STUDIES ON THE TEXTURE OF CRYSTALS

Part I. NaCl, KCl and MgO

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Summary

The X-ray studies on the texture of crystals using internal reflections have been extended to three crystals, *viz.*, rock-salt, magnesium oxide and potassium chloride. The rock-salt specimen was found to be ideally mosaic and showed an increase in the intensity of internal reflection with increase of thickness, as is to be expected theoretically. The melt-grown MgO crystal was nearly perfect, while KCl was mosaic.

Introduction

In spite of the careful attention that has been given to the subject of crystalline texture, there is still no adequate classification of texture types nor any method of stating or measuring quantitatively the features which define the internal texture of a given crystal. Evans, Hirsch and Kellar (1948), Wooster and Macdonald (1948), Guinier and Tennevin (1949) and more recently Ramaseshan and Ramachandran (1953) have investigated this aspect using X-ray diffraction techniques. It has been observed that such studies give some indication of the extent to which the texture departs from that of perfect crystal. In all these studies, which were made on reflection from the surface planes and not from the interior, the emphasis has invariably been on the measurement of the absolute integrated reflection. Absolute measurements are always difficult and it would be preferable to have a method whereby measurements made on a single photograph could be used for obtaining the necessary information. In recent years, the effect of absorption on the intensities of X-rays reflected by crystals has been investigated both theoretically and experimentally. Bragg-reflections (X-rays reflected from a crystal surface) have been discussed by Hirsch and Ramachandran (1950). The case of Laue reflections (X-rays reflected through a crystal) has been studied by Laue (1950), Ramachandran and Kartha (1952), Hirsch (1952) and Zachariasen (1952). Ramachandran (1954) has also worked out the integrated values for the reflected and transmitted beams for perfect and mosaic crystals for an internal reflection.

104

These developments suggest a new method of studying the texture of a crystal. It has been shown theoretically by Ramachandran that in the case of an internal reflection the variation of the integrated reflection with thickness of the crystal would be different for perfect and mosaic crystals. Consequently, from the observed variation of intensity with the thickness of the crystal, one could get some idea about its texture. In actual practice, the intensity variation is photographically recorded in a single photograph by making use of a wedge-shaped crystal the thickness of which varies continuously from top to bottom. This new method has two advantages:— firstly, unlike other methods suggested earlier, the bulk of the reflection in this method occurs in the interior of the crystal so that it gives an idea of the degree of perfection of the crystal as a whole and not of the surface layers alone. Secondly, measurements need be made only on relative intensities on a single photograph and no absolute measurements are necessary. Preliminary investigations were made successfully on a melt-grown lithium fluoride crystal (Padmanabhan. 1953). These have now been extended to some typical cubic crystals. The present paper reports the results obtained in the case of three crystals namely, sodium chloride, potassium chloride and magnesium oxide.

2. Experimental Details

Two of the three crystals studied (MgO and KCl) were grown from melt, while the third one was the naturally occurring rock-salt. The experimental arrangement was the same as that described in the earlier note except that a Shearer tube with Mo target was used as the source of X-rays. A slit was kept close to the window of the X-ray tube which could be made either vertical or horizontal. After passing through a series of apertures, the X-ray beam fell on the crystal the distance between the slit and the crystal being 8 cm. The cleavage plate (010) was used in all the three cases, and the (100) planes used for obtaining the reflections were normal to the surface and vertical. One face of the crystal was ground at an angle so that the wedge tapered from top to bottom. The crystal was mounted on a goniometer capable of rotation about a vertical axis. In all the experiments symmetrical internal reflections were obtained from the 200 planes. The distance of the film from the crystal was 4·0 cm. and the film was always kept normal to the reflected beam. Photographs were first taken with the slit vertical. For studying the intensity variations with thickness the slit was made horizontal. The Bragg reflections were obtained in the form of tapering strips and as the slit was long enough, the intensity at any point along the centre of the strip would actually represent the integrated reflection for the corresponding thickness of the crystal. All the specimens, after

being ground in the form of a wedge were etched and photographs were obtained using both vertical and horizontal slits. The crystals were then dipped in liquid air (so as to render them mosaic by sudden cooling) and photographs were again taken of the reflected beams. The photographs thus obtained are reproduced in Figs. 2 and 3. In addition to the characteristic reflection a faint outline of the whole crystal is also recorded, due to continuous radiation.

The films obtained with the horizontal slit were run through a microphotometer and the curve so obtained was converted into a true intensity scale by making use of standard intensity marks obtained on another film by giving varying exposures for the surface reflection of the same crystal.

