

above and below the normal positions of the layer lines because of a -axis fluctuations on the a -axis oscillation photographs. This explanation of arcing phenomena is contrary to the experimental observations of Lal & Trigunayat (1970, 1971) which showed an increase in arcing or the formation of new arcs on heating the solution-grown CdI_2 crystals at about 270°C . Besides, crystals of lead iodide grown by the gel method also exhibit arcing (Agrawal, Chadha & Trigunayat, 1970). Secondly, the growth of incoherent nuclei on top of a flat surface, resulting in small gaps between different domains of the crystal, would affect the shape and the size of the spots. The arcs might be formed but the spots on them would not be resolved because of random growth of incoherent nuclei. Besides, on irradiating the crystal partially by the X-ray beam, the shape of the diffraction pattern would not change; however, the size of the arcs might be affected; this again is contrary to experimental observations (Agrawal & Trigunayat, 1969a). Furthermore, the arcing is not a surface phenomenon. It is also observed after cleaving off layers from the surface and it is, in general, found that the arcing decreases in a regular fashion from the lower to upper basal surfaces (Gyaneshwar & Trigunayat, 1972). The latter have also found that the closed rings on Laue photographs do change into arcs when crystals are peeled layer by layer. Under no circumstances can the formation of regular hexagonal, trigonal, *etc.* types of rings corresponding to each reflexion be understood on the basis of paracrystalline distortions randomly distributed in the crystals. When a few CdI_2 crystals, grown from solution at room temperature, were re-examined after nearly four years, changes in arcing, streaking and polytypism were observed (Agrawal, 1972); this cannot be explained on the basis of paracrystalline distortions induced during crystal growth. It may be pointed out that the model of a paracrystal is more accept-

able for fibrous crystals, which are in fact built up on a two-dimensional lattice, one of the vectors being parallel to the axis of the fibre, and the other perpendicular. The fluctuations of these vectors arise from essentially different sources, *viz.* the interactions along and between the chains.

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On the form of the fundamental equations of the dynamical theory of X-ray diffraction. By SHIZUO MIYAKE,* *Institute for Solid State Physics, University of Tokyo, Roppongi-7, Minato-ku, Tokyo, Japan* and Y. H. OHTSUKI, *Department of Physics, Waseda University, Tokyo, Japan*

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Property of the fundamental equations of the dynamical theory of X-ray diffraction given in terms of the electric field $\mathbf{E}(\mathbf{r})$, instead of $\mathbf{D}(\mathbf{r})$, is discussed, and some merits of the equations in terms of $\mathbf{E}(\mathbf{r})$ are pointed out.

The fundamental equations of the dynamical theory of X-ray diffraction according to Laue's formulation are usually given by the form

$$\frac{k_{\mathbf{h}}^2 - K^2}{k_{\mathbf{h}}^2} \mathbf{D}_{\mathbf{h}} = \sum_{\mathbf{g}} \chi_{\mathbf{h}-\mathbf{g}} \mathbf{D}_{\mathbf{g}[\perp \mathbf{k}_{\mathbf{h}}]} \quad (1)$$

(Laue, 1960), where $K(=v/c)$ is the wave number of X-rays in the vacuum space, $\mathbf{k}_{\mathbf{h}} = \mathbf{k}_0 + \mathbf{h}$, \mathbf{k}_0 being the wave vector of the primary beam in the Ewald (or Bloch) wave field in the crystal, and \mathbf{h} or \mathbf{g} is the reciprocal-lattice vector;

$\mathbf{D}_{\mathbf{h}}$ is the Fourier component of the field $\mathbf{D}(\mathbf{r})$ which is defined by

$$\mathbf{D}(\mathbf{r}) = \varepsilon(\mathbf{r})\mathbf{E}(\mathbf{r}) \quad (2)$$

where $\mathbf{E}(\mathbf{r})$ is the electric field of the X-ray wave, and $\chi_{\mathbf{h}}$ is the Fourier component of $\chi(\mathbf{r})$, which is a quantity corresponding to 4π times the 'electric susceptibility' and related to the 'dielectric constant' $\varepsilon(\mathbf{r})$ as

$$\varepsilon(\mathbf{r}) = 1 - \frac{1}{\pi} \left(\frac{e^2}{mc^2} \right) \frac{\varrho(\mathbf{r})}{K^2} = 1 + \chi(\mathbf{r}) \quad (3)$$

where $\varrho(\mathbf{r})$ is the electron density in the crystal. The notation $\mathbf{D}_{\mathbf{g}[\perp \mathbf{k}_{\mathbf{h}}]}$ means the component vector of $\mathbf{D}_{\mathbf{g}}$ parallel to a plane perpendicular to $\mathbf{k}_{\mathbf{h}}$.

