

## Radiohalos: Some Unique Lead Isotope Ratios and Unknown Alpha Radioactivity

**Abstract.** *Previously unreported lead isotope ratios, that is, values for the lead-206/lead-207 ratio ranging from about 20 to 60, primarily radiogenic in origin but unsupported by uranium decay, have been determined in the inclusions of certain polonium halos by means of ion microprobe techniques. Evidence for radiogenic lead-208 unsupported by thorium decay may also be inferred from the existence of a composite polonium halo type with rings from the radioactive precursors of lead-208. Several new dwarf halo sizes seem to indicate the existence of unknown, very low-energy alpha-emitters. Furthermore, the three-ring "X halo" also provides evidence for an unknown series of genetically related alpha-emitters with energies in the range from 3 to 7 million electron volts.*

An intriguing aspect of the study of radioactive halos (radiohalos) is the occurrence of unusual halo varieties (1-3), some of which remain unassociated with known  $\alpha$ -emitters. There has been speculation of late that certain of these variant halos may be related to the existence of superheavy elements (4), and, although no definite evidence presently exists in support of this hypothesis, investigations of a variety of radiohalo types have revealed several new results that may be summarized as follows: (i) The ion microprobe mass spectrometer has been utilized to determine the Pb isotope ratios in the halo inclusions of radiohalos previously associated with a Po isotope decay sequence terminating with  $^{206}\text{Pb}$ . Several factors, including values for the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio ranging from about 20 to 60, suggest a unique history for the Pb in the Po halo inclusions as compared to that of previously observed lunar or terrestrial Pb (5, 6). (ii) A new type of composite radiohalo has been found with rings attributable both to the  $^{218}\text{Po}$  decay sequence and to  $^{212}\text{Po}$  and possibly  $^{212}\text{Bi}$  (iii) In addition to the dwarf halos (5.2 and 8.6  $\mu\text{m}$  in radius) found by Joly (1), I have discovered a variety of dwarf halo sizes ranging from 1.5 to 11  $\mu\text{m}$  in radius. If we assume a radioactive origin, the extremely small dwarf sizes (1.5 to 2.5  $\mu\text{m}$ ) correspond to  $\alpha$  energies,  $\alpha$ , of approximately 1 Mev, less than that of any known  $\alpha$ -emitters.

In the past, identification of various radiohalo types was primarily dependent upon a correlation of the halo ring radius with a specific  $\alpha$  energy derived from a range-energy relation for the host mineral in which the halos occurred. This technique was successful in identifying radiohalos that could easily be ascribed to  $\alpha$  decay in the halo inclusion from the  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay chains as well as radiohalos that

apparently matched the sequential  $\alpha$ -decay patterns from  $^{218}\text{Po}$  (three rings),  $^{214}\text{Po}$  (two rings), and  $^{210}\text{Po}$  (one ring). Although the ring structure of this latter group of radiohalos seemed perfectly compatible with the proposed Po isotopic designation (2), some questions have arisen over this identification because of the half-lives of the respective isotopes involved ( $t_{1/2} = 3$  minutes for  $^{218}\text{Po}$ ) and the lack of evidence for a secondary source of Po from  $\alpha$ -decaying precursors in the U decay chain (7). Evidence in support of this identification is now available in the form of mass spectrometric analyses of the Po halo inclusions made with the ion microprobe, a new type of mass spectrometer with a sputtering ion source (8), which makes possible an analysis of the halo inclusions in situ.

