Lamellae and their Organization in Melt-Crystallized Polymers [and Discussion]

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Lamellae and their organization in melt-crystallized polymers†

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Significant advances in knowledge of lamellae and their organization in melt-crystallized polymers have stemmed from the ability to examine internal morphologies systematically with the transmission electron microscope. Spherulites form because the first-forming (dominant) lamellae branch repetitively, often at giant screw dislocations, then diverge substantially creating a skeleton to which later-forming lamellae must accommodate. This sequence promotes chain-folding, invites fractional crystallization and modulates chemical, mechanical and thermal properties of spherulites at the inter-dominant spacing. The key feature of lamellar divergence at screw dislocations is present in individual crystals, probably deriving from pressure of uncrystallized molecular cilia; growing lamellae will also distort very substantially to gain material. If necessary, spacefilling is achieved without lamellar and crystallographic continuity by nucleating new growth at large misorientations. Individual melt-grown crystals have been studied both after extraction from a quenched matrix and *in situ* in thinned specimens. For polyethylene different lamellar profiles have been placed in context while their fine structure provides insights into molecular mechanisms of growth.

1. Introduction

Within the physical texture of solid polymers are messages concerning the history of the system and the ways in which the constituent macromolecules came to be organized. That information is greatest in complex crystalline systems, of which polyethylene is the prototype, but it has to be able to be read. In the first instance one needs to inspect the texture in a controlled manner and recognize patterns in its organization. The origin of these patterns then has to be suggested, leading to formulations and quantitative evaluation. But the inspection of the microstructure is the essential first step. For crystalline polymers successive advances in our ability to reveal and study internal organization have led to substantial corresponding gains in understanding of the making of morphology and the fundamental molecular mechanisms involved.

The history of the subject is full of examples in which microscopic, real-space examination has moved the field significantly forward. Best known is the way in which the long-standing fringed-micelle model had to give way to the revolu-

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tionary concept of lamellar crystals and molecular chainfolding when the former were revealed and studied. Ever since, lamellae have been regarded as building blocks for melt-crystallized systems, though their nature, mutual disposition and interconnection have remained to be established and have sometimes been controversial.

The essential problem, which has hindered the acquisition of textural information, is radiation damage to specimens in the transmission electron microscope (TEM), an instrument which is still much the most productive means of imaging at requisite resolution. Not only is crystallinity destroyed by electrons, but associated mass transport will introduce detail which is not present in the original and may well be related to it in such a complicated way as to be of little use. Nevertheless two complementary means of revealing lamellae and their organization have transformed the situation. Chlorosulphonation of polyethylene plus permanganic (and other) etching of a wide range of polymers have led to substantial new knowledge, often different from previous best guesses, with important advances in understanding.

It has been shown that lamellae are indeed the building blocks of melt-crystallized polymers but that lamellae are not all equivalent, not even when grown isothermally. This stems from a basic phenomenon whereby adjacent layers of a multilayered crystal tend to diverge. Consequently, one lamella will, with repetitive branching at, for example, giant screw dislocations, generate the underlying framework of a spherulite, with the fastest growth direction radial. In certain low-crystallinity systems such a framework, based on individual, so-called dominant, lamellae is all that forms. In others the framework is filled in by subsidiary lamellae, sometimes themselves displaying complex hierarchical organization. Such a morphology invites time-dependent fractionation with later-crystallizing species (usually the shorter and/or more branched molecules) concentrated in subsidiary lamellae. It also introduces a modulation of properties at the interdominant spacing, even for isothermal growth: chemical, thermal and mechanical properties have now been demonstrated to vary systematically between dominant and subsidiary lamellae, i.e. with this periodicity.

Of internal lamellar organization less has been known until recently when means were devised of examining individual lamellae crystallized from the melt in bulk polymer. Lamellae extracted from a quenched matrix or examined directly within such a matrix in a thinned film by diffraction microscopy have revealed that they are not generally single crystals in the crystallographic sense but are usually severely and systematically distorted away from this ideal during growth. The more uniform textures which develop for crystallization at high temperatures and long times, inform on the way folded molecules adjust to a lamellar habit.

2. Morphological studies

(a) *Early days*

The development of morphological studies on polymers is well-known and needs no more than a summary here but it does provide good examples of the way observations of a well-chosen system can remove ambiguities and make significant advances in knowledge and understanding. The first model of the microstructure in crystalline polymers was that of the fringed micelle. It was based on diffraction

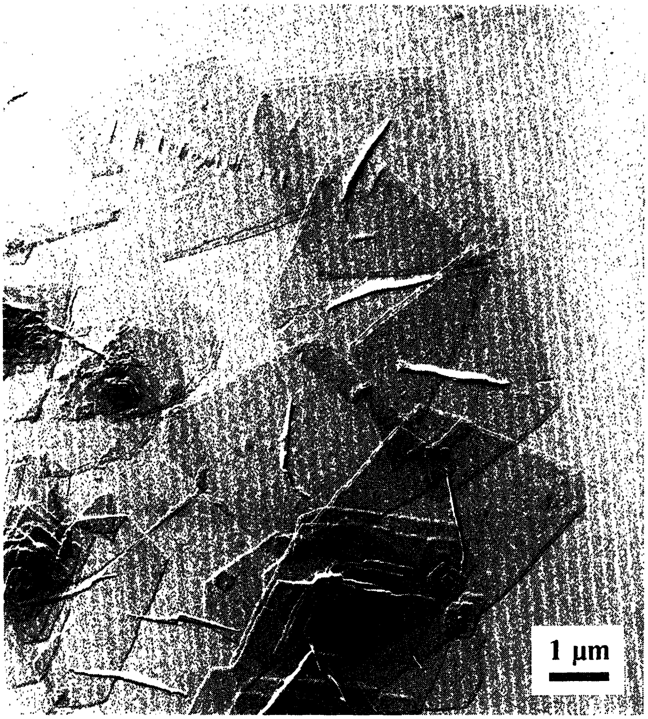

Figure 1. Crystals of linear polyethylene grown from dilute solution in xylene. The double step at their edges shows the crystal thickness is controlled by secondary nucleation while the internal morphology is evidence of initial non-planar habits due to fold interactions.

line-widths and the then new knowledge that polymers were composed of macromolecules. No microscopes could then resolve the postulated *ca.* 10 nm sizes and despite concerns as to how micelles were accommodated in spherulites, to which molecules were typically tangential, no clear alternative was available until the synthesis of linear and stereoregular molecules led to the observation of individual lamellae (figure 1) and the inference of chainfolding (Keller 1957; see also Keller 1958 and Bassett 1981 for reviews).

The consequences of these twin discoveries can hardly be exaggerated. In terms of properties, the fringed-micelle model, which had crystallites embedded in an amorphous matrix, indicated that the degree of crystallinity was the salient controlling quantity. Not so for lamellae: in these systems mutual disposition is all important and the significance of the degree of crystallinity comparatively trivial. This realization triggered searches for ways to change the internal organization, by enhancing the continuity of covalent links in systems of high chain extension, which has led in more recent times to families of high-performance polymers with substantially improved properties, initially mechanical (Frank 1964, 1970) and now conducting.

Study of individual lamellae precipitated from very dilute solution revealed distinct sectors – evidence for regularity in folding – and thermal instabilities (different in different sectors) leading to lamellar thickening, all of which are now basic features of our understanding. When growth was from more concentrated solutions, multilayer crystals developed, which in retrospect, were a much better

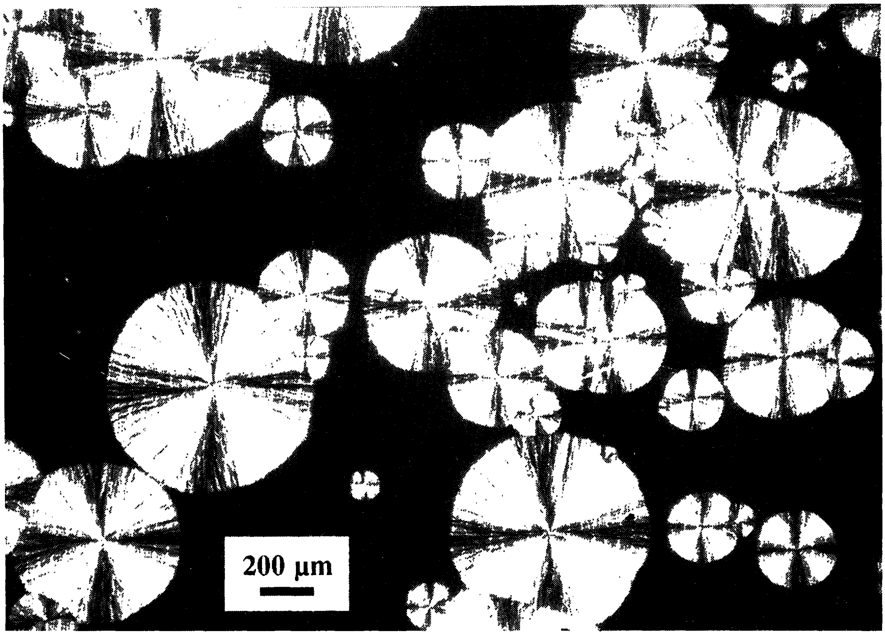

Figure 2. Spherulites of the α form of polypropylene viewed between crossed polars while crystallizing from the melt at 130 °C.

guide to melt-crystallized systems than was appreciated at the time, doubtless because of prevailing preconceptions. Although there were many beautiful illustrations of lamellae in the fracture surfaces of melt-crystallized polymers (Geil 1963), it was then questioned whether lamellae were typical of melt-crystallized polymers and when present if they could be regularly chainfolded. An inherent dichotomy was envisaged between well-ordered solution grown systems and poorly ordered melt grown polymers. Now that we have better evidence the picture which emerges is not black and white but shades of grey.

(*b*) *Spherulitic architecture*

Spherulites (figure 2) were the first morphological entity to receive systematic study. Tantalizingly their fine structure usually crossed the limits of optical resolution but TEM fracture surface studies showed it to consist of a framework of individual lamellae. Nevertheless such open structures were regarded as atypical and the general situation at the solid–melt interface to be that of a wide front, subdivided into lamellae and subject to processes of cellulation such as were known for binary metallic alloys (Keith & Padden 1963). Support appeared to exist for this point of view in blended systems but it has not been consistent with observations at higher resolution made once it became possible to examine the interior of melt-crystallized polymers systematically with the transmission electron microscope.

