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Comments on *The phases of forbidden reflections*, by Post & Ladell (1987)

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Abstract. The observations of Post & Ladell [*Acta Cryst.* (1987), A43, 173–179] of the asymmetry of the strong primary reflection 311 in germanium in the neighborhood of weak reflections such as {222} are shown to be in good quantitative agreement with the predictions of this asymmetry based on first-order perturbation theory of such interactions [Juretschke (1986). *Acta Cryst.* A**42**, 405–406].Since the theory accounting for these data is insensitive to the phases entering in these many-beam interactions, the asymmetries observed in this experimental configuration do not allow any inferences about the phases of forbidden reflections.

In a recent exchange of comments by Templeton (1988) and Post & Ladell (1988) concerning Post & Ladell's (1987, referred to below as PL) reassignment of forbidden reflections in the diamond structure based on an experimentally observed asymmetry of integrated reflections of strong beams in the neighborhood of weak three-beam interaction points, the latter authors try to dismiss the earlier theoretical prediction (Juretschke, 1986, referred to below as HJ) that this type of experiment contains no phase information with the phrase 'As the reader can readily determine, the calculated curves bear no resemblance to any of the 24 experimental curves in Post & Ladell (1987) and, in particular, to the four experimental curves which deal with the cases discussed by Juretschke'. This is a grossly misleading statement, and, as shown below, any careful reader of both theory and experimental data would come to a very different conclusion.

The theory for a strong primary reflection interacting with weak beams is part of a general formalism (Juretschke, 1984) for describing the leading asymptotic contribution to the change of integrated two-beam intensity as an *n*-beam point is approached, *e.g.* by decreasing the azimuthal angular deviation ϕ_T from the *n*-beam point. This contribution varies like

$$\Delta I_{HL}/I_H = -C_{HL}/\phi_T \tag{1}$$

and, being odd in ϕ_T, is asymmetric around the *n*-beam point. C_{HL} is always positive when ϕ_T is outside the Ewald sphere, regardless of the *n*-beam invariant phase of the interaction. Of course, as pointed out explicitly in HJ, for sufficiently small ϕ_T, higher-order terms, of both even and odd powers of $1/\phi_T$, may also give contributions to the intensity, but in most circumstances this occurs at values of ϕ_T overlapping the resolutions of the incident beam and of the detectors, so that there the interpretation of the experimental data becomes complex.

In any case, however, the asymptotic contribution (1) can be extracted from the experimental data by plotting the contribution to the intensity that is *odd* in ϕ_T, *i.e.* $[I(\phi_T) - I(-\phi_T)]/2$ *vs* $1/\phi_T$. The initial slope of such a plot determines C_{HL}.

The well documented data of the figures of PL readily lend themselves to such an analysis. Column *H* of their Table 1 allows the establishment of a relative intensity scale, and each figure designates the exact three-beam point where $\phi_T = 0$.

A sample analysis of this type is shown in Fig. 1. Fig. 1(*a*) is a smoothed-out curve through data points taken from Fig. 3(*a*′) of PL, describing the 311/222 interaction, and plotted using the scales of Fig. 1 of HJ. A purely visual comparison of the two figures shows that the *odd* component of Fig. 1(*a*) is of the same order of magnitude as that given by the theoretical curve of Fig. 1 of HJ. A quantitative analysis of Fig. 1(*a*) along the lines outlined above is given in Fig. 1(*b*). Within the cumulative errors resulting from a noisy signal then approximated by a smooth curve and finally reduced by extraction of an intensity difference in order to obtain the odd component, Fig. 1(*b*) clearly shows the predicted $1/\phi_T$ dependence, and as extending over a relatively wide range of angles. In fact, it allows quite a good determination of $C_{311/222}$, with a value 0.86(15) s. This number should be compared with 0.42 s, that was used to plot the theoretical 311/222 contribution for pure σ polarization in Fig. 1 of HJ.

A similar analysis has been carried out for the curves (*a*), (*b*) and (*b*′) of Fig. 3 of PL. The

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results for all four cases are summarized in the first row of Table 1. There is a systematic difference between the constants for the experimental E (entering) and L (leaving) curves, but since their probable errors overlap anyway, this is of no concern here. Clearly, the relatively high noise in the $\bar{2}22$ data requires more experimental input before a better number can be defined.