The particular halo type analyzed possessed ring structure that correlated with the  $^{218}\text{Po}$   $\alpha$ -decay sequence (Fig. 1A) which terminates with  $^{206}\text{Pb}$  and thus it was suspected that the halo inclusion would reflect an excess of  $^{206}\text{Pb}$  when compared to previously observed isotopic abundances of either common or radiogenic Pb. Such proved to be the case. In general, although variations were noted in the Pb isotope ratios in the U and Th halo inclusions, these ratios were within the range of previously reported values (5). In contrast, the Pb isotope patterns of the Po halos revealed ratios of  $^{206}\text{Pb}$  to  $^{208}\text{Pb}$  of  $\approx 2.2$  and of  $^{206}\text{Pb}$  to  $^{207}\text{Pb}$  of  $\approx 19$  for one inclusion and ratios of  $^{206}\text{Pb}$  to  $^{208}\text{Pb}$  of  $\approx 6$  and of  $^{206}\text{Pb}$  to  $^{207}\text{Pb}$  of  $\approx 60$  for a second inclusion. Uranium was virtually nonexistent in the first analysis and constituted only a small fraction of the Pb content in the second analysis (9). These results were confirmed when a different ion microprobe was used on a third Po halo inclusion (10), and the following ratios were observed:  $^{206}\text{Pb}/^{208}\text{Pb} \approx 33$ ,

$^{206}\text{Pb}/^{207}\text{Pb} \approx 20$ ,  $^{206}\text{Pb}/^{204}\text{Pb} > 1000$ , and  $\text{Pb}/\text{Th}$  and  $\text{Pb}/\text{U} > 5000$ , that is, no  $^{204}\text{Pb}$ , U, or Th was detected.

It is difficult to associate the Pb isotope ratios and Pb/U and Pb/Th ratios in these inclusions with any previously reported sources of terrestrial, meteoritic, or lunar Pb. For example, common Pb, as generally defined, refers to Pb assumed to consist of a primordial component containing the isotopes  $^{204}\text{Pb}$ ,  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$ , and  $^{208}\text{Pb}$ , and a radiogenic component containing the isotopes  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$ , and  $^{208}\text{Pb}$ ; however, the high Pb/U and Pb/Th values characteristic of phases containing common Pb imply that the radiogenic component was not produced by in situ decay of U or Th. Clearly then, the absence of or low abundance of U or Th in the third halo inclusion cannot of itself be taken as an indication of the presence of significant quantities of common Pb, since  $^{204}\text{Pb}$  was not detected. Likewise, this Pb could hardly be characterized as radiogenic Pb in the sense of being derived from the in situ decay of U and Th, because these elements were absent. This Pb is, however, radiogenic in the sense of being derived from Po decay.

Additional evidence for an unusual history of these leads may be seen from the relatively high  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios, which for common Pb is about unity in many terrestrial rock types and for radiogenic Pb [produced by the in situ concurrent decay of  $^{238}\text{U}$  (to  $^{206}\text{Pb}$ ) and  $^{235}\text{U}$  (to  $^{207}\text{Pb}$ )] is in the range of  $\approx 4$  to 15 (5). The theoretical maximum possible radiogenic  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio, on the basis of an instantaneous production of Pb from U decay, would be 21.8. Thus the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios in the Po halo inclusions clearly differ from similar ratios previously observed either in common or in radiogenic Pb from U decay.

Rather than attempting to ascribe these unusual ratios to the general phenomenon of differential movement of U, Th, or Pb, I consider the evidence in these specific cases to provide confirmation for the existence of  $^{206}\text{Pb}$  from Po decay unsupported by U, especially in view of the fact that the highest value of the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio ( $\approx 60$ ) was recorded in the most densely colored Po halo of the three. Further studies of this type may lead to a reevaluation of the basic premise that there is a unique origin for radio-genic Pb, namely, that it originates solely from U and Th decay. Because of the small size of the halo inclusions ( $\sim 2 \mu\text{m}$ ), such variations in Pb isotope ratios would easily have escaped prior detection

with almost any other mass spectrometric techniques. Indeed, such variations may exist only in certain halo inclusions.

