Polaritons in $LiJO_3$

R. CLAUS

Sektion Physik der Universität München
Lehrstuhl Prof. BRANDMÜLLER

(Z. Naturforsch. **25 a**, 306—307 [1970]; received 17 January 1970)

Polaritons in $LiJO_3$ associated with the phonons of symmetry A at 328 and 795 cm^{-1} and symmetry E_1 at 769 cm^{-1} have been recorded using ordinary and extraordinary incident photons.

Using small angles between the wave vectors of the incident and scattered photons it is possible to observe Raman scattering by phonons or polaritons with small wave vectors which are determined by the momentum conservation equation. The photon wave vector inside a crystal with the refractive index n is $k = 2\pi n/\lambda$ where λ is the wave length in vacuum. In uniaxial crystals there are an ordinary and an extraordinary ray with refractive indices n_0 and n, respectively. One can get two different phonon wave vectors at the same scattering angle using ordinary incident and extraordinary scattered photons or vice versa. Fig. 1 illustrates the

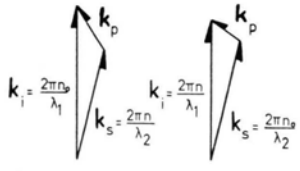

Fig. 1. Dependence of polariton wave vectors on refractive indices of incident and scattered photons.

phenomenon when $n_0 > n$. When making scattering experiments due to non diagonal elements of the Raman tensor this effect can be used in order to get minimum phonon wave vectors [1]. As totally symmetric phonons or polaritons will be observed only due to the diagonal elements, where both incident and scattered photons are ordinary or extraordinary, the method is not adequate in this case. An approximation of the momentum conservation equation $\boldsymbol{k}_i = \boldsymbol{k}_s + \boldsymbol{k}_p$ (i = incident, s = scattered, p = phonon) for small angles φ between $\boldsymbol{k}_i$ and $\boldsymbol{k}_s$, however, shows that the phonon wave vector still is a function of the refractive index [2]:

$$k_p = \left(\frac{n^2}{c^2}\,\omega_p^2 + k_i\,k_s\,\varphi^2\right)^{\frac{1}{2}}$$

(c = velocity of light in vacuum and ω_p = frequency of the phonon). In crystals with strong birefringence as for instance $LiJO_3$ a greater shift of A-polaritons will be observed using photons with minimum refractive index.

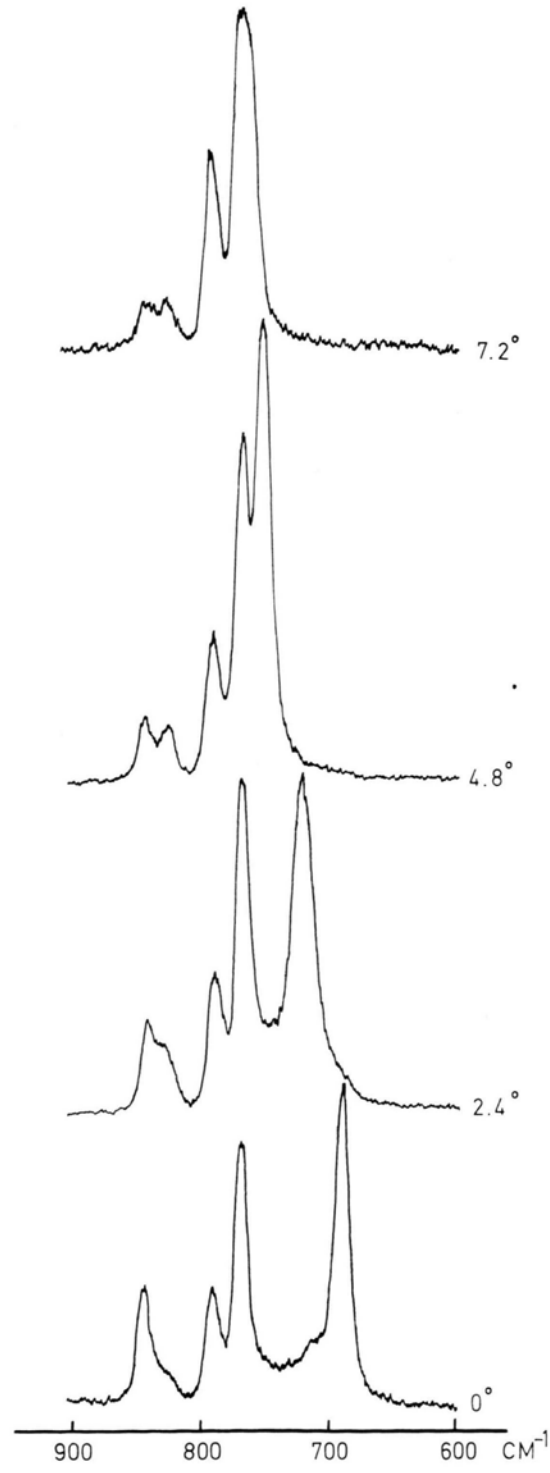

Fig. 2. Polariton associated with the E_1(TO) phonon at 769 cm^{-1} in $LiJO_3$. Scattering angles are those inside the sample. →

Sonderdruckanforderungen an R. CLAUS, Lehrstuhl Brandmüller, *D-8000 München 13*, Amalienstr. 54/IV.

[1] S. P. S. PORTO, B. TELL, and T. C. DAMEN, Phys. Rev. Lett. **16**, 450 [1966].

[2] C. H. HENRY and J. J. HOPFIELD, Phys. Rev. Lett. **15**, 964 [1965].

