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# Crystal Radii of the Heavy Elements*

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Crystal radii for the trivalent and tetravalent ions of the heavy elements from actinium to americium as deduced from crystal structure data are listed and briefly discussed.

THE writer has had the opportunity to carry out extensive crystal structure studies on compounds of elements 89 to 95.

Most of the compounds for which complete crystal structures have been deduced correspond to the trivalent or tetravalent states of these elements. The experimental crystal structure results show that the chemical binding is predominantly ionic for these valence states. For the purpose of crystal chemical considerations it is accordingly useful to introduce crystal radii for these ions.

The variation of lattice periods with atomic number for compounds of the heavy elements is illustrated in Tables I and II, which give the experimental results for the dioxides and the trifluorides. The dioxides have the fluorite type of structure, the trifluorides the tysonite type of structure. The lattice periods given for the trifluorides refer to the hexagonal cell containing two molecules.

TABLE I. Dioxides with fluorite type of structure.

| Compound | $a$ |
|---|---|
| $ThO_2$ | 5.5859±0.0005A |
| $UO_2$ | 5.4568 0.0005 |
| $NpO_2$ | 5.425 0.001 |
| $PuO_2$ | 5.386 0.001 |
| $AmO_2$* | 5.372 0.005 |

* Degree of purity uncertain.

TABLE II. Trifluorides with tysonite type of structure.

| Compound | $a_1$ | $a_3$ |
|---|---|---|
| $AcF_3$ | 4.27 ±0.01A | 7.53 ±0.02A |
| $UF_3$ | 4.138 0.003 | 7.333 0.004 |
| $NpF_3$ | 4.108 0.001 | 7.273 0.004 |
| $PuF_3$ | 4.087 0.001 | 7.240 0.001 |
| $AmF_3$* | 4.071 0.002 | 7.225 0.004 |

* See reference 1.

The crystal radii given in Table III have been corrected for the effect of coordination number[1] and correspond to six coordination. The radii have been deduced from the observed interatomic distances in oxygen and fluorine compounds using crystal radii of 1.33A for $F^-$ and 1.40A for $O^{-2}$. The crystal radii for the lanthanide series of ions are shown for comparison.

Many investigators have predicted that a 5*f* series of elements would begin near the end of the periodic system. The crystal structure studies of the writer provide proof of the correctness of these early predictions. In contrast to the 4*f* series of elements, there is not one but two prominent valence states for the heavy elements so far known. The trivalent state is known for actinium, uranium, neptunium, plutonium, and americium, but not for thorium and protactinium. The tetravalent state has been observed for all the elements in the series from thorium to americium.

Accordingly, one should speak of a thoride series for the tetravalent state, and of an actinide series for the trivalent state,

The slow, monotonic decrease in crystal radius with increasing atomic number, characteristic of the filling of the *f*-shell, is clearly demonstrated in Table III. A similar decrease in the size of the $(XO_2)^{+2}$ radical in going from uranyl to neptunyl to plutonyl compounds has been

TABLE III. Crystal radii.

| Number of *f*-electrons | Thoride series | | Actinide series | | Lanthanide series | |
|---|---|---|---|---|---|---|
| 0 | $Th^{+4}$ | 0.95A | $Ac^{+3}$ | 1.11A | $La^{+3}$ | 1.04A |
| 1 | $Pa^{+4}$ | (0.91) | ($Th^{+3}$ | 1.08) | $Ce^{+3}$ | 1.02 |
| 2 | $U^{+4}$ | 0.89 | ($Pa^{+3}$ | 1.06) | $Pr^{+3}$ | 1.00 |
| 3 | $Np^{+4}$ | 0.88 | $U^{+3}$ | 1.04 | $Nd^{+3}$ | 0.99 |
| 4 | $Pu^{+4}$ | 0.86 | $Np^{+3}$ | 1.02 | $61^{+3}$ | (0.98) |
| 5 | $Am^{+4}$ | 0.85 | $Pu^{+3}$ | 1.01 | $Sm^{+3}$ | 0.97 |
| 6 | | | $Am^{+3}$ | 1.00 | $Eu^{+3}$ | 0.97 |

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* The results given in this article are based on work carried out under Contract Number W-31-109-eng-38 for the Atomic Energy Project at Argonne National Laboratory.