3. RESULTS AND DISCUSSION

The formulæ for the variation of the integrated reflection (ρ) with thickness for an ideally mosaic and perfect crystal are (Ramachandran, 1954)

$$\rho_M \propto Pe^{-P} \text{ (mosaic)}$$

$$\rho_P \propto e^{-P} \left\{ I_0 (KP/G) - 1 + \int_0^{P/G} J_0 (x)\, dx \right\} \text{ (perfect)}$$

where $P = \mu t/v$, $K = \chi_{hi}/\chi_{hr}$, $G = \chi_{oi}/\chi_{hr}$

Here μ is the absorption coefficient, $v = \cos \theta$, χ_{or} χ_{oi}, are the real and imaginary parts of the Fourier components of index zero of 4μ times the polarisability, χ_{hr}, χ_{hi} the coefficients corresponding to the (200) reflection and C polarisation factor. For large values of P/G as are met in the present study $\int_0^{P/G} J_0 (x)\, dx \rightarrow 1$ so that the formula for the perfect crystal reduces to

$$\rho_P \propto e^{-P} I_0 (jP), \text{ where}$$

$j = c \dfrac{\chi_{hi}}{\chi_{oi}}$ the polarisation factor in the case of crystals of NaCl type (Padmanabhan, 1953). An analysis of the formulæ reveal an interesting feature. In the case of an ideally mosaic crystal, if μ and t are sufficiently small, as t increases log ρ instead of decreasing exhibits an increase, reaching a maximum and thereafter show a decrease with further increase of thickness. The perfect crystal formula, on the other hand, shows a fall in intensity with increase of thickness.

Rock Salt.—To test the validity of the above statement, rock-salt being a typical mosaic crystal, was chosen for the study. As stated earlier, it was ground in the form of a wedge with thickness varying from ·4 to 3 mm. The thickness at the thin end could not be made smaller than ·4 mm. as

the crystal got bent due to plastic deformation. The internal reflection was photographed with the slit vertical. A positive reproduction of the photograph is shown in Fig. 2 *a*. For purposes of illustration the internal reflection of a LiF crystal (same dimensions) is also reproduced in Fig. 2 *b*. The log ρ *vs. t* theoretical curves for NaCl mosaic crystal, LiF perfect and mosaic crystals have been drawn. They are reproduced in Fig. 1. It can be seen from these curves that there is a difference in the intensity variations of internal
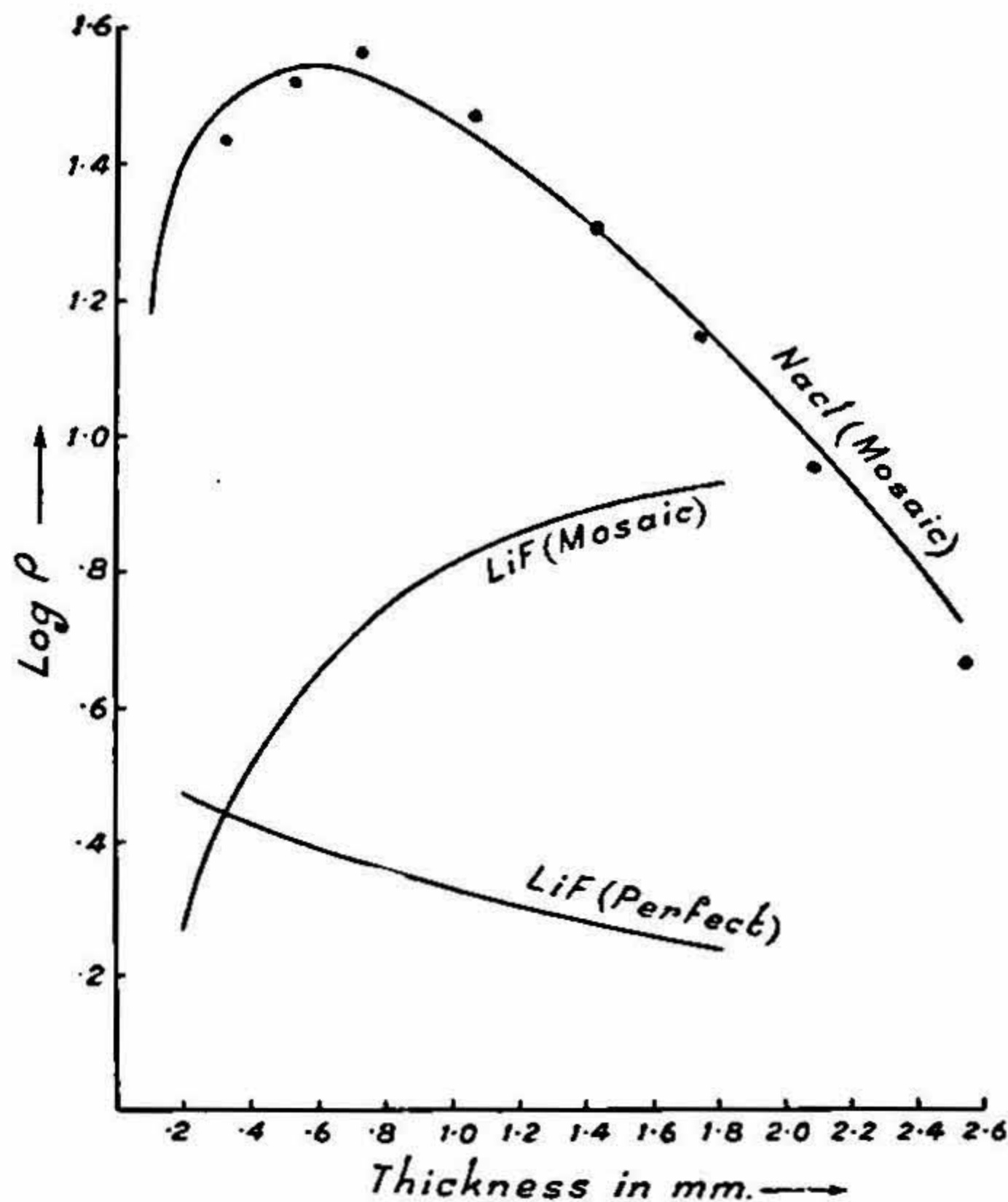

