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SHORT COMMUNICATIONS

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Comments on papers on quantitative phase determination from three-beam diffraction by Chang and Tang (1988). By K. HÜMMER and E. WECKERT, *Institut für Angewandte Physik, Lehrstuhl für Kristallographie der Universität, Bismarckstrasse* 10, *D*-8520 *Erlangen, Federal Republic of Germany*

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Abstract

In two papers [Chang & Tang (1988). *Acta Cryst.* **A44**, 1065-1072 and Tang & Chang (1988). *Acta Cryst.* **A44**, 1073-1078] the authors are confused with respect to the rotation sense of the crystal lattice during a Renninger ψ-scan experiment. This leads to wrong phase determination. We show that the definition of the triple phase sum involved in a three-beam case used by Chang & Tang is not valid if strong anomalous-dispersion effects must be taken into account.

The asymmetry of the integrated ψ-scan profiles scanning through a three-beam position contain information on the phase difference between the directly diffracted wave (primary reflection) generated by diffraction of the incident beam at the lattice planes of the reciprocal-lattice vector (r.l.v.) **G** and the 'Renninger *Umweg*' wave generated by simultaneous diffraction at the lattice plane of the r.l.v.'s **L** (secondary reflection) and **G** − **L**.*

[Parenthetic note: The schematic representation of the three-beam interaction in Fig. 1.1 of the review article of Chang (1987) is wrong. The diffraction condition of the incident beam with respect to the lattice planes **G** − **L** is not fulfilled. In a three-beam case **0**/**G**/**L**, three strong wave fields are excited with wave vectors **K**(**0**), **K**(**G**) and **K**(**L**). A wave field with **K**(**G** − **L**) does not exist.]

It is well known and proved theoretically and experimentally that the asymmetry of a ψ-scan profile depends also on the rotation sense of the reciprocal lattice relative to the Ewald sphere, independent of the special three-beam position selected from the two possible three-beam positions for each individual three-beam case.

To be clear in the nomenclature for the rotation sense, we define the following: A ψ scan through a three-beam position is called an 'in-out' ψ scan when the second r.l.v. **L** lies inside the Ewald sphere at the beginning of the ψ scan and outside at the end of the ψ scan. In Chang's (1987)* nomenclature, this is called an outgoing position. The opposite rotation sense, an out-in scan, is called by Chang the incoming position.

* Here we use the nomenclature of Chang & Tang (1988) and Tang & Chang (1988).

* For simplicity in citation we refer to the review article of Chang (1987), where the previous papers of Chang *et al.* are summarized.

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Table 1. *Calculated (subscript T) and experimentally determined (subscript E) phases of* GaAs *for* $\lambda = 1{\cdot}1236$ Å *with* Ga *at* 000 *and* As *at* $\frac{1}{4}\frac{1}{4}\frac{1}{4}$

For noncentrosymmetric structures we define the moduli of the phases in the range 0 to 180° and we add the sign.

G	L	G−L	$\varphi(\mathbf{G})$ $\varphi(-\mathbf{G})$	$\varphi(\mathbf{L})$	$\varphi(\mathbf{G}-\mathbf{L})$	δ_T	Φ_T	Φ_E
222	113	11−1	+113·0	+51·8	+50·6	−144·6	−10·6*	−39
	115	11−3	+113·0	−40·4	−40·7	+31·9	+165·9	+147
	−1−13	33−1	+113·0	+51·8	+53·3	−141·9	−7·9*	−44
	1−15	13−3	+113·0	+54·8	+53·3	−138·9	−4·9*	−54

In Fig. 3 of the paper on theoretical considerations on quantitative phase determination (Chang & Tang, 1988), calculated dynamical profiles (the solid curves) are shown. These profiles, reading from the left-hand side ($\Delta\varphi$ positive) to the right-hand side ($\Delta\varphi$ negative), must refer to an in-out ψ scan. This can be seen, for example, by looking at the profiles for the triple phase sum $\delta = 0$ and $\delta = 180°$. It has been proven experimentally (Gong & Post, 1983; Hümmer, Weckert & Bondza, 1989) that such profiles refer to in-out ψ scans. For triple phase sums with $\cos\delta > 0$ and for an in-out rotation sense, the so-called 'ideal profiles' (Hümmer, Weckert & Bondza, 1989) or the 'dynamical' profiles in Chang's nomenclature must first show an increase of the two-beam intensity, because of the constructive interference between the directly diffracted and the *Umweg* wave.

In Fig. 5 of the second, experimental, paper about a quantitative phase determination (Tang & Chang, 1988), 'dynamical profiles' are shown which are deduced from the measured 'total profiles'. The asymmetry of the total and dynamical profiles are identical. In agreement with the theoretical paper (Chang & Tang, 1988), the dynamical profiles were exploited to determine the triple phase sum δ as if these profiles refer to an in-out ψ scan, reading them from left to right. However, as is pointed out in several papers by Chang and co-workers (*cf.* Chang, 1987), 'these profiles were obtained at the IN positions' (Chang, 1986); this is also indicated by the positive sign of S_R. Thus, the measured profiles refer to an out-in ψ scan. The experimental rotation sense and the rotation sense assumed for phase determination contradict each other.

