COMMENTS

Comment on the Reported Evidence for Primordial Superheavy Elements

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Proton-induced x-ray studies of selected monazite grains show no evidence for primordial superheavy elements. It is demonstrated that the recently reported evidence for superheavy elements in monazite inclusions in biotite mica showing giant halo formation is due to a misinterpretation of the observed x-ray spectra.

Crystalline monazite inclusions showing giant halo formation in biotite mica were recently investigated by Gentry *et al.*[1] by the method of proton-induced x-ray emission. The resulting x-ray spectra have been interpreted as evidence for primordial superheavy elements with $Z=116$ (or 127), 124, and 126; the deduced abundances of these elements in the inclusions are of the order of 10^2 to 10^3 ppm.

In an attempt to verify these results, we have performed a systematic investigation of monazite crystals (typical grain diameters $\gtrsim 1$ mm) from different occurrences by means of ion-induced x-ray analyses with a low-energy proton beam of $E_p = 2.0$ MeV. In particular, we investigated some of the monazite grains (referred to as M1) left several years ago by Dr. Gentry at the disposal of Professor Kirsten of Heidelberg; according to Dr. Gentry, these grains were picked up in the vicinity of a biotite layer where some giant halos were found.[2] From the intensity of the characteristic x-ray lines (Fig. 1), it can be deduced that the chemical composition of the monazite grains M1 agrees, except for a difference in the uranium content of $\approx 50\%$, with that of the normal and giant halo inclusions investigated by Gentry *et al.*[1]; this result is expected for monazite crystals from the same ore body. However, no evidence for primordial superheavy elements in the monazite samples M1 or in all other monazites investigated has been found. The detection limit for ele-

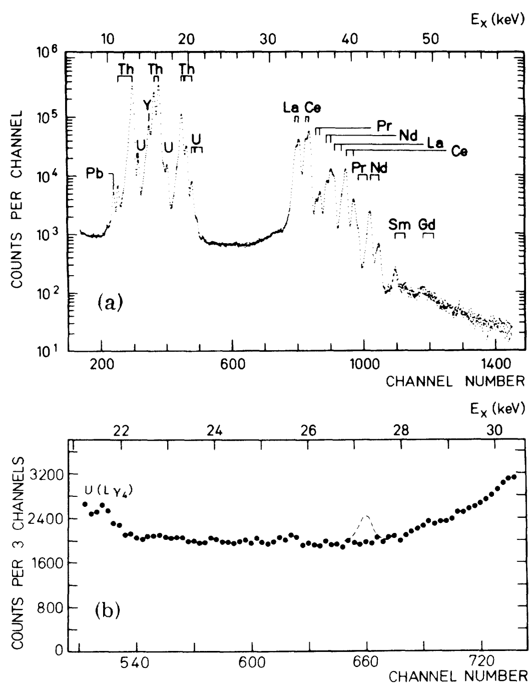

FIG. 1. (a) X-ray spectrum observed in the bombardment of the monazite grains M1 with a 2-MeV proton beam using a 3-mm-thick Si(Li) detector, a lead collimator, and an aluminum absorber of 0.7 mm. (b) The gap region of the spectrum shown in (a) between 21 and 30 keV. The dashed line represents the expected $L\alpha_1$ line of element $Z=126$ assuming an abundance as quoted in Ref. 1 for this element in giant halo inclusion 19 D. Pile-up peaks are expected around 25.9, 28.5, and 29.2 keV.

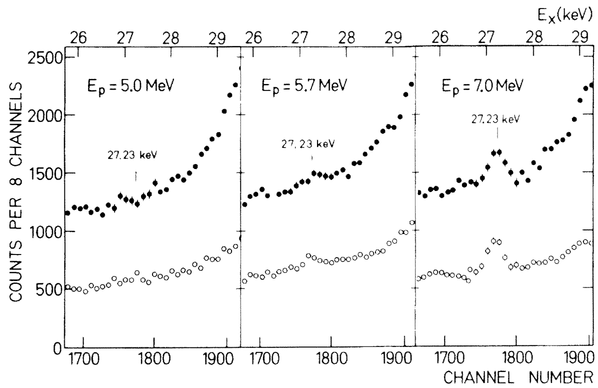

FIG. 2. Portions of the x-ray spectra obtained at three different proton energies from the monazite grains M1 and a thick Ce_2O_2 target using a 3-mm-thick Si(Li) detector and a 0.7-mm-thick aluminium absorber. All spectra were normalized to equal intensities of the Ce $K\alpha_{1,2}$ line measured for the monazite at $E_p = 5.0$ MeV.

ments around $Z = 125$ was of the order of 100 ppm.

