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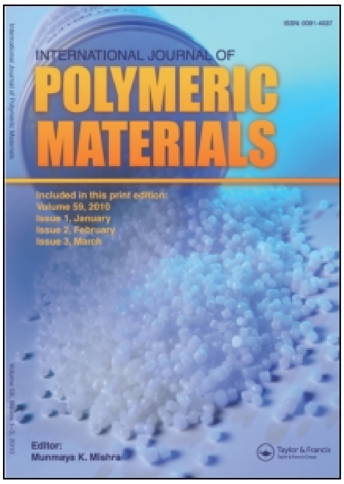

International Journal of Polymeric Materials

Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713647664

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To cite this Article Pietralla, M.(1993) 'Twinning as a Fundamental Process in Polymer Crystal Orientation', International Journal of Polymeric Materials, 22: 1, 245 — 256

To link to this Article: DOI: 10.1080/00914039308012080

URL: http://dx.doi.org/10.1080/00914039308012080

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Intern. J. Polymeric Mater., 1993, Vol. 22, pp. 245–256
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Twinning as a Fundamental Process in Polymer Crystal Orientation

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During a deformation experiment the chain molecules are orientated. The mechanisms of chains orientating producing the crystal orientation differ from orientating of chains within the amorphous phase. Some of them may be active. Twinning processes changing the c-axis direction within the crystallites are possible. They rotate the crystal lattice and change the shape of crystals. This explains easily numerous observed properties previously described in literature. Twinning modes of the orthorhombic and the monoclinic crystal modification of polyethylene, which are believed to be involved, are discussed. Twinning processes observed macroscopically by deforming single crystals of poly(diacetylene), will be of universal importance during the deformation of semicrystalline polymers.

KEY WORDS Twinning, deformation, chain orientation, yield stress, polyethylene.

INTRODUCTION

The deformation mechanisms in semicrystalline polymers have been extensively studied.[1,2] Numerous models have been proposed to explain the observed orientations, however, their physical significance has not always been thoroughly discussed. Thus the general features are well understood but important details are still lacking. A plastic deformation is performed by shear processes. This is also valid for polymer crystals. It is difficult to explain the orientation of crystal lamellae whose chains are initially oriented perpendicular to the stress in a stretching experiment or parallel to the stress in compression experiments. A "rotation of crystals", as was established from the X-ray analysis, is hardly possible. The result and the process should not be mixed up! Crystals in the above mentioned orientation do not have any imposed torque, since shear stresses (in the coplanar case, see later) are zero in these cases. The observation of kink bands[3–6] suggests mechanical twinning as one of the underlying physical processes. During this process the chains are tilted by a stepwise rotation of segments within the crystals into a new crystallographic position ("c-twinning"). This was first proposed[7] in 1976 and then experimentally observed in polydiacetylene single crystals[8] in 1978. Detailed studies of PE deformation[2] still being explained by means of the well established (hk0)-twinning (see below) despite impressive observations pointing to c-twinning. Since no further investigations have been undertaken in this direction the matter was buried and considered as an exercise in a polymer textbook.[9] Much interest has been recently displayed in the processes taking place within crystals. We hope that molecular modelling tools will clear this topic in the near future.

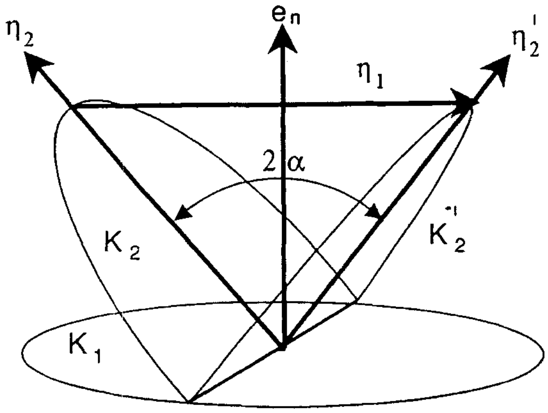

FIGURE 1 The twin elements. Planes K_2 ("undistorted planes") are turned to planes K_2' by a glide in direction η_1, determined by the simple shear s. The angle 2α of K_2 with K_2' is the twin angle.

