

THE CORE STRUCTURE AND THE MOBILITY OF DISLOCATIONS IN ICE

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ABSTRACT. The evidence concerning the velocity of glide of dislocations on the (0001) plane of ice Ih is reviewed and related to atomic processes occurring within or near the dislocation core. The velocity is directly proportional to stress at low stress, with a value of the order of 500 Burgers vectors per second at -18°C and a stress of 10^5 N m^{-2} . The usual idea of a dislocation core is that it is "crystalline" in the sense that the linkages between molecules are as far as possible the same as those in the normal lattice. For such a model the disorder of protons presents an obstacle to dislocation movement, and recent theories predict that dislocations should not be able to glide as fast as they are observed to do. Various ways of avoiding this difficulty within the context of a crystalline core are discussed, but none seems likely to be successful. An alternative model is that the core is "non-crystalline", with a disordered arrangement of mobile molecules within it. The movement of such dislocations should not be seriously impeded by proton disorder. Dislocation mobility would then be limited at least in part by anelastic loss due to the stress-induced order of protons in the surrounding lattice.

RÉSUMÉ. *Structure de coeur et mobilité des dislocations dans la glace.* Les données illustrant le glissement des dislocations dans le plan (0001) de la glace Ih sont évoquées et reliées aux processus atomiques se produisant à l'intérieur ou à proximité du coeur des dislocations. La vitesse des dislocations est directement proportionnelle à la contrainte τ aux faibles contraintes et a une valeur de l'ordre de 500 fois le module du vecteur de Burgers par seconde à -18°C et pour $\tau = 10^5 \text{ N m}^{-2}$. Le coeur d'une dislocation est habituellement considéré comme étant "cristallin" dans la mesure où les liaisons entre molécules sont autant que possible identiques à celles du réseau normal. Dans le cadre d'un tel modèle, le désordre des protons représente un obstacle au mouvement des dislocations mais les théories récentes conduisent à des valeurs de la vitesse plus faibles que les valeurs expérimentales. Diverses hypothèses permettant de surmonter cette difficulté tout en conservant l'idée d'un coeur "cristallin", sont discutées mais aucune ne s'avère satisfaisante. Nous considérons alors une autre possibilité reposant sur l'hypothèse d'un coeur "non-cristallin" où les molécules, arrangées de façon désordonnée, auraient une grande mobilité. Le mouvement d'une telle dislocation ne devrait pas, dans ces conditions, être sérieusement affecté par le désordre des protons. Toutefois, la mobilité des dislocations doit être, au moins pour une part, limitée par l'anélasticité associée à la mise en ordre sous contrainte des protons dans les régions environnant la dislocation.

ZUSAMMENFASSUNG. *Die Kernstruktur und die Beweglichkeit von Versetzungen in Eis.* Das experimentelle Material über die Gleitgeschwindigkeit von Versetzungen auf der (0001)-Ebene von Eis Ih wird nachgeprüft und in Beziehung zu atomistischen Vorgängen gebracht, die innerhalb oder in der Nähe des Versetzungskerns erfolgen. Bei geringen Spannungen ist die Geschwindigkeit direkt proportional zur Spannung und hat bei -18°C und einer Spannung von 10^5 N m^{-2} eine Grösse von etwa 500 Burgersvektoren pro Sekunde. Üblicherweise stellt man sich den Versetzungen "kristallin" vor, in dem Sinne, dass die Verbindungen zwischen den Molekülen so weit als möglich dieselben sind wie im gewöhnlichen Gitter. In solch einem Modell stellt die Protonenunordnung ein Hindernis für die Versetzungsbewegung dar, und neuere Theorien sagen, dass Versetzungen nicht imstande sein sollten, so schnell zu gleiten wie sie in Wirklichkeit beobachtet werden. Es werden verschiedene Wege besprochen, diese Schwierigkeit im Zusammenhang mit dem kristallinen Kern zu vermeiden, aber keiner scheint wahrscheinlich von Erfolg zu sein. Ein anderes Modell ist der "nichtkristalline" Kern, mit einer gestörten Anordnung von beweglichen Molekülen in seinem Innern. Die Bewegung solcher Versetzungen sollte nicht ernstlich durch Protonenunordnung behindert werden. Die Versetzungsbeweglichkeit muss dann zumindest teilweise durch inelastische Reibung begrenzt sein, die von der spannungsinduzierten Ordnung von Protonen im umgebenden Gitter herrührt.

INTRODUCTION

In W. T. Read's classic book *Dislocations in crystals* (1953) the first diagram which he gives of a dislocation is that reproduced here as Figure 1. This elegantly summarizes the puzzling questions about dislocations in ice which are the subject of this review.