The results, extracted directly from PL's experimental data, can be compared with two stages of the theory. In the first stage, we apply equation (4) of HJ to both σ and π polarizations separately, and also list the result expected for unpolarized incident radiation. In the second stage, the theory is refined to include also (a) the usual phase-sensitive contribution, which is positive for 222 and negative for $\bar{2}22$, and (b) a correction for absorption resulting from the right-hand factor in square brackets of equation (3) of HJ. Both stages of the theory are listed in the second and third sections of Table 1. These numbers clearly indicate that there is very good quantitative agreement of the PL data (within their experimental uncertainties) with both stages of the theory. At this level of the experiments, they cannot distinguish between the two stages, nor can they identify the phase-sensitive contributions (about 10%) hidden in the data.

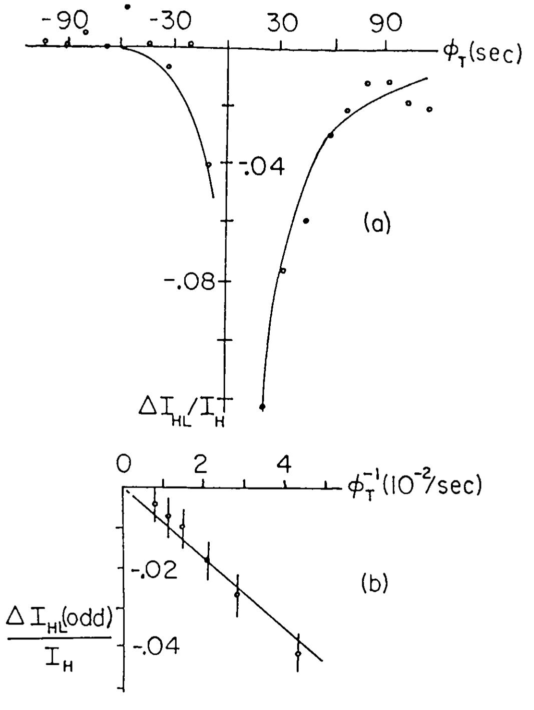

Fig. 1. (a) Smoothed-out representation of data points (circles) taken from Fig. 3(a') of PL, and plotted using the scales of Fig. 1 of HJ. (b) Extraction of the coefficient C_{HL} defined in equation (1) from the curve of (a).

Table 1. *Comparison of the experimental value* $C_{311/L}$ (s) *with two stages of dynamical theory*

	$L = 222$		$L = \bar{2}22$	
	L(eaving)	E(ntering)	L(eaving)	E(ntering)
C_{HL}(exp)	0.60(15)	0.86(15)	0.15(15)	0.29(15)
Theory 1 [equation (4) of HJ]				
$C_{HL}{}^{\sigma}$	0.42		0.07	
$C_{HL}{}^{\pi}$	0.90		0.13	
$C_{HL}{}^{\text{unpol}}$	0.60		0.09	
Theory 2. The above, with corrections for the phase-sensitive term (in parentheses) and for absorption				
$C_{HL}{}^{\sigma}$	0.49[+0.03]=0.52		0.083[−0.008]~0.08	
$C_{HL}{}^{\pi}$	1.18[+0.03]=1.21		0.175[−0.006]~0.17	
$C_{HL}{}^{\text{unpol}}$	0.78		0.11	

However, the ability to obtain such good agreement in the first place speaks for the overall excellence of the experimental data of PL, and it is precisely this excellent experimental work that confirms the asymptotic theory as satisfactorily representing the 'reasonable facsimile of the corresponding physical reality' that they had questioned (Post & Ladell, 1988), on rather superficial grounds. It should be noted that application of the theory is not restricted to perfect crystals (Juretschke, 1988).

A similar discussion applies to the other interactions studied by PL. Since their experimental observations agree so well with a theory of the asymmetry that is insensitive to phase information, it is evident that no phase information can be extracted from the data of PL. To establish whether or not the phase assignments proposed by PL are correct will require another experimental configuration that is sensitive to such information, using criteria such as, for example, equation (5) of HJ.

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