

Electron Microscopy of Some Stony Meteorites [and Discussion]

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Electron microscopy of some stony meteorites

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[Plate 1]

High-voltage (1 MV) electron microscopy has been used to study substructures within grains of the minerals olivine and pyroxene, in stony meteorites of a type that probably originated by aggregation of solid particles in the early Solar System (ordinary chondrites). In specimens that have not suffered later heating or shock-deformation, the substructures were largely formed during cooling after early high-temperature events. The meteorite Saint-Séverin was deformed while slowly cooling, after the rock had acquired a coarse metamorphic texture. The recovered substructures in its olivine are distinguished from cold shock-deformation and from shock-heating effects. In three meteorites (Hedjaz, Parnallee and Chainpur) with relatively low-temperature histories during and after aggregation, inclusions with prior high-temperature histories (chondrules) contain pyroxenes showing effects of relatively rapid cooling. The predominant calcium-poor pyroxenes contain planar defects and submicroscopic twinning. Exsolution lamellae in calcium-bearing pyroxenes, and antiphase domains in one grain of pigeonite, are interpreted in terms of cooling rate by comparison with similar substructures in volcanic pyroxenes. Results of electron microscope microanalysis of pyroxenes in the fine-grained matrix of the low-temperature aggregates indicate that a range of calcium-bearing compositions has been inherited from environments pre-dating aggregation.

1. INTRODUCTION

Meteorites contain a record of events in the early history of the Solar System. Many of them were aggregated at low temperatures and have suffered little subsequent heating. This is true of gas-rich meteorites, which retain noble gases implanted in individual grains from the solar wind (Pellas 1972). Their irradiation history indicates relatively late aggregation, under conditions similar to those in the modern Solar System. Their compaction and cementation to form cohesive rocks is largely due to mild shock deformation (Ashworth & Barber 1976*a*). The present paper is concerned with meteorites aggregated under conditions prevailing at an earlier time. The specimens belong to the commonest class of meteorites, the *ordinary chondrites*. These contain *chondrules*, which are approximately spherical objects *ca.* 1 mm in diameter, widely believed to have originated as liquid droplets (see, for example, Mason 1968; Binns 1970). These are bound together by interstitial *matrix*. In *unequilibrated* ordinary chondrites, a relatively low-temperature history during and after aggregation is indicated by disequilibrium mineral compositions (Dodd, Van Schmus & Koffman 1967). In these, the matrix is usually too fine-grained to be investigated by optical petrographic methods.

There is a gradational series between the most unequilibrated ordinary chondrites and those which are much coarser, and have homogeneous mineral compositions. This series is the basis of the usual classification of these meteorites, in which a number, usually between 3

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(unequilibrated) and 6 (highly equilibrated), is appended to the letter or letters denoting the gross chemical composition (Van Schmus & Wood 1967). The series represents increasingly severe effects of heating, and has been attributed to a rise in temperature after aggregation (Dodd 1969). Alternatively, it has been argued that this *metamorphism* proceeded concurrently with aggregation (Pellas 1972). The high-temperature events were generally ended by a period of slow cooling, typically at rates $\sim 1 \text{ K Ma}^{-1}$ (Pellas & Storzer 1974). This low rate indicates that large masses of material were involved, such as meteorite 'parent bodies' of the size of large asteroids (Wood 1967). The solids may still have been embedded in a slowly cooling gas (Pellas 1972). Gradual condensation of matrix minerals from this gaseous 'solar nebula' has been invoked in a series of theories to explain the chemical compositions of the meteorites, as reviewed by Grossman & Larimer (1974).

The unequilibrated ordinary chondrites contain fragments of pre-existing, relatively coarse rock. These have the same size range as chondrules, but lack the distinctive skeletal or porphyritic crystal morphologies found in chondrules *sensu stricto* (Van Schmus 1969). The implied fragmentation of metamorphosed rocks has been attributed to impacts which, in cases where much kinetic energy was destroyed, could also have generated sufficiently strong local heating to produce the chondrules (Dodd 1971, 1974). Individual chondrules are expected to have had cooling histories before incorporation into the meteorite, as well as sharing in any subsequent metamorphic history.

This paper reports new observations bearing on the cooling histories of several ordinary chondrites or parts thereof. The transmission electron microscope easily resolves the grains in a fine-grained matrix, and substructures within individual crystals can also be studied. Thus we have previously described some dislocation distributions in olivine (Ashworth & Barber 1975*a*) and phase transformations producing planar defects in pyroxene (Ashworth & Barber 1975*b*, 1976*b*), in both cases due to deformation or heating by impact shock. Our observations are made on ion-beam thinned specimens, mostly in a high-voltage (1 MV) electron microscope, giving advantages that have been stated elsewhere (e.g. Ashworth &

DESCRIPTION OF PLATE 1

FIGURE 1. Electron micrographs of chondritic olivines and pyroxenes. (a) Dislocations in olivine in Saint-Séverin, viewed approximately parallel to the [110] direction. The dislocations are partly arranged in (001) arrays. Bright-field, 1 MV. (b) Loops and interacting dislocations in Saint-Séverin olivine, viewed along [210]. Dark-field, operating diffraction vector $g = 004$, 1 MV. (c) Heavily deformed area of Saint-Séverin olivine, with incipient recrystallization in the most deformed area (right), which gives the inset diffraction pattern. Bright-field, 1 MV. (d) Part of a typical large grain of non-calcic clinopyroxene in Parnallee. 1 MV, dark-field using a diffraction vector that is in strongly excited condition in one of the two sets of twin lamellae, I and II, that are represented in the diffraction pattern (inset), so that this set appears bright in the image. Planar (100) defects are visible within the (100) twin lamellae. (e) Coarse pyroxene in Hedjaz with sparse exsolution lamellae approximately on (001), trending from top to bottom of the image, and with dislocation lines. Diffraction pattern (inset) shows predominance of the non-calcic $P2_1/c$ phase, but with maxima having $h+k$ even doubled by the presence of the $C2/c$ lamellae. Bright-field, 1 MV. (f) Calcium-bearing pyroxene with prominent (001) exsolution lamellae (top left) in approximately the same, [110] orientation as the sparsely exsolved non-calcic clinopyroxene with dislocations. Hedjaz. Bright-field, 1 MV. (g) Part of the analysed grain of diopsidic pyroxene in a Hedjaz chondrule, showing fine-scale exsolution, viewed along a direction near $[11\bar{2}]$. Bright-field, 1 MV. (h) Pigeonitic pyroxene with exsolution lamellae, in a Parnallee chondrule. Orientation [010], 100 kV, dark-field, $g = 102$. This reciprocal lattice vector is absent in space group $C2/c$, so the $C2/c$ lamellae appear dark and the more abundant $P2_1/c$ phase appears bright. (i) Antiphase domain boundaries in pigeonite, Chainpur. 100 kV, dark-field, $g = 300$. Orientation approximately [011].

