

Plastic deformation of polyethylene crystals by dislocation motion

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Solvent-free polyethylene single crystals were plastically deformed by depositing the crystals on carbon replicas obtained from the surfaces of deformed copper single crystals. The observations by transmission electron microscopy suggest that plastic shear parallel to the molecular chains of the polyethylene crystals ([001]-direction) occurs by the movement of dislocations the Burgers vector of which is parallel to the [001]-direction.

1. Introduction

The existence of [001]-dislocations in the lattice of polyethylene crystals has been deduced from both experimental studies [1-3] and theoretical considerations [4, 5]. It is the purpose of this paper to report observations by transmission electron microscopy suggesting that plastic shear of polyethylene single crystals parallel to the direction of the molecular chains ([001]-direction) occurs by the motion of [001]-dislocations.

2. Experimental procedures

Single crystals of linear polyethylene (Lupolen 6011 L, branching degree less than 10^{-3} , $M_w = 10^4$) were isothermally grown at 80°C from solutions of 0.005% of the polymer in xylene [6]. The solvent was removed from the crystals by depositing a droplet of the suspension on a water surface. The evaporation of the xylene resulted in solvent-free polyethylene single crystals floating on the water. In order to induce a shear stress parallel to the molecular chains, the solvent-free polyethylene crystals were picked up from the water surface on microscope grids coated with a "stepped" carbon film. The "stepped" carbon film was a carbon replica of the electropolished surface of a copper single crystal deformed in the region of easy glide or at the beginning of state II. Since there are strong adhesion forces between polyethylene crystals and carbon films and since the orientation of the steps of the carbon film was approximately parallel to the direction of molecular chains in the polyethylene crystals, it was hoped that this procedure results in plastic shear of the polyethylene crystals parallel to the

[001]-direction. After evaporating the water, the polyethylene crystals were investigated by dark field transmission electron microscopy. For convenience, the dark field micrographs were generated mostly by using (110) reflections. The electron microscope employed was a Philips EM 300 operated at 60 kV.

3. Results

Fig. 1a shows a typical dark field electron micrograph of a polyethylene crystal deposited on a carbon replica. In addition to the bright and dark background a pattern of pairs of closely-spaced bright and black patches may be noticed. The position of some of the patches is indicated schematically in Fig. 1b. It may be seen from Fig. 1a that the pairs of patches have the following characteristic features:

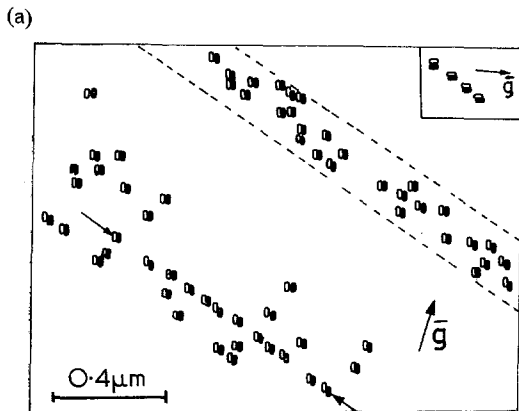
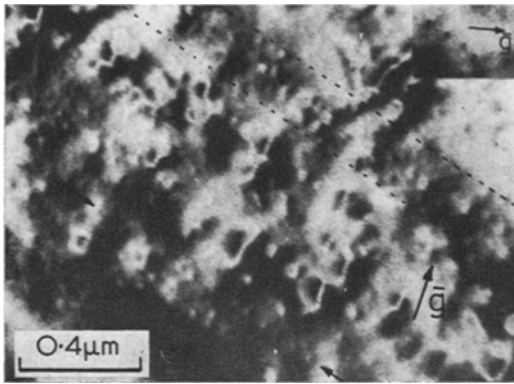
(a) Each bright patch is associated with a dark one and vice versa.

(b) The characteristic distance between the centres of the black and bright patches was about 200 to 400 Å.

(c) The lines connecting the centres of the bright and dark patch was approximately normal to the scattering vector (\vec{g}), if the specimen was oriented according to the Bragg reflection condition.

(d) In the specimen areas with "Bragg orientation" the dark and bright patches had roughly about the same size. With increasing deviation from that orientation, the patches became asymmetrical and unequal in size (cf. the patches of the inserted part of Fig. 1a).

The density and the arrangement of the patches in the polyethylene crystals was found



(a) Dark field electron micrograph ($g = 110$) of a polyethylene crystal deposited on a carbon replica obtained from the surface of a plastically-deformed copper single crystal. In the region indicated by the two arrows a slip step of the carbon replica intersected the polyethylene crystal. It may be seen that the slip step results in the formation of a straight row of patches. The area between the two broken lines indicates the region at which a slip band of the carbon replica intersected the polyethylene crystal. (b) Schematic diagram indicating the position of some of the patches visible in Fig. 1b.

to be correlated as follows with the step structure of the underlying carbon replica.

(e) The density of the patches increased with increasing step density. In other words, increasing plastic deformation of the copper crystal that was used to obtain the carbon replica resulted in a higher patch density.