TWINNING

A twinned crystal consists of both a lattice and its mirror image. It is formally produced by shearing planes parallel to K_1 into direction η_1. K_1 is the mirror plane between parent crystal and twin. This shear turns the planes K_2 by an angle 2α to the planes K_2' (see Figure 1). This type is a reflection twin and the process outlined produces a deformation twin[10] in contrast to widely known growth twins. In cases of restricted shear possibilities mechanical twinning is known to enable plastic deformation. Many of them are known to appear during the deformation of metals and of inorganic crystals.[10,11,16]

In polymers mechanical twins ((hk0)-twins) have been unambiguously observed when the PE bulk[12] and PE single crystals[13,14] are deformed. Their mirror plane, which is called the twinning plane K_1, contains the polymer chain which is thus left undistorted. They are listed in Table I. The process outlined above prevents any plane intersecting the molecules from becoming a twinning plane, otherwise the chains must be scissored by the shear. The important idea is that kinked molecules can orientate stepwise if the kinking proceeds along the chain (Figure 2). If the molecules are arranged within a crystal in such a way that their kinks form a plane, named a kink block, there exist the seed of a twinned crystal. A collective stepwise extension of the kink block, will not remain stable but end up after some reshuffling in a twinned crystal. This process has been called c-twinning. The energy consideration leads to

$$F_{\text{twin}} + \sigma_{\text{twin}} \leq F_{\text{kink}} + \sigma_{\text{kink}}, \tag{1}$$

where F is the free energy of the bulk phase, and σ is the surface energy.

PROPERTIES OF TWINNING

Mechanical twinning is a process which proceeds by the migration of a twin boundary through the crystallite. The remaining parts of the crystallite shear mutually (see Figure 2). This minimizes the defect energy compared e.g. to the rotation of a whole crystal. A point lattice only needs shear for twinning (Figure 3a). A lattice with a base additionally requires

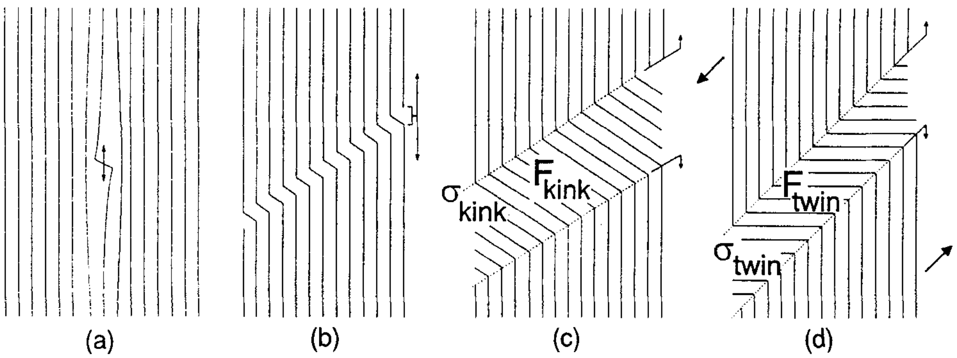

FIGURE 2 Sketch of a possible mechanism of c-twin nucleation. (a) Single kink within a crystal can more along the chain (mechanically activated). (b) The arrangement of kinks within a kink block is energetically favorable. (c) Extending the trans sequences results in a shear deformation of the whole crystal but increases the free energy of the block not being arranged in a crystal lattice. (d) Beyond a certain length of the all-trans part locking to a twinned crystal lattice at the expense of surface free energy is energetically favorable, compare Equation (1).

TABLE I
(hk0)-twinning orthorhombic

K_1	K_2	η_1	η_2	s	2α	k
$(110)_0$	(310)	$[3\bar{1}0]$	[130]	0.25	14.25°	1.13
$(310)_0$	$(1\bar{1}0)$	$[\bar{1}30]$	[110]	0.25	14.25°	1.13

s is the magnitude of the simple shear in the direction η_1. k is the maximum draw ratio attained by twinning. The other symbols are explained in Figure 1.