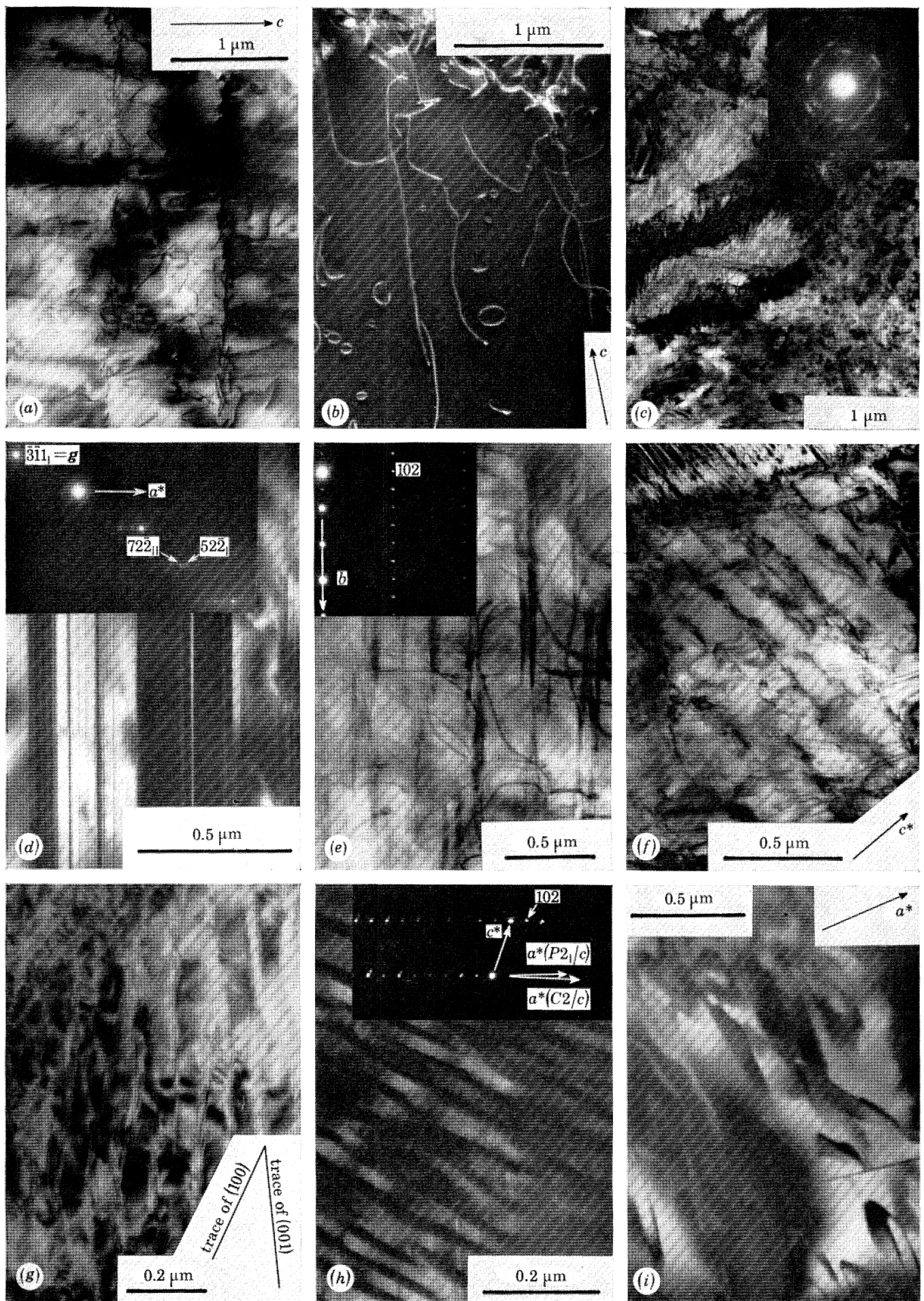


FIGURE 1. For description see opposite.

Barber 1975*b*, p. 149). This paper reports results from the continued study of the commonest minerals in stony meteorites, namely olivine and pyroxenes, but in meteoritic material whose early history has not been obscured by shock events post-dating aggregation and cooling.

Pyroxenes undergo various inversion and exsolution reactions during cooling, with particularly complex results if the cooling is rather rapid. The substructure of olivine, on the other hand, is usually dominated by dislocations and cracks introduced by deformation. As is discussed below in §2*a*, the evidence is that relatively slow cooling is required for these olivine substructures to be strongly modified by annealing processes. Thus we have concentrated our attention on olivine in a slowly cooled, mineralogically equilibrated chondrite (Saint-Séverin), and pyroxenes in some unequilibrated ones. The classification and catalogue numbers (British Museum, Natural History) of the specimens for which new data are reported are as follows: Saint-Séverin, classified as LL6, specimen number BM 1966,493; Parnallee, LL3, BM 34792; Chainpur, LL3, BM 1915,86; and Hedjaz, L4, BM 1925,13 frag. 6.

2. OLIVINE

(*a*) Recovery and recrystallization

We have previously described both low-temperature and high-temperature effects of shock; the former is exemplified by deformational substructures in the olivine in Hedjaz, whereas intense shock-heating has caused coarse recrystallization in the meteorite Goalpara (Ashworth & Barber 1975*a*). We can now describe deformation effects which we deduce have been modified by recovery at intermediate temperatures, with relatively little development of new grains (i.e. recrystallization). These observations were made in Saint-Séverin (figure 1*a-c*). This meteorite is macroscopically inhomogeneous, containing inclusions with no optical indications of deformation (Pellas 1972). Both deformed and undeformed parts are rather coarse-grained and chemically equilibrated, so the meteorite is classified as type 6 (Wasson 1974). The term *brèche de friction* has been applied to chondrites with this type of inhomogeneity, in which the X-ray diffraction patterns from olivine grains in the deformed parts show that the crystal contains regions with slight mutual misorientations (Christophe Michel-Lévy 1971). Our observations are on a small part of the meteorite, in which all olivine appears to be deformed.

