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On Extinction. I. General Formulation

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A new formalism of the dynamical diffraction theory is proposed in order to take into account the statistical nature of lattice distortions. It is postulated that the observed intensity is given by an ensemble average of the intensity field derived from the wave equations of Takagi–Taupin type. The formalism leads to exactly the same results as those of the conventional dynamical theory for ideally perfect crystals and the results of the conventional kinematical theory for ideally imperfect crystals. The general relations are established among the integrated diffraction powers for three types of incident wave; namely, a spherical wave (narrow beam), a plane wave and the wave from an incoherent homogeneous source (wide beam). This paper is a preparation for the following one on secondary extinction [Kato *Acta Cryst.*, (1976) A32, 458–466].

1. Introduction

In the history of diffraction crystallography extinction has been one of the important problems. Darwin (1914*a*, *b*; 1922) proposed two distinct concepts; primary extinction and secondary extinction. Since then, the theory of the former has been developed on the basis of wave equations, whereas the theory of the latter has been worked out on the basis of energy-transfer equations, in which perfect incoherence of the relevant waves is assumed at the beginning. In principle, however, any optical theory ought to be developed without making assumptions about coherence. Apparent incoherence is brought about by taking a statistical average of the medium concerned.

So far, no bridge has been constructed between the two types of extinction. The present paper deals with the fundamental problem of connecting them. Starting from the wave equation of Takagi–Taupin type (Takagi, 1962, 1969; Taupin, 1964; Kato, 1973) and introducing a spatial correlation length τ to characterize the crystalline medium, one can derive the intensity fields for ideally perfect crystals in one extreme case ($\tau \gtrsim$ crystal size) and for ideally imperfect crystals in another extreme case ($\tau \simeq 0$). In the following paper (Kato, 1976) the intermediate cases will be discussed with particular reference to secondary extinction.

The formulation is based on a postulation that the observed intensity is an average of the dynamical intensities over a statistical ensemble of distorted crystals. This approach will open a new field of application of dynamical diffraction to statistically distorted crystals.

In this series of papers, for simplicity, the intensity fields in the Laue cases due to a spherical wave are mainly considered. In the last section of this paper, however, the relations are given among the integrated powers expected when the incident wave is either a spherical wave or a plane wave, as well as when the incident beam is incoherent and homogeneous. The last case corresponds best to practical experiments.

2. Fundamental equations

We shall start with the wave equations of Takagi–Taupin type:

$$\frac{\partial d_o}{\partial s_o} = i\kappa_{-g} \exp iG \,.\, d_g \qquad (1a)$$

$$\frac{\partial d_g}{\partial s_g} = i\kappa_g \exp -iG \,.\, d_o \,, \qquad (1b)$$

where d_o and d_g are the wave fields of the direct and Bragg-reflected waves* respectively, and κ_g is related to the structure factor F_g by

$$\kappa_g = \frac{\lambda C}{v} \frac{e^2}{mc^2} F_g \qquad (2)$$

(λ: wavelength, C: polarization factor, v: unit-cell volume, e, m and c: the physical constants in the conventional usage.) The phase G is related to the displacement $\mathbf{u}$ of the lattice points as

$$G = 2\pi(\mathbf{g} \,.\, \mathbf{u}) \,, \qquad (3)$$

where $\mathbf{g}$ is the reflexion vector concerned. In this paper $\{\exp \pm iG\}$ are called lattice phase factors. The quantities κ_g and κ_{-g} are constant but G is a function of position. A position is specified by the oblique coordinates (s_o, s_g), the axes being taken along the directions of O and G beams.

The details of the derivation of equations (1) and the assumptions involved are described in the papers of Takagi (1962, 1969) and Taupin (1964) and also by Kato (1973).