In a related context it appears that in certain cases small quantities of  $^{208}\text{Pb}$  [and possibly  $^{207}\text{Pb}$  (11)] may have an origin similar to that of the excess  $^{206}\text{Pb}$  referred to in the above analysis. I have found a halo with rings consistent with the  $\alpha$ -decay pattern from  $^{212}\text{Po}$  and possibly  $^{212}\text{Bi}$  as well as the  $^{218}\text{Po}$  decay sequence (see Fig. 1, B and C). This halo and similar halos with rings from the  $\alpha$  decay of  $^{212}\text{Po}$ ,  $^{210}\text{Po}$ , and  $^{212}\text{Bi}$  occur in very low abundance in the micas from the Faraday Mine near Bancroft, Ontario. By analogy with the above results, it would be expected that these composite Po halos would exhibit a Pb isotope ratio pattern high in  $^{206}\text{Pb}$  and  $^{208}\text{Pb}$  without corresponding amounts of the parent nuclides  $^{238}\text{U}$  and  $^{232}\text{Th}$ . Indeed, at this time I cannot rule out the

possibility that some of the  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$  (as well as the  $^{206}\text{Pb}$ ) observed in the ion microprobe analyses might also have originated with Po or Bi decay unsupported by U or Th. Halo rings from the radioactive precursors of  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$  would not necessarily have formed as indicated by the lower abundance of these isotopes. In an earlier study no support was found for a secondary source of Po originating with  $\beta$ -decay precursors, that is, no variation in  $\beta$ -recoil density near the halo inclusions as compared to the background density (7).

Many years ago Joly (1) reported the existence of some very unusual dwarf halos with radii of approximately 5.2 and 8.5  $\mu\text{m}$  in the black micas from the pegmatite quarry at Ytterby (near Stockholm). The halo with a radius of 5.2  $\mu\text{m}$  was later tentatively associated with the decay of  $^{147}\text{Sm}$  ( $E_\gamma = 2.24 \text{ MeV}$ ), whereas the larger halo has never received a satisfactory nuclide identification (12, 13).

The extreme rarity of the occurrence of the dwarf halos apparently has precluded much research on the nature of their origin, and it has been only recently that I have found a few samples of mica from the old Ytterby quarry that contain any significant number of these unique halos for further study. Joly considered the radioactive origin of the halos beyond question and tentatively attributed their bleached appearance to an overexposed, radiation-damaged condition or perhaps to some other phenomena related to the metamorphic history of the mica. I consider the radiogenic origin to have been confirmed when Mahadevan (14) later reported equivalent dwarf halo sizes in an Indian cordierite sample, which also contained Po halos. Generally  $\beta$ -particles from (n,  $\beta$ ) reactions are far too insufficient to produce a halo.

