the following: Both Fox *et al.*<sup>4</sup> and Fletcher<sup>5</sup> published arguments against suggestions that the reaction  $^{140}\text{Ce}(p, n\gamma)$  could wholly account for the  $Z=126$  peak. Therefore, it was highly desirable to examine the original GH inclusions 15, 19A, and 19D in order to remove any vestigial doubts. Unfortunately, GH15 was lost in sample transfer at FSU, and GH19A was lost through overheating at Harwell,<sup>6</sup> leaving only GH19D for confirmation attempts. In the present Letter, we describe a second improved experiment at SSRP in which we studied 19D and numerous other GH samples with record levels of sensitivity, again with negative results for the presence of SHE. In particular, no evidence for SHE ( $Z=126$ ) was found at a sensitivity level 50 to 100 times better than the evidence reported in Ref. 1.

The basic experimental arrangement is similar to that described in Ref. 3 and is shown schematically in Fig. 1. The photon flux generated by electrons circulating in storage rings has several advantages over other excitation sources for x-ray fluorescence analysis. Some of these are as follows: (1) The continuous photon energy spectrum permits selection by monochromatization of photons in a narrow energy band. By choice of a bombarding energy close to the absorption edge, the  $L$  photoionization cross section for a particular SHE can be enhanced over those for lower  $Z$  elements, whereas with protons the cross sections are  $\approx 500$  times smaller and decrease monotonically with  $Z$ . (2) With a variable-

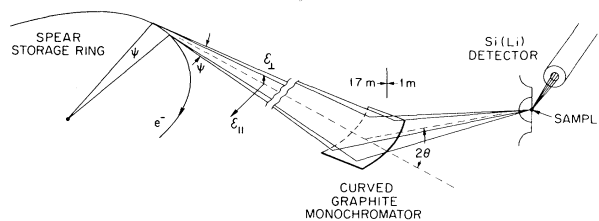


FIG. 1. Schematic layout (not to scale) of the present experiment. The horizontal divergence  $\psi$  was 2 mrad, the vertical divergence 0.14 mrad, the horizontal polarization  $\epsilon_{\parallel} \approx 97\%$  [estimated from the formulas given by A. A. Sokolov and I. M. Ternov, *Synchrotron Radiation* (Pergamon, New York, 1968)], and the scattering angle  $2\theta = 6^\circ$  at 37-keV incident energy.

energy source, the energy of the absorption edge can be correlated with the fluorescence x rays, providing a unique  $Z$  identification. (3) Since synchrotron radiation is highly polarized ( $\approx 97\%$  of its electric vector is in the plane of the ring for 37-keV photons), the unwanted scattered radiation (Rayleigh and Compton) reaching the detector can be greatly reduced by placing the detector at a right angle ( $90 \pm 10^\circ$  in our case) to the incident beam and parallel to the plane of the ring. (4) By choice of a bombarding energy of 37 keV (which is  $\approx 1$  keV above the calculated<sup>7</sup>  $L_{\text{III}}$  absorption edge for  $Z=126$ ), the lanthanide  $K$  x rays can be totally excluded. (5) Overheating of the sample can be avoided in the present photon experiment, where only  $\approx 10^{-3}$  as much energy is deposited in the inclusion as in the proton experiment.<sup>1</sup>

The following modifications were made to the first experiment<sup>3</sup>: The hot-pressed pyrolytic graphite monochromator was made slightly thicker, resulting in an energy spread at 37 keV of 460 eV [full width at half-maximum (FWHM)] instead of 300 eV earlier; all Pb-Sn solder connections in the Si(Li) detector (10 mm diameter, 5.3 mm deep, 0.25-mm Be window) were replaced with pure Pb to reduce Sn  $K\alpha$  contamination; the resistance of the feedback resistor in the detector was decreased to permit higher counting rates at the expense of resolution (450 eV versus 300 eV FWHM at 39.2 keV); the fibers used for mounting the samples were made thinner (0.1–0.6-mg/cm<sup>2</sup> polypropylene replacing 2.54-mg/cm<sup>2</sup> polystyrene strips); and the 27.1-mg/cm<sup>2</sup> Cu filter in front of the detector was replaced by a 210-mg/cm<sup>2</sup> (0.8-mm) Al filter. These modifications resulted in about a factor of 7 improvement in the sensitivity of the present experiment over the initial attempt.

