Search with Synchrotron Radiation for Superheavy Elements in Giant-Halo Inclusions*

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Synchrotron radiation was used to excite x-ray spectra from monazite inclusions taken from the same piece of mica as those recently reported to show evidence of primordial superheavy elements. No evidence was found for the existence of superheavy elements in the range of Z from 105 to 129, and, in particular, no evidence was found for $Z = 126$ at concentrations of $\sim 5\times 10^9$ atoms per inclusion or $\sim$ 1–2 ppm by weight.

Evidence for the existence of several superheavy elements (SHE) between $Z = 114$ and $Z = 127$ has been reported[1] based on proton microprobe excitation of x-ray spectra from monazite inclusions extracted from mica in which the inclusions had generated giant-halo discolorations thought to arise from ~14-MeV α particles.[2] Weak peaks were observed in these spectra with energies and widths that did not correspond well with those expected from known K lines of fifth-period elements. An interpretation of these peaks as L lines of SHE was proposed, although only one line for each element was observed. It was claimed in Ref. 1 that the $L\alpha_1$ line from $Z = 126$ was seen in five out of six inclusions studied. Concentrations were estimated to be as high as several hundred picograms for $Z = 124$ (100 pg $\approx 2\times 10^{11}$ atoms for atomic weights ≈ 310) in one inclusion, raising expectations that a large number of nuclear spectroscopic studies could be carried out. The very existence of primordial SHE, if confirmed, would necessitate drastic revisions of many existing ideas concerning nucleosynthesis and nuclear theory.[3] In this Letter we will present evidence to show that in the eleven giant-halo inclusions which we studied, superheavy elements, if indeed present, are in amounts much smaller than originally estimated.

With proton excitation, possible confusion of SHE L x rays can occur with overlapping K x rays of fifth-period elements, with γ rays produced by nuclear reactions and with the radiative processes from Ce and La which could give lower-energy radiation than the usual characteristic energy.[4] In the case of $Z = 126$, interferences arise from Te K x rays and from a γ ray excited in the reaction[5] $^{140}\mathrm{Ce}(p, n\gamma)$ on Ce present in the inclusions. These uncertainties can be eliminated with a tunable monochromatic photon source, since the incident energy can be varied to bracket the absorption edge of interest. Although the K fluorescence energies may overlap some of the predicted $L\alpha_1$ energies[6] of SHE, the K binding energies are about 4 keV below the SHE L_{III} edges (see Fig. 1), allowing a unique correlation of fluorescence energies with their absorption edges. Radiative processes associated with K-edge excitations of La and Ce or those associated with nuclear photoexcitation processes are negligible, can be avoided, or can be determined by varying the energy of the incident x rays.

Electrons circulating in storage rings provide an intense photon flux in a continuous energy spectrum extending into the x-ray region. An experiment was designed for the Stanford Synchrotron Radiation Project (SSRP)[7] which utilizes the photon flux from a storage ring (SPEAR) operated

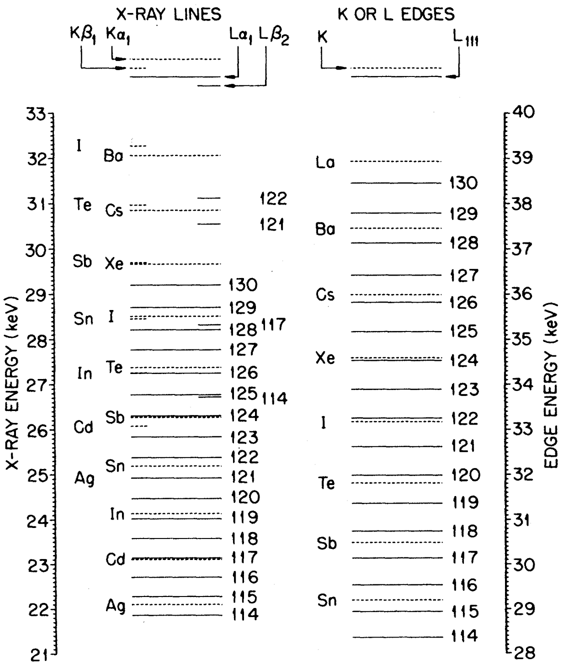

FIG. 1. Absorption edges and fluorescence energies of known elements and proposed SHE (with $Z = 114$ to 130) in the energy region of interest. The positions of $L\beta_2$ lines are shown only for some SHE.