My observations on the dwarf halos

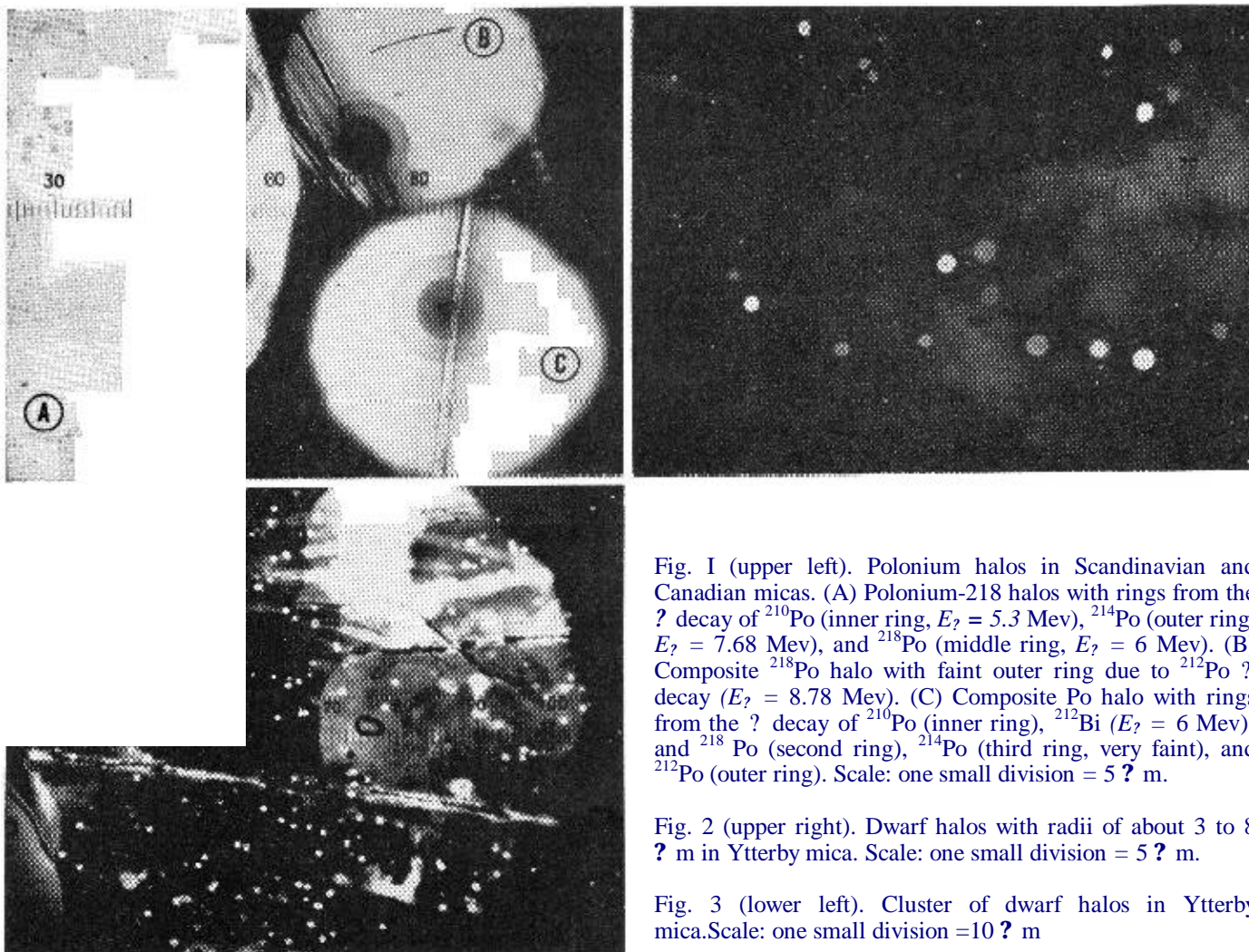


Fig. 1 (upper left). Polonium halos in Scandinavian and Canadian micas. (A) Polonium-218 halos with rings from the  $\alpha$  decay of  $^{210}\text{Po}$  (inner ring,  $E_\gamma = 5.3 \text{ MeV}$ ),  $^{214}\text{Po}$  (outer ring,  $E_\gamma = 7.68 \text{ MeV}$ ), and  $^{218}\text{Po}$  (middle ring,  $E_\gamma = 6 \text{ MeV}$ ). (B) Composite  $^{218}\text{Po}$  halo with faint outer ring due to  $^{212}\text{Po}$   $\alpha$  decay ( $E_\gamma = 8.78 \text{ MeV}$ ). (C) Composite Po halo with rings from the  $\alpha$  decay of  $^{210}\text{Po}$  (inner ring),  $^{212}\text{Bi}$  ( $E_\gamma = 6 \text{ MeV}$ ), and  $^{218}\text{Po}$  (second ring),  $^{214}\text{Po}$  (third ring, very faint), and  $^{212}\text{Po}$  (outer ring). Scale: one small division = 5  $\mu\text{m}$ .

Fig. 2 (upper right). Dwarf halos with radii of about 3 to 8  $\mu\text{m}$  in Ytterby mica. Scale: one small division = 5  $\mu\text{m}$ .