Figure 3 shows features universal to most crystalline synthetic polymers. There is lamellar and, therefore, crystallographic continuity from the centre (e.g. upper right) with branching at, and divergence from, giant screw dislocations – which have been preferentially etched – leading to a space-filling framework of individual

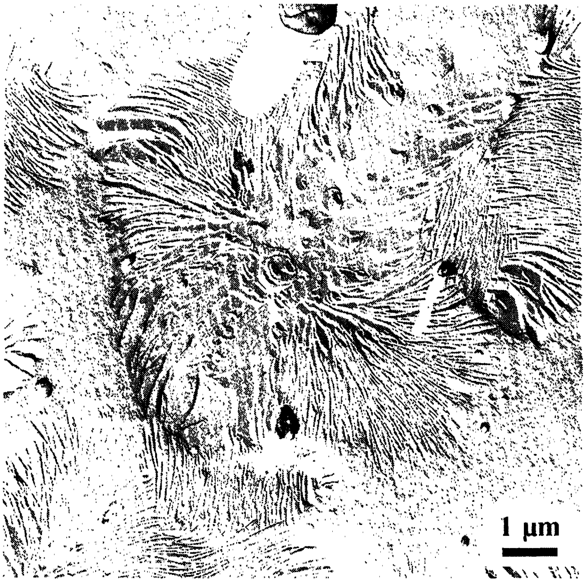

Figure 3. Embryonic spherulites of linear low-density polyethylene grown at 124 °C (from Bassett 1993).

dominant lamellae with the fastest growth direction (***b*** for polyethylene) radial; subsequent growth occurs within this framework as is shown particularly clearly on the left hand side. This pattern is adopted over a very wide range of growth conditions: it is certainly not restricted to low supercoolings. The more open textures found under those conditions seem rather to be a consequence of limited branching because of fewer screw dislocations.

The genesis of screw dislocations can be controlled, for instance by a reduction in growth temperature resulting in dislocations at the resulting interface (figure 4). The asymmetry in growth terraces associated with a screw dislocation would be expected to give a twist because of lamellar divergence. With dislocations of either sign there would be no continuous sense of twist but in banded spherulites of polyethylene the associated dislocations, acting as branch points, all have the same sign, linked to the sense of chain inclination to the lamellar normal. Moreover this is the same sign which alone is able to develop a terrace at low supercoolings (Bassett *et al.* 1988*a*). Although, in polyethylene, the element of twist introduced at each dislocation is amplified by the adoption of curved, frequently S-shaped, profiles (figure 5), the architecture of its banded spherulites is evidently linked to the formation of arrays of isochiral screw dislocations coupled with the innate tendency for adjacent layers to separate.

A tendency to separate was observed long ago for solution-grown crystals. Inspection of melt-grown crystals *in situ* within a quenched matrix, via permanganic etching (figure 6), has now revealed in the clearest way its presence, involvement and importance in generating polycrystalline textures from a single-crystalline precursor. For growth at low supercoolings multilayer crystals frequently adopt a geometry in which planar lamellae are arrayed at a constant angle of divergence to their next neighbours (figure 7). This points clearly to a short-range repulsive force in the region of their contact point for which the strongest candidate is

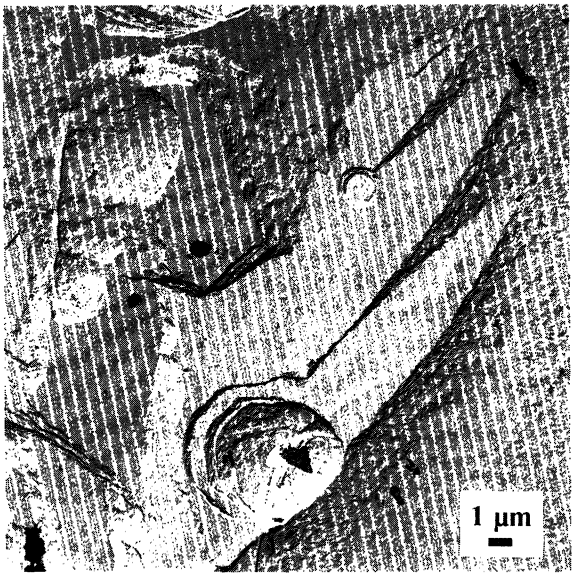

Figure 4. Topological screw dislocations generated in linear polyethylene by a reduction of growth temperature from 130 °C to 128 °C (Patel & Bassett, unpublished work).

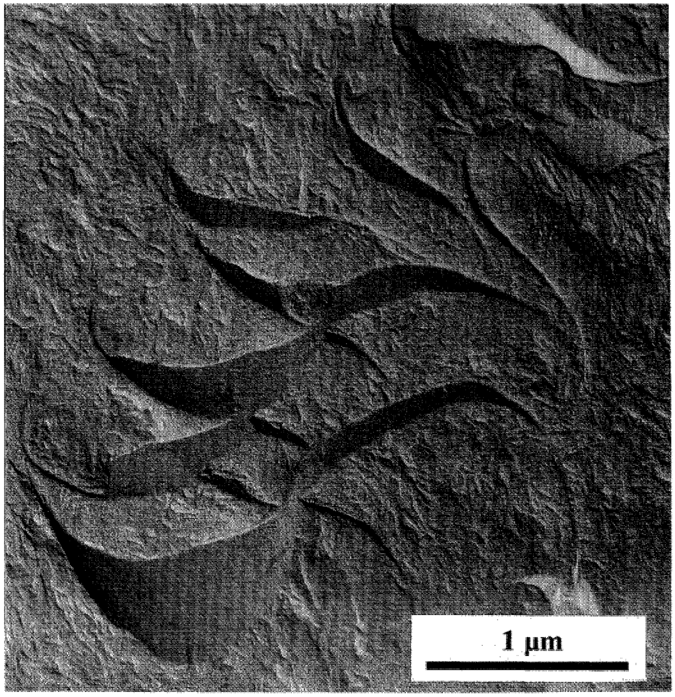

Figure 5. A portion of a continuous lamella of linear low-density polyethylene branching at and diverging from screw dislocations (from Bassett 1989).

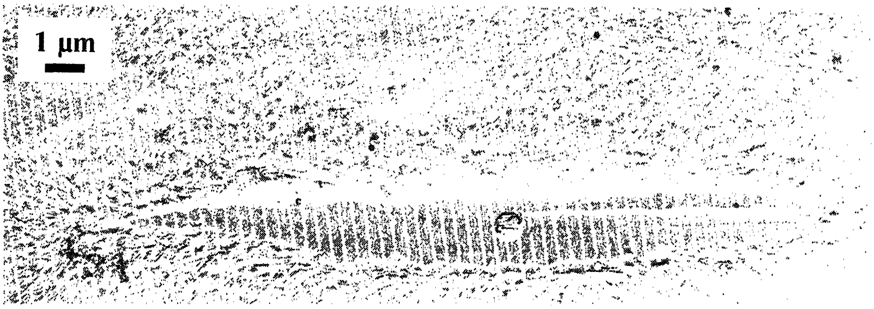

Figure 6. Adjacent layers of a crystal observed within its quenched matrix diverge from the central screw dislocation (Patel & Bassett, unpublished work).

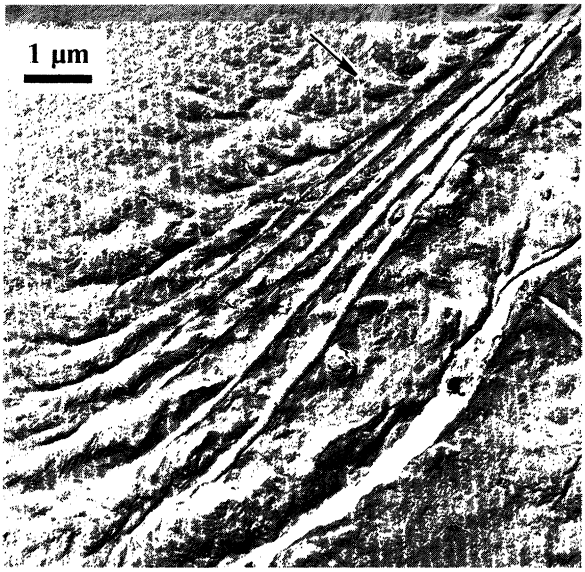

Figure 7. An array of uniformly diverging lamellae in i-poly(4-methylpentene-1) crystallized at 242 °C seen within a quenched matrix. Note the way in which lamellae at the edges (formed on quenching) have bent to become parallel to the material flux (after Patel & Bassett 1994).

the pressure of uncrystallized molecular cilia emerging from the fold surfaces of lamellae.

A discussion of the randomisation of growth directions in space needs also to include effects additional to those cited above as was made explicit in a recent study of melt-crystallized poly(4-methylpentene-1), P4MP (Patel & Bassett 1994). This polymer has a particularly simple square crystal habit but as-grown lamellae are often far from planar. In extreme cases they will bend through a right angle to align their growth direction with that of the flux of material to the growing crystal (figure 7). Such effects and the extent to which geometries are distorted away from those of idealized single-crystalline platelets during spherulitic growth are better studied using linear rather than point nuclei, i.e. in so-called row structures.

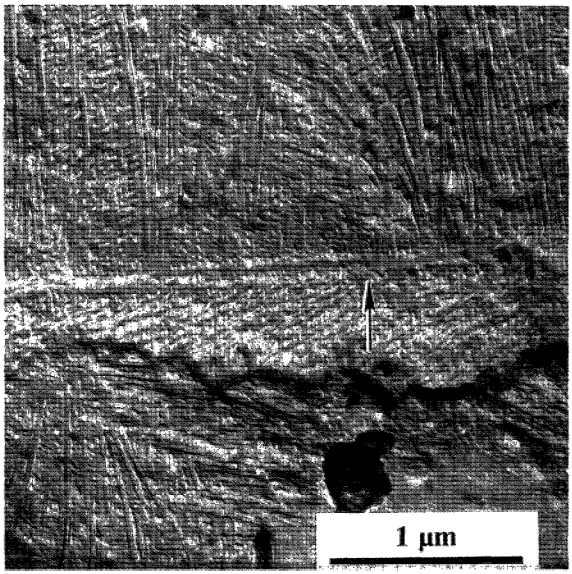

Figure 8. A row structure of isotactic polypropylene grown at 140 °C. Lamellae have nucleated on the central core (arrowed), but have grown separated from their neighbours, either at right angles to the core or curving to one side when that neighbour is missing (White & Bassett, unpublished work).

(c) *Row structures*

Row structures are formed typically under conditions of extensional molecular strain which create linear nuclei parallel to $\boldsymbol{c}$ the chain axis. Subsequent growth shares the same $\boldsymbol{c}$ axis and, occurring with little or no strain, is lamellar. The initial development is as a shish-kebab structure but larger objects reveal the details of crystal growth more clearly than does spherulitic growth which is, essentially, from a point nucleus. This is because of the common initial orientation and reference plane with which one may assess more readily how growth has developed in three dimensions.