by the Stanford Linear Accelerator Center (SLAC). A key feature of the experiment was a hot-pressed (bent to a 10-cm radius) pyrolytic graphite monochromator ($2d = 6.71$ Å) with 0.4° mosaic spread [full width at half-maximum (FWHM)] located 17 m from the source; this monochromator collected 2 mrad of radiation in the horizontal plane and half the total vertical divergence of the source. For 37.5-keV photons (300 eV FWHM), the focused flux contained about 4×10^{10} photons/sec mm^2 when SPEAR operated at an electron energy of 3.4 GeV and a current of ~20 mA.[7] The fluorescence x rays were detected with a Si(Li) detector 16 mm in diameter and 5-mm deep mounted at 90°±15° to the incident beam and parallel to the plane of the synchrotron ring. As the synchrotron radiation is highly polarized with its electric vector in the plane of acceleration, the Rayleigh (elastic) and Compton (inelastic) scatterings from the sample and mounting fiber are minimized at this angle. Specimens were mounted with silicone grease on 2.54-mg/cm^2 polystyrene strips. A Cu filter (27.1 mg/cm^2) was placed between the specimen and detector to reduce the total count rate to $\approx 10^4$ counts/sec. In addition, pulse pile-up was minimized with electronic circuitry.

We use the definition for the total cross section, σ, to derive an expression from which the sensitivity of our experiment for SHE can be determined from observed spectra of known standards. The number of fluorescent radiation events, $N_i{}^f$, neglecting absorption and hole transfer, from element i in the detector is

$$N_i{}^f = I_0 t n_i \epsilon_i \textstyle\sum_k \sigma_{i,k} = I_0 t n_i \epsilon_i (\sum_k \mu_k \omega_k F_k), \qquad (1)$$

where I_0 is the number of incident photons per squared centimeter second, t the counting time, n_i the number of atoms of element i, μ_k the partial photoionization cross section for an electron from the kth shell of element i, ω_k the fluorescence yield, F_k the fraction of these radiative events belonging to the fluorescence energy of interest, and ϵ_i the absolute efficiency of the detector including the solid angle. More than one electron shell can contribute to the fluorescence peak of interest, and the sum over k includes these subshells. From the nominal compositions of the monazite inclusions, we find that the absorption of radiation (~37.4 keV) entering and of fluorescence radiation (~25 keV) emerging from the inclusion would reduce the measured fluorescence intensity by less than $\frac{1}{3}$ for the largest inclusions (≈ 150 μm in diameter).

The sensitivity for the detection of SHE was established by measuring spectra from samples containing small but known quantities ($\approx 10^{11}$–10^{12} atoms) of Cd and Cs. The Cd atoms were loaded into Dow cation exchange resin beads of ~75-μm diam along with an equal amount of U atoms. The U atoms were later assayed through neutron activation yielding results consistent with the original estimates based on elution efficiencies. The number of Cs atoms implanted in an Al foil were also determined by neutron activation.

For known elements, the fluorescence cross section, σ_i in Eq. (1), was determined from evaluated and calculated values for photoelectric cross sections,[8] fluorescence yields,[9] and the relative rates of emission[10]; for SHE, extrapolated[8,9] and calculated[11] values were used. The numerical values used for σ_i in units of 10^{-21} cm^2/atom are 22.2 for Cd $K\alpha$, 3.66 for Cs $K\alpha$, 0.24 for Th $L\gamma_{1,2,3}$, and 8.0 for $L\alpha_1$ of element 126 at 37 keV.