FIG. 1. Log ρ-*t* curves: continuous lines are drawn from theory. Dots represent the experimental points observed in NaCl.

reflection with thickness in the case of NaCl and LiF crystals. In the case of NaCl (as μ is larger than that for LiF) the intensity increases up to $t = \cdot 7$ mm. and thereafter indicates a rapid fall. On the other hand, in the case of LiF mosaic crystal, the intensity increases continuously up to 3 mm. and decreases only after that (only part of the curve is drawn). In the case of a perfect crystal of LiF, the intensity decreases gradually with increasing thickness. The reflection photograph for NaCl (Fig. 2 *a*) clearly shows an increase in intensity as we go down from top to bottom, attaining a maximum and then the intensity falls off as we reach the bottom. Fig. 2 *b* is the reflection of LiF, in which no such increase could be observed. This is to be expected,

a2

as the crystal of LiF is nearly perfect (Padmanabhan, 1953). The photographs of the internal reflections of the chilled specimens of LiF were also taken (not reproduced here) and it was observed that there was no difference between the chilled and etched crystals. Theoretically the mosaic LiF crystal should show an increase in intensity with increasing thickness. The liquid-air treatment might not have made the crystal ideally mosaic and hence the disparity between theory and experiment. The experimental points observed in NaCl are plotted in Fig. 1 and it can be seen that the points fall more or less on the theoretical curve, indicating that the rock-salt specimen is highly mosaic.