In longitudinal view lamellae can be seen to be parallel but regularly spaced apart as are the teeth of a comb (figure 8). However, when there is a gap in the 'teeth' then lamellae will curve into it (figure 8). Both phenomena, the spacing and the curving, are additional evidence for the presence and pressure of uncrystallized molecular cilia at lamellar basal surfaces. It is, however, in transverse view that we gain the opportunity to inspect if and how the system fills space when this cannot be done by maintaining the initial lamellar and crystallographic orientations.

The monoclinic or α form of polypropylene grows as thin primary laths along $\boldsymbol{a}^*$ ie normal to the chain axis $\boldsymbol{c}$ (Lovinger 1983); in addition it develops daughter twinned or cross-hatching lamellae related by a common $\boldsymbol{b}$ axis (Khoury 1966). If growth from a row nucleus is limited to primary laths sharing the $\boldsymbol{c}$ axis of the nucleus and cross-hatching therefrom, then lateral gaps will appear between the outward growing laths. The lathlike habit does not allow these gaps to be filled while maintaining the initial crystallography. One possible means which would maintain lamellar continuity, would be to enter this lateral space either

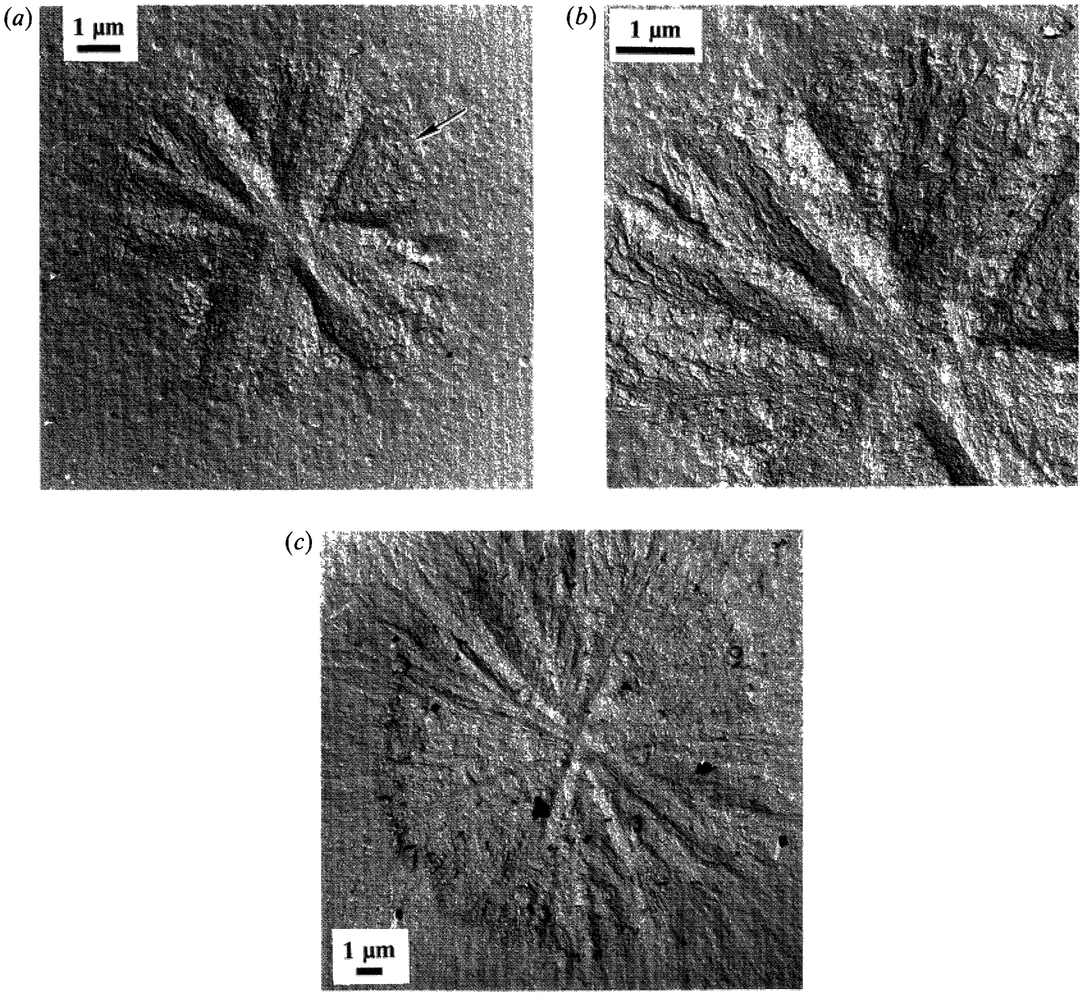

Figure 9. Etched transverse cut surfaces of row structures in isotactic polypropylenes grown at 140 °C (*a*) and detail (*b*); and at 130 °C (*c*). The significant feature is the distinction between those lathlike lamellae which have grown from the central core and neighbouring regions or segments (arrowed) containing lamellae in different orientations. After etching, the former stand proud of the latter (White & Bassett, unpublished work).

by cross-hatching from, or by curving and splaying from, primary lamellae which have twisted around their long axes; such twisting has been observed previously in this polymer (Olley & Bassett 1989). However, in figure 7 one observes a different response namely changes of lamellar orientation bounding filled gaps showing that, at least under these conditions, the system has filled space by nucleating and growing lamellae in appropriate directions but which do not share the ***c*** direction of the initial row.

Inspection of row structures thus shows simply and unequivocally that, at least at moderate supercoolings, spherulitic growth will fill space even if lamellar and crystallographic continuity cannot be maintained. Proximate regions of melt may crystallize using nuclei with large misorientations to the initial radial growth. A corollary is that the ensuing laths cannot have sprung from branch points which were giant screw dislocations. Had they done so lamellar and crystallographic continuity would have been maintained. There must, therefore, be branching mechanisms in spherulitic growth additional to giant screw dislocations.

(*d*) *Intraspherulitic properties*

The divergence of individual dominant lamellae, which imposes two-or-more component textures on polymeric spherulites brings a corresponding spatial variation in properties: chemical, mechanical and thermal. Two principal factors influence the associated lamellar textures. The first is the relative time dependence of formation which invites fractional crystallization between first-forming dominants and later-forming subsidiaries and other infilling growth. This can be both demonstrated and measured if crystallization is interrupted by quenching at an appropriate stage. Then not only can the location of later crystallization be identified by a reduced lamellar thickness but it may well also be possible selectively to dissolve the lower-melting, later-crystallizing component and to measure its molecular characteristics (Mehta & Wunderlich 1975). Data for linear and branched polyethylenes, i-polystyrene and i-P4MP have all shown a tendency for shorter and/or more-branched molecules to crystallize later. For example, in a typical linear polyethylene crystallizing at 128 °C, there is roughly a factor of two between the molecular masses of crystallized and uncrystallized portions at 50% completion, i.e. 120 000 to 60 000 respectively.

The second principal textural factor is that whereas dominant lamellae crystallize freely into the melt, later-forming ones have to do so in increasingly constrained conditions. It has been suggested that this is the origin of systematic spatial variations in melting point even in the absence of fractional crystallization (Bassett *et al.* 1988*b*).

Recent research has highlighted the ways in which properties within spherulites are modulated by the associated lamellar texture, i.e. the separation between adjacent dominant lamellae or interdominant spacing. Early studies using permanganic etching showed differences in relief, with dominant lamellae standing proud. It was subsequently shown that this derived from a differential response to cutting during sample preparation, i.e. from a mechanical rather than an inherently chemical response (Freedman *et al.* 1986). Nevertheless that there is a difference in resistance to etching has been shown explicitly in branched polyethylenes for which, unlike the linear polymers, thinner lamellae are etched faster (Freedman *et al.* 1988) and in other systems such as i-polypropylene and i-P4MP in which dominant lamellae stand proud in specimens grown as films and etched without cutting. A differential mechanical response between dominant and other lamellae has been demonstrated not only as above as an inadvertent consequence of cutting during specimen preparation but also directly during tensile deformation. The matrix of subordinate lamellae in banded spherulites of polyethylene is deformed to lower-melting material before surrounding dominants (Bassett & Freedman 1993).

Subsidiary lamellae also melt before dominant lamellae even in isothermally crystallized polymer because, it is suggested, they are thinner due to isothermal thickening being restricted during growth. Accordingly, when a spherulitic specimen is heated into the temperature range where lamellae begin to thicken (by local melting and recrystallization) it is the subsidiary lamellae which melt and recrystallize first (Bassett & Patel 1994).

In evaluating structure–property relations in polymers the above factors make it clear that, in addition to the more-often-considered interspherulitic boundaries, intraspherulitic texture is an important influence which will contribute to the broad range of properties usually encountered in crystalline polymers.

(*e*) *Melt-crystallized lamellae*

When it became possible systematically to examine the interior morphology of melt-crystallized polymers, first of all polyethylene, it was discovered that lamellae were not simply platelets but had different profiles according to their growth conditions. Thus linear polyethylene viewed down the growth direction $\boldsymbol{b}$ could show ridged facets in which chains were inclined at *ca.* 35° to lamellar normals i.e. approximately $\{201\}$, a single $\{201\}$ plane or, totally unexpectedly, curved profiles, typically S- or C-shaped (Bassett & Hodge 1981). These last are typical of dominant lamellae under most growth conditions and of the branched polymer. None of these profiles is, however, evident in fracture surfaces because of the strong preference for fracture to occur in $\{200\}$ planes. This is the principal bias in viewing morphology via fracture surfaces but, in addition, thinner lamellae may be less well resolved because of their greater tendency to ductility.

Subsequent studies in which the earliest stages of growth were interrupted by quenching have led to the observation and study of individual crystals. Of major importance was the finding (figure 6) that as-grown crystals, still within their matrix had adjacent layers diverging (Bassett *et al.* 1988*a*). Highly significant also (and in agreement with earlier observations in thin films, Labaig 1978) were the curved outlines in plan – elliptic in the botanical sense – of lamellae grown at the highest temperatures, 130 °C. As curved surfaces are necessarily molecularly rough, nucleation on them during crystal growth cannot be in Regime I, unlike the planar growth faces familiar from many solution-grown systems. The consequences of curved growth faces for kinetic theories of polymeric growth continue to be a very active topic (Sadler 1987; Hoffman & Miller 1989; Point & Villers 1992).