In thin-section, the relatively large olivine grains (*ca.* 100 μm –1 mm) have patchy extinction. The extinction position changes, by up to a few degrees, across rather sharp boundaries that tend to be approximately parallel to (001). There is usually no indication of cracking. This type of boundary in unannealed, experimentally deformed olivine has been interpreted in terms of ‘kinking’ (Raleigh 1968), a process which is consequent upon the motion of dislocations in their slip planes and their subsequent alignment into arrays at a large angle to the slip planes. However, the static heating of plastically deformed crystals to promote recovery would produce features with similar optical properties, which are then termed sub-grain-boundaries, instead of kink-band boundaries (Raleigh 1968, p. 5401). Both types of boundary consist of approximately planar arrays of dislocations. The differences between them are likely to lie in their width and in the densities and distributions of the surrounding dislocations. Such boundaries, described as kink-band boundaries, have been reported from optical observations in many ordinary chondrites of types 4 to 6 (Carter, Raleigh & DeCarli 1968). They differ in orientation from the usual extinction-band structure of tectonically deformed terrestrial

olivine, in which the kink-bands or sub-grains are usually aligned approximately parallel to the (100) plane (Raleigh 1968; Boland, McLaren & Hobbs 1971).

In the electron microscope, the olivine in Saint-Séverin is found to contain arrays of dislocations lying approximately parallel to (001) planes (figure 1*a*). Curved sub-grain boundaries at high angles to (001) and healed cracks were also observed. It is noteworthy that the density of dislocations that do not belong to the regular arrays (generally 10^6 – 10^7 mm⁻²) is comparable with the average density of dislocations within arrays. The dislocations that are not in arrays are curved, and often form closed loops within the thin ($\lesssim 1$ μ m) specimen (figure 1*b*). However, there is a tendency for straight segments to occur parallel to the *c* axis (figure 1*a, b*). It is possible to check these segments for screw character by standard contrast-experiment methods at 100 kV (Ashworth & Barber 1975*a*). We find that most, if not all, are screw dislocations; that is their Burgers vector **b** is [001]. It is known from experiments that predominance of this Burgers vector is characteristic of relatively high strain-rate and low temperature (Green 1976); mild shock deformation generally produces dislocations with this Burgers vector (Ashworth & Barber 1975*a*), and their predominance distinguishes Saint-Séverin olivine from that deformed by slow terrestrial processes, in which dislocations with **b** = [100] predominate (Green 1976). Thus the substructure of Saint-Séverin olivine is attributed to impact rather than tectonic deformation.

In this olivine, the most obvious effect of a history at a relatively high temperature is the recovered nature of the dislocation distributions. The shapes of dislocation lines generated by slip in olivine at low temperatures are strongly crystallographically controlled, with the result that most segments are straight and are of screw character, the edge segments having been depleted, perhaps because they have greater mobility during slip (Green 1976); thus, cold shock deformation produces mostly straight [001] dislocation lines (Ashworth & Barber 1975*a*). The curved and interacting dislocations in Saint-Séverin olivine indicate that the mineral has been affected by climb (motion of the dislocations out of their slip planes, a process that is accommodated by the diffusion of point defects). In experiments of duration ~ 1 h, climb effects become clearly apparent in olivine at about 1300 K under both static and dynamic conditions (Goetze & Kohlstedt 1973; Phakey, Dollinger & Christie 1972). In both cases, loops and sub-grain boundaries are formed. Recovery has probably also affected the (001) arrays illustrated in figure 1*a*, since the dislocation arrangement is much more regular than that described by Blacic & Christie (1973) in an experimentally deformed olivine showing (001) extinction bands. It appears to be possible that the (001) arrays in Saint-Séverin are sub-grain boundaries produced entirely during static recovery, although their initiation as deformational kink-band boundaries is suggested by the observation of Goetze & Kohlstedt (1973) that annealing of material containing [001] screw dislocations leads to a substructure consisting predominantly of (100) arrays.

The extent of deformation is spatially variable within the large olivine grains. As is found in the unrecovered olivine of Hedjaz (Ashworth & Barber 1975*a*), the most intensely deformed parts show mosaicism due to pervasive fracture (cataclasis), and have exceptionally high dislocation densities. But in Saint-Séverin most of the cracks are healed, and there is often evidence for incipient recrystallization in the heavily-deformed material (figure 1*c*), producing small new grains containing very few dislocations or cracks. Electron diffraction patterns show that these grains have a preferred orientation inherited from that of the parent grain.

The recovery and partial recrystallization of deformed olivine represent a stage in the meta-

morphic history of Saint-Séverin. The observed deformation, and its modification by partial recovery, post-date the climax of metamorphism, since they are superimposed on the coarse texture which is observed optically. On the other hand, these processes must pre-date the final stages of slow cooling of the meteorite, as recorded by increasing fission-track retention down to temperatures of about 300 K (Pellas & Storzer 1974); it is well known that the annealing conditions required for the fading of charged-particle tracks (Haack 1972; Macdougall, Rajan, Hutcheon & Price 1973) are less severe than those for recovery of dislocation distributions in olivine (Goetze & Kohlstedt 1973; Green 1976). The progressive climb of dislocations into sub-grain boundaries was studied by Goetze & Kohlstedt (1973). The time t required for a given degree of recovery was estimated as a function of temperature T . Extrapolating from their results according to the relation $\ln t = Q/RT + A$, where Q is the activation energy for climb, R is the gas constant, and A is a constant for a given degree of recovery, suggests that no recovery would be expected during the age of the meteorite (approximately 4.6 Ga) at temperatures below about 730 K, even using the lowest Q value compatible with the experimental data, $Q \approx 420 \text{ kJ mol}^{-1}$. According to the cooling equation for Saint-Séverin given by Pellas & Storzer (1974), $T = 923 \exp(-2.23t)$ where t is in Ga, complete recovery by the mechanisms studied by Goetze & Kohlstedt (1973) would be possible only during the first 30 Ma or thereabouts. On the assumption that the recovery processes studied in the laboratory remain predominant on the enormously longer geological time-scale, extrapolation from the experimental results gives an estimate of the lowest temperature on the cooling curve at which significant recovery would be proceeding. It is thus estimated that the observed partial recovery probably occurred above 770 K, implying that the deformation probably occurred during the first 100 Ma of the history discussed by Pellas & Storzer (1974).

