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Relation between habit and structure of liquid single crystals of blue phases (Reply to the preceding Comment by P. Pieranski and P. E. Cladis)

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REPLY

Relation between habit and structure of liquid single crystals of blue phases

(Reply to the preceding Comment by P. Pieranski and P. E. Cladis)

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In the preceding Comment [1] Pieranski and Cladis assert that the blue phase space groups derived in our Invited Article [2] are not consistent with the growth forms observed for our BP single crystals (*cf.* table 3 of [2]). We wish to reply as follows.

(i) The space groups given for the BP I (I 4_132) and the BP II (P 4_232 or P 23) in [2] have not been derived solely from the morphology of our BP single crystals but additionally from results of Bragg scattering and electric field-induced distortions of both BPs (*cf.* [3]) as can be seen by a careful study of §4.2.4 in [2]. The Donnay–Harker rule has only been used for a preselection of possible space groups.

(ii) Pieranski and Cladis [1] claim that the form {100} is absent in BP I and BP II. In our three-dimensional BP I single crystals [4] we derived {100} from the measured crystallographic angles by means of a stereographic projection (see figure 1). As the elevation angle could not be obtained experimentally we cannot decide strictly if several sides of our rhomb-like BP I single crystals (*cf.* figure 19 in [2]) are due to {100}

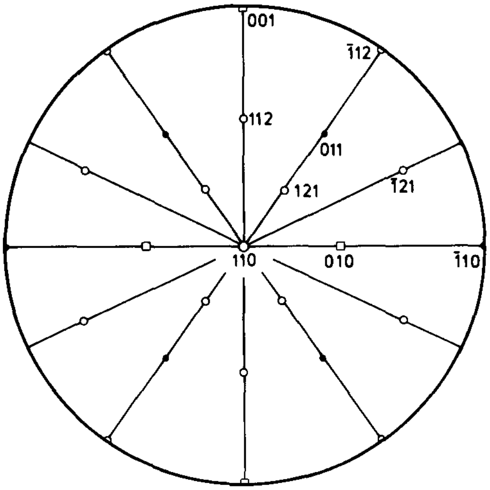

Figure 1. Wulff net for the stereographic projection of a three dimensional BP I single crystal.

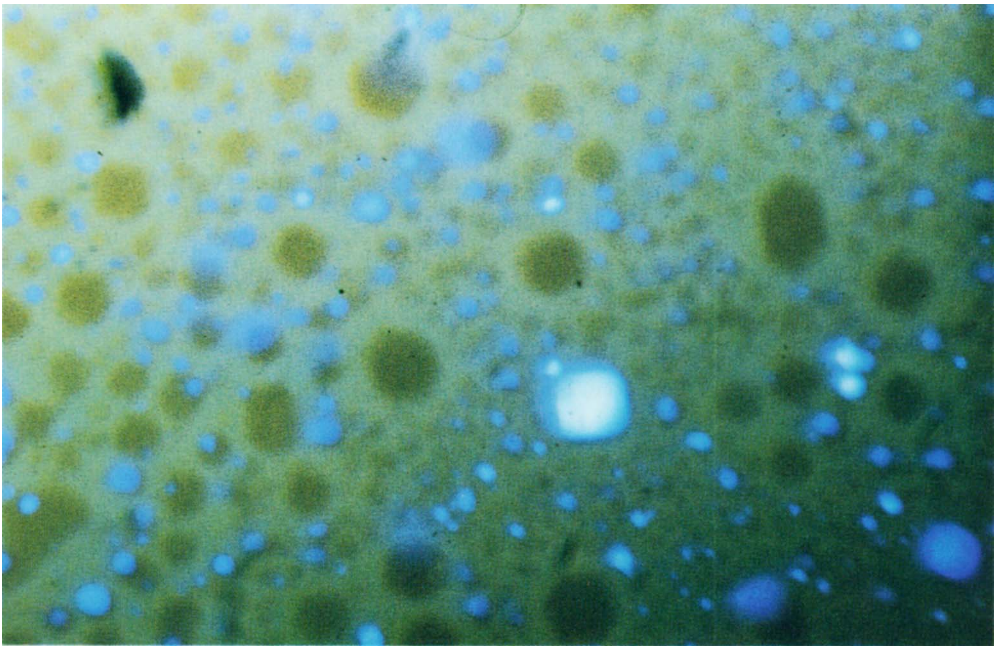

Figure 2. Microphotograph of BP II single crystals of the system cholesteryl benzoate/4-*n*-octyl-4′-octyloxybenzoate with 37·4 mol per cent CB; 102·05°C, sample thickness 60 μm.

or caused by sections of, for example, (112) with the projection plane (see figures 18 and 20 in [2]).

All *BP II single crystals* which grow rapidly from the fog phase BP III occur in a quadratic shape in (100) orientation. Supposing that the observed squares are limited by (110) faces then crossing medial lines should occur within the squares. As we never observed such crossed lines in rapidly growing BP II crystals their habit is governed by the {100} form. In dimorphic BP systems we observed very slowly growing BP II single crystals with hexagonal as well as quadratic habits. In figure 2 we show the BP II crystals of a mixture cholesteryl benzoate (CB)/4-*n*-octyl-4′-octyloxy benzoate (8OBE) with 37·4 mol per cent CB. These crystals are indeed limited by the form {110}. We were able to detect medial lines across the blue squares indicating a rhombic dodecahedral crystal in (100) orientation. The BP II hexagons in figure 2 are very difficult to detect by polarizing microscopy because they do not reflect visible light. Only by turning the polarizers slightly out of the crossed position does a small contrast to the environment occur. Obviously, the wavelength of the Bragg scattering at the (110) planes is in the UV.

(iii) The contribution of the {100} form to the habit of both BPs cannot be excluded generally. This is not in contradiction with the Donnay–Harker rule which only concerns the *geometry* of a mass-point lattice of *solid* crystals. The morphological aspect observed for the *liquid* BP I crystal by Pieranski *et al.* [5] is indeed the Donnay–Harker form sequence for I $4_1 32$. Actually, the physical process of crystal growth is governed by the interfacial free energy of the faces involved in the equilibrium polyhedron. Only by this concept given very early by J. W. Gibbs can a similar relation between habit and structure in *solid* as well as in *liquid* single crystals be understood. It must be taken into account that the equilibrium polyhedron usually differs from the growth polyhedron because the growth velocity of the faces is not constant but depends on the environment of the crystal. Thus, it is not surprising that different habits of the BP crystals have been observed in systems with different chemical compounds and compositions (*cf.* [2] and [5–7]). The influence of the environment also explains the appearance of additional forms during the growth of our BP I single crystals [2, 4], whereas in solid crystals a depletion of forms occurs on crystal growing. Due to large biphasic regions BP I/isotropic the composition of the isotropic liquid changes during the growth which causes a change in the interfacial energy.

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