In Fig. 2, we have compared the x-ray spectra excited by 5.7-MeV protons at FSU<sup>1</sup> with those

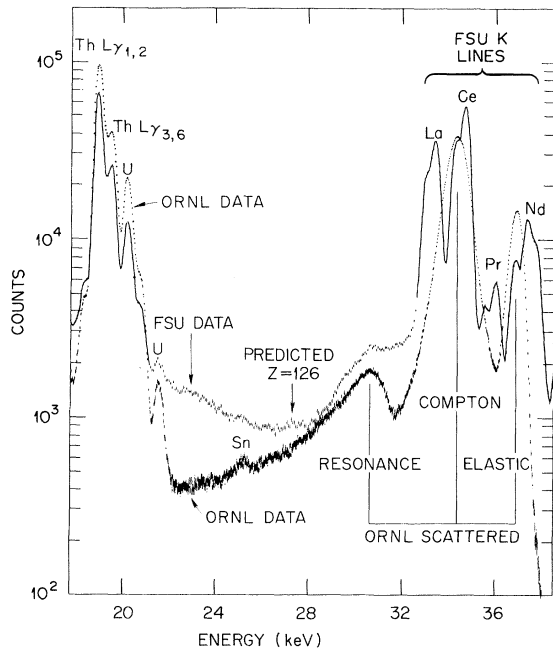


FIG. 2. Comparison of our photon-induced x-ray fluorescence data (ORNL) on 19D with proton-induced data (FSU) reported in Ref. 1.

excited by 37-keV photons at Stanford in the present ORNL experiment. Our spectra were recorded at 20 eV per channel and the FSU spectra at 61.8 eV per channel. Both spectra were taken with  $\approx 5$ -mm-thick Si(Li) detectors and with  $\approx 0.8$ -mm Al filters, thus undergoing similar attenuation. The number of fluorescent radiation events,  $N_i^f$ , from element  $i$  in the detector is

$$N_i^f = I_0 t n_i \epsilon_i \Omega_i a_i \sum_k \sigma_{i,k}; \quad (1)$$

$$\sum_k \sigma_{i,k} = \left( \sum_k \mu_k \omega_k F_k \right)_i,$$

where  $I_0$  is the number of incident photons (or protons) per square centimeter second,  $t$  the counting time,  $n_i$  the number of atoms of element  $i$ ,  $\epsilon_i$  the detector efficiency,  $\Omega_i$  the detector solid angle,  $a_i$  the attenuation factor for the exciting radiation into and the fluorescence radiation out of the sample,  $\mu_k$  the partial photoionization cross section for an electron from the  $k$ th shell of element  $i$ ,  $\omega_k$  the fluorescence yield, and  $F_k$  the fraction of radiation events contributing to the fluorescent energy of interest. With Eq. (1), we can calculate the number of SHE 126  $L\alpha_1$  (27.25 keV) events per atom entering the detector relative to the Th  $L\gamma$  ( $\gamma_{1,2,3,6}$ ) events as

$$\frac{N_{126}^{L\alpha_1}/\text{atom}}{N_{\text{Th}}^{L\gamma_{1,2,3,6}}/\text{atom}} = \frac{\epsilon_{126} a_{126} \left( \sum_k \mu_k \omega_k F_k \right)_{126}}{\epsilon_{\text{Th}} a_{\text{Th}} \left( \sum_k \mu_k \omega_k F_k \right)_{\text{Th}}}. \quad (2)$$

The numerical values used to evaluate Eq. (2) are given in Table I. The cross-section values are based on extrapolated experimental subshell cross sections for protons,<sup>8</sup> theoretical subshell photoionization cross sections for photons,<sup>9</sup> evaluated fluorescence and Coster-Kronig yields,<sup>10</sup> and theoretical x-ray emission rates.<sup>11</sup> The observed counts were extracted from the respective data.