Individual polyethylene crystals have been extracted selectively by dissolution of their surrounding matrix, then examined by TEM using methods developed initially for solution-grown lamellae (Bassett *et al.* 1988 *a*). Diffraction patterns from lamellae tilted around their $\boldsymbol{b}$ axes confirmed the average chain tilt to be *ca.* 35° to the lamellar normal but diffraction microscopy showed only patchy contrast in nominally planar lamellae (figure 10). On the other hand, ridged crystals did show uniform diffraction contrast for each ridge side. However, while the latter crystals were stiffened by the ridging and did not collapse during extraction, this was not the case for the 'planar' lamellae. During specimen preparation these developed cracks, which were not present beforehand, indicative of non-planarity in as-grown lamellae, which reconstruction (by closing all cracks) supported. The desirability of examining crystals before collapse was, therefore, reinforced.

A recent achievement, in a further development of etching technique, is to control the thinning of specimens. In this way the matrix can be removed to the stage where a chosen crystal breaks the surface, at which point the emergent

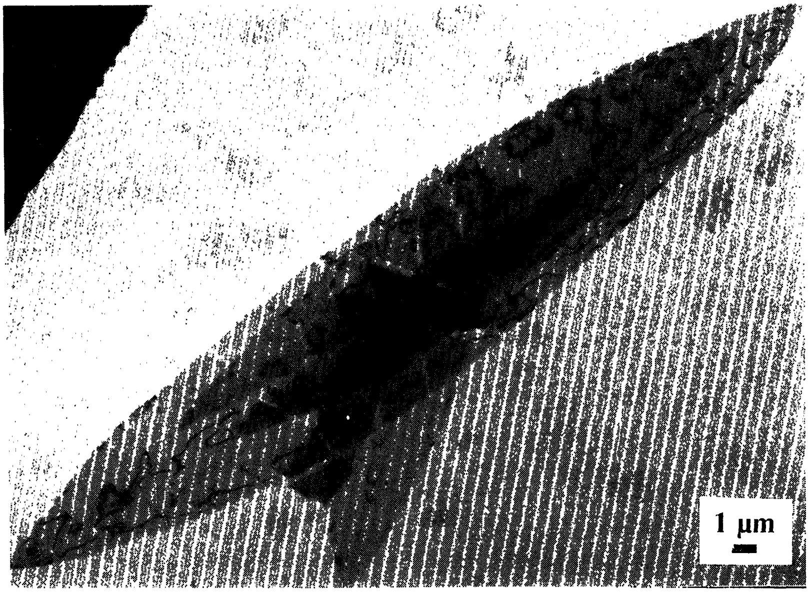

Figure 10. Diffraction contrast in a polyethylene lamella grown at 130 °C then extracted from a quenched matrix (Patel & Bassett, unpublished work).

crystal can be metal shadowed to emphasize relief but also can be studied in transmission by diffraction TEM.

Figure 11 shows an application of this technique to the study of S-profiled lamellae in polyethylene. The movement of the Bragg diffraction fringe as the sample is rotated about its long (***b***) axis reveals a twist around this axis of order $5^\circ\,\mu\mathrm{m}^{-1}$. Moreover, the propensity to generate screw dislocations by shear in $\{200\}$ – the planes of preferred fracture – arises naturally: not only is the stress relieved but the dislocations generated are isochiral with the same sign as that self-selected at high growth temperatures and which occurs in banded spherulites (Bassett *et al.* 1988*a*).

In figure 11 one is, therefore, seeing the initiation of processes whose iteration would lead to a banded spherulite of polyethylene. The strains giving the non-planarity in this crystal are greatest in its centre, i.e. the oldest part. They are, very likely, due to a movement of the local lamellar normal away from the ***c*** axis with time because of developing fold interactions. This is a theme which is reinforced by other observations. For example, crystallization at 130.5 and 131 °C, which requires very long times, tends to give bisectored and singly ridged elliptic crystals (Patel & Bassett, unpublished work). This habit occurs only sparsely at 130 °C, when elliptic crystals are mostly nominally planar but locally non-uniform according to figure 10, while ridging is the norm for lower mass material, crystallizing at a lamellar thickness where there are about two folds and three stems per molecule (Cotton 1987) and, accordingly, less hindrance to molecular mobility.

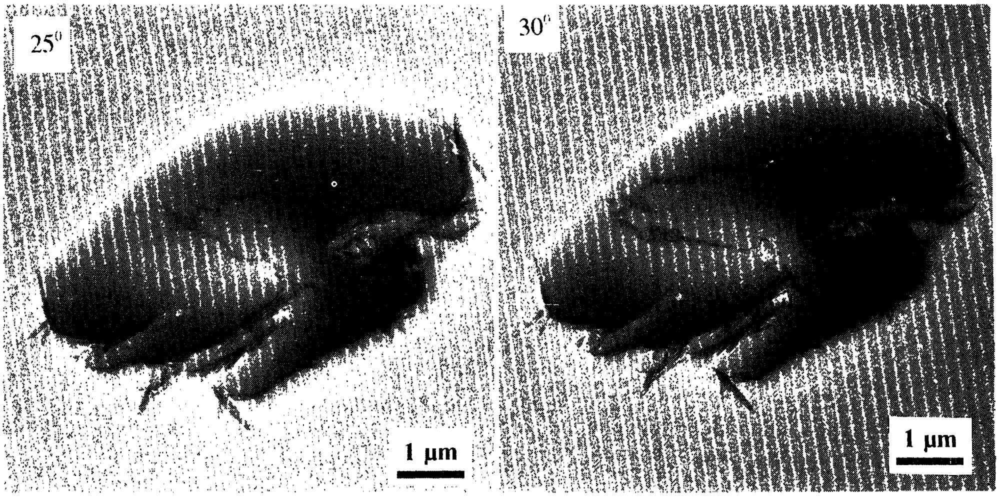

Figure 11. A lamella at the surface of a thinned film of linear low-density polyethylene grown at 123 °C, metal shadowed then viewed in transmission in the electron microscope. Note the movement of the Bragg fringe for different angles of orientation about the long (***b***) axis (Patel & Bassett, unpublished work).

3. Résumé

The systematic examination of the morphology of melt-crystallized polymers in real space continues to enlighten on important features of these complex systems which are not readily accessible in other ways. Knowledge and understanding of the architecture, genesis and related properties of spherulites, including those which are banded, have been advanced significantly. The location of particular molecular species, because of fractionation during crystallization, may be measured. The study of individual crystals grown in bulk has been made possible and used to reveal the innate tendency of adjacent lamellae to diverge and the fundamental importance of this phenomenon. Study of the prevalent S-profiled lamellae of polyethylene has highlighted the importance of molecular adjustments during growth due to developing fold interactions.

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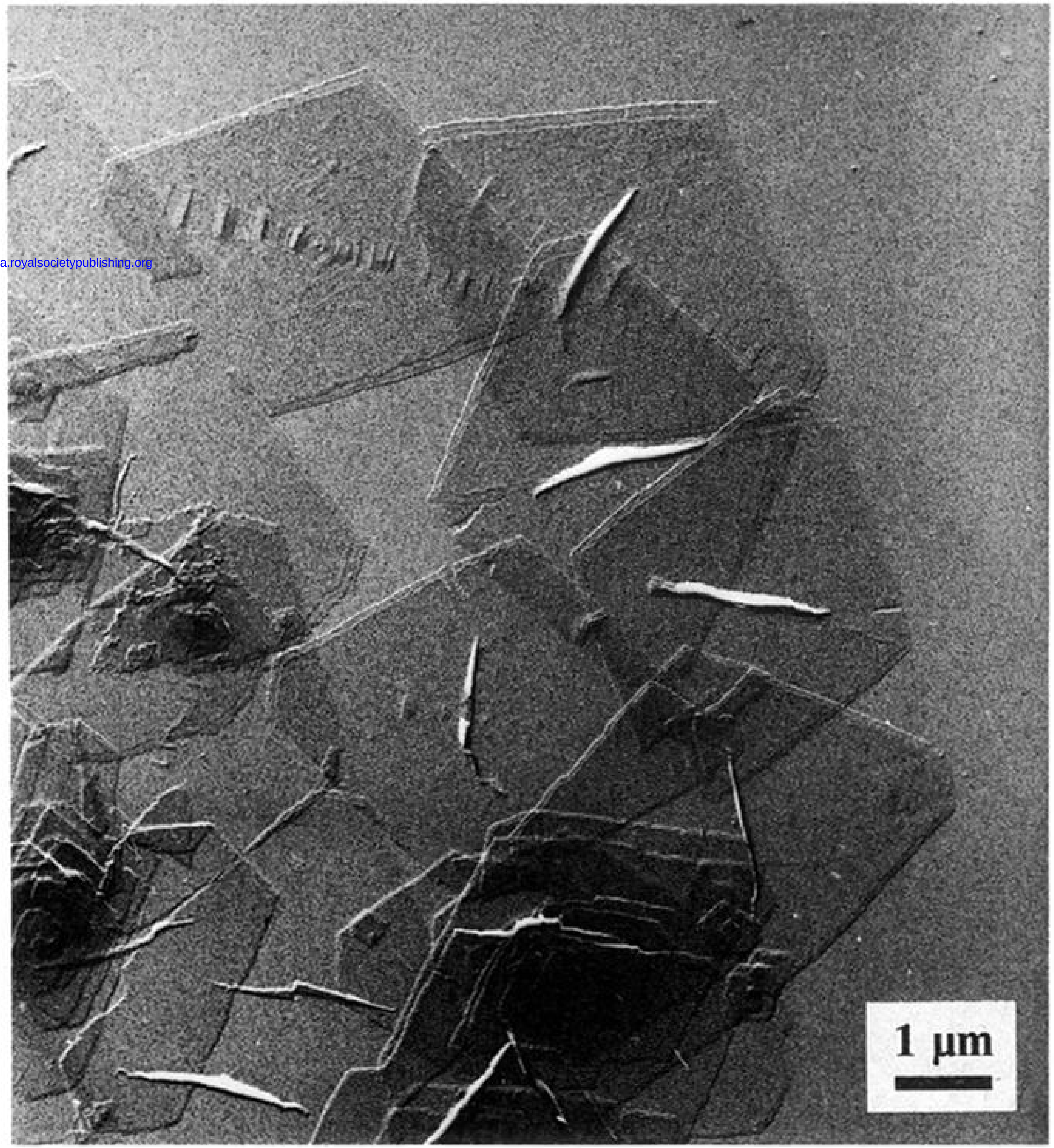
Discussion

F. C. Frank (*University of Bristol, U.K.*). Some of your pictures suggest that they are near to an understanding of the origin of twisted spherulites. Bernauer (*Gedrilte Kristalle* (1920)) shows that all sorts of different crystaline materials, including sodium chloride, can give twisted spherulites. He pursues, and in turn rejects, all sorts of suggested causations. We need a general explanation.