The eleven monazite giant-halo inclusions (~45–150 μm in diameter) studied were extracted from the same piece of mica as those examined by proton excitation[1] but were different inclusions. Spectra were also obtained from five regular-halo inclusions in which the halos are proposed to arise from the U and Th α-decay chains.[2] The incident x-ray beam was approximately 0.25 mm× 1 mm, an area sufficient to irradiate the entire inclusion in every case. Typical results are shown in Figs. 2(a) and 2(b). The spectra are dominated by the L x rays from Th atoms which make up about 1% of the ~5×10^{16} atoms in a typical inclusion. Emission lines ($L\alpha_1$, $L\beta_1$, $L\beta_2$, etc.) from SHE (with $Z = 105$–129), if present, should appear in the 22- to 31-keV region. The spectrum ob-

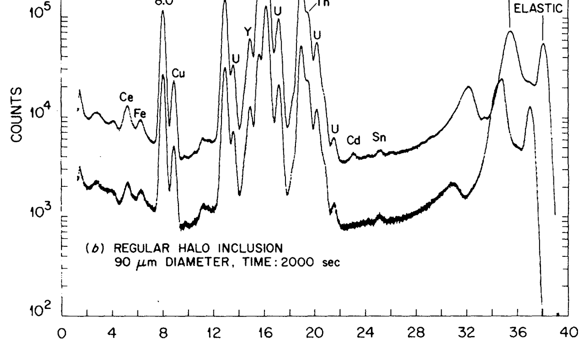

FIG. 2. Typical fluorescence spectra from (a) giant-halo and (b) regular-halo inclusions excited by synchrotron radiation.

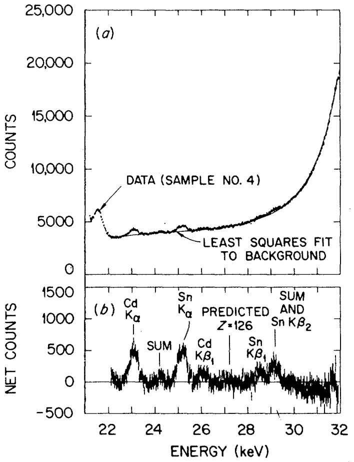

FIG. 3. (a) Expanded spectrum in the 21–32-keV region and (b) the difference between the actual counts and those from the background fit.

TABLE I. Summary of results.

Samples[a]	Th counts[b] (10^{-5})	Th atoms[c] (10^{-14})	S[d] (10^{11})	D[e] (10^{-9})
1	38.5	12.6	3.4	6.6
2	21.2	0.9	8.9	1.4
3	2.4	0.9	9.2	2.4
4	67.7	9.0	3.5	4.2
5	29.7	4.1	10.4	3.0
6	13.3	13.1	2.3	11.3
7	31.1	3.0	9.3	2.5
8	8.9	0.6	15.4	1.4
9	9.6	1.3	12.9	2.3
10	6.9	1.6	7.8	4.0
11	6.2	1.4	2.8	2.8
12 (Cd)			3.7	
13 (Cs)			7.8	

[a] Samples 1–11 refer to monazite inclusions characterized by giant halos. Samples 12 and 13 refer to Cd and Cs standards with $(1.8\pm0.7)\times10^{11}$ and $(4.0\pm1.0)\times10^{12}$ atoms, respectively.

[b] Obtained by integrating the $L\gamma_1$ and $L\gamma_{2,3}$ peaks.

[c] From the size and nominal composition of the inclusion and from an assumed density of 5 g/cm^3, typical of monazite.

[d] Sensitivity of the present experiment for detecting $Z=126$ $L\alpha_1$ peak (27.25 keV) in units of counts/sec atom.

