

On the Possibility of Stress Determination by Consideration of Pendellösung Effect

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Z. Naturforsch. **37a**, 626—632 (1982); received February 8, 1982

Dedicated to Prof. G. Hildebrandt on the occasion of his 60th birthday

In this paper a simplified method of stress estimation is proposed. The calculation is based on the Pendellösung effect (i.e. beat of two Bloch waves). It is assumed that lattice distortion causes a local change of fringe spacing. The results of the calculations appear to be consistent with results based on the consideration of separated curved trajectories of two Bloch waves related to both branches of the dispersion surface.

I. Introduction

The dynamical theory of X-ray diffraction deals with the interaction of X-rays with crystals. The theory considers the energy flow in perfect as well as deformed crystals while diffraction is taking place. This energy flow is very sensitive to small lattice deformations. Hence, the diffracted beam leaving the crystal contains information about local strain fields.

In X-ray topographical methods the outgoing beam is registered on a photographic film. Under the proper experimental conditions considered by Kato [1], [2] the interference fringes may be observed on the section topogram of the flat crystal plate (Figure 1a). Deformation of the interference pattern appears as a result of lattice deformations inside the investigated plate.

Kato, Patel, and Ando [3a, b, c] proposed the method of stress determination based on an analysis of the interference pattern. According to the theory which is a base for calculations, two conjugated Bloch waves having a common trajectory in the perfect crystal are separated in a deformed crystal. The reason for such separation is that X-ray trajectories related to two branches of dispersion surface are bent in the opposite directions in the distorted lattice area. In the paper of Patel and Kato [3b] the force S representing stress field at the edge of 2000 Å oxide film on the surface of the

silicon plate has been evaluated as

$$S = 0.8 \times 10^5 \text{ dyn/cm} = 80 \text{ N/m}.$$

Filscher [4, 5] introduced a correction to the computational procedure proposed by Kato et al. [3]. He obtained a similar result: $S = 76 \text{ N/m}$. He also performed very good simulations of topographs (Fig. 1 in the previous paper [6]).

In this paper a simplified procedure of stress evaluation is proposed. The main steps of my consideration leading to this procedure can be specified as follows:

(i) According to the spherical wave diffraction theory developed by Kato [1], [2] the fringe pattern is a result of the interference between two conjugated waves: $\mathbf{k}_{0,g}^{(1)}$ and $\mathbf{k}_{0,g}^{(2)}$; flowing in the direction \mathbf{v} . This effect occurs in each \mathbf{v} direction of the Borrmann fan (Figure 2). The fringe spacing depends on the direction of energy flow. For a chosen direction it is determined by a difference of conjugated wave vectors:

$$\{\Delta \mathbf{k}(\mathbf{v}) \cdot \mathbf{v}\} = [\mathbf{k}_{0,g}^{(1)}(\mathbf{v}) - \mathbf{k}_{0,g}^{(2)}(\mathbf{v})] \cdot \mathbf{v}.$$

(ii) Using this result as well as the analysis of the beat phenomenon (Crawford [7], Appendix), it seems reasonable to represent the wave field excited in a perfect crystal as a fan of the "beat waves" (or "pendellösung waves") flowing in all directions \mathbf{v} of the Borrmann fan. The following notation has been taken:

$$\Lambda(\mathbf{v}) = \frac{2\pi}{\Delta \mathbf{k}(\mathbf{v}) \cdot \mathbf{v}} = \begin{array}{l} \text{wavelength of "beat"} \\ \text{wave"} \\ \text{= fringe spacing.} \end{array}$$

$$\mathcal{H}(\mathbf{v}) = \{\Delta \mathbf{k}(\mathbf{v}) \cdot \mathbf{v}\} = \text{beat vector.}$$