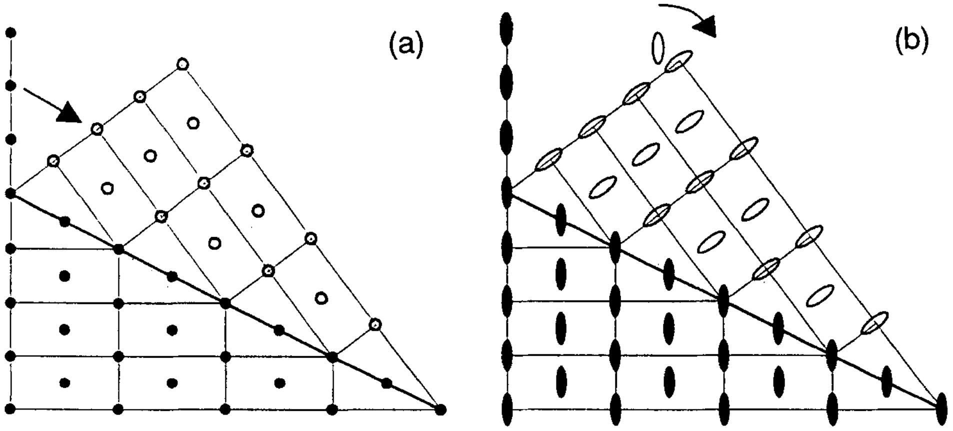

FIGURE 3 (a) Simple shear of a point lattice produces a twin lattice according to Figure 1. (b) A lattice with base additionally needs reshuffling mechanisms to obtain a twinned lattice.

reshuffling mechanisms to restore the exact orientation of the motifs within the twinned part (Figure 3b). The only rotation is that of local motifs. In the case of chain molecules these local rotations are assumed to be crankshaft-like in the first step. This means that the initial movement is not directed to the maximum shear stress.

TABLE II
(hk1)-twinning orthorhombic

K_1	K_2	η_1	η_2	s	2α	k	λ_{eff}	Φ_{kb}	Φ_{sb}
$(110)_0$	(110)	$[49\bar{1}3]$	[001]	1.24	63.5°	1.80	1.75	53°	58.3°
$(210)_0$	(100)	$[10\bar{2}]$	[001]	1.37	68.87°	1.90	1.87	50°	56°
$(021)_0$	(010)	$[01\bar{2}1]$	[001]	2.06	91.61°	2.47	2.47	38.6°	?

λ_{eff} is the maximum draw ratio normal to the c-axis which is attained by these twin modes. Φ_{kb} is the angle of a kink band with the chain axis in a crystal. Φ_{sb} is the shear band angle with the c-axis for a crystallinity $V_c = 0.8$ (see Figure 7).

TABLE III
(hk1)-twinning monoclinic

K_1	K_2	η_1	η_2	s	2α	k	λ_{eff}	Φ_{kb}	Φ_{sb}
$(201)_m$	(100)	?	[001]	1.11	58.07°	1.70	1.63	55.9°	61°
$(011)_m$	(110)	?	[001]	1.33	66.64°	1.85	1.81	51.3°	56.7°

The meaning of the symbols is the same as in the Tables I, II.

Twinning modes in PE determined by classical crystallography have been given in Reference 15. Our intention was to see how the chains might behave. Thus we obtained twinning modes which have not be considered in this classification on account of their high shear. Our results are given in Table II. Additionally we calculated the twinning modes for the monoclinic crystal modification which has been observed during the deformation process. They are given in Table III. The general rule that modes with the least shear are preferable may be modified in polymers. The chains must accommodate to both lattices, and the twin boundary energy must be minimized. Shears up to ~ 1 have been observed in inorganic crystals of high symmetry ($s = 0.707$ for Cu (fcc) and Fe (bcc), $s = 0.908$ for TiO_2 (tetragonal) $s = 1.066$ for Mg (hexagonal)).[11] Thus the shears of our listed twinning modes appear to be reasonable for the chain molecules crystalizing in low symmetry. The monoclinic modification has one important advantage over the orthorhombic one, i.e. all chains have the same setting angle. Thus, all the necessary reshuffling is the same for all chains contrary to the orthorhombic lattice, this evidences in favor of a transformation sequence orthorhombic → monoclinic → c-twinning → monoclinic → orthorhombic in the case of c-twinning.

