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Visibility of the asymmetry effect in multiple diffraction experiments in benzil. Wavelength dependence.

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Abstract

The observability of the asymmetry effect in multi-beam diffraction on benzil is discussed in the light of the objections raised by Post, DiMarco & Kiszenick [*Acta Cryst.* (1989). **A45**, 654-656]. It is shown that, while their data with Cu $K\alpha$ do not show convincingly the asymmetry effect, those with Cr $K\alpha$ do, thereby confirming our previous assertion that long-wavelength photons are needed when dealing with light atoms and large unit cells.

Following our initial studies of multiple diffraction in benzil in relationship to the phase problem (Shen & Colella, 1986, 1987, 1988), similar work has been pursued in other laboratories (Hümmer, Weckert & Bondza, 1989; Post, DiMarco & Kiszenick, 1989). In the last and most recent reference mentioned, hereafter described as PDMK, exception was taken in regard to a statement we had made in our earlier papers, when we reported our inability to detect the asymmetry effect in the *Umweganregung* pattern taken with Cu $K\alpha$ ($\lambda = 1{\cdot}54$ Å) and Cr $K\alpha$ ($\lambda = 2{\cdot}29$ Å).

We did later detect, indeed, the asymmetry effect by using longer wavelengths (3·5 Å), and explained the different results obtained in terms of the different widths of the asymmetric profiles in the azimuthal plots,* when compared with the mosaic spread of the crystal, of the order of 2′. We then concluded, on the basis of the results obtained with synchrotron radiation, that it would have been impossible to observe the asymmetry effect in benzil with 1·54 Å X-rays, even with the best instrumental resolution attainable.

This statement has been alledgedly contradicted in the PDMK paper, in which some experimental results are presented to support the authors' viewpoint.

Our statement was based on an experiment done at the National Synchrotron Light Source, at Brookhaven National Laboratory, using a large high-quality parallel-plate benzil single crystal. (The details of the crystal preparation technique are given in our 1986 and 1988 papers.) The results are presented in the three figures accompanying this paper. The wavelength used was 1·55 Å. In Fig. 1 we show a portion of the azimuthal scan for the 505, a rather weak reflection.

We only show a limited portion because preliminary work showed that the 400 was the strongest *Umweg*, and therefore it appeared to be the best candidate for observing the asymmetry effect, on the basis of our previous experience.

The experimental width of the 400 *Umweg* peak (0·03°) can be taken to be close to the mosaic spread, since the beam was highly collimated, probably within 0·004°, in a plane perpendicular to the diffraction plane.

Fig. 2 shows in detail the pedestal of the 400 *Umweg* peak. It is clear that no asymmetry effect is visible.

In Fig. 3 we show a *calculated* azimuthal profile, using the perturbation theory developed by one of us (Shen, 1986). The peak value is not significant because this theory breaks down at the exact three-beam excitation point. Fig. 3 is calculated assuming zero mosaic spread. It is clear from the profile shown that the whole action takes place within

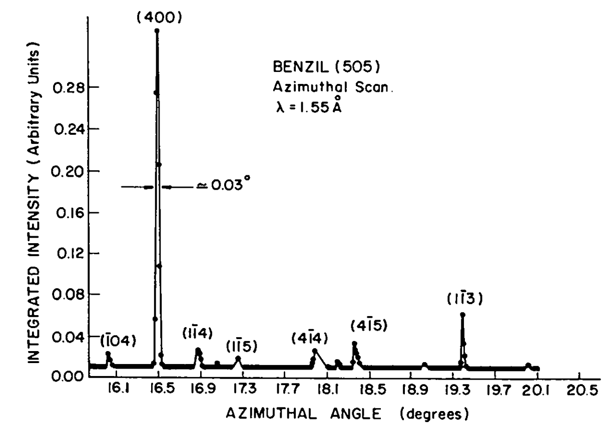

Fig. 1. Integrated intensity of the 505 reflection as a function of the azimuthal angle. The zero on the angular scale corresponds to the (1̄20) lying in the diffraction plane, mostly antiparallel to the incident beam. Data taken using synchrotron radiation at NSLS (Brookhaven).