MgO.—The wedge used in the study had its thickness varying from ·4 to 3 mm. Fig. 3 *a* is the internal reflection after the crystal was etched. The edges alone appear and the internal portion is very feeble indicating that the bulk of the crystal is nearly perfect. This was verified by microphotometric study of the film obtained with the slit horizontal. In Fig. 4, the
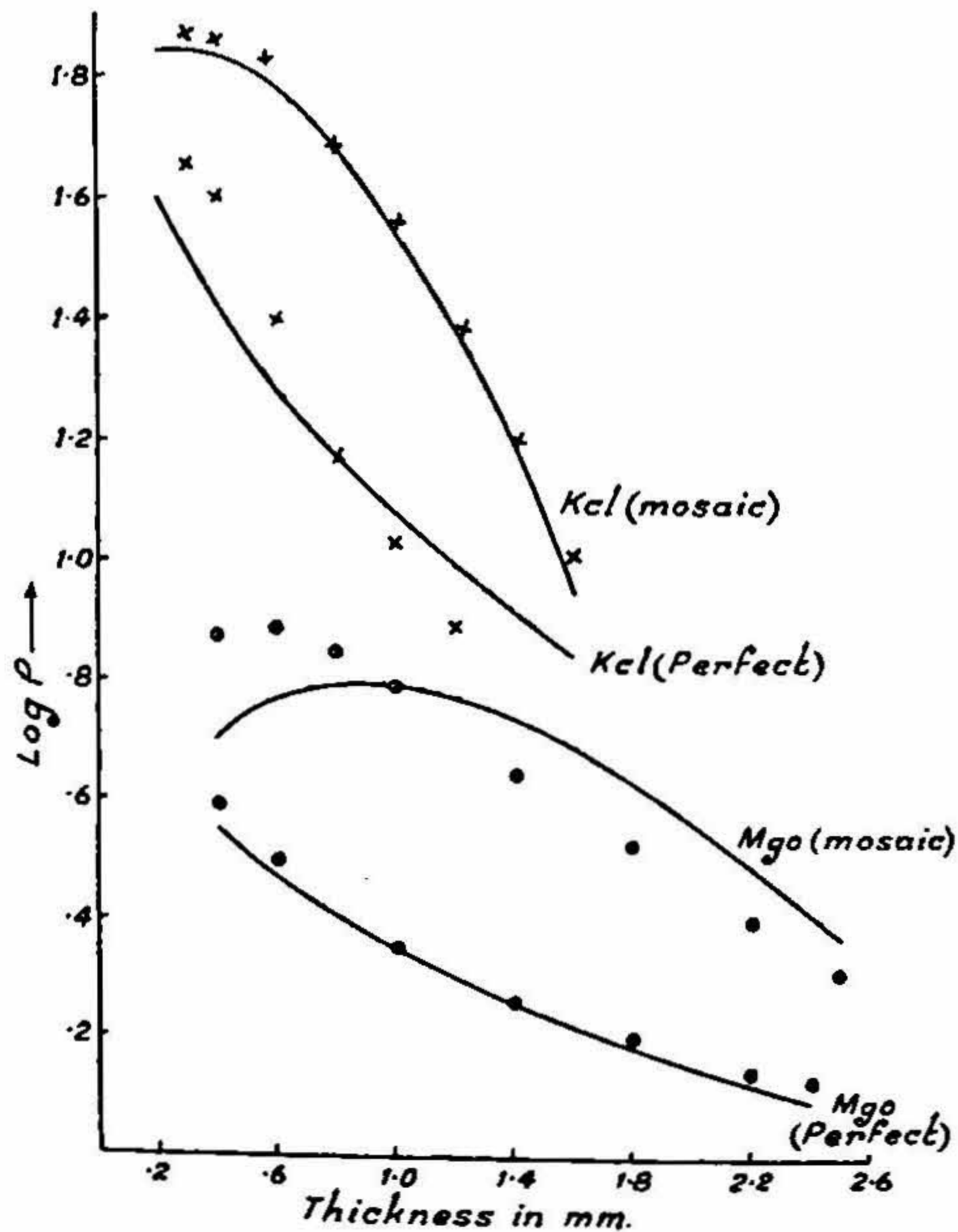

FIG. 4. Log *p-t* curves: continuous lines are drawn from theory. Circles with central dot and crosses represent the experimental points in MgO and KCl respectively.

theoretical log ρ *vs. t* curves are drawn for MgO perfect and mosaic crystals. The crystal was then dipped in liquid air, and Fig. 3 *b* is the internal reflection after the chilling treatment. Except for a slight increase in intensity there is practically no change between the two reflections, reproduced in Fig. 3 *a* and 3 *b*. The reflection obtained with horizontal slit is subjected to microphotometric study and the experimental points are plotted in Fig. 4. The points observed in the etched condition follow the theoretical curve closely, while points of the chilled crystal do not coincide with the theoretical curve of the mosaic crystal indicating thereby that the melt-grown MgO crystal is nearly perfect.

KCl.—A wedge of thickness ·4 to 1·8 mm. was used for the investigation. Fig. 3 *c* is the typical positive reproduction of the internal reflection of 200 planes in the etched condition. As in contrast to that of MgO, the bulk of the crystal also gives reflection and in addition the intensity falls off very rapidly with increase of thickness. The crystal was then dipped in liquid air and the reflection is reproduced in Fig. 3 *d*. No marked changes could be noticed between Figs. 3 *c* and 3 *d* except for a slight increase in intensity. The theoretical curves for the perfect and mosaic crystal are drawn in Fig. 4 along with the experimental points in the etched and chilled conditions. The experimental points of the etched crystal do not coincide with the theoretical perfect crystal curve, while the points of the chilled crystal follow the theoretical curve of the mosaic crystal very closely. Further the etched crystal points indicate a curve the shape being the same as the theoretical mosaic crystal curve. Hence it can be concluded that the melt-grown KCl crystal is mosaic.

4. Concluding Remarks

Although the increase in the intensity of reflection with thickness is shown clearly in the case of NaCl, yet it would be interesting to study a crystal where there is a continuous increase, when the thickness increases from say ·5 to 3 mm. For that μ has to be small and should be less than 5 cm.[-1] The absorption coefficient for LiF crystal is small enough but it did not show this behaviour as the crystal did not respond to chilling treatment. It is quite likely that other crystals may be more promising and work in this direction is in progress.

V. M. PADMANABHAN

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