PHYSICAL REVIEW
LETTERS

VOLUME 3 DECEMBER 15, 1959 NUMBER 12

NEUTRON AND FISSION FRAGMENT DAMAGE IN ZIRCONIA

J. Adam and B. Cox
United Kingdom Atomic Energy Authority, Harwell, England
(Received November 23, 1959)

Wittels and Sherrill[1] reported recently that the irradiation-induced phase transformation observed in natural zirconia[2] does not take place when synthetic ZrO_2 crystals or powders are exposed to neutrons. Their analysis indicates that considerable quantities of uranium are present in the natural material and it is suggested that the phase transformation is due to fission spikes.

Our work[3] confirms the conclusion that pure ZrO_2 does not undergo phase transformation under neutron bombardment but we did observe a monoclinic to cubic phase change in oxidized zirconium alloys, containing up to 5 weight percent of either vanadium, chromium, or tantalum under moderate neutron doses. Materials used in the preparation of these alloys were of commercially pure quality and later analysis did not reveal uranium contamination. This suggests that the phase transformation in ZrO_2 takes place only in materials containing impurities which may provide nuclei of a cubic or tetragonal phase. Uranium is known to stabilize tetragonal ZrO_2,[4] and, although fission fragments undoubtedly accelerate the transformation rate, without the stabilizing effect of the uranium impurity they would not produce the transformation. The evidence for this comes from our experiments in which we irradiated spectroscopically pure ZrO_2 with fission fragments from an outside source.

It was reported at the Second Geneva Conference[5] that phase transformation can be produced in thick oxide layers formed on zirconium alloys by bombarding them with fission products from a uranium oxide film placed opposite the ZrO_2 layers at a distance of about 1-2 mm. These experiments were subsequently repeated with oxides on sponge zirconium and on spectroscopically pure ZrO_2. The transformation was observed in all cases but it was also found that ZrO_2 was heavily contaminated with uranium which "evaporated" from the film. This "evaporation" of uranium is associated with the escape of fission fragments[6] and, although it cannot be avoided, uranium may be prevented from reaching the surface of ZrO_2 if a very thin aliuminum foil is placed between the fission fragment source and the specimen. The absorption of fission fragment energy in the aluminum foil is small when the latter is less than 1 micron thick.

Experiments with aluminum foils 0.7 micron thick showed that pure monoclinic ZrO_2 did not transform into cubic at doses which, in the absence of the aluminum foil, would have produced 100% transformation. A further experiment was performed in which the gap between the source and target was reduced from 2 mm to zero. By this means any effect due to the absorption of fission fragment energy in the aluminum foil was offset by elimination of the absorption in the air gap. A very small percentage transformation was found following this experiment, but this was ascribed to failure of the aluminum foil since a small α count indicating the presence of uranium was obtained from the target.

---

[1]M. C. Wittels and F. A. Sherrill, J. Appl. Phys. 27, 643 (1956).

[2] M. C. Wittels and F. A. Sherrill, Phys. Rev. Letters 3, 176 (1959).
[3] J. Adam and B. Cox, J. Nuclear Energy A11, 31 (1959) and Atomic Energy Research Establishment, Harwell Report M415, April, 1959 (unpublished).
[4] G. N. Wolten, J. Am. Chem. Soc. 80, 1772 (1958).
[5] Dawson, Cox, Murdoch, and Sowden, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958 (United Nations, Geneva, 1958), Paper P/46.
[6] B. V. Ershler and F. S. Lapteva, J. Nuclear Energy 4, 471 (1957).

SELF-ABSORPTION AND TRAPPING OF SHARP-LINE RESONANCE RADIATION IN RUBY

F. Varsanyi, D. L. Wood, and A. L. Schawlow
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received November 12, 1959)

As an aid in planning a microwave-optical double resonance experiment,[1,2] we have examined by high-resolution optical spectroscopy the details of the sharp-line fluorescence of ruby (Cr^{3+} in Al_2O_3).