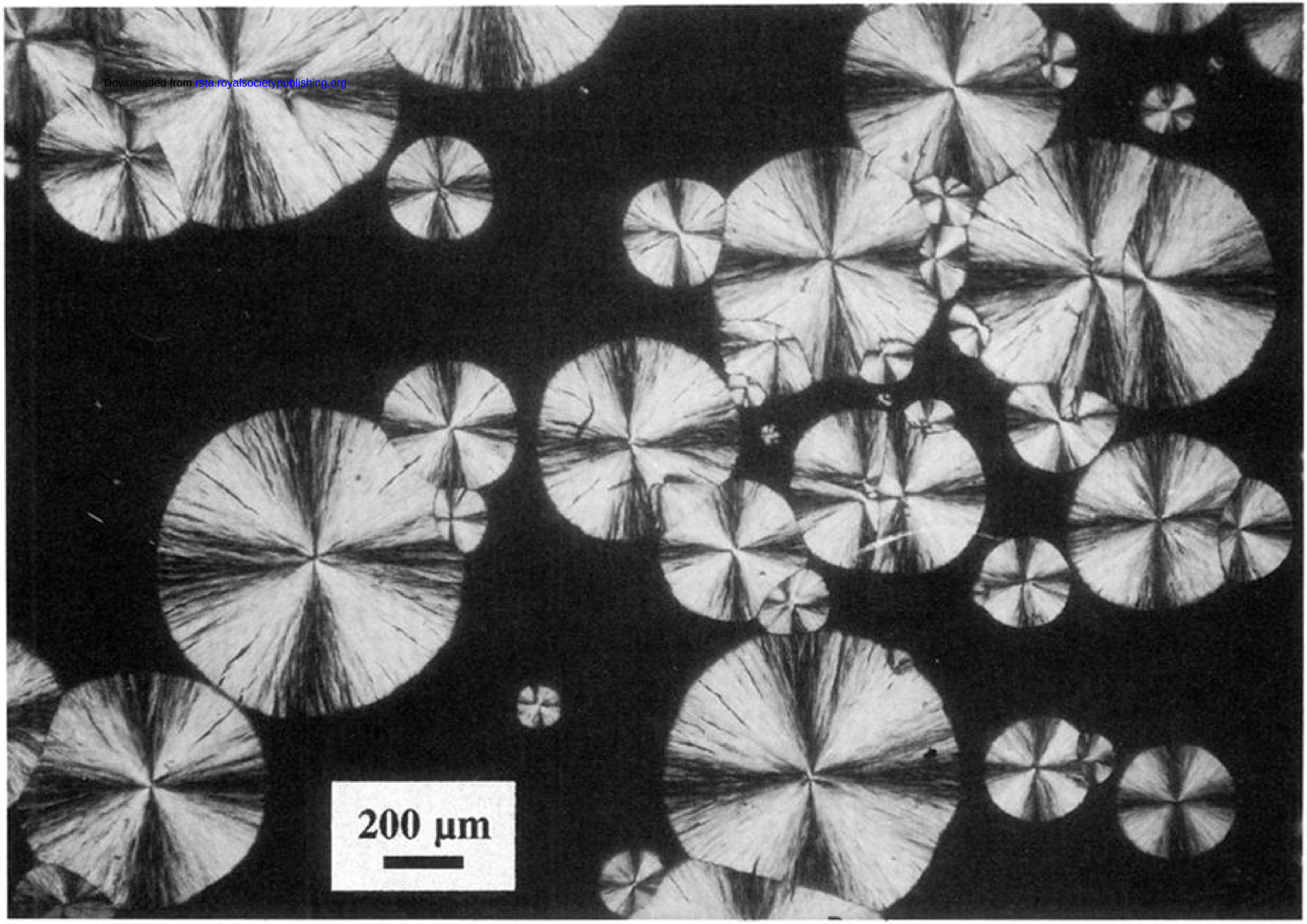
I wrote recently on the breaking of chiral symmetry in zinc sulphide whiskers tht develop an increasingly giant twist as they grow (zinc sulphide has no intrinsic spherality). The same effect as that discovered by Eshelby, that the interaction of a screw dislocation parallel to the axis of cylinder with a twist of the cylinder that it induces causes the screw dislocations to be attracted onto the axis, and also has the effect that when one or more screw dislocations of one hand are present and another pair, right-handed and the other left-handed, are added the alien screw is ejected and that of the same hand as the indigenous population retained. I wonder whether a similar effect can operate in a sphere: although I can solve the problem in a cylinder, I don't know how to deal with it in the spheres.

D. C. Bassett. I am very conscious of the need to look for general as opposed to particular explanations for spherulitic growth. So far as polymers are concerned, we have looked at many systems and all show a commonality of behaviour. In terms of banding polyethylene is perhaps the only material for which we have the relevant detailed microstructural knowledge. What are we finding (figure 9) is a way of generating isochiral dislocaitons successively with the chirality linked to the sense of chain tilt to the lamellar normal. In addition, very recent results obtained by D. Trifonova on banded growth in polypropylene also show increments of twist in association with isochiral giant screw dislocations though in this case chains are normal to lamellae. It is my belief that banding in general may well be generated by isochiral screw dislocations and the twist associated with them but that isochirality could arise in different ways in different materials to produce the same overall effect.

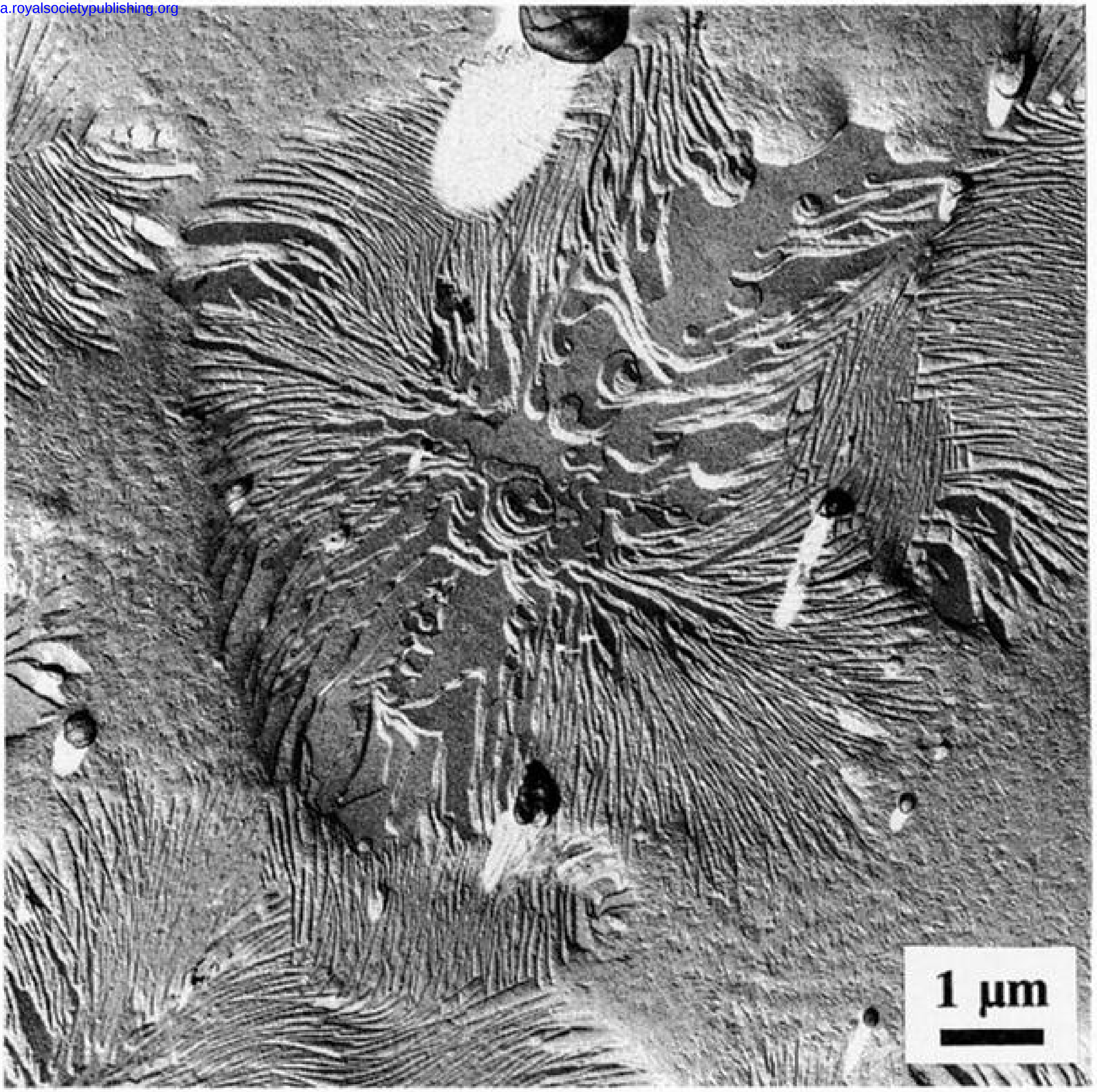
A. Keller (*University of Bristol, U.K.*). In a recent paper by A. Toda & A. Keller (*Colloid Polymer Sci.* **271**, 328–342 (1993)) we extracted and studied lamellar crystals of polyethylene as grown from the melt. Of specific relevance is the finding that there are two categories of S shaped crystal. In addition to the one featuring in your work association with banded spherulites, there is a second type that corresponds to the cross-sectional view of chair type crystals. Such chair crystals are the melt grown counterparts of those originally identified in solution crystallized products (D. C. Bassett *et al.*, *Phil. Mag.* **8**, 1753–1783 (1963)). Their significance lies in the fact that by the interpretation adopted they arise through the regularization of the fold surface subsequent to the growth of the crystal. Accordingly, when grown initially the folds will be disordered, but will regularize subsequently giving rise to well-defined sector structures with specific basal plane (the fold surfaces) obliquities. If there is no correlation between this regularization in the diametrally opposed sectors, then the chair configuation has a chance to arise. This then accounts not only for certain shapes observed, but also throws light on aspects of crystallization mechanism and is pertinent to some controversial issues surrounding this subject. It is significant that this feature, so far confined to solution crystallization, has now emerged also in crystallizations from the melt and this, as the above quoted work shows, in a pronounced and representative manner.