To make certain that we had not misunderstood the papers of Chang and co-workers, we also measured the three-beam ψ-scan profile of Fig. 5, using the special ψ-circle diffractometer installed at DORIS II at HASYLAB, DESY, Hamburg. With our special ψ-circle diffractometer, any error in fixing the rotation sense is excluded, because the rotation sense can be read directly from the rotation sense of the separate ψ axis. We were anxious to use the same setting of GaAs as Tang & Chang did, being aware that in noncentrosymmetric structures with roto-inversions $\bar{4}$ (space group of GaAs $F\bar{4}3m$) a twofold ambiguity exists with respect to the absolute axis assignment (Jones, 1986; Burzlaff & Hümmer, 1988). The calculated phases given by Tang & Chang are only consistent with the setting Ga at 000 and As at $\frac{1}{4}\frac{1}{4}\frac{1}{4}$. As a result we measured the same asymmetries consistent with the results of Tang & Chang for an out-in scan. We also calculated the intensity profiles for the three-beam cases investigated by Tang & Chang (1988), the asymmetries are consistent with the experimental results.

So in earlier papers by Chang and co-workers (*cf.* Chang, 1987), all is correct and it is stated in several papers that the intensity profiles refer to the incoming position; to re-iterate, this is in our nomenclature an out-in ψ scan, reading the profiles from left to right.

Why did the authors change the rotation sense in the last paper (Tang & Chang, 1988)? In Chang & Tang (1988), in equation (38), a new definition is given for S_R. However, this relation makes no sense to us, as $S(l^2 - \mathbf{l}\,.\,\mathbf{g})$ is positive or negative dependent on the mutual orientation of the r.l.v.'s $\mathbf{l}$ and $\mathbf{g}$. Its sign is independent of the rotation sense.

In several papers (*cf.* Chang, 1987), the running direction of the paper chart is indicated on the ψ-scan profiles. It should be realized that the running direction of the paper chart without further detail does not give any information on the rotation sense of the crystal.

There is reasonable doubt whether in the case of strong anomalous dispersion the triple phase sum $\delta = \varphi(-\mathbf{G}) + \varphi(\mathbf{L}) + \varphi(\mathbf{G}-\mathbf{L})$ is the relevant phase relationship which can be deduced from the ψ-scan profiles. The φ's represent the individual reflection phases. This relation is used in the papers commented on. In GaAs for $\lambda = 1{\cdot}1236$ Å strong anomalous-dispersion effects occur. In particular, the phase of the primary reflection $\mathbf{G} = 222$ is affected. It is shifted from 188·2° for $\lambda = 1{\cdot}5405$ Å (Cu $K\alpha_1$) to +113·0° for $\lambda = 1{\cdot}1236$ Å. Calculating these phases, we used the corrections for the atomic scattering factors due to anomalous dispersion from the data file based on the algorithm of Cromer & Liberman (1981).

To discuss this point in more detail we consider the result of the Bethe solution for three-beam cases, *i.e.* a modified two-beam approximation, exploited by several authors (Juretschke, 1984; Shen, 1986; Chang & Tang, 1988). To simplify matters we write (*cf.* Hümmer & Billy, 1986)

$$D(\mathbf{G})/D(\mathbf{0}) \sim \alpha F(\mathbf{G}) + \beta R(\mathbf{L}) F(\mathbf{L}) F(\mathbf{G}-\mathbf{L})$$

with

$$R(\mathbf{L}) = K(\mathbf{L})^2/[K^2 - K(\mathbf{L})^2].$$

α and β stand for scalar products of the different modes of polarization.

As in a ψ-scan experiment, the relative change of the integrated two-beam intensity $I(\mathbf{G})$ is measured, we rewrite

$$D(\mathbf{G})/D(\mathbf{0}) \sim \alpha F(\mathbf{G})\{1 + (\beta/\alpha) R(\mathbf{L}) \times [F(\mathbf{L}) F(\mathbf{G}-\mathbf{L})/F(\mathbf{G})]\}.$$

From the term $F(\mathbf{L})F(\mathbf{G}-\mathbf{L})/F(\mathbf{G})$, it can be seen that the triple phase sum which governs the three-beam interference is given by

$$\Phi = \varphi(\mathbf{L}) + \varphi(\mathbf{G}-\mathbf{L}) - \varphi(\mathbf{G}).$$

In the case of anomalous dispersion, the triple phase sums δ and Φ give different values, because Friedel's law is no longer valid (*cf.* Table 1).

To be clear, we list in Table 1 the individual reflection phases and the triple phase sums δ and Φ for the three-beam cases under discussion for $\lambda = 1{\cdot}1236$ Å. Looking at the ψ scan profiles of Fig. 5 in the second paper (Tang & Chang, 1988), one can immediately see that the asymmetries correspond with Φ and are contradictory to δ, bearing in mind that the measured profiles must refer to an out-in ψ scan. For the cases marked with an asterisk in Table 1, cos Φ is positive, for the fourth case cos Φ is negative. As a consequence, Φ is the relevant phase relationship.

Assuming that the phase-determination process proposed by Chang & Tang (1988) is applicable, then the experimentally determined δ_E (Tang & Chang, 1988) can be used to estimate the triple phase sum Φ_E for the right rotation sense, namely for an out-in ψ scan. It is known that the intensity profiles for Φ and $180° - \Phi$ are related by a mirror line through $\Delta\psi = 0$, that is to say, the asymmetry of an in-out ψ-scan profile for Φ is equivalent to the asymmetry of an out-in ψ scan for $180° - \Phi$. So, Φ_E for the right rotation sense is given by (*cf.* Table 1)

$$\Phi_E = 180 - \delta_E.$$

The values of Φ_E confirm that in the case of anomalous dispersion the triple phase sum Φ and not δ can be deduced from the ψ-scan profiles.

There is another point to be mentioned. In several previous papers, Chang stated (*cf.* Chang, 1987) that without anomalous dispersion only the cosine of δ (cos δ) can be determined. This statement is disproved by our measurements (Hümmer, Weckert & Bondza, 1989). We are surprised to see that in the theoretical paper (Chang & Tang, 1988) it is stated that the sign of δ can also be determined without any discussion about anomalous dispersion.

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