The phonon spectrum of $LiJO_3$ which belongs to the crystal symmetry class C_6 has been reported earlier [3]. In extension of these measurements the author has made polariton scattering using the effects due to refractive indices described above. An argon laser with $\lambda = 5145$ Å was used to excite the spectra. The dimensions of the crystal sample were $\sim 10 \times 10 \times 12$ mm³. Using ordinary incident and scattered photons a shift of the polariton associated with the phonon at 795 cm^{-1} could be observed from 795 to 770 cm^{-1}. This result was already reported [3]. However, using extraordinary incident and scattered photons, the polariton could be observed moving to 747 cm^{-1} at $\varphi = 0° = x(z\,z)x$. The same scattering arrangement showed, that in addition a polariton associated with the A-phonon at 328 cm^{-1} moved to 264 cm^{-1} at $\varphi = 0{,}9°$ inside the sample. Furthermore the polar TO-phonon at 769 cm^{-1} showed polariton behaviour using extraordinary incident and ordinary scattered photons. A shift from 769 to 690 cm^{-1} at direct forward scattering $0° = x(z\,y)x$ was recorded. No shift of this polariton could be observed using ordinary incident and extraordinary scattered photons [3]. Fig. 2 shows spectra of the E_1(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 cm^{-1} could not be eliminated. Furthermore it is interesting to see that the E_2 phonon at 822 cm^{-1} will appear in the spectra recorded at greater scattering angles, as there is some $z(x\,y)x$-scattering in addition due to slightly changed geometry.

I want to thank Prof. S. HAUSSÜHL, Cologne, who has grown the crystals and prepared the samples, and the Deutsche Forschungsgemeinschaft for financial support.

[3] R. CLAUS, H. W. SCHRÖTTER, H. H. HACKER, and S. HAUSSÜHL, Z. Naturforsch. **24 a**, 1733 [1969].

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

A. WYTTENBACH

Eidg. Institut für Reaktorforschung, Würenlingen

(Z. Naturforsch. **25 a**, 307—308 [1970]; received 9 December 1969)

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 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 (α-decay or spontaneous fission), the detection of α-decay or fission products, and the detection of fission tracks. An experiment of this sort has been undertaken by THOMPSON et al. [4], who looked for prompt neutrons acompanying 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 PERELYGIN [5].

Beside these direct methods there are several indirect techniques. One of the most sensitive could be the detection of neutron-induced fission of the superheavy isotopes. WESOLOWSKI et al. [6] tested placer platinum for the emission of fission fragment pairs whose total kinetic energy is greater than the 172 MeV originating from ^{235}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}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/cm² sec (fast flux: $1.8 \cdot 10^{12}$ n/cm² 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/μg ^{235}U, while the total background (including the irradiated sample container) over the same counting period was 3.5 counts [7]. Sample weight was between 3 and 10 grams.

The minerals tested are

Cerussite ($PbCO_3$) from Amada, Arizona, USA,

Wulfenite ($PbMoO_4$) from Los Lamentos, Mexico, and

Galena (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 $PbSO_4$, dissolved and reprecipitated as $PbCrO_4$, assuming suf-

Sonderdruckanforderungen an Dr. A. WYTTENBACH, c/o Eidg. Institut für Reaktorforschung, *CH-5303 Würenlingen*, Schweiz.

[1] 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].

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

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

[4] S. G. THOMPSON, R. C. GATTI, L. G. MORETTO, H. R. BOWMAN, and M. C. MICHEL, UCRL-18667, 227 [1969].

[5] G. N. FLEROV and V. P. PERELYGIN, Atomnaya Energiya **26**, 520 [1969].

[6] J. W. WESOLOWSKI, W. J. R. JEWELL, and F. GUY, Phys. Letters **28** B, 544 [1969].

[7] A. SCHUBIGER, A. COMUNETTI, J. MOLLES, and A. WYTTENBACH, Interner Bericht TM-CH-103 [1969].

ficient chemical similarity between the hypothetical eka-lead and lead to prevent a separation in these operations [8]. Furthermore, following the suggestion of JØRGENSEN and HAISSINSKY [8] 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 intervall and the fission cross section.

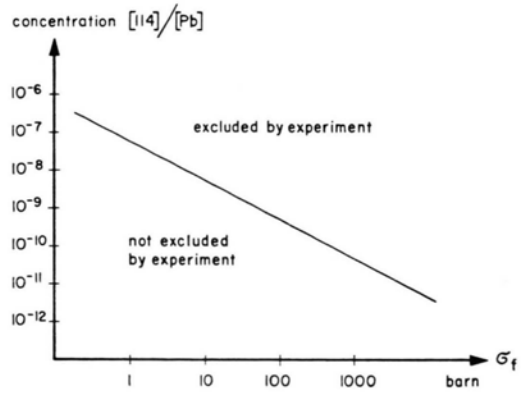

Fig. 1. Experimental upper limit on fissionable nuclides in lead.

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 [9]), 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 [10] 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 semiepirical relation between E_f and the fissility Z^2/A for $Z > 97$ (see [11]); if this relation is assumed to hold up to $^{298}114$, it gives approximatly 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 σ_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 livetimes 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 an $5 \cdot 10^{-8}$ for a fission cross section of 1 barn.

Thanks are due to Prof. TAUBE (EIR) and to Prof. OSWALD (University of Zurich) for helpful discussions; the latter was also of great help in the choice and procurement of the lead minerals.

[8] C. K. JØRGENSEN and M. HAISSINSKY, Radiochem. Radioanal. Letters **1**, 181 [1969].
[9] J. R. NIX, Phys. Letters **30** B, 1 [1969].
[10] A. PRINCE, BNL-50168, 25 [1969].
[11] A. PRINCE, NBS Special Publ. **229**, 951 [1968].