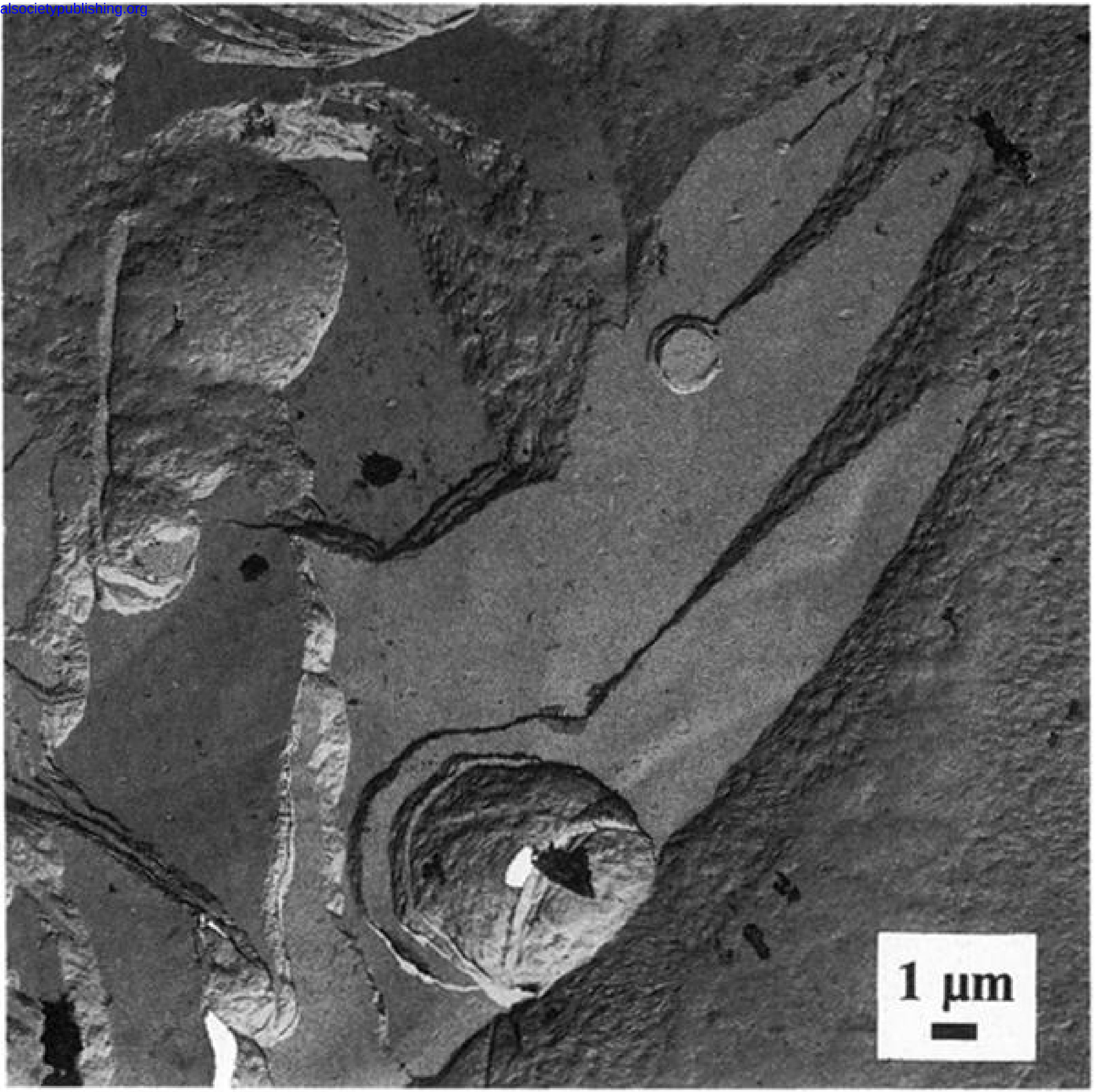
gure 1. Crystals of linear polyethylene grown from dilute solution in xylene. The double step their edges shows the crystal thickness is controlled by secondary nucleation while the internal orphology is evidence of initial non-planar habits due to fold interactions.



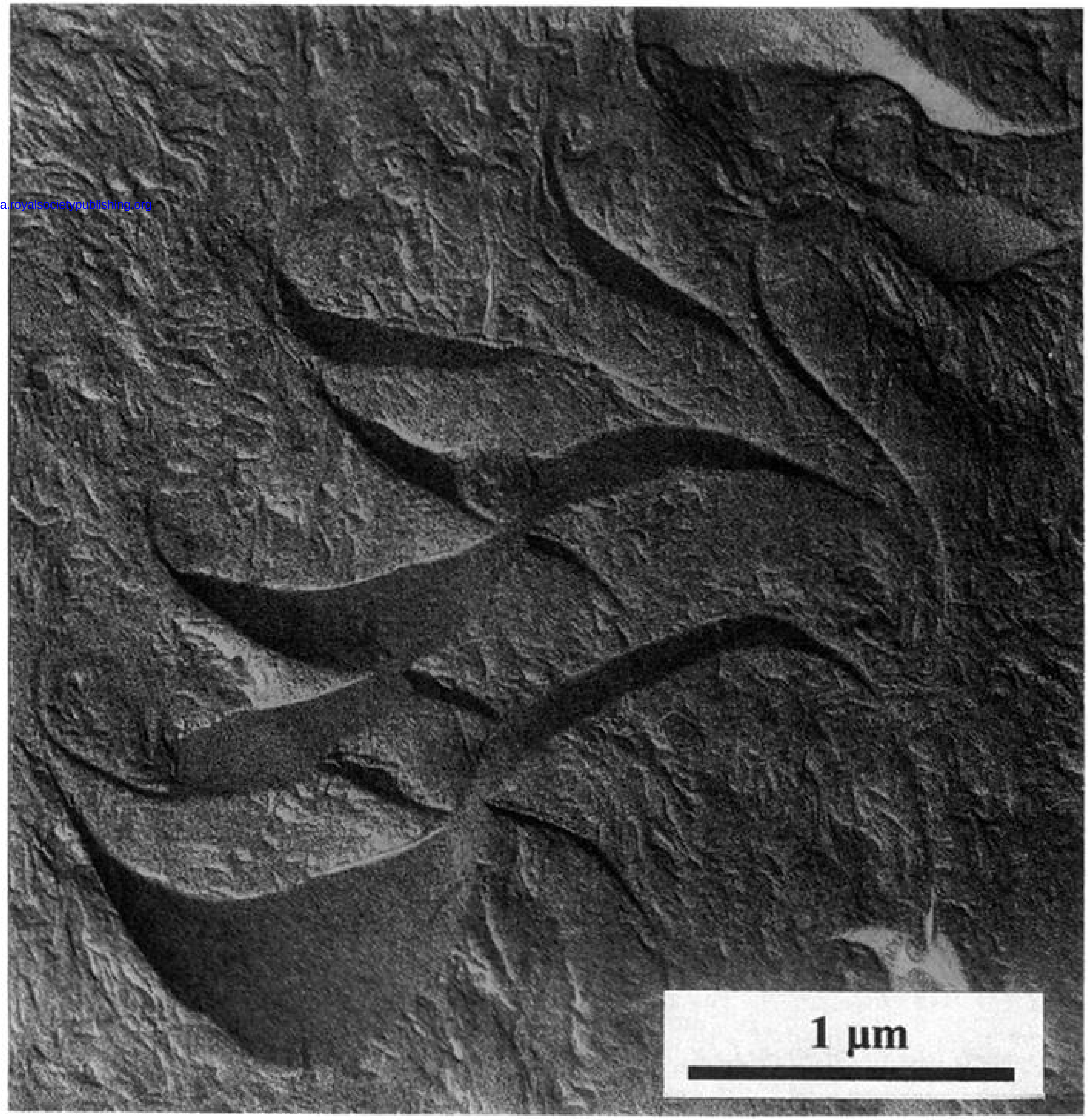
igure 2. Spherulites of the α form of polypropylene viewed between crossed polars while crystallizing from the melt at 130 °C.



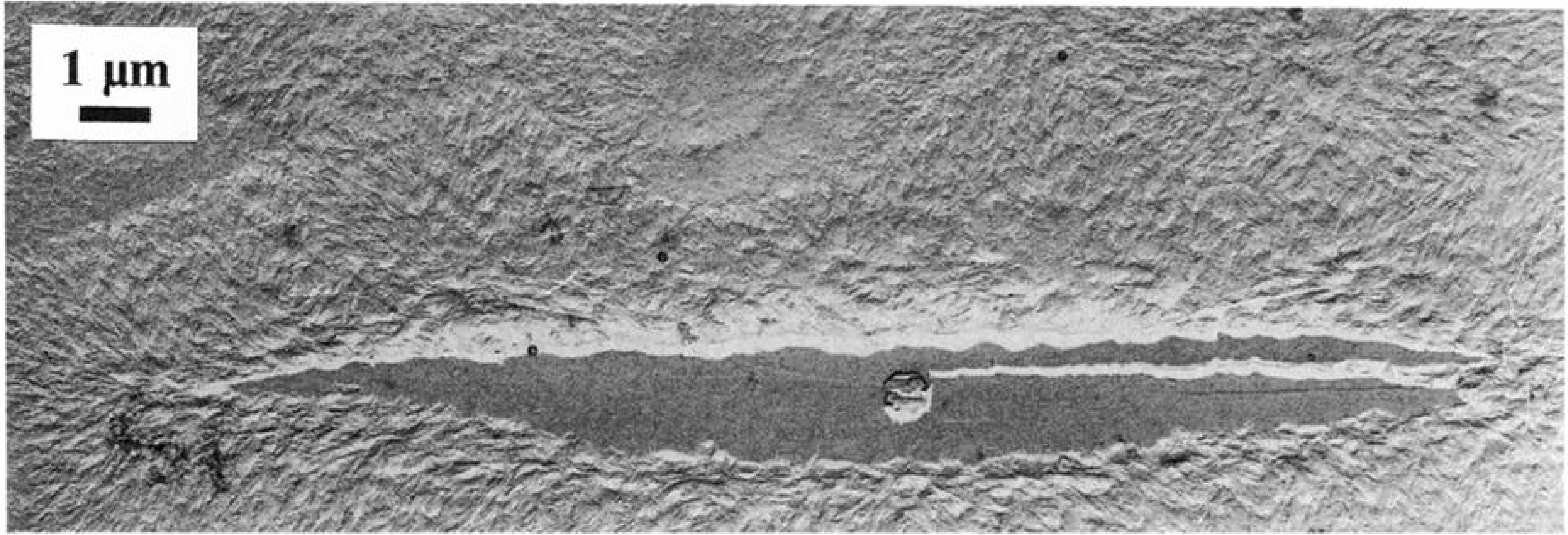
igure 3. Embryonic spherulites of linear low-density polyethylene grown at 124 °C (from Bassett 1993).