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III. Theory

Energy flow in a perfect crystal

In the spherical wave theory given by Kato [1, 2, 8, 9] the incident wave is represented in the direct space by a bundle of coherent waves and in the reciprocal space by a wide region of the vacuum dispersion surface. Each tie point of this surface represents the Bloch wave propagated in the direction \mathbf{v} in the real space. The directions \mathbf{v} are confined by the directions of the direct and the diffracted beams (Figure 2). So, in a crystal excited by a spherical wave, a pair of coherent Bloch waves is propagated in each direction \mathbf{v} . They interfere, resulting in a beat*. To describe the consequences of the beat it is convenient to use the terms "O" wave and "G" wave instead of "direct" wave and "Bragg diffracted" wave (Kato [2, 9]). In these terms, each Bloch wave can be written as a superposition of two coupled waves: "O" and "G". As a consequence of beat between two Bloch waves, the energy of "O" wave is transferred to the "G" wave and back over the distance of $\Lambda(\mathbf{v})$ called "fringe spacing". It is written in [2] as:

$$\Lambda_{o,g}(\mathbf{v}) = \frac{2\pi}{[\mathbf{k}_{o,g}^{(1)}(\mathbf{v}) - \mathbf{k}_{o,g}^{(2)}(\mathbf{v})] \cdot \mathbf{v}} = \frac{2\pi}{\{\Delta\mathbf{k}(\mathbf{v}) \cdot \mathbf{v}\}}. \quad (1)$$

The difference $\{\Delta\mathbf{k}(\mathbf{v}) \cdot \mathbf{v}\}$ has been written by Kato [3a] as

$$\{\Delta\mathbf{k}(\mathbf{v}) \cdot \mathbf{v}\} = A \cos \theta / \sqrt{1 - p^2}, \quad (2)$$

where:

A is a distance between the apex points of the two branches of dispersion surface. Hence A can be written as $2\pi/\Lambda_0$,

θ is an angle between \mathbf{v} and the net planes,

p is a parameter specifying the direction \mathbf{v} :

$$p = \tan \theta / \tan \theta_B. \quad (3)$$

In the approach proposed in this paper, the value of the fringe spacing $\Lambda(\mathbf{v})$ (the wavelength of the "beat wave") is treated as a basic parameter for the description of the energy flow in a perfect as well as in a deformed crystal. In the calculations it is more convenient to use the reciprocity of fringe

* The beat observed in case of diffraction is an analogical phenomenon to the beat observed in a system of two coupled pendulums (Appendix).

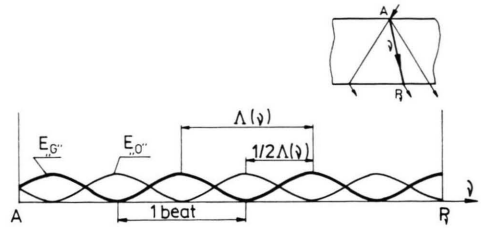


Fig. 3. Symbolic representation of the "beat wave" propagated in the direction \mathbf{v} in the perfect crystal. Two Bloch waves, $\mathbf{k}_{o,g}^{(1)}(\mathbf{v})$ and $\mathbf{k}_{o,g}^{(2)}(\mathbf{v})$, flow in this direction. Each of them is composed of two coupled waves: "O", wave and "G" wave. The beat of two Bloch waves leads to energy oscillation. Energy is transferred from one of the coupled waves to the other and back. A cycle of energy oscillation occurs over the distance $\Lambda(\mathbf{v})$. The resulting oscillating form of the energy flow suggests to introduce the term "beat wave" to describe the energy flow. The drawing has been made following Crawford's "Waves" [7].

spacing, called here "beat vector": $\mathcal{K}(\mathbf{v}) = 2\pi/\Lambda(\mathbf{v})$. It is then useful to express both parameters as a function of the deviation parameter η ; an important parameter of dynamical theory. This is possible by replacing θ in formula (2) by η . According to the relation used by Authier [10] the parameter η can be written:

$$\eta_p^2 = \frac{\tan^2 \theta / \tan^2 \theta_B}{1 - \tan^2 \theta / \tan^2 \theta_B}. \quad (4)$$

The letter "p" indicates the value of η corresponding to the direction \mathbf{v} in a perfect crystal. Values of Λ and \mathcal{K} related to perfect crystals will be also indicated by "p". They have been found from (1), (2) and (4):

$$\begin{aligned} \Lambda_p(\mathbf{v}) &= \Lambda_0(1 + \eta_p^2 / \cos^2 \theta_B)^{1/2}; \\ \mathcal{K}_p(\mathbf{v}) &= \mathcal{K}_0(1 + \eta_p^2 / \cos^2 \theta_B)^{-1/2}. \end{aligned} \quad (5)$$

Λ_0 is a fringe spacing along diffracting planes and $\mathcal{K}_0 = 2\pi/\Lambda_0$. The parameter η plays an important role in the theory. It can be used instead of θ and specifies a direction of energy flow in a perfect crystal. In the deformed crystal the parameter η is used to characterize the lattice deformation as it is proportional to the deviation from the Bragg angle.