The energy-transfer equations on which the recent theories of secondary extinction were developed can

* Hereafter, they will be called O and G waves.

be written in the form (Hamilton, 1957; Werner & Arrott, 1965; Werner, Arrott, King & Kendrick, 1966; Zachariasen, 1967*a*, *b*; Becker & Coppens, 1974),

$$\frac{\partial I_o}{\partial s_o} = -\bar{\sigma} I_o + \bar{\sigma} I_g \qquad (4a)$$

$$\frac{\partial I_g}{\partial s_g} = -\bar{\sigma} I_g + \bar{\sigma} I_o \,, \qquad (4b)$$

where I_o and I_g are the intensity fields of O and G beams and $\bar{\sigma}$ is the coupling constant. It is defined by

$$\bar{\sigma}(\varepsilon) = \int \sigma(\varepsilon+\eta) W(\eta) \mathrm{d}\eta \,, \qquad (5a)$$

where $\sigma(\theta)$ is the angular distribution of the diffracted intensity due to a mosaic block and $W(\theta)$ is the angular distribution of the mosaic blocks within a specimen. Zachariasen (1967*b*) defined two special cases, namely

Type I: W-dominant case; $\bar{\sigma}(\varepsilon) \sim Q W(\varepsilon)$ (5*b*)

Type II: σ-dominant case; $\bar{\sigma}(\varepsilon) \sim \sigma(\varepsilon)$, (5*c*)

where Q is the integrated power per unit volume owing to the kinematical theory, which is given by

$$Q = \frac{\lambda^3 C^2 |F_g|^2}{v^2 \sin 2\theta_B} \left(\frac{e^2}{mc^2}\right)^2 . \qquad (6)$$

It is worth noting the similarities and differences between the mathematical structures of equations (1) and (4). To see this, the enhanced intensities

$$J_o = \exp\{\bar{\sigma}(s_o+s_g)\} I_o \qquad (7a)$$

$$J_g = \exp\{\bar{\sigma}(s_o+s_g)\} I_g \qquad (7b)$$

are introduced. Then one can obtain the relations

$$\frac{\partial J_o}{\partial s_o} = \bar{\sigma} J_g \qquad (8a)$$

$$\frac{\partial J_g}{\partial s_g} = \bar{\sigma} J_o \,. \qquad (8b)$$

The coupling constants in equations (1) are complex and depend upon position, whereas in equations (4) and (8) they are real and usually assumed to be constant throughout the crystal.

In the both equations (1) and equations (4), the normal photoelectric absorption has been eliminated at the stage of formulating the differential equations. The intensity fields including the absorption can be written down as follows when d_o and d_g are obtained:

$$I_o^{(a)} = \exp -\mu_0(s_o+s_g) \,.\, |d_o|^2 \qquad (9a)$$

$$I_g^{(a)} = \exp -\mu_0(s_o+s_g) \,.\, |d_g|^2, \qquad (9b)$$

where μ_0 is the linear absorption coefficient. Similarly, if J_o and J_g are given, the intensities can be written in the form:

$$I_o^{(a)} = \exp -(\mu_0+\bar{\sigma})(s_o+s_g) \,.\, J_o \qquad (10a)$$

$$I_g^{(a)} = \exp -(\mu_0+\bar{\sigma})(s_o+s_g) \,.\, J_g \,. \qquad (10b)$$

For these reasons we shall not bother hereafter about the normal absorption factor, $\exp -\mu_0(s_o+s_g)$.

In the following, we shall start with equations (1). In general cases, $\kappa_g \kappa_{-g}$ has a small imaginary component. This part accounts for Borrmann absorption. In equations (4), however, the imaginary part of $\kappa_g \kappa_{-g}$ does not appear explicitly. The conventional theory of secondary extinction cannot take into account the Borrmann absorption, unless some *ad hoc* assumptions are introduced.

3. The formal solution of equations (1)

It is more convenient to rewrite equations (1) with the recurrence formulae

$$d_o(n+1,m) = d_o(n,m) + i(\kappa_{-g} a) \exp iG(n,m) \,.\, d_g(n,m) \qquad (11a)$$

$$d_g(n,m+1) = d_g(n,m) + i(\kappa_g a) \exp -iG(n,m) \,.\, d_o(n,m) \,, \qquad (11b)$$

where the arguments (n,m) are the abbreviations of the coordinates $(na,ma) \equiv (s_o,s_g)$ and a is unit increment of the coordinates, which will be made infinitesimal in the final expression for the intensity. The relations among the field components are illustrated in Fig. 1. In the following the kinks of the lines corresponding to $d_o \rightarrow d_g$ and $d_g \rightarrow d_o$ will be called (*a*)-type and (*b*)-type respectively.