Fig. 3 (lower left). Cluster of dwarf halos in Ytterby mica. Scale: one small division = 10  $\mu\text{m}$ .

are, in general, in accord with those of Joly, except that I have found several additional sizes that are somewhat difficult to account for on the basis of  $\alpha$ -decay systematics of known radioactive nuclides. I find that the smallest dwarf halos range from only 1.5 to about 2.5  $\mu$ m with associated  $\alpha$  energies in the range of approximately 1 Mev. The half-lives of known  $\alpha$ -radioactive nuclides are in excess of  $10^{13}$  years for  $\alpha$ -decay energies of 2 Mev or less and thus normally correspond to such weakly active nuclides as to almost escape detection and would hardly be expected to produce a halo at all. These considerations do not seem in accord with the appearance of both some very small and some intermediate-sized dwarf halos, which show intense bleaching or reversal effects characteristic of a highly radiation-damaged region. The inference is that, whatever the difference in the half-lives of the nuclides responsible for the various dwarf halo sizes, the half-lives of the nuclides involved do not produce measurable differences in the degree of halo development.

Further, I have found other dwarf halos in the size range from 3 to 11  $\mu$ m (see Figs. 2 and 3) corresponding to  $\alpha$  energies ranging from approximately 1.1 to 3.4 Mev. The dwarf halos in this range do reflect some coloration differences which tentatively may be attributed to varying concentrations of parent radionuclides in the halo inclusions. Uranium and Th halos in a given hand specimen often exhibit similar effects for this reason. Although the dwarf halos which exhibit the most intense reversal effects have proved very satisfactory for radius measurements ( $r \sim 0.25 \mu$ m), the variety of sizes has thus far precluded the construction of a discrete radius distribution. (Some overlap in the distribution may result from the appearance of the same type of halo in later stages of development.) Generally speaking, the radius of an ordinary halo in the initial stages of development may vary a micrometer or two from that in later stages, possibly as a result of an increased ionization effect near the terminal range of the  $\alpha$ -particle in the mineral, and a reduced effect of this nature may be operating in the dwarf halos. Interestingly, there is a close correspondence between some of these halo sizes and some very low-energy  $\alpha$ -emitters ( $E_\alpha = 1.1$  to 1.3 Mev, 1.8 to 2.0 Mev, 2.5 to 2.7 Mev, and 3.1 to 3.3 Mev) of unknown origin observed over the last several decades by Schintlmeister

(15), Bruki et al. (16), and Gysae (17). Whether there is a causal relation between the dwarf halos and these previously reported  $\alpha$  activities is presently open to question. The significant result is that  $\alpha$  activities in the range of a few million electron volts apparently exist apart from the naturally occurring, rare-earth  $\alpha$ -emitters with low decay energy (16, 17).

Further observations on the dwarf halos have shown some with an elliptical cross section as opposed to the ordinary circular cross section seen in thin section under the microscope. In certain cases this ellipticity arises from the fact that the long ( $> 10 \mu$ m) filament-like halo inclusion is inclined somewhat with respect to the cleavage plane. In other cases a shearing-type deformation of the crystal may have produced this effect, although it is not certain that either of these observations is the full explanation of this effect. Another observation I consider important is the presence of dual-ring dwarf halos, which must be considered similar in nature to other radiohalo types which exhibit ring structure, that is, U or Th halos. I believe that the rarity of the dual-ring dwarf halos implies that the nuclides involved are not necessarily genetically related in a decay sequence.

A most significant point is that the bleached regions immediately surrounding the inclusion etch extremely rapidly, a characteristic of highly radiation-damaged regions. The bleaching phenomenon exists in the same micas in overexposed U and Po halos. After etching thousands of these minute halos, I have found a few which exhibit fission tracks with a range of only about one-half to three-fourths the normal fission-track length. I was at first inclined to consider these tracks the result of lower-energy fission events but am as yet unable to rule out the possibility that they are U fission tracks that have been annealed somewhat through a metamorphic episode in the rocks. Since, however, fission-track measurements on these dwarf halos generally reveal a low content of U in the inclusions, an attempt was made to resolve their origin by searching for additional short-range tracks in unetched samples by means of high-voltage electron microscopy. Results have thus far been negative (18).