A spherical wave incident on the crystal causes the excitation of the crystal area represented by a Borrmann triangle. For a description of the excitation state, the parameter $\Lambda(\mathbf{v})$ plays a significant role as it specifies a distance of energy transfer between the direction of the direct beam and that of the diffracted beam. The energy transfer occurs

along every direction \mathbf{v} . $\Lambda(\mathbf{v})$ is the function of direction, determined by (4) and (5).

The aim of this paper is to show that it is convenient to describe the excitation state as a fan of "beat waves" carrying energy along directions \mathbf{v} . Wavelengths of "beat waves", $\Lambda(\mathbf{v})$ are dependent on direction. In the perfect crystal, the wavelength of the beat wave is constant along the specified direction and equal to $\Lambda_p(\mathbf{v})$.

Energy flow in a deformed crystal

It seems to be reasonable to assume that in a slightly deformed crystal the beat waves also exist, but their wavelengths change under the influence of lattice deformation. Then the model of the fan of beat waves could be applicable also in a deformed crystal. By this assumption (5) is still valid, even in deformed crystals, but η_p has to be replaced by a new value η_d dependent on deformation. This dependence can be expressed by writing η_d as function of parameters (p, l, S) . Here p specifies the direction of \mathbf{v} (Fig. 2), l determines the position of the considered point along this direction and S represents a stress field. A value of $\eta_d(p, l, S)$ is proposed to be:

$$\eta_d = \eta_p + \Delta\eta, \quad (6)$$

where η_p is given by (4) and the change of the deviation parameter, $\Delta\eta$, is synonymously related to the change of the Bragg angle. From the definition of both parameters (Tanner [11]) it follows similarly the symmetrical Laue case that

$$\Delta\eta = (\Lambda_0/d) \Delta\theta, \quad (7)$$

where d is a spacing of net planes.

The value of $\Delta\theta$ expresses a change of diffraction conditions. This change can be caused by bending of the diffracting planes and change of their spacing (Blech and Meieran [12]):

$$\Delta\theta = (\partial u / \partial z) + \tan \theta_B (\partial u / \partial x), \quad (8)$$

where u is a component of the displacement vector. Both derivatives are functions of (p, l, S) . They can be evaluated basing on formulae of the theory of elasticity.

The value of η_d calculated by this means allows one to find local values of fringe spacing $\Lambda_d(\mathbf{v})$ along a direction \mathbf{v} in the deformed lattice area:

$$\begin{aligned} \Lambda_d(\mathbf{v}) &= \Lambda_0(1 + \eta_d^2 / \cos^2 \theta_B)^{1/2} \quad \text{or} \\ \mathcal{K}_d(\mathbf{v}) &= \mathcal{K}_0(1 + \eta_d^2 / \cos^2 \theta_B)^{-1/2}. \end{aligned} \quad (9)$$

IV. Interpretation of Section Topographs

Perfect crystal

The fringe pattern observed on the section topograph (Fig. 1) is determined by the intensity distribution of a diffracted beam leaving the crystal. Hence the intensity distribution depends on the distribution of the phases of "G" waves coming to the exit crystal surface. A dark fringe is observed in the P_r^f point on a film placed on the way of the diffracted beam (Fig. 2) if the intensity of the "G" wave reaches a maximum in the point P_r^c on the exit crystal surface. According to the conclusions of the spherical wave theory, maxima of "G" waves form a set of hyperbolas inside the Borrmann triangle. The distance between the maxima of the "G" wave intensity is equal to the fringe spacing $\Lambda(\mathbf{v})$. The intersection of the hyperbola set by the exit crystal surface is imaged as a fringe pattern on a section topograph.

In order to interpret a fringe pattern using the concept of the fan of beat waves, the energy flow along an arbitrary \mathbf{v} direction in the Borrmann triangle has to be considered. According to the conclusions of the spherical wave theory, a pair of coherent Bloch waves is propagated along each \mathbf{v} direction. These Bloch waves, specified by vectors $\mathbf{k}^{(1)}(\mathbf{v})$ and $\mathbf{k}^{(2)}(\mathbf{v})$, are called conjugated waves. Due to the beat between the conjugated waves, the energy flowing along the way $\overline{AP_r}$ through the crystal is periodically transferred from "O" wave to "G" wave and back again. A fringe spacing $\Lambda(\mathbf{v})$ is a period of this process and is equal to one beat.