Ice Ih can be deformed on various slip systems, the simplest being glide on the basal plane (0001) for which the Burgers vector \mathbf{b} is one of the three lattice constants $\frac{1}{3}a\langle 11\bar{2}0 \rangle$. This paper will discuss only dislocations of this type, although similar considerations apply to all types of dislocation which can glide in the ice structure.

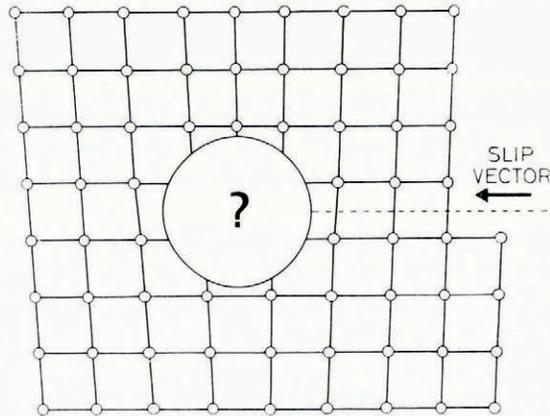


Fig. 1. An edge dislocation in a square lattice (after Read, 1953).

Macroscopic deformation of a monocrystal or within a grain of a polycrystalline sample occurs by the movement of many dislocations. For a single slip system (an unattainable ideal) the shear strain-rate $\dot{\gamma}$ is related to the density ρ and the velocity v_d of the mobile dislocations by the equation

$$\dot{\gamma} = \rho v_d b. \quad (1)$$

The velocity v_d depends on the appropriately resolved local shear stress τ , and the dislocation density ρ depends on the previous growth and deformation history of the crystal. It is not possible to separate the two terms in the product ρv_d by measuring only macroscopic parameters, but values of $v_d(\tau)$ for individual dislocations can be obtained from separate experiments to be described in the following section. One of the most fundamental problems in understanding the deformation of ice is to find a model which can quantitatively account for the dependence of v_d on τ in terms of parameters which have meaning on the molecular level.

The general theory of dislocations requires that away from the core the molecules should fit together normally as illustrated schematically in Figure 1; the lattice will be distorted in a manner that can be treated by the standard theory of elasticity. The arrangement of molecules close to the core is less easily predicted, and yet it is on just these molecules and on the way they move that we must concentrate attention if we are to account for the dependence of v_d on τ . The aim of this review is to compare our knowledge about dislocation mobility in ice with current ideas about the structure of the core. A satisfactory model has not yet been developed, but a great deal can be said about the features a final model should contain.

Similar problems arise in all crystalline materials, and have received a great deal of attention since the first calculations by Peierls (1940). However, the problem in ice has certain unique and specially interesting features. In some materials such as copper and zinc, dislocations move very easily giving high values of v_d ($> 10^{-2} \text{ m s}^{-1}$ at $\tau = 10^5 \text{ N m}^{-2}$). In others such as silicon iron, molybdenum, or the alkali halides, negligible movement occurs below some critical stress of 10^6 to 10^8 N m^{-2} , and above that stress the velocity increases very rapidly with stress. Such behaviour is characteristic of the need to overcome some kind of obstacle by the combined action of stress and thermal activation. In contrast the movement of dislocations in ice has been found to be directly proportional to stress at low stresses but the velocity is very small (of the order of 10^{-7} m s^{-1} at $\tau = 10^5 \text{ N m}^{-2}$ and -18°C) compared with the soft metals. The most comparable behaviour is probably that of dislocations in germanium which at temperatures of 500 – 600°C exhibit an approximately linear dependence of velocity on stress for stresses of the order of 10^7 N m^{-2} with a constant of proportionality similar to that for

ice (Patel and Chaudhuri, 1966). Germanium has not been studied at stresses as low as those used for ice.

There is clearly some mechanism in ice which inhibits the movement of dislocations while maintaining the linear dependence of v_d on τ . In looking for this mechanism attention has been drawn to the disorder of the protons. Glen (1968) showed that this leads to a difficulty over moving the core, and Weertman (1963) pointed out that it also leads to an anelastic loss in the surrounding lattice. These models and their consequences constitute the major part of this review.

More wide-ranging reviews of dislocations and the plastic deformation of ice have recently been written by Weertman (1973), Glen (1974, 1975), and Maï and others (1977).

