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THERMAL EQUILIBRIUM BETWEEN F AND M CENTERS IN POTASSIUM CHLORIDE

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Various models have been proposed for the M centers in alkali halides. According to Seitz[1] an M center consists of an F center associated with a neutral vacancy pair. A modification, having inversion symmetry, was put forward by Knox.[2] Van Doorn and Haven[3] have proposed a model consisting of a pair of associated F centers (F_2 model). From these models it is clear that the formation of M centers from F centers requires one F center for each M center formed in the case of the Seitz-Knox model and two in the case of the F_2 model.

It might be possible to distinguish between these cases by investigating (a) the kinetics of the reaction $F \rightarrow M$ and (b) the thermal equilibrium $F \rightleftharpoons M$. For the Seitz-Knox center the equilibrium can be written as

$$F + \text{vacancy pair} \rightleftharpoons M,$$

$$K = [M]/[F][\text{vac. pairs}];$$

and for the F_2 center as

$$2F \rightleftharpoons M, \qquad K' = [M]/[F]^2.$$

In the first case the M concentration will be proportional to the F concentration at a fixed temperature (the concentration of vacancy pairs being constant), whereas in the second case the M concentration will vary quadratically with the F concentration.

Theisen and Scott[4] found the thermal bleaching of F centers at about 300°C to be a second-order reaction, indicating that two F centers combine to form a new center. The thermal equilibrium between F and M centers was studied by Smith and Scott.[5] They colored a KCl crystal inhomogeneously by heating for a short time in potassium vapor in order to obtain a range of F concentrations. It was assumed that the attainment of the equilibrium between F and M centers is rapid compared with the diffusion of the centers in the crystal. The F and M concentrations in different regions of the crystal were measured optically, using a thin beam of monochromatic light. A linear dependence between F and M centers was found up to a concentration of 10^{17} F centers/cm^3. This experiment is, however, not conclusive because (a) no true equilibrium existed and (b) the high M/F ratio found indicates that the crystals were not properly quenched. Therefore, the following experiment was performed. KCl was purified by crystallizing it twice according to the Kyropoulos method. From the single crystal thus obtained, plates were cut having the dimensions $4 \times 5 \times 1.3$ mm^3. These were colored by heating to 697°C in potassium vapor of various pressures in a nickel tube. A heating time of 10 minutes was found to be sufficient to obtain equilibrium, as no further increase in optical absorption was observed with longer heating times. After quenching in carbon tetrachloride to room temperature, which was done in the dark to avoid photochemical reactions, the relative F and M concentrations were determined by measuring the extinction of the absorption bands at 77°K. With the apparatus used for coloring (to be described in a forthcoming publication) it took less than one second to transfer the crystal from the furnace to the quenching bath, thus ensuring freezing-in of the high-temperature equilibrium. All crystals had the same dimensions in order to obtain equal quenching rates.

The results are shown in Fig. 1. It is seen that the M concentration varies quadratically with the F concentration, which is inconsistent with the Seitz-Knox model but agrees well with the F_2 picture.

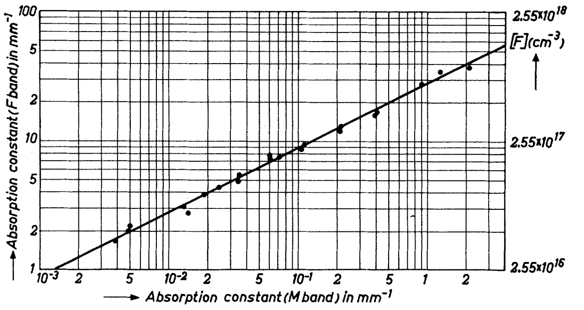

FIG. 1. Double-logarithmic plot of the concentrations of F and M centers in equilibrium at 697°C, expressed as absorption constants at 77°K of the F and M bands, respectively. The number of F centers per cm^3 is calculated from the Smakula formula with $f = 0.81$ and a half-value width 0.195 ev.

A rough determination of the equilibrium constant K' at other temperatures showed the temperature dependence to be small, corresponding to a heat of formation of the M center of about 0.01 ev.

I would like to thank Mr. A. T. Vink for his assistance.

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NUCLEAR RESONANCE FLUORESCENCE IN Au^{197}†

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We have observed the Mössbauer effect[1] in the 77-kev gamma-ray line of Au^{197}. A resonance curve from which the level lifetime may be deduced was obtained by moving a Pt^{197} source relative to a Au absorber. Resonance was found using as sources either the β^- parent, Pt^{197}, or the electron capture parent, Hg^{197}. We also find that the effect can be either enhanced or diminished by placing the source and/or absorber atoms in suitable crystal lattices. All measurements were performed at a temperature of 4°K.

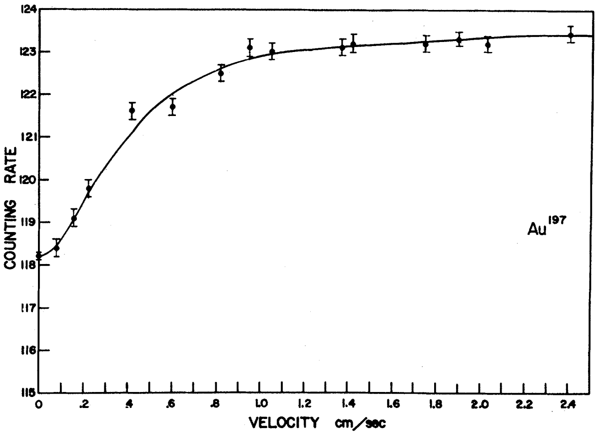

FIG. 1. Nuclear resonance absorption vs the magnitude of the source velocity in the 77-kev gamma ray of Au^{197}.

The shape of the resonance line was determined using a Pt^{197} source prepared by neutron activation of 60 mg of natural Pt. Pulse-height spectra taken at intervals for several days following activation showed a prominent gamma line at 77 kev, decaying with the mean life of about 26 hours, which is characteristic of Pt^{197}. Figure 1 shows the counting rate (corrected for source decay during measurement) versus the velocity of the source relative to a 200-mg/cm^2 Au absorber. No distinction between positive and negative velocities was made. The velocity half-width obtained is 3.5 ± 0.5 mm/sec. No evidence of subsidiary resonances was seen up to the highest velocity used, 24 mm/sec. The total resonance absorption effect is 5.5 ± 0.5 % after a correction (not shown in the figure) is made for background in the gamma-ray spectrum.

Using a source prepared by activation of enriched Pt (65.9 % Pt^{196}), measurements of resonance absorption were made on three Au absorber foils of thickness 100, 200, and 400 mg/cm^2. The net absorptions were, after correction for background (including the unresolved 60-kev x ray), 4.0 ± 0.6 %, 7.7 ± 0.5 %, and 11.5 ± 0.7 %, respectively. The velocity curve was similar