

## Effect of X-Ray Irradiation on the Self-Diffusion Coefficient of Na in NaCl†

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The self-diffusion coefficient of the sodium ion in sodium chloride is decreased if the crystal is exposed to x-rays during the diffusion process. This effect was observed at temperatures under 550°C and was not observed at higher temperatures.

THE self-diffusion coefficient for the sodium ion in sodium chloride has been measured for samples which were subjected to x-ray irradiation during the diffusion process. The results indicate that the rate of diffusion is decreased by exposure to x-rays while diffusion is occurring.

The diffusion measurements were made using a radioactive tracer technique which was described in an earlier paper.<sup>1</sup> For the present work a small furnace was constructed to hold two crystal samples at the same temperature and permit radioactive sodium to diffuse from an evaporated film of sodium chloride on one face of each crystal. One crystal was situated about one inch from the window of a Machlett, type A-2, x-ray tube (molybdenum target) so that the evaporated face intercepted about 80 percent of the emergent x-ray beam. The x-ray tube was operated at 48 kv and 18 ma. The other sample was well shielded from the x-rays, and the two samples were located symmetrically in the furnace to minimize the possibility of dissimilar temperature gradients in the samples. The temperature was measured with thermocouples embedded in each sample. The temperature of the crystals was constant within  $\pm 1^\circ\text{C}$  during an experiment. The samples were cleaved from single crystal sodium chloride supplied by the Harshaw Chemical Company.

After the radioactive sodium had been allowed to diffuse the samples were sectioned, and the distribution of radioactivity as a function of distance from the evaporated face was compared for the two samples.

Solution of the diffusion equation for the one-dimensional case approximated in these experiments shows the concentration of radioactive sodium to vary with depth  $x$  and time of diffusion  $t$ , according to the relation

$$C = C_0 / (\pi Dt)^{1/2} \exp(-x^2/4Dt), \quad (1)$$

where  $C_0$  is a constant and  $D$  is the diffusion coefficient. Thus the points in a plot of logarithm of radioactive intensity versus distance squared should lie along a straight line whose slope is proportional to the diffusion coefficient. The data for one run have been plotted in this way in Fig. 1, and it will be noted that the points for the unirradiated sample do, in fact, fall

along a straight line as do the deepest points for the irradiated sample. It can also be seen from Fig. 1 that the diffusion rate for the irradiated sample is substantially smaller in a thin layer next to the evaporated face, since the slope in this region is steeper and also the general level of radioactivity below this region is lower than in the case of the unirradiated sample. Below the layer in which the diffusion rate is retarded, the diffusion rate for the irradiated sample is approximately the same as for the unirradiated sample. The same behavior was observed when the experiment was repeated with the crystals interchanged with respect to irradiation. Data taken at 527°C, 496°C, and 462°C showed the same effect illustrated in Fig. 1.

The occurrence of this effect depends upon the temperature at which the diffusion takes place. For sodium chloride as for the other alkali halide crystals there is a fairly well defined temperature above which the acti-

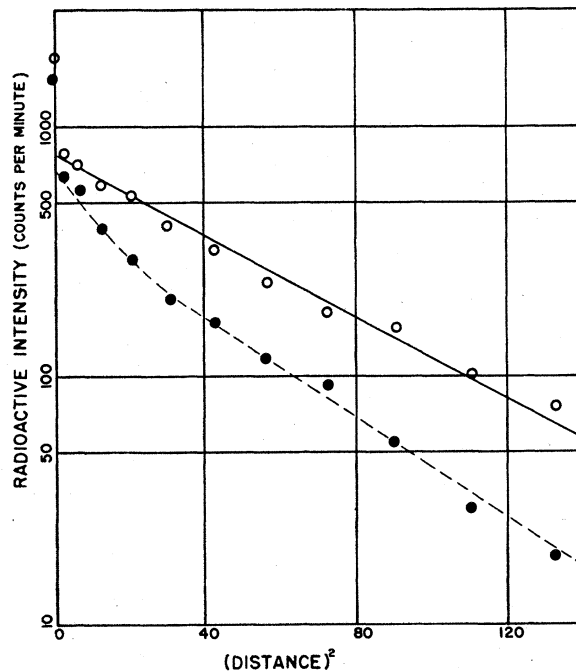


FIG. 1. Semilogarithmic plot of radioactive intensity versus square of distance below surface of crystal. Temperature of crystal 427°C. Diffusion time 3.75 days. Diffusion constant of unirradiated crystal  $1.0 \times 10^{-11}$  cm<sup>2</sup>/sec. Unit of length is  $4.84 \times 10^{-4}$  cm. O Unirradiated crystal. ● Irradiated crystal.