The only transition which appears in the fluorescence of dilute ruby at 4°K is from $^2E(\bar{E})$ to $^4A_2$ and appears at 6934 A. Selection rules for this transition have been calculated,[3] and partially verified for absorption.[4] The Zeeman levels to be discussed here are those occurring for a magnetic field parallel to the symmetry axis and are shown in Fig. 1.

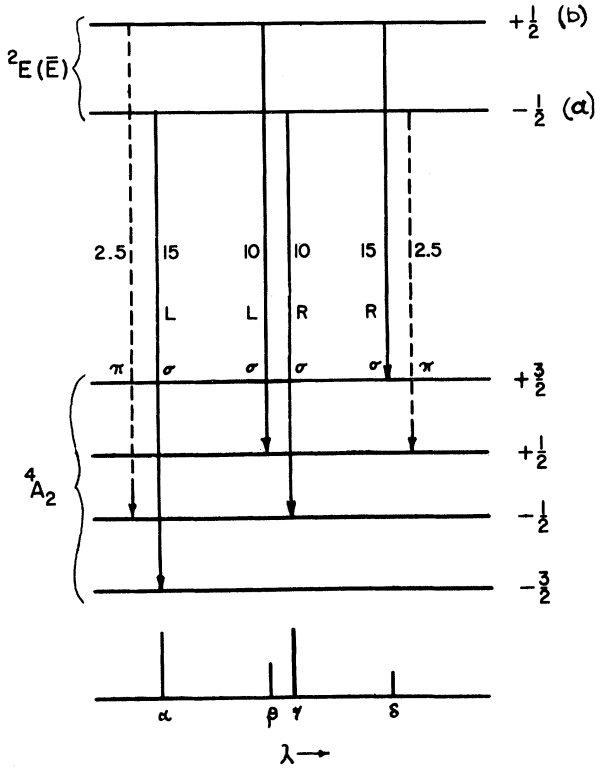

FIG. 1. Energy levels and transition probabilities for Zeeman levels involved in the resonance lines of ruby. $H \| c$ axis.

Figure 2(a) gives the predicted and observed intensities for a dilute ruby containing about 10^{-6} Cr per Al, and showing no color. For the theoretical patterns complete thermalization among the Zeeman levels is assumed in both ground and excited states. The figure shows that the selection rules predicting a 3:2 ratio of components α to γ and of δ to β are confirmed. Moreover, use of the α to δ and γ to β ratios shows that the population of atoms in the upper Zeeman level of the excited state is only slightly above that of a Boltzmann distribution. The effective spin temperature is 1.77°K. If it is assumed (although this is doubtful) that the excita-

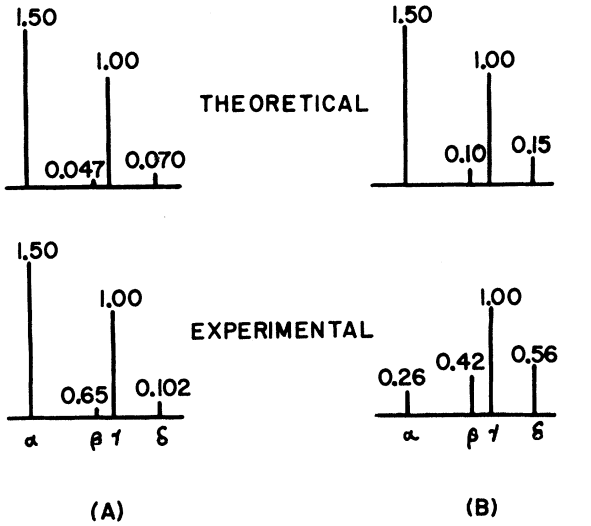

FIG. 2. (a) Theoretical and observed intensity ratio of Zeeman components in very dilute ruby sample (white sapphire). $H = 30\,000$ gauss; $T = 1.58$°K. (b) Theoretical and observed intensity ratio of Zeeman components in ruby containing 0.05% Cr_2O_3. $H = 30\,000$ gauss; $T = 2.1$°K.