Equally as rare as the dwarf halos are the X halos reported by Joly (1) and later referred to by other investigators (5, 9, 12).

I include a summary of Joly's measurements here. The inside ring may be somewhat diffuse and measures about 8.5 to 9.8  $\mu$ m in radius. The bleached rings extend out to a radius of approximately 14 to 15  $\mu$ m with an adjacent dark ring at about 17  $\mu$ m, whereas the outer wide band extends to approximately 28  $\mu$ m. Actually, two different types of X halos exist, one with and one without a bleached band. This particular halo is due neither to U nor to Th decay in spite of some close similarities with certain portions of the Th decay sequence. Rings in the X halo may be attributed to  $\alpha$  energies of approximately 2.9 to 3.2 Mev (inner ring), 4.5 to 5.2 Mev (middle rings), and 6.8 to 7.0 Mev (outer ring). The chart of the nuclides reveals no genetically related  $\alpha$  sequence corresponding to these energies, and it is extremely difficult to concoct a mixture of  $\alpha$ -emitters that would fit the above pattern without additional rings also being present.

Although an earlier suggestion relating the inner ring of the X halo to the spontaneous fission of U (19) is not tenable, the possibility of a combined fissionogenic and  $\alpha$ -derived origin for the inner ring involving other nuclides must not be overlooked. It seems clear that both the X halos and the dwarf halos may be evidence of presently unidentified  $\alpha$  activity. Detailed ion microprobe studies of the halo inclusions may very well elucidate the nature of this activity.

*Note added in proof:* I recently visited several European research centers to investigate earlier reports of low-energy  $\alpha$  activity cited in (15–17). F. Hergner, Radium Institute, University of Vienna, carefully reviewed his collaborative efforts with Schintlmeister. Several of their unpublished reports written during World War II (20) give additional evidence of genetically related  $\alpha$  energies of about 3.1 and 4.5 Mev—an interesting pattern when compared with the X-halo ring structure.

G. Herrmann, University of Mainz, called my attention to the work of R. Coppens, University of Nancy, France, whom I subsequently visited. The low-energy  $\alpha$  activity found by Coppens (21) and other earlier investigators (15–17) may be related to the radioactive origin of the dwarf halos.