To obtain a concrete solution, one needs the boundary conditions for the wave fields. Here, we shall consider the Laue case excited by the incident wave $D_o(s_o,s_g) = A\delta(s_g)$. In order to fit the present formalism, the δ-function is replaced by the discrete function in the way,

$$D_o(0,m) = A/a \qquad m=0 \qquad (12a)$$

$$= 0 \qquad m \neq 0 \,. \qquad (12b)$$

No wave arrives at the line $m=0$ in the direction of G beam, so that one can put

$$D_g(n,0) = 0 \,. \qquad (12c)$$

If conditions are matched, the crystal waves $d_o(0,m)$ and $d_g(n,0)$ must satisfy the same conditions as equations (12).

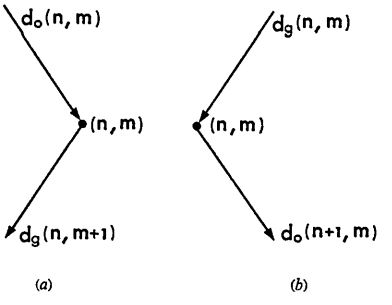

Fig. 1. Bragg reflexion; the kink points (n,m) denote reflexions of types (*a*) and (*b*).

We shall be interested in the wave fields at an observation point specified by (N,M). If one looks at equations (11) or Fig. 1, the wave fields are composed of zigzag routes starting from the entrance point $(0,0)$ and arriving at the observation point (N,M). Thus, one can write the wave fields as

$$d_o(N,M)=\sum_R A_R \exp iP_R \tag{13a}$$

$$d_g(N,M)=\sum_R B_R \exp iQ_R\,, \tag{13b}$$

where R denotes symbolically an individual route which is specified by the kink points. Explicitly the amplitudes A_R and B_R are given by

$$A_R=\left(\frac{A}{a}\right)(-\kappa_g\kappa_{-g})^r a^{2r} \tag{14a}$$

$$B_R=\left(\frac{A}{a}\right)(i\kappa_g)\,(-\kappa_g\kappa_{-g})^r a^{2r+1}, \tag{14b}$$

where the integer r is the total number of (b)-type kinks. As can be seen in Fig. 2, the number of kinks of type (a) must be identical to r for A_R and to $r+1$ for B_R.

The phases P_R and Q_R depend on the individual positions of the kinks. They have the forms

$$P_R=-G(n_1,0)+G(n_1,m_1)\ldots -G(n_r,m_{r-1})+G(n_r,m_r)\;(m_r=M) \tag{15a}$$

$$Q_R=-G(n_1,0)+G(n_1,m_1)\ldots +G(n_r,m_r)-G(n_{r+1},m_r)\;(n_{r+1}=N) \tag{15b}$$

where (n_l,m_l) specifies the position of a kink (b) and (n_l,m_{l-1}) that of a kink (a). One obtains the formal solutions for the wave fields $d_o(N,M)$ and $d_g(N,M)$ by inserting equations (14) and (15) into equations (13).

4. The observed intensity fields

Here we postulate that the observed intensity is the average of the intensity over a statistical ensemble of the displacement $\mathbf{u}$ at the kink points. Then one can obtain immediately the expressions for the observed intensities as

$$\langle I_o\rangle=\sum_R\sum_{R'} A_R A_{R'}^*\langle\exp i(P_R-P_{R'})\rangle \tag{16a}$$

$$\langle I_g\rangle=\sum_R\sum_{R'} B_R B_{R'}^*\langle\exp i(Q_R-Q_{R'})\rangle\,. \tag{16b}$$

Here use is made the fact that the amplitudes A_R and B_R are independent of the ensemble assumed. In this section we shall calculate explicitly the intensity for two extreme cases, namely ideally perfect and ideally imperfect crystals.