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Laue's formulation is based on the electromagnetic theory in *vacuum*, in which $\mathbf{E}(\mathbf{r})$ is a basic microscopic quantity. It is, therefore, valuable to write down the fundamental equations in terms of $\mathbf{E}(\mathbf{r})$. Using the Maxwell equations

$$\left. \begin{aligned} \text{rot } \mathbf{E}(\mathbf{r}) &= -2\pi i K \mathbf{H}(\mathbf{r}) \\ \text{rot } \mathbf{H}(\mathbf{r}) &= 2\pi i K \mathbf{D}(\mathbf{r}) \end{aligned} \right\} \quad (4)$$

and the expansion

$$\mathbf{E}(\mathbf{r}) = \sum_{\mathbf{h}} \mathbf{E}_{\mathbf{h}} \exp[-2\pi i(\mathbf{k}_{\mathbf{h}} \cdot \mathbf{r})], \quad (5)$$

we can readily derive the equations

$$\frac{\mathbf{k}_{\mathbf{h}}^2 \cdot \mathbf{E}_{\mathbf{h}}[\perp \mathbf{k}_{\mathbf{h}}] - K^2 \cdot \mathbf{E}_{\mathbf{h}}}{K^2} = \sum_{\mathbf{g}} \chi_{\mathbf{h}-\mathbf{g}} \mathbf{E}_{\mathbf{g}}. \quad (6)$$

The appearance of the form $\mathbf{E}_{\mathbf{h}}[\perp \mathbf{k}_{\mathbf{h}}]$ in the left-hand side is due to the fact that the field $\mathbf{E}(\mathbf{r})$ is not purely a transverse wave, because of its property that $\text{div } \mathbf{E}(\mathbf{r}) \neq 0$ in general (see also Battermann & Cole, 1964).

Between $\mathbf{E}_{\mathbf{h}}$ and $\mathbf{D}_{\mathbf{h}}$, there is a relation

$$\mathbf{E}_{\mathbf{h}} = \mathbf{D}_{\mathbf{h}} - \sum_{\mathbf{g}} \chi'_{\mathbf{h}-\mathbf{g}} \mathbf{D}_{\mathbf{g}} = \mathbf{D}_{\mathbf{h}}(1 - \chi'_0) - \sum_{\mathbf{g} \neq \mathbf{h}} \chi'_{\mathbf{h}-\mathbf{g}} \mathbf{D}_{\mathbf{g}} \quad (7)$$

where χ' is the quantity defined by

$$\frac{1}{\varepsilon(\mathbf{r})} \equiv \frac{1}{1 + \chi(\mathbf{r})} = 1 - \chi'(\mathbf{r}). \quad (8)$$

Considering the fact that $\chi_{\mathbf{h}}$ or $\chi'_{\mathbf{h}}$ is a very small quantity of the order of $10^{-4} \sim 10^{-6}$, we may disregard the perpendicular components of $\mathbf{D}_{\mathbf{g}}$ to $\mathbf{D}_{\mathbf{h}}$ in the summation terms in (7). Then, $\mathbf{E}_{\mathbf{h}}$ is approximated to be parallel to $\mathbf{D}_{\mathbf{h}}$ and, accordingly, perpendicular to $\mathbf{k}_{\mathbf{h}}$. Equation (6) is, thus, approximated as

$$\frac{\mathbf{k}_{\mathbf{h}}^2 - K^2}{K^2} \mathbf{E}_{\mathbf{h}} = \sum_{\mathbf{g}} \chi_{\mathbf{h}-\mathbf{g}} \mathbf{E}_{\mathbf{g}}. \quad (9)$$