Using the data of Table I, we draw the following conclusions:

(1) For 37-keV photon excitation, a 126  $L\alpha_1$  event/atom is (3765/265) or 14.2 times more likely to occur than a Th  $L\gamma$  event/atom, whereas this factor is only (6.7/8) or 0.84 for 5.7-MeV proton excitation.

(2) The Th  $L\gamma$  events in the ORNL spectrum of GH 19D are  $2.71 \times 10^6 / 0.53 \times 10^6$  or 5.1 times those in the FSU spectrum. Therefore, the ORNL

TABLE I. Analysis of ORNL and FSU data pertaining to GH19D.

Fluorescence line	$\sum \sigma_{i,k}$ (b/atom) <sup>a</sup>		Observed counts	
	37-keV photons	5.7-MeV protons	ORNL data <sup>b</sup>	FSU data <sup>c</sup>
Th $L\gamma_{1,2,3,6}$	265 ± 30	8 ± 2	$2.71 \times 10^6$	$0.53 \times 10^6$
Z = 116 $L\alpha_1$	2690 ± 400	12 ± 400	< 400	290
Z = 124 $L\alpha_1$	3500 ± 550	7.0 ± 3.0	< 465	260
Z = 126 $L\alpha_1$	3765 ± 600	6.7 ± 3.0	< 496	500
Z = 127 $L\alpha_1$	3950 ± 630	6.5 ± 3.0	< 510	350
3 × (background in 800 eV at 27 keV) <sup>1/2</sup>			496	315

<sup>a</sup>Quoted uncertainties in the cross-section values are our estimates.

<sup>b</sup>Upper limit corresponds to  $3\sigma$  above background.

<sup>c</sup>Values given in Ref. 1 for SHE have been reduced by  $\approx 40\%$  in the re-evaluation of background (Ref. 5).

spectrum would have contained  $5.1 \times 14.2 \times (1/0.84)$  or  $86 \pm 15$  events for every event attributed to  $126 L\alpha_1$  in the FSU spectrum.

(3) Using the  $3\sigma$  (98% confidence) criterion for detection above background, the ORNL detectability would be  $86 \times (315/496)$  or  $55 \pm 10$  times that of FSU for  $126 L\alpha_1$  events.

(4) In Ref. 1, the signal observed for  $126 L\alpha_1$  was  $928 \pm 90$  counts of which  $\approx 65$  counts could come from the reaction  $^{140}\text{Ce}(p, n\gamma)$ .<sup>4</sup> A re-evaluation of the background in the FSU spectrum by Fletcher<sup>5</sup> further reduced the signal to  $\approx 500$  counts. If these were genuine  $126 L\alpha_1$  events, our spectrum should have contained  $86 \times 500$  or 43 000 counts. Such a peak would be 24 times larger than the Sn  $K\alpha$  peak (see Fig. 2) in our spectrum which contained 1800 counts ( $\approx 1000$  from Sn in the sample and the remainder from the sample chamber and the detector).

(5) For the other fluorescence lines referred to in Ref. 1, we conclude that the ORNL spectrum would have contained 34 times as many  $116 L\alpha_1$  events, 77 times the  $124 L\alpha_1$  events, and 94 times the  $127 L\alpha_1$  events. No evidence for these SHE was observed in our data.

(6) Though the FSU experiment used a proton beam of  $\approx 30 \mu\text{m}$  diameter to fluoresce the  $\approx 60\text{-}\mu\text{m}$ -diam inclusion 19D, it was acknowledged in Ref. 1 that the proton beam wandered during the long periods of data accumulation. If, however, we assume nonhomogeneous distribution for SHE and a highly stable proton beam irradiating a fixed area on the sample, the FSU experiment could at best be capable of detecting  $\frac{1}{5}$  the number of SHE atoms used in the comparison above. Even if this were so, the ORNL spectrum of 19D would have given 17 times the number of  $L\alpha_1$  events attributed to  $Z = 126$  FSU spectrum.