The deformation event itself probably did not contribute noticeably to the heating of the bulk of the rock. The intensity of the pervasive deformation is comparable to that in Hedjaz. In the latter case, the deformation occurred after cooling from metamorphic temperatures, and did not produce marked thermal effects (Ashworth & Barber 1975*a*). Both Hedjaz and Saint-Séverin were deformed when in the form of rather coarse, non-porous rocks. They therefore lack the effects of localized heating, at the points of impact between loose grains, which have been observed in specimens that were porous when subjected to mild shock (Ashworth & Barber 1976*a*). In the latter, recrystallization is observed, but only in the fine-grained matrix and at the edges of the larger grains. The interiors of large olivine crystals were not heated sufficiently to cause recovery of dislocation distributions or the erasure of charged-particle tracks. The incipient recrystallization within large grains in Saint-Séverin is quite distinct, not only from matrix recrystallization, but also from the intrusion of veins along cracks in large grains during the shock-lithification of the porous aggregates (Ashworth & Barber 1976*a*, figure 3*c*). The veins in the shock-lithified material contain several minerals, some of them opaque to light, and appear black in thin-section. Black veins are not found in the Saint-Séverin specimen, in which the incipiently recrystallized parts of olivine grains are transparent in the transmitted-light microscope, although their appearance is confused because of the presence of unresolved cracks and grain boundaries. Shock intense enough to initiate recrystallization by heating the olivine could be expected to produce pervasive optical mosaicism in unrecrystallized areas (Carter *et al.* 1968), which is not observed. It is inferred that the deformation is unrelated to the initiation of the cooling described by Pellas & Storzer (1974). Rather, it seems that the deformed parts of Saint-Séverin suffered mild shock during their

gradual cooling. The observations are consistent with the hypothesis that the chondritic *brèches de friction* were formed by low-velocity, accretionary collisions involving masses of hot rock (Pellas 1972).

(b) *Olivine in chondrules and rock-fragments*

The absence of cold-shock features in Saint-Séverin indicates that it has not suffered significant recent impact-deformation. Moreover, the absence of deformational features in some parts of the carbonaceous chondrite Allende has the same implication for that meteorite as a whole (Ashworth & Barber 1975*a*); those inclusions which have been strongly deformed were in that condition before their incorporation in the aggregate. We now find that this is also true of the unequilibrated ordinary chondrite Parnallee, in which undeformed olivine has isolated, often curved sub-grain boundaries. These probably formed during the initial growth of the grain from non-crystalline matter; this process of crystallization is distinct from the recrystallization discussed above. The same origin seems possible for curved sub-grain boundaries in Saint-Séverin, in both olivine and pyroxene, which are associated with small crystalline inclusions and voids. Dislocations 'decorated' in this manner have previously been reported, without any evidence for their being associated with deformation, in Allende (Ashworth & Barber 1975*a*, figure 2*d*).

In our specimen of Parnallee, the olivine in chondrules usually appears undeformed, with dislocation densities $< 10^6 \text{ mm}^{-2}$. Chondrules are identified as those inclusions containing faceted olivine and/or pyroxene grains between which is a much finer-grained mesostasis (in Parnallee, this generally has a grain-size $\lesssim 500 \text{ nm}$). These objects probably originated in liquid form. It is sometimes difficult to distinguish them from unmelted fragments of pre-existing rock, but some coarse objects in Parnallee are clearly shock-deformed rock-fragments. One coarse ($\sim 1 \text{ mm}$ diameter) olivine grain was found to have straight [001] dislocation lines at densities 10^6 – 10^7 mm^{-2} , between areas of much higher dislocation density associated with open cracks that in places produce intense mosaicism. These observations indicate cold shock deformation post-dating growth of the large grain, and are consistent with the idea of repetitive cycles of coarsening and impact-fragmentation (Dodd 1971). The unrecovered nature of the substructure implies a relatively low temperature for the final stages of lithification, which involved growth of small grains in the matrix (§3*b* below).

3. PYROXENES

(a) *Classification of chondritic pyroxenes*

Most meteoritic pyroxenes are approximately ternary solid solutions of the components MgSiO_3 (En), FeSiO_3 (Fs) and CaSiO_3 (Wo). They are classified by chemical composition and crystal structure (Deer, Howie & Zussman 1966). *Orthopyroxenes* have space group Pbc_a. Their crystal structure cannot accept much CaSiO_3 in solid solution; they rarely contain more than 3 mol % Wo, and can be subdivided simply according to the ratio of En to Fs (Deer *et al.* 1966), the common orthopyroxenes in chondritic meteorites being the Mg-rich varieties *enstatite*, defined as having $\text{Fs}/(\text{Fs} + \text{En})$ between 0 and 0.12, and *bronzite*, 0.12 to 0.30. These are the most abundant pyroxenes in those chondrites having a mineral assemblage that has approached equilibrium at high temperatures subsequent to the aggregation of the rock (Mason 1968; Dodd 1969; Van Schmus 1969; Binns 1970). On the other hand, in the unequilibrated chondrites that are the subject of this section, the orthopyroxenes are subordinate in

amount to clinopyroxenes (with space group $P2_1/c$) having the same compositional range, and referred to as *clinoenstatite* and *clinobronzite*. These will collectively be called *non-calcic clinopyroxene*.