(1) *Perfect crystals*

Equations (16a, b) can be rewritten in the form

$$\langle I_o\rangle=\sum A_R\sum A_{R'}^* +\sum\sum A_R A_{R'}^*\langle\exp i(P_R-P_{R'})-1\rangle \tag{17a}$$

$$\langle I_g\rangle=\sum B_R\sum B_{R'}^* +\sum\sum B_R B_{R'}^*\langle\exp i(Q_R-Q_{R'})-1\rangle\,. \tag{17b}$$

In perfect crystals, $G(n,m)$ are constant throughout the crystal so that the averages $\langle\,\rangle$ are zero. Thus, it is enough to calculate $\sum A_R$ and $\sum B_R$. This can be done by a combinatorial calculation. Since A_R and B_R are determined only by the kink number as shown in equations (14), the intensity fields are given by

$$I_o(N,M)=|A/a|^2\quad(M=0) \tag{18a}$$

$$=\Big|\sum_{r=1}\alpha_r(-\kappa_g\kappa_{-g})^r a^{2r}\Big|^2|A/a|^2\quad(M>0) \tag{18b}$$

$$I_g(N,M)=|\kappa_g|^2\Big|\sum_{r=0}\beta_r(-\kappa_g\kappa_{-g})^r a^{2r+1}\Big|^2|A/a|^2, \tag{18c}$$

where α_r and β_r are the possible number of routes having r kinks of type (b). Here the average sign $\langle\,\rangle$ of I_o and I_g is omitted.

First we shall consider α_r. Since r kinks of type (a) are distributed over N possible lines of the G direction and $r-1$ kinks of type (b) over $M-1$ possible lines of the O direction [the last kink must be always on the line $m_r=M$; see equation (15a)], one can see that

$$\alpha_r=\frac{N!}{r!(N-r)!}\cdot\frac{(M-1)!}{(r-1)!(M-r)!}\,. \tag{19a}$$

By similar arguments it is not difficult to see that

$$\beta_r=\frac{N!}{r!(N-r)!}\cdot\frac{M!}{r!(M-r)!}\,. \tag{19b}$$

Denoting the observation point by the coordinates $(s_o,s_g)=(Na,Ma)$, and letting a go to zero, one obtains

$$\alpha_r=\frac{(s_o/a)^r(s_g/a)^{r-1}}{r!(r-1)!} \tag{20a}$$

$$\beta_r=\frac{(s_o/a)^r(s_g/a)^r}{r!r!}\,. \tag{20b}$$

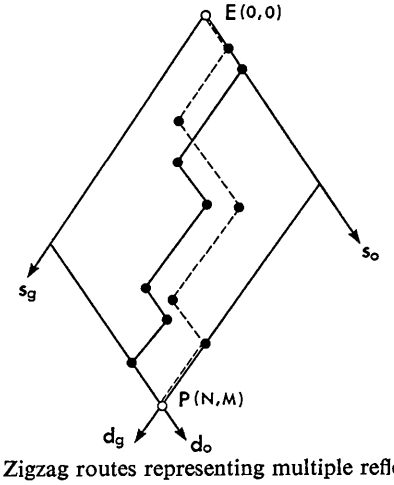

Fig. 2. Zigzag routes representing multiple reflexions.

Inserting these into equations (18), one obtains

$$I_o(s_o,s_g)=|\sum_{r=1}^{\infty}\frac{1}{r!(r-1)!}(-\kappa_g\kappa_{-g})^r s_o^r s_g^{r-1}|^2|A|^2 \quad (21a)$$

$$I_g(s_o,s_g)=|\kappa_g|^2|\sum_{r=0}^{\infty}\frac{1}{r!r!}(-\kappa_g\kappa_{-g})^r s_o^r s_g^r|^2|A|^2 . \quad (21b)$$

It is to be noticed that the results are independent of the choice of the coordinate increment a. By consulting the definitions of the Bessel functions J_0 and $J_{-1}=J_1$, finally, one obtains the intensity fields

$$I_o(s_o,s_g)=|\kappa_g\kappa_{-g}|\frac{s_o}{s_g}|J_1[2(\kappa_g\kappa_{-g})^{1/2}(s_o s_g)^{1/2}]|^2|A|^2 \quad (22a)$$

$$I_g(s_o,s_g)=|\kappa_g|^2|J_0(2(\kappa_g\kappa_{-g})^{1/2}(s_o s_g)^{1/2}]|^2|A|^2. \quad (22b)$$

The results are identical to those originally obtained by the spherical-wave theory (Kato, 1960, 1961, 1968) under equivalent boundary conditions to those described by equations (12).