The result of beat is schematically sketched in Figure 3. According to this sketch and to the definition of the fringe order applied in Kato's papers [3, 9], the dark fringe of n -th order is recorded in point P_r^f on the film, when the distance $\overline{AP_r^c}$ contains n times the distance $\Lambda(\mathbf{v})$ plus a part of $\Lambda(\mathbf{v})$ dependent on the boundary conditions:

$$n \cdot \Lambda(\mathbf{v}) + b \cdot \Lambda(\mathbf{v}) = \overline{AP_r^c}. \quad (10)$$

This part of $\Lambda(\mathbf{v})$, called "initial phase" or "residual distance", has been estimated by Kato [9] as $b = 1/4$ for the case of spherical waves.

The formula, equivalent to (10), which contains the beat vector has the following form:

$$2\pi n + 2\pi b = \mathcal{K}(\mathbf{v}) \cdot \overline{AP_r^c}. \quad (11)$$

In the case of a perfect crystal the product $\mathcal{K}(\mathbf{v}) \cdot \overline{AP_r^c}$ may be treated as a phase $F(\mathbf{v})$ reached by

the "G" wave at point P_r^c . In a perfect crystal this formula leads to the simple relation determining positions of dark fringes on a section topograph (the positions of fringes are specified by means of the parameter p ; $p = \tan \theta / \tan \theta_B = x/t_0 \tan \theta_B$).

From (1), (2) and (11) it follows that

$$2\pi(n+b) = \mathcal{K}_0 t_0 \sqrt{1-p^2}, \quad (12)$$

where $\mathcal{K}_0 = 2\pi/\Lambda_0$. Hence \mathcal{K}_0 is a constant parameter determined by material constants and diffraction conditions:

$$\mathcal{K}_0 = 2 \frac{\lambda}{V} \frac{e^2}{m c^2} \cos \theta_B |F_g|. \quad (13)$$

Here F_g is the structure factor, V the volume of the unit cell and λ the wavelength of the X-ray radiation.

Distorted Crystal

According to the assumption proposed in Sect. III, the beat vector $\mathcal{K}(\mathbf{v})$ (or the fringe spacing $\Lambda(\mathbf{v})$) can be treated as a variable parameter characterizing an energy flow along a constant direction \mathbf{v} on going through the slightly deformed crystal. Then, in the case of small deformations (11) is still valid but $\mathcal{K}(\mathbf{v})$ is not constant for a specified \mathbf{v} direction. The beat vector has only a local meaning in the deformed crystal. It is a function of the local deformation. According to (6), (7), (8), and (9) the local values $\mathcal{K}_d(\mathbf{v})$ can be written as functions of (p, l, S) when both derivatives $\partial u/\partial z$ and $\partial u/\partial x$ are found as functions of (p, l, S) based on the assumed deformation model.

In case of a deformed crystal the product $\mathcal{K}(\mathbf{v}) \cdot \overline{AP_r^c}$ in (11) should be replaced by the integral $\int_A^{P_r^c} \mathcal{K}_d(\mathbf{v}) dl$. The positions of the fringes on the topograph of the deformed part of the crystal can be calculated from the relation

$$2\pi(n+b) = \int_A^{P_r^c} \mathcal{K}_d(\mathbf{v}) dl \quad (14)$$

$$= \mathcal{K}_0 \int_A^{P_r^c} (1 + \eta_d^2 / \cos^2 \theta_B)^{-1/2} dl.$$

The integral $\int_A^{P_r^c} \mathcal{K}_d(\mathbf{v}) dl$ may be treated as a phase $F(\mathbf{v})$ reached by the "G" wave at point P_r^c in the case of a deformed crystal. The formula (14) describes the fringe distribution along the line ab of the section topograph of the deformed part of the crystal plate (Figure 1).