It should be pointed out, in this connexion, that equation (1) also involves the approximation which is of the same order as that assumed in deriving (9). Namely, in deriving equation (1) an approximation is introduced that

$$\frac{1}{1 + \chi(\mathbf{r})} \simeq 1 - \chi(\mathbf{r}) \quad (10)$$

in place of the exact equation

$$\frac{1}{1 + \chi(\mathbf{r})} \equiv 1 - \chi'(\mathbf{r}) = 1 - \chi(\mathbf{r})\{1 - \chi(\mathbf{r})\} + \dots \quad (11)$$

The strict form of the equations for $\mathbf{D}_{\mathbf{h}}$ should, therefore, be given by replacing $\chi_{\mathbf{h}}$ in (1) by $\chi'_{\mathbf{h}}$.

Equation (9) has the following formal merits:

(a) Its form is simpler than (1), especially in the fact that K^2 appears in the denominator of the left-hand side term, instead of $\mathbf{k}_{\mathbf{h}}^2$ in (1).

(b) Its form is quite similar to the scalar fundamental

equations for electron diffraction, which are written as

$$\frac{\mathbf{k}_{\mathbf{h}}^2 - K^2}{K^2} \psi_{\mathbf{h}} = \sum_{\mathbf{g}} \chi_{\mathbf{h}-\mathbf{g}}^{(e)} \psi_{\mathbf{g}} \quad (12)$$

where $\psi_{\mathbf{h}}$ is the amplitude of a component of the electron Bloch wave, and $\chi_{\mathbf{h}}^{(e)}$ is the Fourier component of $\chi^{(e)}(\mathbf{r})$ which is defined by

$$\chi^{(e)}(\mathbf{r}) = \frac{eV(\mathbf{r})}{E}, \quad (13)$$

E being the total energy and $V(\mathbf{r})$ the periodic potential in the crystal (Takagi, 1969).

(c) If all $\mathbf{k}_{\mathbf{h}}$ in an n -wave problem are included in the same plane and if we are concerned with the perpendicular mode $\mathbf{E}^{\perp}(\mathbf{r})$ of $\mathbf{E}(\mathbf{r})$ to this plane, then $\mathbf{E}_{\mathbf{h}} \perp \mathbf{k}_{\mathbf{h}}$ [refer also to equation (7)]. In this problem, equation (9) is as strict as is equation (6) (Batterman & Cole, 1964).

(d) The boundary conditions for (9) can be expressed in a simple form as that with respect to (1).

An example showing a merit due to the simple form of (9) may be found in the fact that the equation of the dispersion surface in the two-wave problem for the perpendicular mode of $\mathbf{E}(\mathbf{r})$ can be obtained most honestly from (9) in the form

$$\zeta_0 \zeta_{\mathbf{h}} = \frac{1}{4} \left(\frac{K^2}{\kappa_0} \right)^2 \chi_{\mathbf{h}} \chi_{-\mathbf{h}} = \frac{1}{4} \left(\frac{K}{n_0} \right)^2 \chi_{\mathbf{h}} \chi_{-\mathbf{h}} \quad (14)$$

where $\zeta_0 = k_0 - \kappa_0$, $\zeta_{\mathbf{h}} = k_{\mathbf{h}} - \kappa_0$, $\kappa_0^2 = K^2(1 + \chi_0)$ and n_0 (the mean refractive index) $= (1 + \chi_0)^{1/2}$. In most texts the dispersion equation has been given dropping the index n_0 , probably not only because of the very small deviation of n_0 from unity, but also because of some arbitrariness introduced in the approximation in dealing with the rather cumbersome form of (1).

For most practical purposes, equations (1) and (9) give essentially the same results, and the numerical difference between them is trivial. However, in addition to the formal merits of (9), since $\mathbf{E}(\mathbf{r})$, being directly related to the vector potential $\mathbf{A}(\mathbf{r})$, is a more basic microscopic quantity than $\mathbf{D}(\mathbf{r})$, we may state at least that there is no positive necessity to stick specifically to the equation (1) in terms of $\mathbf{D}(\mathbf{r})$. The theoretical meaning of equation (9) in terms of $\mathbf{E}(\mathbf{r})$ has already been discussed briefly in a previous paper by one of us (Ohtsuki, 1964). The present note is to stress again this statement by adding some amendments.

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