[1] W. H. Zachariasen, Zeits. f. Krist. **80**, 137 (1931).

observed. This would, of course, correspond to a uranide contraction. In the subnormal valence states the variation of crystal radius with increasing atomic number shows irregularities which are not understood at present.

The new compounds listed in Tables I and II were prepared by various chemists as follows: $NpO_2$ by T. J. LaChapelle and L. B. Magnussen, $PuO_2$ by P. Kirk; $AmO_2$ and $AmF_3$ by Sherman Fried, $AcF_3$ by Sherman Fried and F. Hagemann; $UF_3$ by J. Karle, $NpF_3$ by Sherman Fried, A. E. Florin, and N. R. Davidson; $PuF_3$ by A. E. Florin and R. E. Heath.

Detailed accounts of the various crystal structure investigations upon which the results given in this article are based will appear in Division IV of the National Nuclear Energy Series (Manhattan Project Technical Section).

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# The Question of Non-Singular Solutions in the Generalized Theory of Gravitation

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It has been already proved by Einstein and Pauli that in the relativistic theory of gravitation the field equations do not admit any non-singular static solution representing a field of non-vanishing total mass. In this paper it is shown that this theorem is also valid in the generalized theory of gravitation with non-symmetric $g_{ik}$.

## 1

It has been proved first by Einstein[1] and then by Einstein and Pauli[2] that in the relativistic theory of gravitation (with symmetric $g_{ik}$) the field equations do not admit any non-singular static (i.e., with all components $g_{ik}$ independent of $t$) solution which would represent a field of non-vanishing total mass. In the second paper the proof has been given for a space with a number of dimensions $n \geqslant 4$ in order to extend the validity of the theorem to the 5-dimensional theory of Kaluza.

The question arises whether this theorem is also valid in the generalized theory of gravitation with non-symmetric $\mathfrak{g}_{ik}$.[3] It will be shown in this paper that this question can be easily answered by using another proof of the theorem established by Einstein and Pauli. This second proof can be extended to the generalized theory, thus leading us to the result that *the same theorem is also valid in the generalized theory of gravitation.*

## 2

In this section we recapitulate the main points of the proof given by Einstein and Pauli for the theory with symmetric $g_{ik}$. This proof is based on the assumption that the solution is embedded in a Euclidean space, i.e., that at large distances the tensor $g_{ik}$ tends to the metrical tensor $\gamma_{ik}$ of flat space:

$$g_{ik} \rightarrow \gamma_{ik} \quad \text{for} \quad r \rightarrow \infty, \tag{1}$$

where

$$\begin{gathered} \gamma_{11} = \gamma_{22} = \gamma_{33} = -1, \quad \gamma_{44} = 1, \\ \text{all other } \gamma_{ik} = 0. \end{gathered} \tag{2}$$

Then from the field equations

$$R_{ik} = 0$$

the following relation is shown to be valid in the case of static fields:[4]

$$(\mathfrak{g}^{4i}\Gamma_{4i}{}^{k})_{,k} = 0.$$

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[1] A. Einstein, Revista (Universidad Nacional de Tucuman) **A2**, 11 (1941).

[2] A. Einstein and W. Pauli, Ann. Math. **44**, 131 (1943).

[3] A. Einstein and E. G. Straus, Ann. Mathematics **47**, 731 (1946); E. Schrödinger, Proc. Roy. I. A. **A51**, 163 (1947).

[4] This relation has been derived in the paper of Einstein and Pauli in a rather complicated way. But it follows immediately from the field equations, and even in the stronger form

$$(\mathfrak{g}^{li}\Gamma_{4i}{}^{k})_{,k} = 0,$$

if one takes into account that

$$\mathfrak{g}^{li}{}_{;k} \equiv \mathfrak{g}^{li}{}_{,k} + \mathfrak{g}^{si}\Gamma_{sk}{}^{l} + \mathfrak{g}^{ls}\Gamma_{ks}{}^{i} - \mathfrak{g}^{li}\Gamma_{ks}{}^{s} = 0,$$