Let us consider some features of such twinning processes. Of special importance was to know up to which draw ratio the crystal is capable to follow the macroscopic deformation. For this purpose we examined a sphere which transforms into an ellipsoid of equal volume.[16] The ratio of the long axis and the radius of the sphere defined the local draw ratio k.

$$k = \frac{s}{2} + \sqrt{\left(\frac{s}{2}\right)^2 + 1}. \tag{2}$$

Since it embraced the apparent rotation of the ellipsoid, we observed the projection to the direction of the stress defining the macroscopic draw ratio λ_{max}. It is

$$\lambda_{max} = k\cos\phi, \qquad \phi = \frac{\pi}{4} - \frac{\alpha}{2}. \tag{3}$$

Both are listed in the tables.

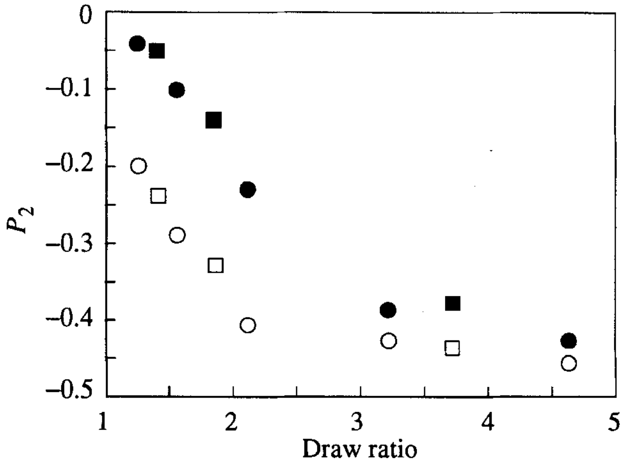

FIGURE 4 Orientation parameters P_2 of the *a*-, *b*-, and *c*-axes determined by X-ray scattering.[18] The open symbols represent the *a*-axis, the filled symbols represent the *b*-axis. Squares are from HDPE, circles are from LDPE.

OBSERVATIONS DURING DEFORMATIONS

There are many peculiarities observed during the deformation of semicrystalline polymers which have to be explained. Let us refer to the results obtained with PE, because it is the most thoroughly investigated polymer. First the overall measure of crystal orientation, the orientation parameter P_2 of the *c*-axis, does not follow the affine deformation scheme which appears for passive elements within the deforming rubber-like matrix. The orientation of the *c*-axis generally proceeds faster than affine. Whereas lower orientations compared to the affine one are easily explained by the appearance of a slip, the only possibility of higher orientations is provided by internal deformation processes of the crystals. The corresponding orientation parameters of the *a*- and *b*-axes are not equal like in the affine scheme but show a markedly different behavior,[17] (see Figure 4). The same behavior has been observed at alternating copolymers of ethylene and fluoroethylene.[18]

The orientation density distribution of the crystallites *c*-axes reveals the most striking difference as compared to the affine deformation scheme. The latter develops the maximum in the draw direction, the experiments[2,18–20] always show the maximum starting at about 30–35°, (see Figure 5).

There are other evidences of structural changes within the deformed polymer. The development of the long period shows the changing crystal thickness and distance. The observations of the long period are found in Reference 21. They are completed measurements of the crystal coherence dimensions in the crystallographic directions.[22] The general features of all measurements are similar. The long period exhibits a transition behavior from a lamellar to a fibrillar structure at about $\lambda \sim 2$. The main effects occur during the first 100% of deformation. Afterwards the long period or the crystal thickness in the chain direction remains more or less constant, the difference in the orientation parameters of the *a*- and *b*-axes decreasing.

Unexpected data were obtained when rolling single crystal mats with chain axes per-

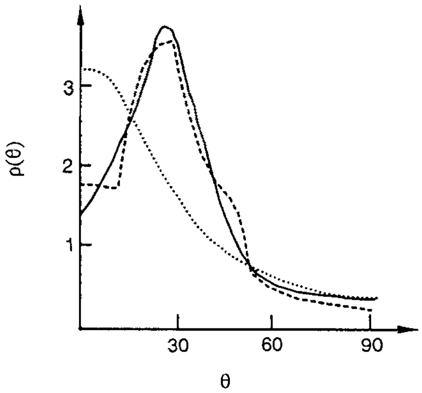

FIGURE 5 The c-axis density distribution of an uniaxially stretched PE[19] (solid curve) compared to the result of the affine deformation (dotted curve) and a theoretical one (dashed curve), calculated with the assumption of (111)-twinning.[7]

TABLE IV
Sequence of (hk0)(111)-twinning

(hk0)	δ_a	δ_b
(110)	51.5°	−9.65°
(310)	53.5°	1.25°

pendicular to the sample plane. The thickness diminished gradually whereas the crystal lattice rotated through a finite angle.[23] Similar observations have been made by stretching highly oriented samples normal to the orientation axis.