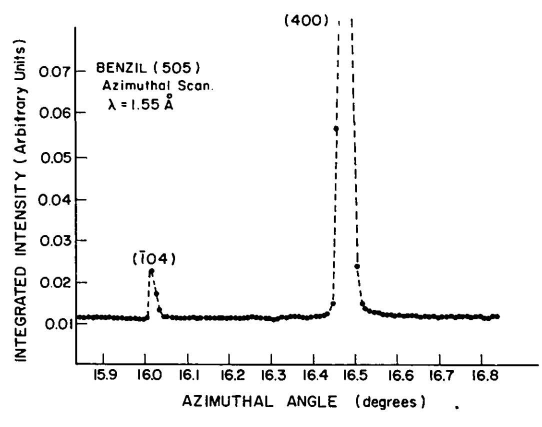

Fig. 2. Detail of Fig. 1, in the neighborhood of the 400 *Umweg*.

* By 'azimuthal plot' we mean a plot of integrated intensity (with respect to θ, angle of incidence on the lattice planes) as a function of φ, azimuthal angle.

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a range of 0·03°, just about the mosaic spread of the crystal. In such a situation it is clearly impossible to observe the asymmetry effect, unless a perfect benzil crystal is available, with zero mosaic spread, an unlikely possibility.

The same calculated profile, of the kind shown in Fig. 3, would be expanded by a factor of ten, approximately, at 3·5 Å, which is what prompted us to explore the use of long wavelengths for these experiments.

In regard to the results reported by PDMK, it is impossible to make any comments on the Cu $K\alpha_1$ profiles (Fig. 1 of PDMK) since no information is given about the Miller indices of the *Umweg* reflections, no angular values are indicated for the abscissae, and the authors do not make any comparison with theory. The only thing we can say is that, for the 006 scan with Cu $K\alpha_1$, there are 758 *Umweg* peaks over a range $\Delta\varphi$ of 60°, corresponding to an average of 12·6 peaks per degree. Most likely, the features present in Fig. 1 of PDMK, purporting to show the asymmetry effect, are artifacts due to unresolved *Umweg* reflections occurring very close to the main peaks.

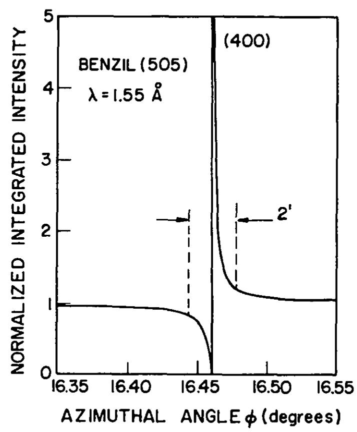

Fig. 3. A three-beam calculation for the benzil 505–400 multiple reflection with $\lambda = 1{\cdot}55$ Å, using Shen's (1986) perturbation theory. Each point in this plot is an integrated intensity with respect to θ, the angle of incidence on the (505) lattice planes. The intensities are for a perpendicularly polarized incident beam and have been normalized to the two-beam value. It shows that the asymmetry effect is confined within a very small angular region, comparable to the mosaic spread of the crystal. The peak value, corresponding to full excitation of the 400, is not plotted on this graph.

The results obtained with Cr $K\alpha_1$, on the contrary, do indeed show genuine asymmetry effects. The most convincing case is peak no. 2 in Fig. 2 (of PDMK), which could be reproduced by our computer simulation, based on our earlier theoretical work (Colella, 1974; Shen, 1986), since in this case the Miller indices and the angular values on the abscissae were given.

There is no real contradiction between these results and our previous statements. Essentially, by resorting to Cr $K\alpha_1$ radiation ($\lambda = 2{\cdot}29$ Å), PDMK agree with us that use of long-wavelength X-rays is the key for observing the asymmetry effect in crystals with low electron density such as benzil. The reason we were not able to observe the asymmetry effect in benzil with Cr $K\alpha$ (Shen & Colella, 1986) is that we did not have high resolution in a plane perpendicular to the diffraction plane. In PDMK's work, high resolution (45″ divergence) was present in all directions, which made it possible to see the asymmetry effect.

We can then reach the following conclusions. Our original statement regarding the non-visibility of the asymmetry effect in benzil using $\lambda = 1{\cdot}54$ Å was referred to a particular choice of main and simultaneous reflections (505 and 400). On the basis of our experience and theoretical calculations, we believe that this combination of *hkl*'s is the best candidate, and that most likely our statement is true for all other choices of *hkl*'s. This statement has not been contradicted by PDMK. On the other hand, PDMK show that by proper use of high resolution a medium-long wavelength such as Cr $K\alpha$ ($\lambda = 2{\cdot}29$ Å) the asymmetry effect in benzil is made visible without going to the extreme case of $\lambda = 3{\cdot}5$ Å, which necessitates the use of synchrotron radiation.

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