So far we have considered only the Laue cases. If the crystal is bounded by a pair of plane surfaces, one can obtain the wave fields $\sum A_R$ and $\sum B_R$ under the geometrical conditions of Laue–(Bragg)^n and (Bragg)^m cases* with the boundary conditions equivalent to equation (12). They were treated by Uragami (1969, 1970, 1971) on the basis of the Riemann function and independently by Saka, Katagawa & Kato (1971*a*, *b*, 1972) on the basis of Fourier transformation. Thus, in principle, one can write down the intensity fields for perfect polyhedral crystals. This point has been discussed in the last paper of Saka *et al.* mentioned above.

(2) *Imperfect crystals*

Equations (16*a*, *b*) can also be rewritten in the form

$$\langle I_o\rangle=\sum_R A_R A_R^*+\sum_R{\sum_{R'}}' A_R A_{R'}^*\langle\exp i(P_R-P_{R'})\rangle \quad (23a)$$

$$\langle I_g\rangle=\sum_R B_R B_R^*+\sum_R{\sum_{R'}}' B_R B_R^*\langle\exp i(Q_R-Q_{R'})\rangle \quad (23b)$$

where $\sum'$ means summation omitting the term $R=R'$. In ideally imperfect crystals, the random-phase approximation can be used so that only the first term in each case remains. Again a combinatorial calculation similar to case (1) can be used. From equations (14), we have

$$I_o(N,M)=|A|^2\sum_{r=1}\alpha_r|\kappa_g\kappa_{-g}|^{2r}a^{4r-2} \quad (24a)$$

$$I_g(N,M)=|A|^2|\kappa_g|^2\sum_{r=0}\beta_r|\kappa_g\kappa_{-g}|^{2r}a^{4r}. \quad (24b)$$

As the increment a is decreased to zero, $I_o(N,M)$ tends to zero except for $M=0$, where $I_o(N,M)$ goes to infinity. On the other hand, I_g tends to $|A|^2|\kappa_g|^2$. These results are identical to those expected from kinematical theory, as will be shown in equation (28*b*). The kinematical solution is independent of whether the geometrical conditions are those of the Laue cases or of the Bragg cases.

* For the new terminology specifying the various cases, see Azaroff, Kaplow, Kato, Weiss, Wilson & Young (1974).

5. The integrated powers

The present formulation is based on the spherical-wave theory, whereas conventional diffraction theory is based on the plane-wave theory. The former is adequate for the case where the crystal is sufficiently large and part of the crystal is illuminated by a narrow incident wave. In practice, however, we often encounter the case where the crystal is bathed in the incident beam as shown in Fig. 3. In this section we shall first consider the relation of the integrated powers in the spherical and plane-wave theories and then the integrated power illuminated by the homogeneous incident beam.

In the spherical-wave theory, the incident wave can be written to a good approximation in the form (Kato, 1961; Saka, Katagawa & Kato, 1972)

$$D_o(s_o,s_g)=A\delta(s_g) \quad (25a)$$

$$=A\sin 2\theta_B\delta(x_o), \quad (25b)$$

where x_o and x_g (which will be used soon) are the coordinates perpendicular to the O and G beams respectively, and they are (s_g,s_o) times sin $2\theta_B$. A plane wave can be represented by

$$\exp i(K_x x_o)=\int\exp i(K_x x_o')\,.\,\delta(x_o-x_o')\mathrm{d}x_o', \quad (26)$$

where the irrelevant part $\exp i(K_y y+K_z z)$ is omitted.

If the incident wave, $A\delta(s_g)$, excites the wave field $d_g^s(x_o,x_g)$, the plane-wave solution must be

$$d_g^p(x_o,x_g;K_x)=(A\sin 2\theta_B)^{-1}\int\exp i(K_x x_o')\times d_g^s(x_o-x_o',x_g-x_o')\mathrm{d}x_o', \quad (27a)$$

where (x_o',x_g') is an entrance point, so that x_g' is a function of x_o' depending on the shape of the crystal. Con-

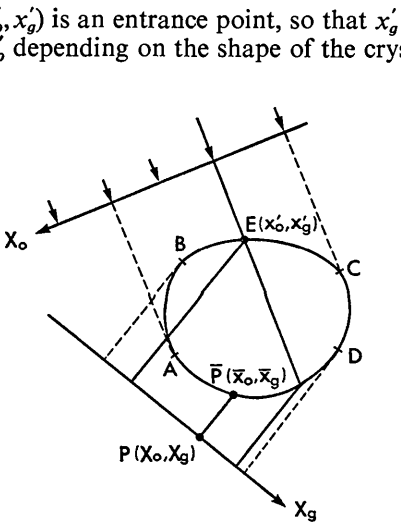

Fig. 3. The case of wide-beam illumination. *ABC*: The entrance surface, *BAD*: The exit surface. [From Fig. 1 of Hamilton (1957) and Fig. 2 of Werner & Arrott (1965).]