(f) Long straight rows of patches (for example the row indicated by the two arrows in Fig. 1a) were observed when a very fine slip line of the carbon replica intersected the polyethylene crystal.

(g) In the region where a slip band intersected the polyethylene crystal, bands with a high density of patches were observed (cf. the patches in the area between the two broken lines in Fig. 1a).

The major difficulty in observing the strips (rows) of high patch density in all parts of a crystal came from the fact that the lattice of a polyethylene crystal deposited on a carbon film was buckled [7]. Therefore the electron diffraction conditions and hence the visibility of the patches varied appreciably between different areas of the same crystal (cf. Fig. 1). Nevertheless in practically all cases (about 300 crystals were investigated) it was possible to tilt the crystals so that the electron diffraction by the (110) planes was sufficiently uniform to reveal the strips (rows) of high patch density extending all the way across the polyethylene crystals intersected by slip bands (slip lines) of the carbon replicas.

4. Discussion

The characteristic features of the patches reported coincide with the contrast effects observed and predicted [8-10] for screw dislocations in thin crystals seen end-on by transmission electron microscopy. It is therefore proposed that the patches represent screw dislocations with a Burgers vector parallel to the [001]-direction of the polyethylene crystals. The observed correlation between the slip-line structure of the carbon replicas and the arrangement of the patches in the polyethylene crystals, may be interpreted by assuming that the polyethylene crystals have adjusted to the steps on the surface of the carbon replicas by plastic shear, which occurred by the movement of [001]-dislocations in the polyethylene crystal lattice.

It follows from the theory of dislocations [11] that the polyethylene crystals can adjust, by plastic deformation, to the step pattern on the surface of the carbon replica by introducing the appropriate number of [001]-dislocations. In order to generate a step (height h) parallel to the [001] direction* in the polyethylene crystal, h/b dislocations are required if b is the Burgers vector of the [001]-dislocations in polyethylene. Hence a band of locally high step density in the carbon replica (for example the replica of a slip band in the copper crystal) results in a band of high dislocation density in the polyethylene crystal. The band of high dislocation density is

*It is assumed that the step terminates somewhere in the polyethylene crystal. In order to generate a step extending over the whole crystal a dislocation has to be moved across the whole crystal.

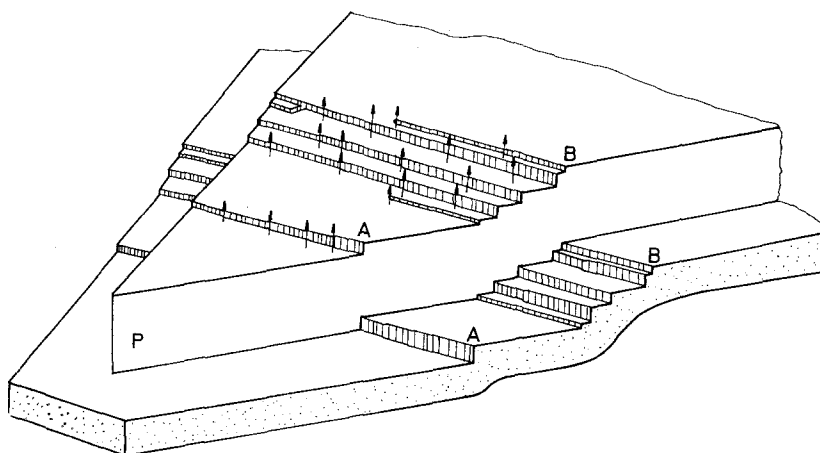


Figure 2 Schematic drawing of a polyethylene crystal (P) deposited on the "stepped" surface of a carbon replica. In order to fit the polyethylene crystal properly to the slip steps, dislocations have to be introduced in the crystal. The location and the Burgers vector of the dislocations is indicated by the arrows. A is a single slip step; B is a slip band.

located in the region at which the slip band in carbon replica intersects the polyethylene crystal that is deposited on the replica. This is schematically shown in Fig. 2 for the slip band B and agrees with the high density of patches observed in the region where a slip band intersects the polyethylene crystal (Fig. 1a, the region between the broken lines). In order to see how a straight row of dislocations in the polyethylene crystals (cf. the row indicated by the two arrows in Fig. 1a) may be explained in terms of that model, let us assume that there was a straight slip step of varying step height on the surface of the copper crystal (slip step A, Fig. 2). A slip step of this kind may for example result from a set of parallel dislocations in the copper crystal residing on the same slip plane, and with Burgers vectors normal to the free surface. If we produce a replica of that slip step and deposit a polyethylene crystal on that replica, the polyethylene crystal may adjust by plastic deformation to the slip step on the replica if a straight row of parallel $[001]$ -dislocations is introduced in the polyethylene crystal lattice along the line of intersection between the slip step and the polyethylene crystal. Hence both the observed straight rows of patches and the bands of high patch density (Fig. 1a) may be interpreted in terms of the proposed dislocation model suggesting that plastic shear of polyethylene crystals parallel to the $[001]$ -direction occurs by the motion of $[001]$ -dislocations.

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