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## References and Notes

1. J. Joly, *Proc. Roy. Soc. Ser. A Math. Phys. Sci.* 102, 682 (1923).
2. G. H. Henderson, *ibid.* 173, 250 (1939).
3. E. Wiman, *Bull. Geol. Inst. Univ. Upps.* 23, 1 (1930); R. V. Gentry, *Appl. Phys. Lett.* **8**, 65 (1966); *Earth Planet. Sci. Lett.* **1**, 453 (1966); *Science* **169**, 670 (1970); S. Imori and J. Yoshimura, *Sci. Pap. Inst. Phys. Chem. Res. Tokyo* 5, 11 (1926).
4. H. Meidner and G. Herrmann, *Z. Naturforsch. A* 24, 1429 (1969); H. Meier, W. Albrecht, D. Bdsche, W. Hecker, P. Menge, A. Ruckdeschel, E. Unger, G. Zeitler, E. Zimmerhacki, *ibid.* 25, 79 (1970).
5. B. Doe, *Lead isotopes* (Springer-Verlag, New York, 1970).
6. L. T. Silver, in *Proceedings of the Apollo 11 Lunar Science Conference*, A. A. Levinson, Ed. (Pergamon, New York, 1970), vol. 2, p. 1553.
7. R. V. Gentry, *Science* 160, 1228 (1968).
8. C. A. Andersen, J. R. Hinthorne, K. Fredrikson, *Proceedings of the Apollo 11 Lunar Science Conference*, A. A. Levinson, Ed. (Pergamon, New York, 1970), vol. 1, p. 159.
9. R. V. Gentry; paper presented at the Apollo 12 Lunar Science Conference, January 1971. A small U signal was observed in this analysis, originating in the Po halo inclusion or in another inclusion nearby. This is in accord with my recent observation that fission tracks emanate from some Po halo inclusions, which is perhaps not surprising in view of the ubiquitous nature of U. Even in these cases, however, the U content is anywhere from ten times to several thousand times less than the requisite amount of Po necessary for threshold coloration. The halos were in pegmatitic mica samples from Norway (Iveland district). These analyses were performed on the ion microprobe at Applied Research Laboratories, Goleta, California.
10. These analyses were performed by Dr. J. McHugh on the new ion microprobe mass analyzer at the Knolls Atomic Power Laboratory. This mica sample was from a Canadian pegmatite.
11. R. V. Gentry, *Nature* **213**, 487 (1967). (Radio nuclides attributed to the fluorite halos are in error.)
12. G. Hevesy and M. Pahl, *ibid.* **131**, 434 (1933); J. H. J. Poole, *ibid.*, p. 654. Association of the 5.2- $\mu$  m halo with  $^{147}\text{Sm}$   $\alpha$ -decay is unconfirmed.
13. E. E. Picciotto and S. Deutsch, "Pleochroic Halos" (Comitato Nazionale per l'Energia Nucleare, Rome, 1960).
14. C. Mahadevan, *Indian J. Phys.* **1**, 445 (1927); see also J. S. van der Lingen, *Zentralbl. Miner. Abt. A* **1926**, 177 (1926). Through the courtesy of D. Lal and A. V. Bagayatkhar, Tata Institute of Fundamental Research, Bombay, I have recently received some cordierite samples from the location where Mahadevan obtained his material but have not had opportunity to examine these samples in detail as yet.
15. J. Schintlmeister, *Oesterr. Akad. Wiss. Math. Naturwiss. Kl. Sitzungsber. Abt. IIa* **144**, 475 (1935); *ibid.* 145, 449 (1936).
16. A. Brul, F. Hergner, H. Hilbert, *ibid.* 160, 129 (1951).
17. B. Gysae, *Z. Naturforsch. A* 5, 530 (1950).
18. Work was carried out at U.S. Steel's Edgar C. Bain Laboratory for Fundamental Research, Monroeville, Pennsylvania.
19. G. N. Flerov, *J. Phys. USSR* 6, 49 (1942).
20. J. Schintlmeister and F. Hergner, "On a Previously Unknown Alpha Emitting Chemical Element," *German Rep. G-55, Part 1* (June 1940); "Further Chemical Investigations of the Element with Alpha Rays of 1.8 Centimeters Range," *German Rep. G-h12, Part 11* (May 1941); J. Schintlmeister, "The Position of the Element with Alpha Rays of 1.8 Centimeters Range in the Periodic System," *German Rep. G-111, Part lii* (May 1941); Prospects for Energy Production by Nuclear Fission of the 1.8 Centimeter Alpha Emitter," *German Rep. G-186* (February 1942).
21. R. Coppens, *Compt. Rend.* 243, 582 (1956).
22. Research sponsored by the U.S. Atomic Energy Commission under contract with Union Carbide Corporation and by Columbia Union College, Takoma Park, Maryland, with the assistance of grants from the National Science Foundation (grant GP-29510) and the J. F. Schneider Memorial Foundation. I thank J. A. Mandarino and R. I. Gait, Royal Ontario Museum, Toronto; L. Moyd, National Museum of Canada, Ottawa; and I. Bryhni, Mineralogical and Geological Museum, Oslo, for supplying some of the mica samples.

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