EXPLANATIONS

We will show at least qualitatively how the most unexpected features can be explained by the c-twinning.

Figure 2 provides explanation of the results obtained.[24] The crystal lattice "rotates" stepwise by the twinning angle 2α whereas the change of the thickness follows continuously the transformed portion of the crystals. The unit cell rotates around the b-axis in accordance with the action of (201)-twinning or a sequence (111)(hk0). The latter produces the angles of the a- and b-axis with the plane of the sample listed in Table IV. If a crystal lamella is pressed along the c-axis (uniaxial rolling of single crystal mats) a sequence of twinning modes may appear. To illustrate it we show the angles δ_a, δ_b of the crystals a- and b-axis with the lamellar surface (plane of the sample). It is obvious that the b-axis seems to be a rotation axis of the unit cell. In the case of the monoclinic phase the relations are not known.

A different behavior of the a- and b-axis orientation parameter during uniaxial drawing is easy to understand. The first (hk0)-twinning tends to turn the b-axis into the draw direction, and the a-axis perpendicular to it. The c-axis remains unchanged. (111)-twinning affects the a-axis less than the b-axis when orienting them perpendicular to the draw. (201)-

twinning turns the a-axis perpendicular to the draw and retains the b-axis. Thus the different behavior of the two axes is obvious. The final orientation is accomplished by the normal c-slip deformation. A more detailed analysis must incorporate different orientations of the unit cells prior to deformation as well as the appearance of the monoclinic modification.

The orientation density distribution with its striking maximum at ~ 30° has been calculated already in Reference 7. One result is shown in Figure 5. The important feature is the non continuous "rotation" of the crystal lattice. It makes the orientation faster than in the case of the affine orientation. This property has been roughly accounted for by slip models.[24] When regarding pure numerical relations we find them to show a sufficient fit to the experimental data. However, note should be made that just for these slip mechanisms there exists no shear stress for the crystals in perpendicular orientation.

ANISOTROPIC YIELDING

The phenomenon of anisotropic yielding of uniaxially oriented samples was investigated years ago.[25–27] In this case the orientation is not random, and twinning depends on the relative orientation of the sample and the applied uniaxial stress. A usual criterion for the appearance of yielding at the stress σ_y is the maximum resolved shear stress τ within a glide system which must exceed some critical value τ_c. The general form is given by

$$\tau_c = \sigma_y \cos\beta_1 \cos\beta_2, \tag{4}$$

where β_1 denotes the angle between the normal $\boldsymbol{e}_n$ on the glide plane and the applied stress σ, and β_2 denotes the angle of the glide direction g with the trace of the $(\sigma \boldsymbol{e}_n)$-plane in the glide plane (see Figure 6a). In the mostly assumed coplanar case the directions of $\boldsymbol{e}_n$, σ, and g lie in one plane resulting in

$$\tau_c = \sigma_y \sin\beta \cos\beta. \tag{5}$$

In this case the maximum shear stress acts in the 45° direction. When parallel and perpendicular to the applied stress, the shear stress vanishes. This poses a severe problem in understanding the orientation of crystals oriented normal to the draw direction. There is no shear and hence no torque to orient the c-axis. When considering twinning (see Figure 6b), we should not identify the directions g and η_1. The particle displacement during the crankshaft motion starts into another direction than it ends, compare the bended arrow in Figure 6c. We regard the beginning movement of the motifs as the "shear direction" g. Hence, we are dealing with the general case. The angles β_1, β_2 according to Figure 6a are found using rotation matrices.[28] The result for the critical shear stress now is

$$\tau_c = \sigma_y(a\sin 2\theta + b\sin^2\theta - c) \tag{6}$$

with the constants given by

$$a = \cos\Psi\cos\hat{\Psi}\cos 2\alpha - \cos\alpha\sin\Psi\sin\hat{\Psi}$$

$$b = \sin\alpha[\cos\alpha\cos\hat{\Psi}(\cos^2\Psi + 1) - \sin\hat{\Psi}\sin\Psi\cos\Psi] \tag{7}$$