It is in the chondrules that the well-known non-calcic clinopyroxenes are found as relatively large grains with optically visible lamellar twinning on (100). Their Ca content is usually less than 1 mass % (Mason 1968; Binns 1970; Dodd 1969, 1971). Figure 1*d* is an electron micrograph of a typical area, with twinning occurring on a finer scale than that which can be seen optically. Much smaller amounts of other clinopyroxenes are also found in chondrules. In unequilibrated chondrites, these *calcium-bearing pyroxenes* occur as rims on non-calcic pyroxene grains or as small independent crystals (Binns 1970). Compositionally they can be divided into pigeonitic and diopsidic types (cf. Van Schmus 1969; Dodd 1968, 1969, 1971). *Pigeonite* has space group $P2_1/c$ and generally has less than 15 mol % Wo; pyroxenes with higher Ca contents have space group $C2/c$. Much of their compositional field is covered by the term *augite*, but compositions close enough to $\text{CaMg}(\text{SiO}_3)_2$ to be called *diopside* (Deer *et al.* 1966) are found in meteorites. In the unequilibrated chondrites, the pigeonitic type varies in Ca content up into the subcalcic augite field (Binns 1970), and the diopsidic type is usually slightly less calcic than a true diopside and should strictly be called *endiopside* or *augite* (Binns 1970; Dodd 1971).

The boundary between calcium-poor pigeonite and clinoenstatite or clinobronzite has not been clearly defined, but the validity of the distinction is supported by a bimodal distribution of Ca contents in pyroxene in the Sharps chondrite (Dodd 1971), suggesting that the boundary should be drawn at about 1 weight % CaO (corresponding to approximately 2 mol % Wo). This is slightly lower than the Ca contents of some orthopyroxenes in equilibrated chondrites (Dodd 1969).

In the present study, differences between non-calcic and calcium-bearing pyroxenes were noticed in the electron microscope. In particular, the calcium-bearing pyroxenes usually exhibit exsolution effects. There is no exsolution in the typical non-calcic clinopyroxene of chondrules (figure 1*d*). Areas with sparse lamellae of $C2/c$ pyroxene approximately parallel to (001) planes (figure 1*e*) presumably have slightly higher bulk Ca content, but complete gradational transitions to typical pigeonite were not observed; calcic pyroxenes with more abundant exsolution lamellae are separated from the larger non-calcic grains by sharp interfaces, although the crystallographic orientations of adjacent grains of the two types are often approximately the same (figure 1*f*). Exsolved pyroxene was also found to be a major constituent of the fine-grained matrix in the unequilibrated chondrites Hedjaz and Parnallee.

The compositions of selected grains were assessed by electron microscope microanalysis (e.m.m.a.). The 100 kV electron beam was focused in the form of a probe of diameter 200 nm–1 μm , depending on the grain size, and using a probe larger than the scale of any exsolution substructure present in order to obtain an estimate of bulk composition. Characteristic X-rays from 2 chemical elements were simultaneously detected using 2 spectrometers. The counting time was 100 s. Each pair of counts can be corrected to give the estimated concentration ratio of the 2 elements, after measurement of intensities from standards of known composition under standard conditions (Jacobs 1973). The molar percentages of Wo and Fs in the pyroxene were then estimated from the Ca/Si and Fe/Si ratios respectively, assuming that the only other important component present was En. Ca/Mg or Fe/Mg was also determined, and was always consistent with the assumption that amounts of components other than En, Wo and Fs are negligible at the level of accuracy attained, which is estimated from counting statistics as being approximately ± 2 mol % for each component.

The results are plotted in figure 2, in the En-rich part of the conventional pyroxene quadrilateral. Figure 2 illustrates the distinction between the non-calcic coarse clinopyroxenes and the subordinate calcium-bearing pyroxenes of the chondrules.

The pigeonites in figure 2 are more magnesian than terrestrial pigeonites. Mg-rich pigeonite probably has a stability field at high temperatures and low pressures (Kushiro 1972; Yang & Foster 1972). Pigeonites of similar composition occur in lunar basalts (e.g. Ghose, Ng & Walter 1972). One of the small grains analysed in a chondrule in Hedjaz transpires to be diopsidic (figure 2).

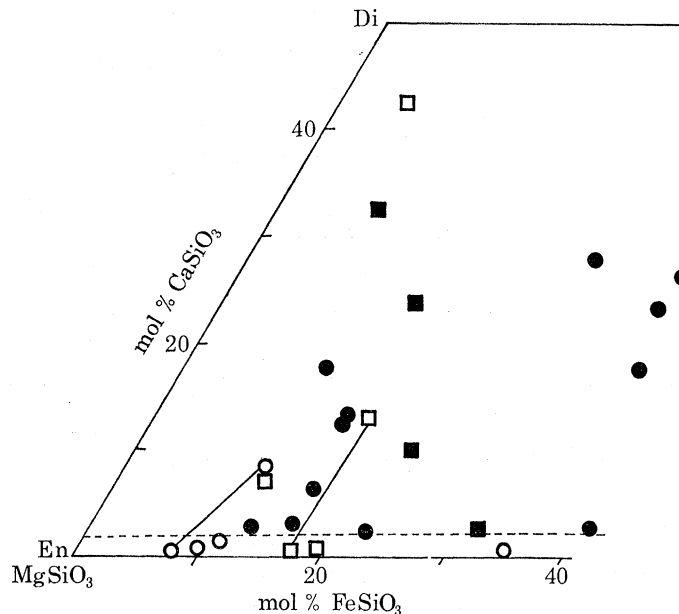


FIGURE 2. E.m.m.a. results for pyroxene grains in two unequilibrated ordinary chondrites. Tie-lines join epitactically related pyroxenes. Pecked line separates coarse (non-calcic) chondrule pyroxenes from other varieties.

	Parnallee	Hedjaz
Chondrule	○	□
matrix	●	■

(b) *Pyroxene compositions in unequilibrated matrix*

Matrix areas were identified optically in specimens after ion-beam thinning. The areas selected had an average grain-size less than 10 μm and were within a few tens of micrometres of the sharply defined boundary of a coarse-grained chondrule or rock-fragment. It is possible that some of these areas lie within fragments of pre-existing fine-grained rock which, however, should not disqualify them as samples of unequilibrated aggregates.