In similar experiments performed at higher proton energies ($E_p \geq 5.0$ MeV, a γ-ray line at $E_\gamma = 27.23 \pm 0.04$ keV has been observed, which also appears in the x-ray spectra obtained by bombarding a thick cerium target (Fig. 2). This γ ray, being observed recently also by Fox *et al.*[3] in the proton bombardment of an isotopic enriched ^{140}Ce target, is very likely due to the γ decay of the first excited state of ^{140}Pr (Peker, Sigalov, and Kharitonov[4]) produced in the reaction $^{140}Ce(p, n)\,^{140}Pr^+$; within experimental errors, its energy is identical with the energy of the line tentatively interpreted in Ref. 1 as being due to the $L\alpha_1$ x ray of element $Z = 126$. The intensity ratio observed at a proton energy of $E_p = 5.7$ MeV for the monazite grains M1 as well as for the thick Ce_2O_3 target amounts to $I(27.23\ \text{keV}):I(\text{Ce}\ K\alpha_{1,2}) = (2.5 \pm 0.5) \times 10^{-4}$.

In view of our negative result as to the presence of superheavy elements expecially in the monazite crystals M1, and because of the occurrence of a γ-ray line for proton energies $E_p \gtrsim 5$ MeV which may interfere with the observation of the $L\alpha_1$ line of element $Z = 126$, we re-examined the x-ray spectra for the normal and giant halo inclusion published in Ref. 1 and came to the following conclusions: (i) There are no statistically significant differences between the structures observed in the "gap" region between $E_x = 23$ keV and $E_x = 29$ keV of the x-ray spectra obtained for the normal halo inclusion 19 B and the giant halo inclusion 19 D [Figs. 1 and 2(a) of Ref. 1; the spectra were recorded using a proton beam of $E_p = 5.7$ MeV[5]]. The differences quoted in Ref. 1 are due to the normalization of the underlying background, which is unjustified since there is no reason to assume that the background in the gap region for 19 D is equal to that for 19 B if the two spectra are normalized to equal counts in the La $K\alpha$ lines. In fact, the differences disappear if the background in the gap region for 19 D is assumed to be enhanced by 6–8% compared to the background observed for 19 B. Such an increase of the background is consistent with the background enhancement observed at x-ray energies $\lesssim 50$ keV (see Fig. 1 of Ref. 1). Note, furthermore, that Fig. 2(c) of Ref. 1 is obviously inconsistent with Figs. 1 and 2(a) of Ref. 1. (ii) If the background is property subtracted, the intensity of the line around 27.2 keV observed by bombarding the giant halo inclusion 19 D as well as the normal halo inclusion 19 B with 5.7-MeV protons is of the order of 300 counts, which is in agreement with the estimated intensity of the 27.23-keV line from the γ decay of $^{140}Pr^*$ using the measured intensity ratio $I(27.23):I(\text{Ce}\ K\alpha_{1,2})$ and considering the finite thickness of the monazite inclusions *as well as* the slightly different detection efficiencies. Thus, we conclude that the 27.23-keV line which constituted the main evidence for primordial superheavy elements in giant halo inclusions[1] has to be ascribed to the reaction $^{140}Ce(p, n\gamma)^{140}Pr$. This conclusion is in disagreement with that of Ref. 3 which states that this γ ray is too weak to change significantly the results of Ref. 1. However, their intensity argument is based on the analysis of Ref. 1 where the intensity of the 27.23-keV line is grossly overestimated because of the unjustified background subtraction. All other structures in the gap region are either not satistically significant or can be interpreted without stretching the point as K x rays of known elements.

Therefore, we conclude that the experiments of Ref. 1 do not present any evidence for the existence of superheavy elements in nature.[6]

We wish to acknowledge the permission of Dr. Gentry, Oak Ridge National Laboratory, to use the monazite samples M1 for the present investigation. We are, furthermore, indebted to Professor Herrmann, Mainz-Darmstadt, and Professor Ramdohr, Heidelberg, for stimulating discussions.

[1]R. V. Gentry, T. A. Cahill, N. R. Fletcher, H. C.