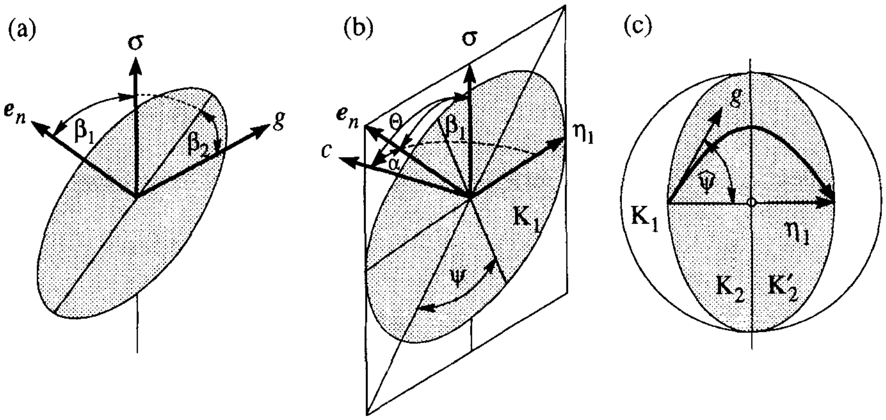

FIGURE 6 (a) A general glide system. Applying uniaxial stress σ, the maximum resolved shear stress τ into the direction g within the glide plane with normal e_n has to be determined. It is given by Equation (4). The angles β_1 and β_2 are obvious. (b) The glide system proposed for c-twinning. K_1 is the glide plane. The intersection between K_2 and K_2' (the rotation axis of the chains) makes an angle Ψ with the trace of the (σe_n)-plane in the K_1-plane. The chains make angles θ and α with the stress and the normal e_n of K_1, respectively. (c) Projection of the crankshaft movement onto the glide plane K_1 visualizing the meaning of the angle $\hat{\Psi}$. The glide direction is formally given by η_1 but the glide starts into the direction g which makes an angle $\hat{\Psi}$ with η_1.

$$c = \cos\alpha \sin\alpha \cos\hat{\Psi}.$$

The meaning of the angles is obvious from Figure 6. This result differs from the equation used to describe the experimental observations only by lacking the constant term. Besides, $c = 0$, i.e. $\hat{\Psi} = \pi/2$, means that the motion starts perpendicular to the plane spanned by the c-axis and the draw direction. We fitted our equation to the published results. Additionally a coplanar process has been added accounting for the normal slip processes. This combination of processes explains the observed behavior. The best fit is achieved at $\hat{\Psi} = 70°$ when accepting the crankshaft mechanism and

$$\tau_{[001]\mathrm{s}} \approx 4.7\tau_{(111)\mathrm{t}} \tag{8}$$

when preferring twinning to slip. However, the ratio of the critical shear stresses have to be temperature dependent.

FINAL REMARKS

To follow an arbitrary deformation there must be five independent shear systems.[10,16] Otherwise processes like twinning, lattice transformations etc. will become active, which is well known from the physics of metals. Polymer crystals must follow the same general rules.

Since c-twinnings have been observed in macroscopic single crystals of polydiacetylene they cannot be treated as simple speculations. The user of ultraoriented fibers suffers from the fact that the transverse yield strength is low. One reason for that is the occurrence of twinning, which is obvious from the anisotropic yield behavior in the above discussion. In single crystals this process will be reversible as long as the twin boundaries exist. It is

