

The phonon spectrum of  $\text{LiJO}_3$  which belongs to the crystal symmetry class  $C_6$  has been reported earlier<sup>3</sup>. In extension of these measurements the author has made polariton scattering using the effects due to refractive indices described above. An argon laser with  $\lambda = 5145 \text{ \AA}$  was used to excite the spectra. The dimensions of the crystal sample were  $\sim 10 \times 10 \times 12 \text{ mm}^3$ . Using ordinary incident and scattered photons a shift of the polariton associated with the phonon at  $795 \text{ cm}^{-1}$  could be observed from  $795$  to  $770 \text{ cm}^{-1}$ . This result was already reported<sup>3</sup>. However, using extraordinary incident and scattered photons, the polariton could be observed moving to  $747 \text{ cm}^{-1}$  at  $\varphi = 0^\circ = x(zz)x$ . The same scattering arrangement showed, that in addition a polariton associated with the A-phonon at  $328 \text{ cm}^{-1}$  moved to  $264 \text{ cm}^{-1}$  at  $\varphi = 0,9^\circ$  inside the sample.

<sup>3</sup> R. CLAUS, H. W. SCHRÖTTER, H. H. HACKER, and S. HAUS-SÜHL, *Z. Naturforsch.* **24 a**, 1733 [1969].

## A Search for Element 114 in Lead Minerals via Neutron-induced Fission

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Recently the possibility has been raised that isotopes of superheavy elements (with  $Z$  between 110 and 114) might have half-lives of the order of  $10^8 \text{ y}^{1-3}$ . It might therefore be possible to detect these isotopes in earthly matter, although their present amount is of course not only governed by their half-life, but also by the amount originally formed.

The most direct methods for this search comprise the detection of the radioactivity ( $\alpha$ -decay or spontaneous fission), the detection of  $\alpha$ -decay or fission products, and the detection of fission tracks. An experiment of this sort has been undertaken by THOMPSON et al.<sup>4</sup>, who looked for prompt neutrons accompanying spontaneous fission events in a Pt-ore; they found an upper limit for the concentration of spontaneously fissioning isotopes of  $10^{-9}$  to  $10^{-6}$ , depending on the half-life assumed. Some inconclusive results on fission tracks in lead-containing materials have been published by FLEROV and PERELGIN<sup>5</sup>.

Beside these direct methods there are several indirect techniques. One of the most sensitive could be the detection of neutron-induced fission of the super-

Furthermore the polar TO-phonon at  $769 \text{ cm}^{-1}$  showed polariton behaviour using extraordinary incident and ordinary scattered photons. A shift from  $769$  to  $690 \text{ cm}^{-1}$  at direct forward scattering  $0^\circ = x(zy)x$  was recorded. No shift of this polariton could be observed using ordinary incident and extraordinary scattered photons<sup>3</sup>. Fig. 2 shows spectra of the  $E_1(\text{TO})$  polariton. Due to microscopic impurities in the sample backward scattering of the  $E_1$  phonon and some diffuse scattering of the strong A phonon at  $795 \text{ cm}^{-1}$  could not be eliminated. Furthermore it is interesting to see that the  $E_2$  phonon at  $822 \text{ cm}^{-1}$  will appear in the spectra recorded at greater scattering angles, as there is some  $z(xy)x$ -scattering in addition due to slightly changed geometry.

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heavy isotopes. WESOŁOWSKI et al.<sup>6</sup> tested placer platinum for the emission of fission fragment pairs whose total kinetic energy is greater than the  $172 \text{ MeV}$  originating from  $^{235}\text{U}$ ; they set an upper limit of  $6 \cdot 10^{-12}$  on the abundance of superheavy nuclides in platinum, assuming a thermal fission cross section equal to that of  $^{235}\text{U}$ .

We looked for neutron-induced fission in 3 lead minerals by the method of delayed neutron counting. The samples were exposed for 60 seconds to a flux of  $7.2 \cdot 10^{12}$  thermal neutrons/ $\text{cm}^2 \text{ sec}$  (fast flux:  $1.8 \cdot 10^{12} \text{ n/cm}^2 \text{ sec}$ ) by means of a pneumatic transfer system. After a cooling period of 25 seconds the samples were counted in a neutron counter for 60 seconds. This procedure gave a sensitivity of  $16'000$  counts/ $\mu\text{g } ^{235}\text{U}$ , while the total background (including the irradiated sample container) over the same counting period was 3.5 counts<sup>7</sup>. Sample weight was between 3 and 10 grams.

The minerals tested are

Cerussite ( $\text{PbCO}_3$ ) from Amada, Arizona, USA, Wulfenite ( $\text{PbMoO}_4$ ) from Los Lamentos, Mexico, and Galena ( $\text{PbS}$ ) from Treece, Kansas, USA.

Upon irradiation, all minerals showed delayed neutron emission which was suspected to be due to their uranium content. Therefore, the minerals were dissolved and lead was precipitated as  $\text{PbSO}_4$ , dissolved and reprecipitated as  $\text{PbCrO}_4$ , assuming suf-

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<sup>1</sup> S. G. NILSSON, C. F. TSANG, A. SOBICZEWSKI, Z. SZYMANSKI, S. WYCECK, C. GUSTAFSON, I.-L. LAMM, P. MÖLLER, and B. NILSSON, *Nucl. Physics A* **131**, 1 [1969].