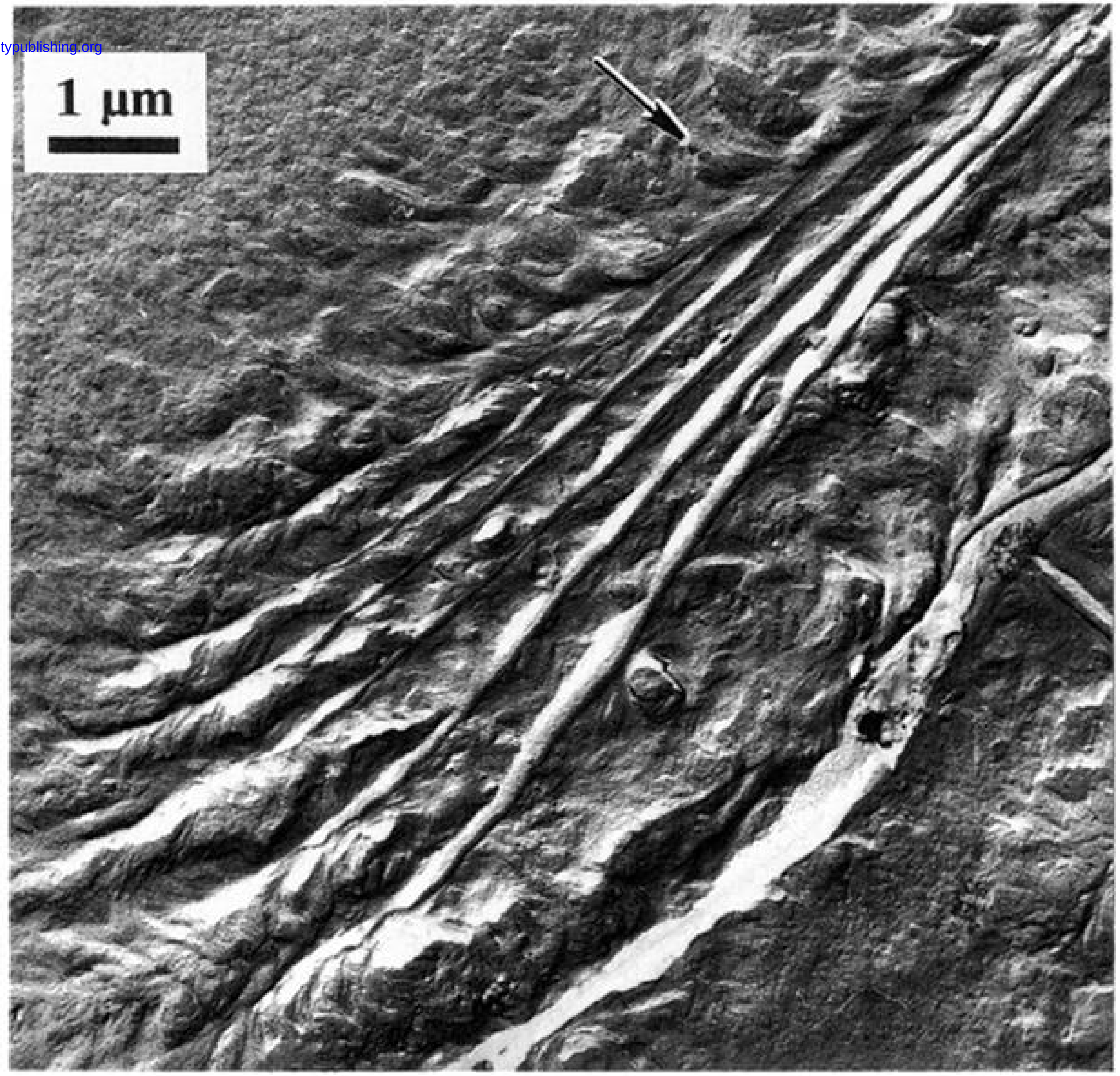
igure 4. Topological screw dislocations generated in linear polyethylene by a reduction of growth temperature from 130 °C to 128 °C (Patel & Bassett, unpublished work).



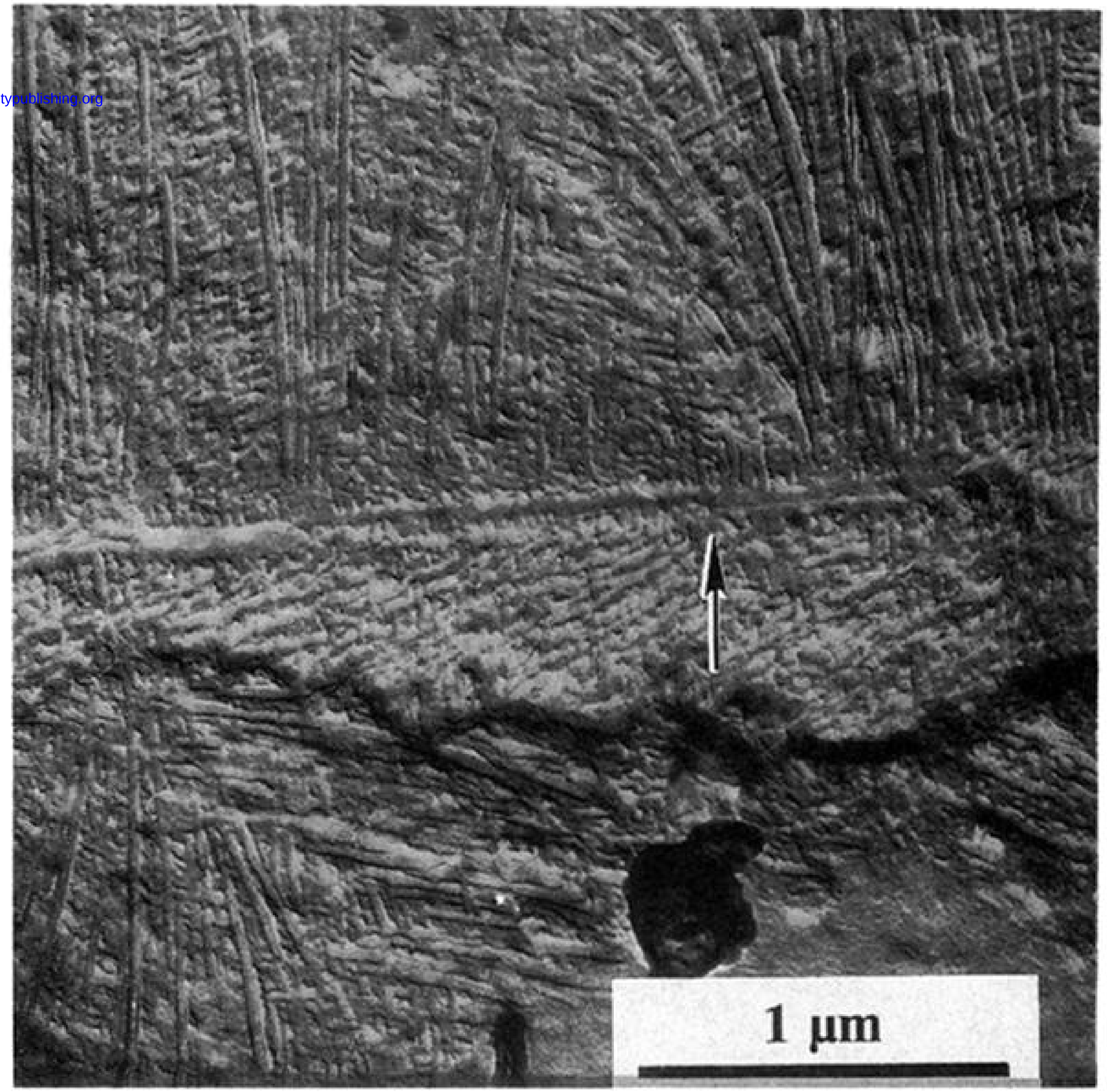
igure 5. A portion of a continuous lamella of linear low-density polyethylene branching at and diverging from screw dislocations (from Bassett 1989).



igure 6. Adjacent layers of a crystal observed within its quenched matrix diverge from the central screw dislocation (Patel & Bassett, unpublished work).



igure 7. An array of uniformly diverging lamellae in i-poly(4-methylpentene-1) crystallized at 42 °C seen within a quenched matrix. Note the way in which lamellae at the edges (formed on uenching) have bent to become parallel to the material flux (after Patel & Bassett 1994).



igure 8. A row structure of isotactic polypropylene grown at 140 °C. Lamellae have nucleated n the central core (arrowed), but have grown separated from their neighbours, either at right ngles to the core or curving to one side when that neighbour is missing (White & Bassett, npublished work).