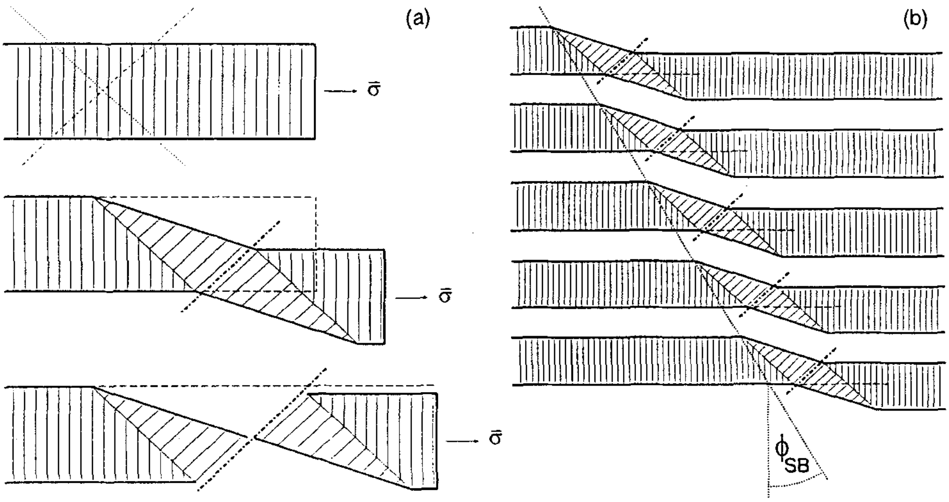

FIGURE 7 (a) A twin starting within a lamella prepares it for c-slip. (b) Within a stack of lamellae the process must run cooperatively, producing a shear band the angle Φ_{sb} of which depends on crystallinity (see Equation (9)).

clear that this process is compatible with any kind of fold surface. Many questions remain unanswered: what can be the real structure of the twin boundary which must not be the mirror plane, how large is the defect energy and by what molecular mechanisms the twin transformation takes place? Hopefully molecular modelling tools will provide some more insight into these problems. Calculations of (hk0)-twins have been presented in Reference 29. Defect energies of bent single chains have been calculated using semi-empirical potentials.[30] The angles may be quite high and close to the twinning angles whereas the energy is typically smaller than 1.6 kcal mol^{-1}. Such energetically favorable angles may not only lead to twinning but also to general deformation kinking[31] in noncrystalline polymers with a short range order. From Figure 8 representing twinning which may proceed within a stack of lamellae, it is obvious that it must be a highly cooperative process. Moreover, we see the occurrence of kink bands in the superstructure. Their angles depend on the twinning angle and crystallinity because the amorphous thickness, and thus the volume, have to be maintained constant.

$$\Phi_{sb} = \frac{\pi}{2} - \arctan\left(\frac{1}{V_c}\tan\alpha\right). \tag{9}$$

These angles are also listed in Tables II, III.

A c-twinning is seen to prepare the crystallites for the action of subsequent c-slip processes. Hence, the slip on (110) planes which is found to be a prominent process embraces (111)-twinning. Note should be made that other processes will also lead to kink and shear bands. In chain extended polymers however the twinning angle may be directly inferred from the structure kinking. The angle between chain axis and kink band should be given by $\Phi_{kb} = \pi/2 - \alpha$. Indeed the statistics of kink bands in such PE samples reveals a broad skewed distribution of chain axes with the plane. The maximum is at 62° with a hump at 55° (see Reference 32). The theoretical angles of c-twinning are listed in Tables II, III.

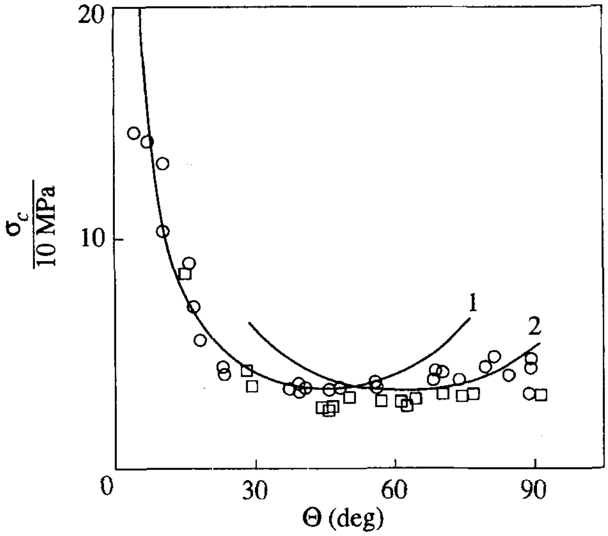

FIGURE 8 Direction depending yield stress of oriented PE. The experimental values are taken from Reference 26 (o) and Reference 27 (□). The solid curves are calculated for *c*-slip (curve 1) and for (111)-twinning (curve 2) using Equations (6) and (7), with $\Psi = 0°$, $\hat{\Psi} = 70°$, and the fit parameter (8).

We see that already the monoclinic phases reveal the same angles. The distribution and the skewness is due to the projection to the film plane when taking a photomicrograph.