Figure 2 shows the range of compositions found among matrix pyroxene grains. The matrix population is not dominated by non-calcic pyroxene, but is widely scattered in composition. Although some of the analysed grains may have originated as fragments from chondrules, the data as a whole suggest that the matrix mineralogy has been modified by chemical processes in an environment distinct from that which produced the coarse, chondrule pyroxenes. The matrix areas studied have low porosity and a simple grain morphology, indicating that grain growth occurred during or after aggregation (Ashworth & Barber 1976c). The pyroxene compositions should clearly be related to this growth. In the Hedjaz specimen, and perhaps in Parnallee, the matrix has been coarsened by slight metamorphism, but the pyroxene com-

positions indicate that diffusion in the solids was insufficient to equilibrate mineral compositions. In this respect, the present results supplement the established observation that coarser grains of olivine and pyroxene within any unequilibrated ordinary chondrite show a wide range of Fe/Mg ratios (Dodd *et al.* 1967); the matrix analyses show that this scatter persists in the finest-grained material, with the additional complication of widely variable Ca contents in pyroxene. The compositional heterogeneity must be inherited from a stage pre-dating any metamorphism of the aggregated rock. Since other evidence suggests that a gas may have been present, either as a result of vaporization of rock during chondrule-forming impacts (Dodd 1972) or in the form of remnants of a solar nebula, enveloping the accreting planetesimals (Pellas 1972; Dodd 1975), it seems plausible to suggest that small grains grew by deposition of material from the gas phase, after the high-temperature events that produced chondrules. Grains formed at various times and in various places, and hence with various compositions, probably came together gradually to form the matrix. The data indicate that evolutionary models in which it is assumed that matrix grains remained homogeneously in equilibrium with ambient gas, down to a well-defined temperature of aggregation (Grossman & Larimer 1974) are severely oversimplified.

(c) *Twinned clinobronzite and associated chondrule pyroxenes*

The analysed non-calcic pyroxenes (figure 2) fall in the usual range of coarse, twinned, chondrule clinopyroxenes. These are usually interpreted as products of a martensitic inversion undergone by the high-temperature polymorph, protopyroxene, during a cooling stage that was so rapid that the relatively slow reaction to the stable low-temperature form (orthopyroxene) could not occur (Mason 1968; Binns 1970). The fine structure observed in the present study is consistent with this hypothesis. Typically, the pyroxene is twinned on (100) planes on a fine scale, and also has a high density of (100) planar defects (figure 1*d*), giving rise to continuous streaks parallel to a^* in electron diffraction patterns (Ashworth & Barber 1976*b*, Figure 3*d*). This type of defect substructure would be expected in clinopyroxene formed by the martensitic transformation under conditions similar to those pertaining to laboratory X-ray diffraction studies of the quenching of protoenstatite (Brown & Smith 1963; Smyth 1974). Although there are indications that the protopyroxene stability field may be restricted to compositions with Fs < 15 mol percent (cf. Smyth 1974; Reid, Williams & Takeda 1974), there are also indications that protopyroxenes with a much wider range of compositions can crystallize under unusual laboratory conditions (Clarke & Biggar 1972). In view of the incomplete nature of the experimental data and the possibility that many reactions in chondrules may occur metastably, it seems to remain plausible to interpret the twinned clinobronzite as inverted protobronzite.

This twinned non-calcic clinopyroxene is distinct from the untwinned clinopyroxene regions in orthopyroxene grains described elsewhere (Ashworth & Barber 1976*b*) and attributed to deformation by the partial-dislocation mechanism studied by Coe & Kirby (1975). Unrecovered deformation of this type is seen in the orthopyroxene coexisting with recovered olivine in Saint-Séverin. A deformational origin also seems probable for the dislocation distributions found in clinopyroxene in Hedjaz (figure 1*e, f*), since this specimen is known to have been lightly shocked while cold, and since glide dislocations are known to result from deformation of clinoenstatite (Coe & Kirby 1975).

Orthopyroxene with (100) planar defects is found as an intermediate stage in the deformational transformation to clinopyroxene. However, it has now also been observed in association

with *twinned* non-calcic clinopyroxene, in the absence of any deformation features, in a chondrule in Parnallee. In this case it probably either formed from the melt, or inverted from protopyroxene during a cooling stage that was gradual enough for nucleation and growth of the orthopyroxene to compete with the martensitic transformation. In either case, the indication is that the chondrule was annealed for at least several hours at temperatures just below the protopyroxene stability field, rather than being instantaneously quenched. The work of Smyth (1974) indicates that non-calcic pyroxene could probably attain the observed condition by cooling at a few kelvins per hour through the approximate range 1300 to 900 K.

Pigeonite which is related orientationally to the non-calcic pyroxene occurs in chondrules of both porphyritic and radiating types in the classification discussed by Van Schmus (1969). It forms rims on large grains of non-calcic pyroxene, or occurs between laths of the latter in the radiating chondrules. The pigeonite areas, as observed in the electron microscope, have dimensions of several micrometres, and their boundaries with the non-calcic pyroxenes are not crystallographic planes but are irregular in shape. Since it is very unlikely that such large, irregular areas could form by exsolution during the inferred rapid cooling of the chondrules, the pigeonite is interpreted as an epitactic overgrowth on the non-calcic pyroxene.

Twinning and (100) planar defects are consistently much less abundant in the pigeonite than in coexisting non-calcic clinopyroxene. This suggests that the overgrowth formed before the inversion of the protopyroxene, since an overgrowth formed after the martensitic inversion had produced the (100) features might be expected to show some continuity with this substructure. The orientational relationship appears to be consistent with overgrowth on protopyroxene, in that the two clinopyroxenes appear to share the set of crystallographic directions that are unaffected by the inversion, namely a^* , b and c . It is difficult to be certain whether this relation is precise, because there is usually a crack at the boundary. The crack may itself have formed as a result of the large decrease in volume during the inversion of protopyroxene (Brown & Smith 1963), which is believed to have produced optically visible arrays of cracks in the coarse non-calcic clinopyroxenes (Binns 1970). Thus the observations are consistent with overgrowth of pigeonite on protopyroxene followed by inversion of the latter.