V. Computations and Results

A section topograph performed in "parallel geometry" allows one to observe parallel shifts of the Kato fringes (Figure 1). For the interpretation of this kind of fringe pattern it is convenient to apply the concept of the fan of beat waves. The force S can be evaluated from the topograph just after determination of the positions p and p' of the chosen fringe. Let p' denote the fringe position related to the perfect part of the plate and p the position related to the deformed part (Figure 1). The value of an initial phase, $2\pi b$ may be assumed as independent on p , according to the considerations of Kato [9]. Hence, the value of a lateral force S can be fitted by using the formula resulting from comparing (12) and (14):

$$t_0 \sqrt{1-p'^2} = \int_A^{P_r^c} (1 + \eta_d^2 / \cos^2 \theta_B)^{-1/2} dl, \quad (15)$$

where $\eta_d(p, l, S)$ is given by (6). The derivatives $\partial u/\partial z$ and $\partial u/\partial x$, necessary for the calculation of η_d , are functions of the position in the crystal and the value of S . The particular form of both derivatives can be found from elasticity theory if a particular deformation model is assumed.

For the computations it is convenient to use the coordinates (p, z) (Fig. 2) and to perform the integration along z instead of along l . Formula (15) takes then the form:

$$t_0 \sqrt{1-p'^2} = \sqrt{1+p^2 \tan^2 \theta_B} \quad (15a)$$

$$\cdot \int_0^{t_0} (1 + \eta_d^2 / \cos^2 \theta_B)^{-1/2} dz,$$

where t_0 denotes the thickness of the crystal plate and

$$\eta_d = \frac{p}{\sqrt{1-p^2}} + \frac{\Lambda_0}{d} \left(\frac{\partial u}{\partial z} + \tan \theta_B \frac{\partial u}{\partial x} \right). \quad (16)$$

The deformation model applied by Blech and Meieran [12] was used for the computation performed in this work. The formulae for $\partial u/\partial z$ and $\partial u/\partial x$ given in the paper of Blech, Meieran, were transformed to the coordinates (x, z) of Figure 2. After inserting them into (16), $\eta_d(p, z, S)$ has been found in the form

$$\eta_d = \frac{p}{\sqrt{1-p^2}} + \frac{\Lambda_0}{d} W B(p, z) S, \quad (17)$$

where d is the lattice spacing, W a term containing

elasticity constants and

$$W = \frac{2(1 + \sigma)}{\pi E},$$

(σ = Poisson ratio, E = Young's modulus).

$B(p, z)$ has the form

$$B(p, z) = \frac{N(t_0 - z)^3 - Qx(t_0 - z)^2 + Rx^2(t_0 - z) + Tx^3}{[x^2 + (t_0 - z)^2]^2}$$

for $x = p \cdot z \cdot \tan \theta_B$; $N = 1 - \sigma$; $Q = \sigma \cdot \tan \theta_B$; $R = 2 - \sigma$; $T = (1 - \sigma) \tan \theta_B$.

The procedure based on (15a) and (17) has been used to evaluate the lateral force S , acting at the

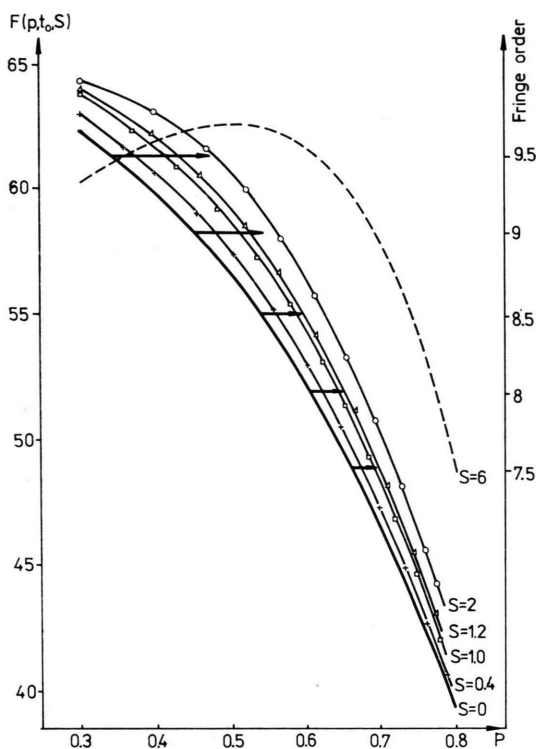


Fig. 4. Phases of "G" waves coming to the exit surface of a deformed crystal area are plotted as a function of the parameter p ($p = 0.5$ corresponds to the line lying in the distance of about $100 \mu\text{m}$ from the oxide edge). The curves are calculated for different values of a force S , (in $\text{dyn}/\mu\text{m} = 10 \text{ N/m}$). The phases F were calculated from (14). Horizontal arrows represent shifts of interference fringes read out from the topograph shown in Figure 1. The curve for $S = 6 \text{ dyn}/\mu\text{m}$ shows that in the case of higher stresses the assumed deformation model is not proper up to the distance of about $100 \mu\text{m}$ from the edge.