More indirect figures are frequently found when investigating the deformation process of bulk material. An interesting photomicrograph has been made by W. Wulff[33] at a draw ratio of $\lambda = 1.4$. It is reproduced in Figure 9. Probably it shows a stack of lamellae in the first steps of destruction by *c*-twinning, compare Figure 8. (This figure and the deformation range proposed by us for the maximum number of existing twins definitely encouraged W. Wulff to perform this experiment.)

RESUME

(i) We have shown that *c*-twinning—the details of which still need to be elucidated—is a well defined crystallographic process. It is compatible with any kind of fold surface. Its known properties obey conventional geometrical and energetic rules. This twinning is probably joined to the orthorhombic-monoclinic lattice transformation.

(ii) This process results in a gradual deformation combined with a large orientational step.

(iii) *c*-twinning is active prior to the structure transformation in uniaxial stretching experiment. It covers the range of draw ratios up to $\lambda \sim 1.8$. It tilts the chains within the lamellae, begins to destroy them, and brings the crystallites into orientations suited for *c*-slip (induction of micronecking and necking).

(iv) In proper deformation experiments (e.g. shear or compression of oriented material) it leads to well defined kink or deformation bands.

Despite the fact that no unambiguous direct *c*-twinning in PE has been observed, there is little doubt that this process is of general importance for any type of chain molecule.

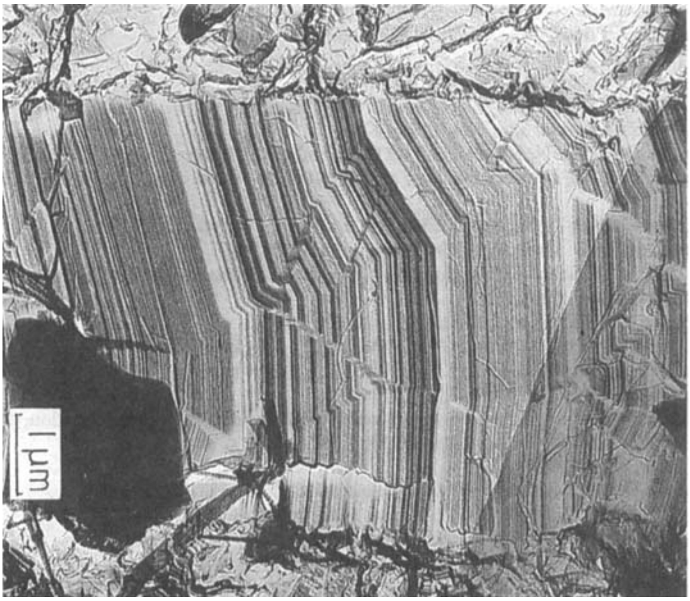

FIGURE 9 Kink bands in chain extended PE (courtesy of D. C. Bassett). The angles observed are compatible with c-twinning.

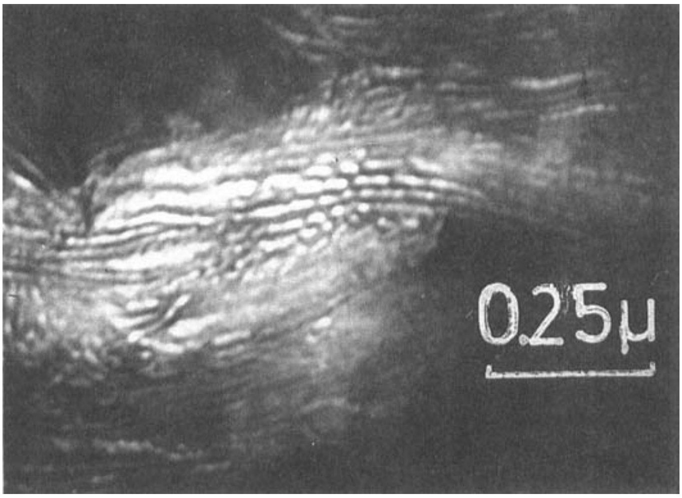

FIGURE 10 Photomicrograph of a stack of lamellae in linear PE, with crystallinity $V_c = 0.82$. The cooperative distortion of the stack is possibly caused by twinning. The draw direction is horizontal, the draw ratio is 1.4. (Courtesy of W. Wulff).[34]

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