The compositional gap between analysis points a few micrometres apart in epitactically related pyroxenes (figure 2) bears some resemblance to the protoenstatite–pigeonite miscibility gap in the most recent experimentally-based phase diagrams (Warner 1975), but it is unlikely that crystallization proceeded close to equilibrium. Compositional zoning, indicating fractional crystallization, was noted by Binns (1970) in clinobronzite in a Parnallee chondrule. And in the present work it was observed that the sparse exsolution lamellae in some coarse clinopyroxenes in Hedjaz (figure 1*e*) vary in abundance, their number generally increasing towards the edge of the grain. Such behaviour indicates a gradual increase in bulk Ca content. It is likely that this fractional crystallization trend culminated in the late crystallization of the more calcic phases. Both the gross morphology of pyroxene in radiating chondrules, and the frequency of epitaxy, suggest that difficulty of nucleation was an important control on crystallization behaviour.

In summary, the observations on chondrule pyroxenes in Parnallee and Hedjaz are consistent with fractional crystallization from a liquid. Calcic pyroxenes often grew epitactically on the non-calcic pyroxene, which has substructures interpreted in terms of metastable inversion from protopyroxene during rather rapid cooling. Some further light will be shed on cooling rates by a consideration of the diffusive solid-state reactions that produce exsolution lamellae.

(d) Exsolution in calcium-bearing pyroxenes

The analysed diopsidic pyroxene has sets of fine exsolution lamellae approximately parallel to (100) and (001) (figure 1g). The complex contrast between the lamellae probably arises from strain due to the lattice misfit between the two phases. This substructure is comparable in scale and geometry with that reported in diopside in a terrestrial volcanic rock (Liddell, Phahey & Hobbs 1974).

In the pigeonitic pyroxenes, the usual exsolution feature is a set of well-defined lamellae of C2/c pyroxene, usually approximately parallel to (001). Figure 1h shows a typical area. The C2/c and P2₁/c phases have *b* and *c** directions in common. This lattice orientation and the (001) lamellar habit are presumably related to strain energy considerations (Morimoto & Tokonami 1969*a*). In the [010] electron diffraction pattern, the C2/c phase produces distinct maxima (figure 1h). Corresponding P2₁/c and C2/c maxima are joined by weak streaks approximately parallel to the 102 reciprocal lattice vector. The difference in β crystallographic angle between the two phases is approximately $\Delta\beta = 2^\circ$.

Similar exsolution effects are common in the pigeonites of lunar basalts (Champness & Lorimer 1971; Nord *et al.* 1973). The regularity in lamella width and spacing (figure 1h) is consistent with an origin by coarsening after spinodal decomposition (Champness & Lorimer 1971). In principle, quantitative inferences about cooling history could be made from more detailed electron microscopic observations if experimental data on the rates of the relevant processes were available (McConnell 1975). Qualitatively it can be said that the chondrule containing the pigeonite was not quenched from the temperature of crystallization, but probably had a cooling history comparable with those of lunar volcanic rocks having similar exsolution features in pigeonites of similar composition. The typical Parnallee example (figure 1h) is intermediate between the extremes observed in lunar basalts, in both its coarseness and its $\Delta\beta$ value, the latter being a measure of the compositional separation between the phases (Lally, Heuer, Nord & Christie 1975). The exsolution is also comparable with that in pigeonites from a terrestrial shallow intrusion (Dunham, Copley & Strasser-King 1972), but these are relatively Fe-rich, and exsolution behaviour probably depends strongly on pigeonite composition (Nord *et al.* 1973).

(e) Antiphase domains in pigeonite

Antiphase domain structure is a well-known feature of many pigeonites. It results from inversion, the high-temperature structure being C2/c. The domains are responsible for diffuseness of X-ray diffraction maxima for which $h+k$ is odd, called class (b) reflexions. The pigeonites in which presence of domains was first inferred, from X-ray data, are relatively Fe-rich terrestrial and achondritic specimens (Morimoto & Tokonami 1969*b*), but lunar magnesian pigeonites have since been found to contain domains of the same type (see, for example, Champness & Lorimer 1971; Ghose *et al.* 1972; Nord *et al.* 1973). The domains are revealed in the electron microscope by dark-field imaging using a class (b) reflexion. Figure 1i shows domains observed in the present study in a small grain of chondrule pyroxene, which does not show exsolution but is identified as pigeonite because it lacks the twinning and high density of (100) defects present in adjacent coarse non-calcic pyroxene. The domains are coarser than in most lunar volcanic pigeonites (Ghose *et al.* 1972). Other things being equal, slower cooling rates should result in larger domains (Morimoto & Tokonami 1969*b*), but domain size almost certainly also depends strongly on the availability of nucleation sites and on inversion

temperature (Nord *et al.* 1973); in turn, inversion temperature varies strongly with composition (Prewitt, Brown & Papike 1973).

The grain shown in figure 1*i* is particularly favourable for the observation of antiphase domains, because of the absence of confusing contrast from other substructures. It is probable that the chondritic pigeonites generally contain similar domains, which are difficult to observe because they are slightly coarser than the exsolution effects, which give stronger contrast. Antiphase domains were not observed in the non-calcic clinopyroxenes, but presence of widely spaced domain boundaries cannot be ruled out, as they would be obscured by the finer-scale substructures of (100) planar defects and dislocations.

The chondrule of figure 1*i* is in the highly unequilibrated chondrite Chainpur. The coarseness of the domains indicates that the chondrule was not cooled suddenly from the high-temperature field in which pigeonite has C2/c structure. On the other hand, failure to exsolve into two phases suggests more rapid cooling than for the Parnallee example. Low temperatures at the aggregation stage are indicated by the fact that the matrix around the Chainpur chondrule is essentially fragmental, with little evidence of grain growth (Ashworth & Barber 1975*c*). This suggests that 'blanketed' cooling of the chondrule occurred before aggregation, when the ambient medium was probably a gas.

4. CONCLUSIONS

Electron microscope observations within individual grains of olivine and pyroxene are relevant to the interpretation of the cooling history following high-temperature events. At present our inferences are qualitative, but the substructures are complex enough to yield quantitative information if investigated further.