<sup>2</sup> S. G. NILSSON, S. G. THOMPSON, and C. F. TSANG, *Phys. Letters* **28 B**, 458 [1969].

<sup>3</sup> H. MELDNER and G. HERRMANN, *Z. Naturforsch.* **24 a**, 1429 [1969].

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<sup>6</sup> J. W. WESOŁOWSKI, W. J. R. JEWELL, and F. GUY, *Phys. Letters* **28 B**, 544 [1969].

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ficient chemical similarity between the hypothetical eka-lead and lead to prevent a separation in these operations<sup>8</sup>. Furthermore, following the suggestion of JØRGENSEN and HAISSINSKY<sup>8</sup> that eka-lead would show certain chemical properties similar to  $Tl^{+1}$ , 50 mg of Tl were added to the solution of the minerals and the Tl separated as  $TlJ$ . When these separated Pb- and Tl-fractions were tested for delayed neutron emission, the count obtained did not exceed background. It was therefore concluded that the counts of the original minerals were indeed due to their uranium content (1 to 2 ppm for Cerussite and Wulfenite, and 0.01 ppm for Galena).

This negative result allows us to set an upper limit on the concentration of eka-lead in these minerals (Fig. 1). Contrary to the methods based on radioactive decay, the half-life of the nuclide sought does not enter into the relationship between recorded events and concentration with the method of delayed neutron counting. However, two other variables have to be considered: the number of delayed neutrons emitted per fission during the measuring interval and the fission cross section.

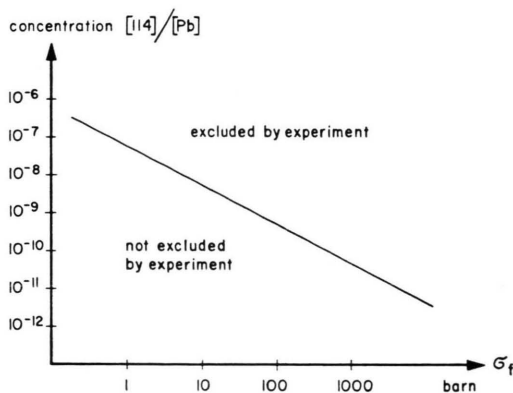


Fig. 1. Experimental upper limit on fissionable nuclides in lead.

The number of delayed neutrons counted with the irradiation and measuring cycle used in this experiment is in the case of  $^{235}U$  mainly governed by the fission yield of  $^{137}I$ , which contributes about 50% of the observed counts; further significant contributors are  $^{88}Br$  (about 28%) and  $^{87}Br$  (about 18%). It is certain that

in fission of  $Z=114$  the fission yield of the isotopes  $^{137}I$ ,  $^{88}Br$ , and  $^{87}Br$  will change. However it is difficult to foresee the amount of change. When it is assumed that for a fissioning nucleus  $^{298}114$  there will be ternary fission and  $\bar{\nu}$  (the average number of prompt neutrons emitted per fission) will be 10 (see<sup>9</sup>), then mass 137 will be near the center of the middle peak of the fission yield curve with a corresponding high yield; further, since the neutron/proton ratio of  $^{298}114$  is about that of  $^{235}U$ , the distribution of the independent yields within the mass chain should be about the same. Both arguments let us hope that the fission yield of  $^{137}I$  in  $^{298}114$  will not be depressed by more than an order of magnitude as compared to  $^{235}U$ .

As to the fission cross section, PRINCE<sup>10</sup> has suggested a qualitative correlation between the thermal fission cross section and  $(B_n - E_f)$ , where  $B_n$  is the neutron binding energy and  $E_f$  the threshold energy for fission. He has also given a semiempirical relation between  $E_f$  and the fissility  $Z^2/A$  for  $Z > 97$  (see<sup>11</sup>); if this relation is assumed to hold up to  $^{298}114$ , it gives approximately 4 MeV for  $E_f$ . A crude estimate indicates a value of 5 MeV for  $B_n$ ; this combination of  $(B_n - E_f)$  would indicate a thermal fission cross section of the same order of magnitude as  $\sigma_f$  for  $^{235}U$ . However, if 184 neutrons are indeed a magic number (and this is after all the assumption that leads to the expectation of longer lifetimes in this mass region),  $B_n$  should be lowered by about 2 MeV; this in turn would imply a thermal fission cross section below 1 barn. The fission cross section in the MeV-range is expected to be about 1 barn.

This uncertainty of the fission cross section entails a corresponding uncertainty in the upper limit on the concentration as indicated in Fig. 1. (Fig. 1 has been drawn with the assumption that the fission yields of the nuclides contributing to the count rate are the same as for  $^{235}U$ ). Based on the rather conservative assumption that an effect as big as the background would have been detected, the upper limit of  $[114]/[Pb]$  in the three tested minerals is  $9 \cdot 10^{-11}$  for a fission cross section of 580 barn and  $5 \cdot 10^{-8}$  for a fission cross section of 1 barn.

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<sup>10</sup> A. PRINCE, BNL-50168, 25 [1969].

<sup>11</sup> A. PRINCE, NBS Special Publ. **229**, 951 [1968].