Even though microscopic examination of 19D showed that the size and shape of this inclusion remained substantially unchanged, an argument may still be made that perhaps any SHE present in GH 19D were evaporated during the FSU run. However, we have studied eighteen GH inclusions in addition to the eleven studied earlier,<sup>3</sup> all taken from the same piece of mica as 19D. These results are given in Table II. We have achieved detectability levels as low as  $\approx 5 \times 10^8$  atoms for several inclusions. It is improbable that  $Z = 126$  would be present in five out of six GH inclusions studied in Ref. 1 and yet not be present at levels 10 to 100 times lower in 29 other GH inclusions extracted from nearby sites in the same mica.

We conclude that the previously reported evi-

TABLE II. Upper limits for the number of  $Z = 126$  atoms in each giant-halo inclusion.

GH inclusions	Th $L\gamma$ counts <sup>a</sup> ( $10^{-6}$ )	$I_0$ <sup>b</sup> ( $10^{-10}$ )	Th atoms <sup>c</sup> ( $10^{-14}$ )	S <sup>d</sup> ( $10^{11}$ )	D <sup>e</sup> ( $10^{-9}$ )
1	2.16	0.5	5.0	1.8	4.1
2	1.14	0.5	4.4	1.2	9.7
3	3.42	1.5	1.7	3.4	2.2
4	0.62	1.5	0.45	4.3	0.74
5	0.41	1.5	0.57	3.5	2.2
6	2.65	4.0	0.97	11.2	0.78
7	1.91	17.9	0.46	50.0	0.51
8	0.14	15.7	0.08	10.4	0.60
9	0.49	19.3	0.12	49.8	0.45
10	0.75	4.0	0.88	4.9	1.4
11	0.28	4.6	0.11	5.0	5.0
12	1.46	3.5	0.84	9.9	0.82
13	0.45	2.8	0.14	7.7	0.60
14	0.43	2.8	0.13	7.3	0.56
15	0.67	3.9	0.25	10.0	0.83
16	0.22	3.6	0.082	8.5	0.52
17	0.06	4.5	0.016	12.7	0.57
18	4.11	4.5	0.94	10.4	1.2
19D	2.71	9.8	0.28	8.7	0.43

<sup>a</sup>Obtained by integrating the  $L\gamma_{1,2,3,6}$  peaks.

<sup>b</sup>Number of incident photons/[ $(0.45\text{-mm}^2) \cdot \text{sec}$ ], determined from Cd standards with  $(1.8 \pm 0.7) \times 10^{11}$  and  $(4.6 \pm 1.7) \times 10^9$  atoms and from National Bureau of Standards certified coal standard SRM-1632 and scaled to the energy and current of the circulating electron beam.

<sup>c</sup>Determined using Eq. (1).

<sup>d</sup>Sensitivity of the present experiment for detecting  $Z = 126 L\alpha_1$  peak (27.25 keV) in units of count/sec  $\cdot$  atom.

<sup>e</sup>Detectable number of  $Z = 126$  atoms at 98% confidence level.

dence for SHE is invalid.

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<sup>1</sup>R. V. Gentry *et al.*, Phys. Rev. Lett. **37**, 11 (1976).

<sup>2</sup>F. Bosch *et al.*, Phys. Rev. Lett. **37**, 1515 (1976), and Z. Phys. **A280**, 39 (1977); C. Stéphan *et al.*, Phys. Rev. Lett. **37**, 1534 (1976); B. H. Ketelle *et al.*, Phys. Rev. Lett. **37**, 1734 (1976); N. A. Jelley *et al.*, Nature (London) **265**, 35 (1977); J. Kantele, University of Jyväskylä, Finland, Research Report No. 5, 1976 (unpublished); H. J. Annegarn *et al.*, Phys. Rev. C **16**, 379 (1977); R. Middleton *et al.*, Phys. Rev. C **16**, 477 (1977).