sequently, the plane-wave solution outside the crystal can be given by

$$D_g^p(X_o, X_g; K_x) = (A \sin 2\theta_B)^{-1} \int \exp i(K_x x_o') \times D_g^s(X_o - x_o', X_g - x_g') \mathrm{d}x_o' . \quad (27b)$$

Here, capital letters are used for the wave fields and the position variables in order to emphasize that they refer to the observation point outside the crystal. Except for the phase factor, $D_g^s(X_o, X_g)$ is identical to $d_g^s(\bar{x}_o, \bar{x}_g)$ in which the coordinates $(\bar{x}_o, \bar{x}_g)$ represent the point at the exit surface corresponding to the observation point (X_o, X_g) as shown in Fig. 3. It is significant that D_g^p is proportional to the Fourier transform of D_g^s, the results having been obtained previously in a different way (Kato, 1961, 1968).

The integrated power in the plane-wave theory can be defined by

$$P_g^p = K^{-1} \int\int\int |D_g^p|^2 \mathrm{d}K_x \mathrm{d}X_g \mathrm{d}Y , \quad (28a)$$

where $\mathrm{d}K_x/K = \mathrm{d}\theta$ is an angular differential and the coordinate Y refers to the axis perpendicular to the reflexion plane. The integration $\iint \mathrm{d}X_g \mathrm{d}Y$ must be taken over the exit surface. Thus, from Parseval's theorem on the Fourier transform, we see that

$$P_g^p = \lambda (A \sin 2\theta_B)^{-2} \int\int\int |D_g^s|^2 \mathrm{d}x_o' \mathrm{d}X_g \mathrm{d}Y . \quad (28b)$$

The result is useful for calculating the integrated power P_g^p from the spherical-wave solution discussed in the previous sections.

As the simplest example, let us consider the kinematical case in which $|D_g^s|^2$ is given by $|A|^2|\kappa_g|^2$ as discussed below equation (24*b*). With the use of the expression (2), and remembering that $\mathrm{d}x_o' \mathrm{d}X_g \mathrm{d}Y = \sin 2\theta_B \mathrm{d}(\text{volume})$ one can obtain the integrated power per unit volume, equation (6), based on the conventional kinematical theory.

The integrated power due to a spherical wave of the form (25*a*) is defined by

$$P_g^s = A^{-2} \int\int |D_g^s|^2 \mathrm{d}X_g \mathrm{d}Y . \quad (29)$$

Equation (28*b*), therefore, can be rewritten as

$$P_g^p = \lambda (\sin 2\theta_B)^{-2} \int P_g^s(x_o') \mathrm{d}x_o' . \quad (28c)$$

This is the relation between P_g^p and P_g^s. The integral

$$P_g = \int P_g^s(x_o') \mathrm{d}x_o' \quad (30)$$

means the integrated power when spherical-wave sources are homogeneously distributed with a unit intensity per unit length on the plane X_o in Fig. 3 without any phase relation and when each source has the form (25). This is the most realistic case for diffractometry, in which the crystal is illuminated by a wide homogeneous beam.

6. Conclusions

The present paper is a preparation for the following one. The approach adopted here gives a way to take the ensemble average of the intensity based on the wave equation. For two extreme cases, namely ideally perfect and imperfect crystals, the theory can predict exactly the same results as those of the conventional theories. No *ad hoc* assumption is made regarding wave coherence. The varieties of the intensity depend entirely on the correlation of the lattice phase, *i.e.* on the nature of the medium.

The present theory is based on the spherical-wave theory. It is, however, worth mentioning that the present formalism is not limited to such a special case. As described in § 5, if one obtains the intensity field for a spherical wave, mathematically speaking for an impulse of δ-function type, the integrated power can be obtained by means of equations (28) or equations (30) for the incidence of a plane wave or an incoherent wide beam respectively.

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