edge of the 2000 \AA oxide layer. $S = 15 \text{ N/m}$ has been found as a fitting value explaining the shift of the fringe indicated by arrows in Figure 1a.

For more a detailed analysis the shifts of all visible fringes should be considered. This can be done by using of (14), which allows one to calculate functions $F(p, t_0, S)$ (i.e. the distribution of the phases of the "G" waves at the exit crystal surface) for different values of S (Figure 4). The curve calculated for $S = 0$ represents the behaviour of this function related to the case of a perfect crystal. The horizontal arrows represent the shifts of the interference fringes read out from the topograph shown in Figure 1. The ends of the arrows indicate positions of fringes related to the deformed part of the crystal. They should be placed on one of the calculated curves. It can be seen that the experimental points manifest the tendency of systematical deviation from the calculated curve. A similar tendency can be observed on the graphs presented in the paper by Patel and Kato [3b]. This effect suggests that the applied deformation model does not work very well.

VI. Conclusions

The method of stress estimation proposed in this paper is based on the "parallel geometry" of performing section topographs. Simultaneously, the concept of the fan of "beat waves" was applied in the interpretation of images of perfect as well as deformed crystals. The proposed method allows one to estimate the stresses connected with the edge of the thin layer by means of simple calculations. Simplification was possible due to application of the concept of "beat waves" propagated along straight paths with changing wavelengths in the deformed crystal area. Its application leads to similar results as the "optical" model anticipating curved trajectories.

The proposed model can be applied only in the case of spherical waves falling on the crystal, i.e. when the whole Borrmann triangle is excited.

Finally, it is worth mentioning that the parameter $\{\Delta \mathbf{k}(\mathbf{v}) \cdot \mathbf{v}\}$ which has been taken in this method as the length of a beat wave vector, $\mathcal{K}(\mathbf{v})$, is a spatial equivalent of an energy gap which determines properties of solids in relation to excitations at different energies.

Acknowledgements

The author like to express his appreciation to Professor J. Auleytner for constant encouragement

and helpful discussions. It is also pleasure to thank to Mr. M. Tomala for performing the computations. The specimens were prepared by Dr. M. Liberadzka from the Institute of Electron Technology.

Appendix

A Comparison of the Beat in a System of Two Coupled Pendulums and the Pendellösung Effect.

Phenomenon in time:

Two coupled pendulums or oscillators

a and b .

Two modes of oscillation have nearly the same *frequencies*

ω_1 and ω_2 .

The motion of each pendulum exhibits beats. Total energy is constant and that flows back and forth from pendulum a to pendulum b at the beat frequency of two modes [7].

A beat period $T = \frac{2\pi}{\omega_1 - \omega_2}$.

A beat frequency

$\omega_{\text{beat}} = \omega_1 - \omega_2$

Phenomenon in space:

Two coupled waves

“O wave” and “G wave”.

Two Bloch waves have nearly the same *wave vectors*

$\mathbf{k}_{0,g}^{(1)}$ and $\mathbf{k}_{0,g}^{(2)}$.

Interference between two waves result in beats. Energy of “O wave” is transferred to “G wave” and back to “O wave” in the distance equal to fringe spacing [2].

Fringe spacing $\Delta(\mathbf{v}) = \frac{2\pi}{[\bar{\mathbf{k}}_{0,g}^{(1)}(\mathbf{v}) - \mathbf{k}_{0,g}^{(2)}(\mathbf{v})]\mathbf{v}}$

and a length of

a beat vector

$\mathcal{K}(\mathbf{v}) = [\mathbf{k}_{0,g}^{(1)}(\mathbf{v}) - \bar{\mathbf{k}}_{0,g}^{(2)}(\mathbf{v})]\mathbf{v}$.

are analogous parameters.

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