<sup>3</sup>C. J. Sparks, Jr., *et al.*, Phys. Rev. Lett. **38**, 205 (1977).

<sup>4</sup>J. D. Fox *et al.*, Phys. Rev. Lett. **37**, 629 (1976).

<sup>5</sup>N. R. Fletcher, Phys. Rev. Lett. **38**, 479 (1977).

<sup>6</sup>J. A. Cookson *et al.*, FSU Tandem Accelerator Laboratory Annual Report, 1976 (unpublished), p. 73.

<sup>7</sup>T. A. Carlson *et al.*, Nucl. Phys. **A135**, 57 (1969); C. C. Lu *et al.*, Nucl. Phys. **A175**, 289 (1971).

<sup>8</sup>C. V. Barros Leite *et al.*, Phys. Rev. A **15**, 943 (1977); T. L. Hardt and R. L. Watson, At. Data Nucl. Data Tables **17**, 107 (1976).

<sup>9</sup>J. H. Scofield, Lawrence Livermore Laboratory Report No. UCRL-51326, 1973 (unpublished); D. T. Cromer and D. Libermann, Los Alamos Scientific Laboratory Report No. LA-4403, 1970 (unpublished).

<sup>10</sup>W. Bambynek *et al.*, Rev. Mod. Phys. **44**, 716 (1972), and data published in the open literature since this compilation.

<sup>11</sup>J. H. Scofield, Phys. Rev. A **10**, 1507 (1974), and At. Data Nucl. Data Tables **14**, 121 (1974); R. Anholt and J. Rasmussen, Phys. Rev. A **9**, 585 (1974).

## Muon Capture by the Triton

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Muon capture by the triton is investigated. "Exact" ground-state wave functions extracted from realistic nucleon-nucleon interactions are incorporated in the calculations. The treatment involves non-energy-weighted sum rules and explicit introduction of the three-neutron final state. Beside results on muon capture by  $\text{He}^3$ , a lower limit is established for the muon-capture rate in  $\text{H}^3$ .

Muon capture by the triton presents two features which make this process quite worthy of detailed investigation. These features are (i) the existence of only one open outgoing channel—to wit, the three neutrons; (ii) a significant inhibiting Pauli effect which induces an outstanding reduction of the capture rate in  $\text{H}^3$  by comparison to the free-proton case. While the uniqueness of the final-state channel simplifies the theoretical analysis, hindrance of the process suggests the possibility of investigating mesonic exchange contributions (MEC) possibly quite significant in such a reaction.<sup>1</sup>

This Letter presents results on this reaction, i.e.,  $\mu^- + \text{H}^3 \rightarrow \nu_\mu + n + n + n$ , obtained with the nucleon-only impulse approximation (NOIA) in view of investigations to come on MEC in this process. While the operator is the usual effective-muon-capture Hamiltonian  $H_{\text{WI}}^{\text{eff}}$ ,<sup>2</sup> the initial state is the  $\text{H}^3$  ground state obtained by Laverne and Gignoux

(LG) through a solution of the Faddeev equations in configuration space.<sup>3</sup> The final three-neutron state can also be obtained through the results of these authors together with those of Merkuriev on the scattering three-body problem within configuration space again.<sup>4</sup> The direct connection of the LG wave function with the realistic nucleon-nucleon interaction makes the  $A=3$  wave function especially adequate for calculations of MEC. To fix ideas, we take the "exact" wave function obtained with the Reid soft core (RSC) and the de Tourreil and Sprung supersoft core (SSC) nucleon-nucleon interaction.<sup>3,5</sup> Weights  $P_S, P_{S'}, P_D$  of the main components of the corresponding wave functions  $\psi = \psi_S + \psi_{S'} + \psi_D$  are given in Table I. As usual,  $\psi_S$  stands for the  $L=0$  part, spatially symmetric in the interchange of any pair of particles;  $\psi_{S'}$  and  $\psi_D$  stand for the  $L=0$  and  $L=2$  parts of mixed symmetry. In a first step, results on muon capture by  $\text{He}^3$  are displayed in