[e] Detectable number of atoms of $Z=126$ at 98% confidence level.

served in the 11- to 22-keV region is qualitatively similar to that in Ref. 1; lanthanide K x rays are absent in our data since the incident energy was kept below the K absorption edges of these elements in most of our spectra. The broad peak near 31 keV results from an inelastic resonance scattering process.[12]

The 21–32-keV region is shown more clearly in Fig. 3(a). After excluding the regions where peaks might be present, the data were fitted by nonlinear least squares with an expression of the form

$$y=(a_0+a_1x)\exp(a_2+a_3x+a_4x^2+a_5x^3+a_6x^4), \qquad (2)$$

where y is the counts per channel, x is the channel number, and the coefficients a_0 to a_6 were determined by the least-squares fit. The range of values for χ^2 per degree of freedom in these fits was 0.9–1.4 for 220 degrees of freedom. The difference between the actual counts and those predicted by a fit is shown in Fig. 3(b). Peaks from trace amounts of Cd were seen in spectra from two different giant-halo inclusions. All spectra contained Sn peaks attributable to trace amounts of Sn present in the Cu sample chamber and to the Sn solder in the vicinity of the detector diode. Apart from these peaks and weak sum peaks arising from the three intense peaks labeled with energies in Fig. 2(a), no other peaks were detectable above background.

The integrated peak intensities of the Th $L\gamma_{1,2,3}$ lines from the inclusions and of the $K\alpha_1$ lines from the Cd and Cs standards were combined with known cross sections to deduce the number of $Z=126$ atoms that would have been detected in each spectrum. This information is given in Table I. The sensitivity, S, is defined as N_i^f/tn_i given in Eq. (1). The detectable number of $Z=126$ atoms, D, is taken as $3\sqrt{N_b}/tS$ where N_b is the total number of background counts in an energy spread of 800 eV (~2.3 FWHM of the detector) centered at 27 keV. Variations in S result from fluctuations in the current stored in the ring, compositional differences among the inclusions, and corrections made for self-absorption of the Th $L\gamma$ x rays relative to 27.2-keV x rays. The counting statistics for each spectrum ($t=30$ to 150 min) also affect D.

It was reported in Ref. 1 that $Z=126$ was present in five out of six giant-halo inclusions studied, implying its presence in nearby giant-halo inclusions as well. Yet, with a sensitivity of $(2-12)\times10^9$ atoms per inclusion (see Table I), we did not detect $Z=126$ in any of the eleven inclusions studied. Our results show, at 98% confidence level, that none of the SHE (with $Z=105$ to 129) are present in the giant-halo inclusions studied at levels which are at least a factor of 10 less than that reported in Ref. 1.

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Fine Structure of the Magnetic Dipole States in ^{208}Pb

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The splitting of the magnetic dipole states in ^{208}Pb due to the admixture of two-particle, two-hole configurations is calculated. The qualitative structure of a one-particle, one-hole calculation remains but about 30% of the $M1$ strength is distributed over many levels above 8 MeV. The influence of the spin-dependent part of the interaction on the results is critically discussed.

For more than ten years experimentalists and theorists have investigated magnetic dipole states in nuclei. Originally most of the work was done for light nuclei.[1,2] Recently, however, these investigations have been extended also to the heavier ones. From a comparison of the experimental data with the theoretical results one expects information about the spin-dependent part of the interaction.[3] Among all the heavy nuclei, ^{208}Pb is the best example for such a comparison because the assumptions of the theoretical treatment are most reliable here. The structure of the magnetic dipole resonances in ^{208}Pb is mainly given by the two spin-orbit partners $\pi 1h_{9/2}$-$\pi 1h_{11/2}{}^{-1}$ and $\nu 1i_{11/2}$-$\nu 1i_{13/2}{}^{-1}$. Therefore, in a one-particle, one-hole (1p-1h) approximation [Tamm-Dancoff approximation, random-phase approximation (RPA)] one expects to obtain two 1^+ states which practically exhaust the total $M1$ transition strength to the ground state. The relative strengths and the energies of the two states, however, depend sensitively on the particle-hole interaction used.[4,5]