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<sup>1</sup> Mapother, Crooks, and Maurer, J. Chem. Phys. 18, 1231 (1950).

vation energy for diffusion has a distinctly higher value than in the lower temperature region. Above this temperature (which in the present case is about 550°C) the concentration of vacant lattice sites is believed to be determined by the temperature of the crystal and independent of the presence of impurity ions.<sup>1</sup> It is significant that the effect of x-ray irradiation shown in

Fig. 1 was not present when the same experiments were performed at 567°C and 604°C.

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## Inelastic Scattering of Neutrons by Cd<sup>111</sup>†

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Neutrons with an energy spread of  $\pm 40$  kev bombarded a disk of natural cadmium; among the products of bombardment was metastable Cd<sup>111m</sup> produced by inelastic scattering. The yield of Cd<sup>111m</sup> was measured from its threshold at 396 kev up to 1300 kev. Abrupt changes in the slope of the yield curve are interpreted as showing the onset of  $(n, n')$  excitation of higher levels. Such levels occur at 720 and 1150 kev. Below 720 kev, the Cd<sup>111m</sup> was excited only by direct transition from the ground state. We have measured at 720 kev the cross section for this  $(n, n')$  event, which involves known changes of energy, spin, and parity. The cross section is about 10 millibarns.

THE existence of nuclear isomerism provides in some cases a practicable means of studying the inelastic scattering of neutrons. We have exploited this possibility in the case of Cd<sup>111</sup>, a nuclide with an isomer, or metastable state, that has convenient half-life<sup>1,2</sup> and known spin.<sup>3</sup>

When a sufficiently energetic neutron is scattered from a nucleus, it may lose energy and leave the nucleus in an excited state. If this state is metastable, a gamma-ray or a conversion electron will be emitted later; detection of this radiation affords a means of tallying the inelastic scattering event. If the scattering leaves the nucleus in a state higher than the metastable one, the nucleus may make a transition to the metastable state before returning to the ground state. A curve

showing the excitation of the metastable state as a function of neutron energy will therefore have a change of slope whenever a new excited state becomes energetically possible, provided that this state can decay to the metastable one. Moreover, if the neutron energy is such that the metastable state can be excited directly but not by decay, all metastable excitations will be caused by an inelastic scattering that leaves the nucleus in a known state. By measuring the cross section for such creation of the metastable state we measure the cross section for a *particular* inelastic scattering process.

Wiedenbeck<sup>4</sup> excited the 49-minute state of Cd<sup>111</sup> by bombardment with x-rays and with electrons; he interprets these excitation curves as evidence for the existence of four excited levels above 1 Mev. Waldman and Miller<sup>5</sup> report two other levels in the same region. Hole,<sup>6</sup> using cadmium irradiated by neutrons, showed that the resulting metastable cadmium decays by the emission of two gamma-rays in cascade. Isotopically enriched cadmium showed<sup>7</sup> that the 49-minute state belongs to Cd<sup>111</sup>. The energies of the gamma-rays were measured by Hole<sup>6</sup> and also by McGinnis,<sup>8</sup> who gives the gamma-ray energies as 149.6 kev and 246 kev. The 49-minute level is therefore 396 kev above the ground state. Deutsch and Stevenson<sup>9</sup> found that the 246-kev

TABLE I. Decay of metastable Cd<sup>111</sup>.

Activity	49 min	$8 \times 10^{-8}$ sec
Conversion coefficient	2.2	0.06
K/L ratio	2.0	5.1
$\gamma$ energy	150 kev	246 kev
K conversion energy	123 kev	219 kev
L conversion energy	146 kev	242 kev
K x-ray	23 kev	23 kev

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<sup>1</sup> M. Dodé and B. Pontecorvo, *Compt. rend.* **207**, 287 (1938).

<sup>2</sup> M. L. Wiedenbeck, *Phys. Rev.* **66**, 36 (1944); *Helmholtz*, Hayward, and McGinnis, *Phys. Rev.* **75**, 1469 (1949).

<sup>3</sup> C. L. McGinnis, *Phys. Rev.* **81**, 734 (1951); *Phys. Rev.* **83**, 686 (1951); A. W. Sunyar, *Phys. Rev.* **83**, 864 (1951).

<sup>4</sup> M. L. Wiedenbeck, *Phys. Rev.* **67**, 92 (1945); see also reference 5.

<sup>5</sup> B. Waldman and W. C. Miller, *Phys. Rev.* **82**, 305 (1951).

<sup>6</sup> N. Hole, *Arkiv Mat. Astron. Fysik* **34B**, No. 19 (1947); also **36A**, No. 9 (1949).

<sup>7</sup> M. Goldhaber and C. O. Muehlhause, *Phys. Rev.* **74**, 1559 (1948).

<sup>8</sup> *Helmholtz*, Hayward, and McGinnis, *Phys. Rev.* **75**, 1469 (1949); C. L. McGinnis, *Phys. Rev.* **81**, 734 (1951).

<sup>9</sup> M. Deutsch and D. T. Stevenson, *Phys. Rev.* **76**, 184 (1949).