Our data for an equilibrated ordinary chondrite (Saint-Séverin), and 3 unequilibrated ones, confirm and supplement previous deductions from observations by other techniques. Saint-Séverin suffered mild deformation at high strain-rate, probably due to impact, during its metamorphic history. The deformation produced [001] screw dislocations in olivine and (100) planar defects in orthopyroxene. The subsequent slow cooling of the rock permitted widespread recovery and, in places, partial recrystallization of the olivine. The resulting substructures are distinct from both the effect of mild shock after metamorphism, and shock-heating effects such as occur locally in shock-lithified porous aggregates. In the unequilibrated ordinary chondrite Parnallee, olivine in a rock-fragment retains unrecovered effects of cold shock deformation pre-dating its incorporation in the meteorite. In nearby chondrules, olivine without these deformational features is accompanied by pyroxene assemblages consistent with fractional crystallization from a melt. Calcic pyroxenes are often epitactic on earlier non-calcic pyroxene, which has substructures interpreted as the result of inversion from protopyroxene during relatively rapid cooling. However, the chondrules cannot be regarded as simply 'quenched'. Orthopyroxene, with (100) planar defects, grew in some cases. Exsolution in pigeonitic and diopsidic pyroxenes in Hedjaz and Parnallee, and antiphase domains in a pigeonite in Chainpur, are comparable with those in lunar and terrestrial volcanic rocks, suggesting that the chondrules had comparable cooling histories. The scatter of matrix pyroxene compositions in Hedjaz and Parnallee indicates that matrix grain growth was not accompanied by extensive diffusion. Variations in mineral compositions may reflect variations in ambient conditions prior to aggregation, when matrix grains may have been reacting with a gas.

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Discussion

G. M. BROWN, F.R.S. (*Department of Geological Sciences, Durham University*). Do the matrix pyroxene compositions plotted on the Di–Hed–En–Fs quadrilateral relate to bulk compositions? If so, their subcalcic augite compositions suggest, by analogy with terrestrial pyroxene assemblages, metastable crystallization through rapid cooling of the melt.

J. V. SMITH (*Department of the Geophysical Sciences, University of Chicago, Chicago, Illinois 60637, U.S.A.*). In the Hallimond Lecture, I emphasized the rôle of disintegrative capture, and am contemplating writing a paper on the origin of chondrules during such an event. Would you care to comment on how your interesting results might bear on the validity or otherwise of such an origin? Particularly I would like to draw attention to the high Na content of some of the matrix in unmetamorphosed ordinary chondrites (e.g. Mezo-Medaras), and to the olivine- and pyroxene-rich chondrules. Perhaps the different components (including the iron-rich metals) can be interpreted as the remnants of a differentiated planetesimal.

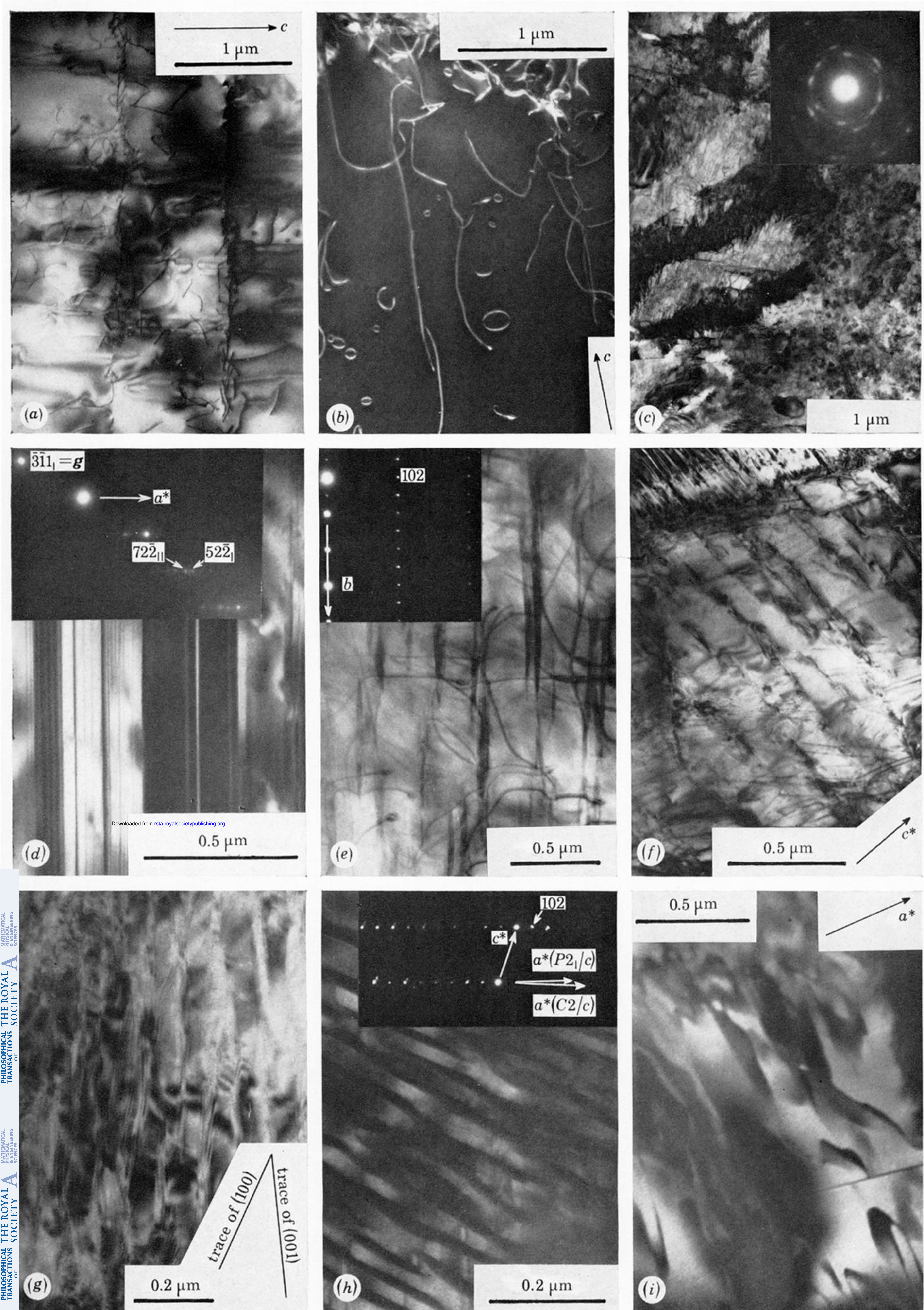


FIGURE 1. For description see opposite.