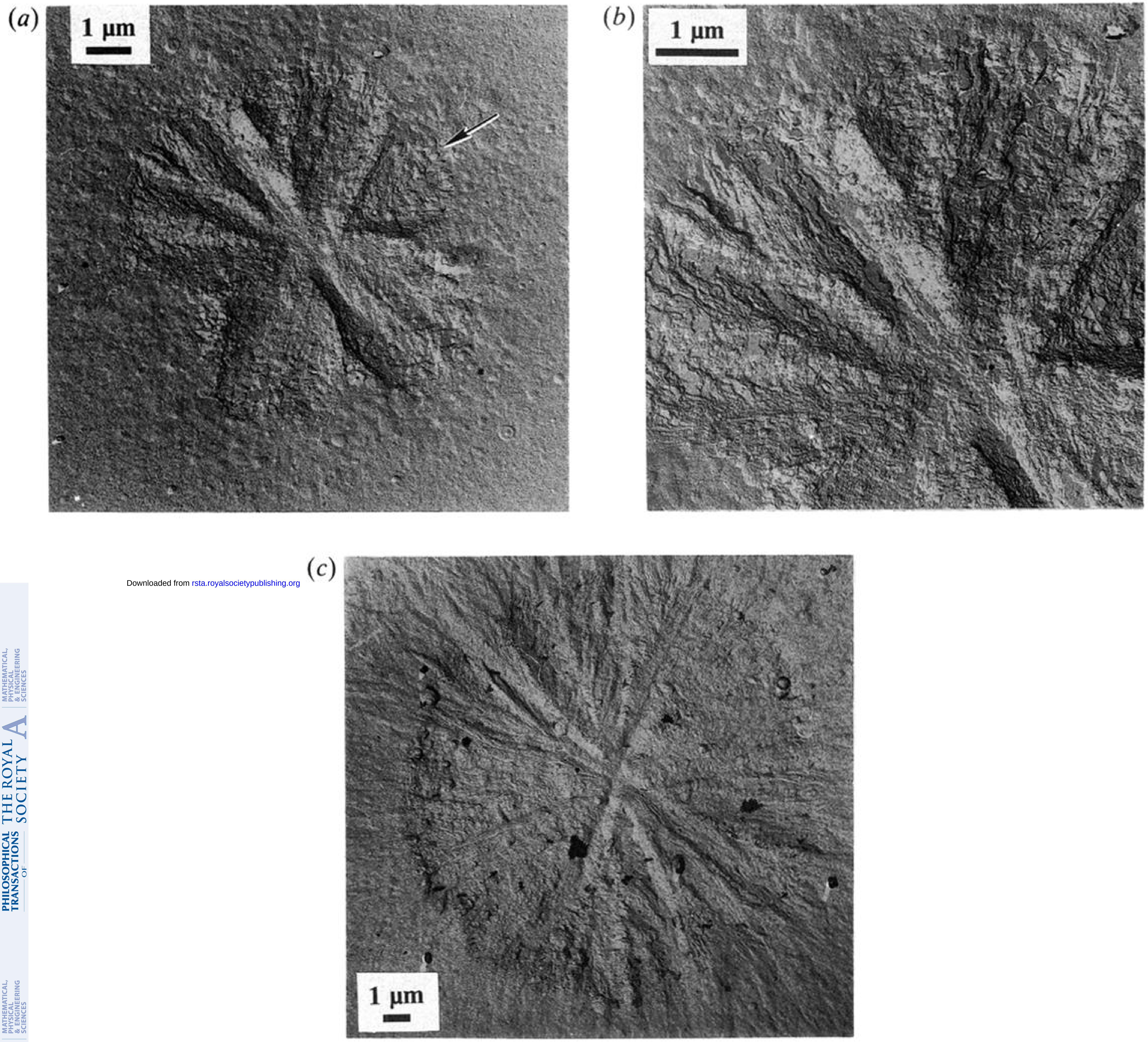
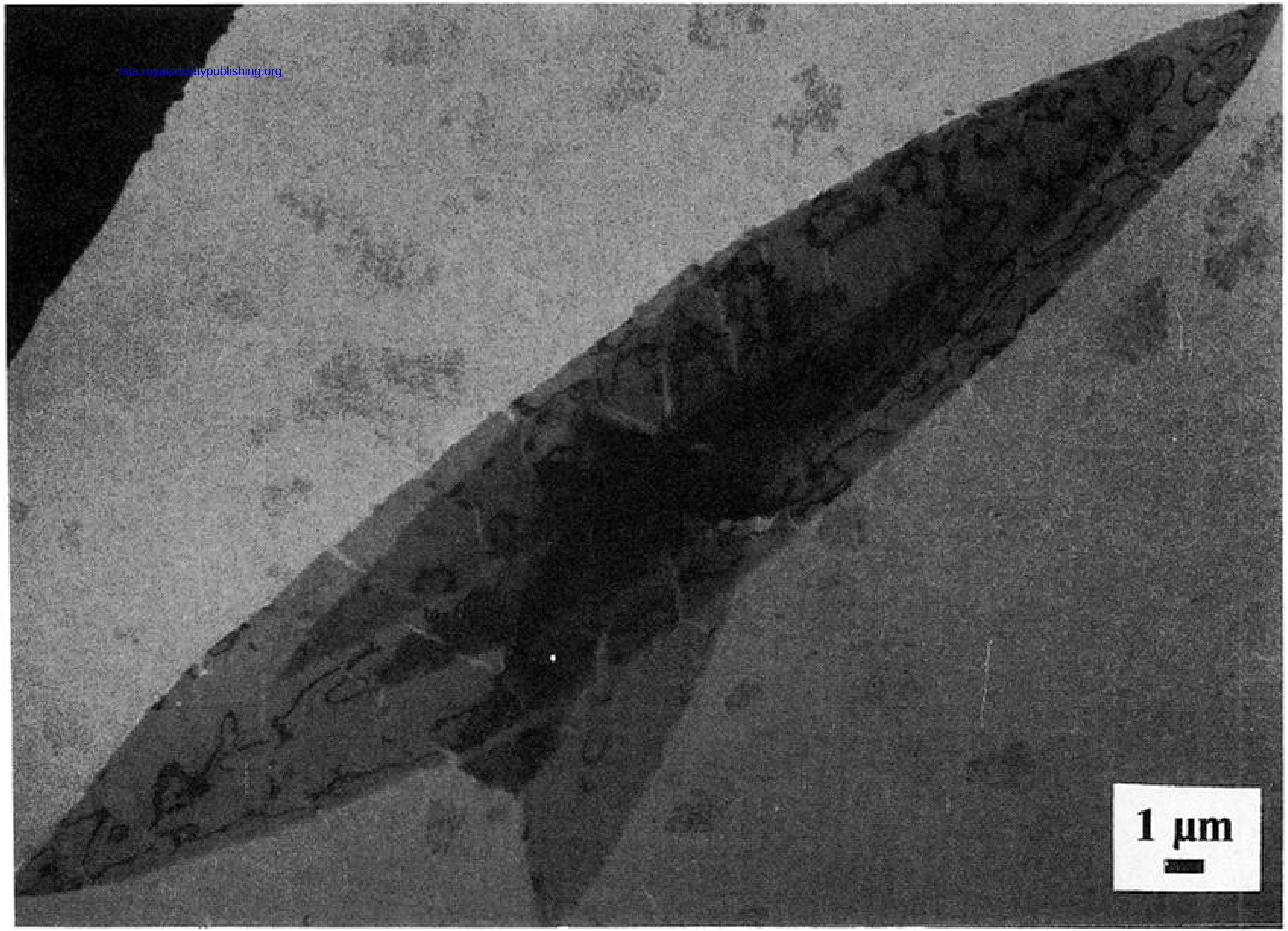


Figure 9. Etched transverse cut surfaces of row structures in isotactic polypropylenes grown at 140 °C (a) and detail (b); and at 130 °C (c). The significant feature is the distinction between those lathlike lamellae which have grown from the central core and neighbouring regions or segments (arrowed) containing lamellae in different orientations. After etching, the former stand proud of the latter (White & Bassett, unpublished work).



igure 10. Diffraction contrast in a polyethylene lamella grown at 130 °C then extracted from a quenched matrix (Patel & Bassett, unpublished work).

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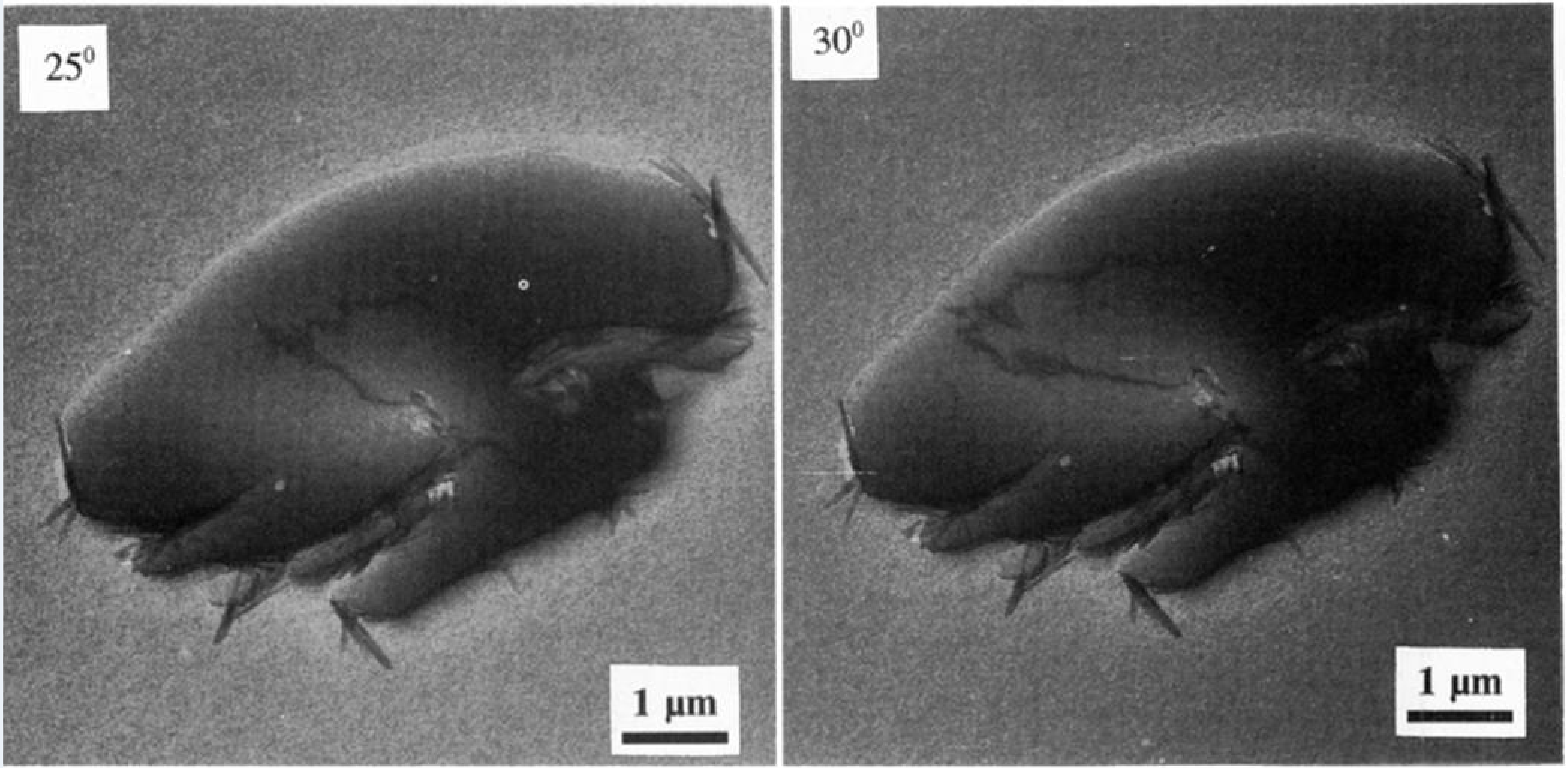

igure 11. A lamella at the surface of a thinned film of linear low-density polyethylene grown t 123 °C, metal shadowed then viewed in transmission in the electron microscope. Note the novement of the Bragg fringe for different angles of orientation about the long (***b***) axis (Patel Bassett, unpublished work).