Kaufmann, L. R. Medsker, J. W. Nelson, and R. G. Flocchini, Phys. Rev. Lett. 37, 11 (1976).

[2]In contrast to his former information, Gentry recently informed us that the monazite samples (M1) are from the same ore body, but more than 10 km away from where the biotite with giant halos have been found.

[3]J. D. Fox, W. J. Courtney, K. W. Kemper, A. H. Lumpkin, N. R. Fletcher, and L. R. Medsker, Phys. Rev. Lett. 37, 629 (1976).

[4]L. K. Peker, V. M. Sigalov, and Yu. I. Kharitonov, Nucl. Data Sheets 12, 343 (1974).

[5]J. W. Nelson, private communication.

[6]A more detailed account of our investigations will be published elsewhere.

Commensurate Ordering in Tetrathiafulvalene-Tetracyanoquinodimethane

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Is is suggested that the first-order instability in tetrathiafulvalene-tetracyanoquinodimethane [(TTF)-(TCNQ)] at 38 K is related to the fourth-order umklapp coupling which concerns the wave vectors of the star involved in the three-dimensional periodic ordering. Within the anharmonic TCNQ interchain-coupling model, we predict that the configuration of chain deformation changes abruptly at 38 K, provided that this coupling is attractive.

The recent structural investigations[1] on (TTF)-(TCNQ) have revealed the existence of a peculiar three-dimensional ordering of the Peierls deformations ($q_b = 0.295b^*$) in the temperature range between 54 and 38 K.

Between 54 and 49 K, the most unstable wave vector has a component in the direction of alternating chains equal to $a^*/2$; below 49 K, this component starts to decrease. This effect was thoroughly discussed in the recent work by Bak and Emery,[2] where it was attributed to the bilinear coupling of the intrinsically different Peierls deformations on TCNQ and TTF chains. A similar explanation was also mentioned, but only briefly, in an earlier paper by Šaub, Barišić, and Friedel.[3] Here we accept such an interpretation of the 54-K and 49-K transitions, but for the third transition at 38 K we wish to suggest an alternative explanation to that proposed in Ref. 2.

At 38 K, q_a jumps to the commensurate value $q_a = a^*/4$ and remains pinned to this value at lower temperatures. In Ref. 2, this effect was attributed to the fourth-order umklapp coupling in which the $(a^*/4, q_b)$ wave on TCNQ chains, taken to the third power, is umklapp-coupled to the $(a^*/4, -3q_b)$ wave of the TTF chains. However, the full crystal symmetry also allows the fourth-order umklapp coupling of the waves involved in the star $(\pm a^*/4, \pm q_b)$ [Fig. 1(a)]. In principle, four coupled waves can belong either to TCNQ or to TTF chains and mix together in the fourth-order invariants. Obviously, this alternative does not imply any additional scattering at 38 K such as that expected at $-3q_b = 0.115b^*$ for the mechanism of Ref. 2.

We have performed the actual calculations in the model which retains only the coupling of the

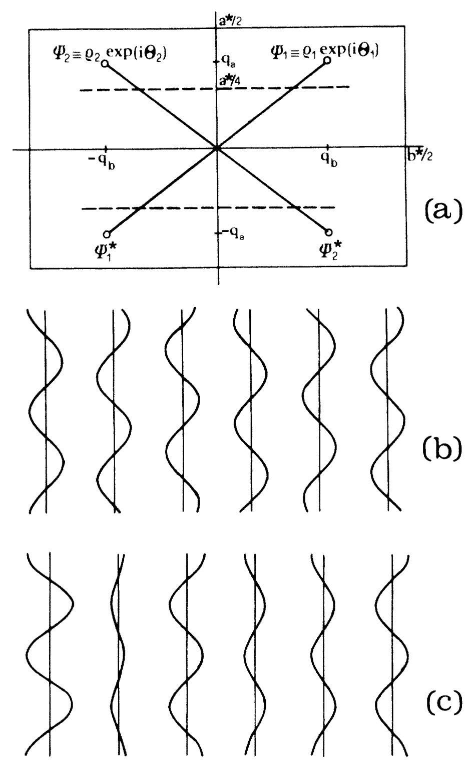

FIG. 1. (a) The star of wave vectors $(\pm q_a, \pm q_b)$. (b) The phase-shift chain deformations corresponding to the solution with only two waves ψ_1, ψ_1^* being different from zero. (c) The "amplitude-wave" configuration involving all four waves in the star with the same amplitude.