EXPERIMENTAL EVIDENCE CONCERNING DISLOCATION MOBILITY

The only technique by which the velocities of individual dislocations have been successfully studied in ice is X-ray topography. The most recent and comprehensive results are those of Maï (1976) on relatively straight fresh dislocations of mixed orientation in pure ice; his results are shown in Figure 2. At small stresses the velocity v_d is directly proportional to the stress τ but at larger stresses and at temperatures above about -10°C v_d rises more rapidly than linearly with τ . Within the linear range the results can be fitted to an equation of the form

$$v_d = A\tau \exp(-U/kT), \quad (2)$$

where A is a constant, T is the absolute temperature, and k is Boltzmann's constant. The value given by Maï for the activation energy U is 0.55 ± 0.05 eV, but the value depends on how allowance is made for the non-linearity, and somewhat higher values may also be compatible with the data.

Fukuda and Higashi (1973) also performed X-ray topographic experiments but at the single temperature of -18°C . They found that freshly created and pre-existing dislocations

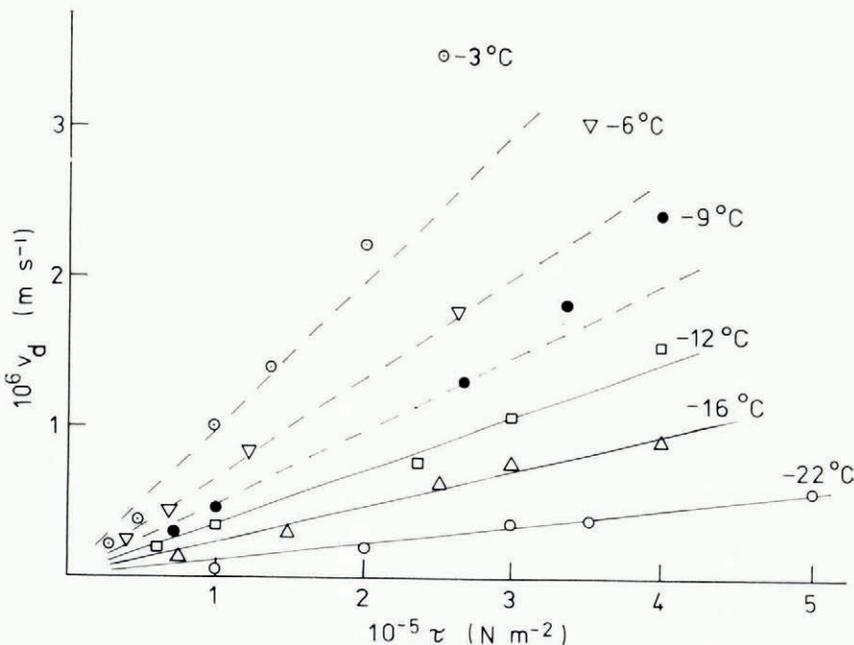


Fig. 2. Velocities of dislocations in ice as measured by Maï (1976) using X-ray topography.

had similar velocities, and that curved dislocations moved two or three times faster than straight ones. Jones and Gilra (1973) performed similar experiments at -15°C but at a single lower value of stress. All the available data in the range -15° to -18°C are plotted on a log-log graph in Figure 3.

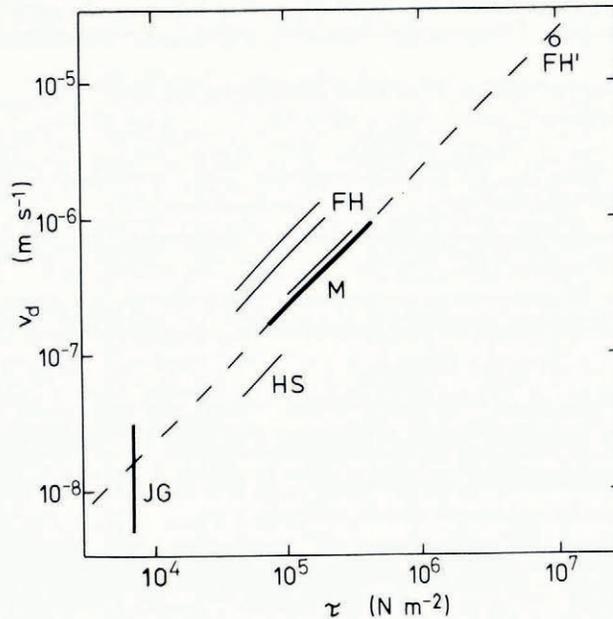


Fig. 3. Collected measurements of dislocation velocity v_d as function of stress τ at temperatures of -15° to -18°C .

- M*: Mai (1976), X-ray topography, -16°C .
FH: Fukuda and Higashi (1973), X-ray topography, -18°C .
JG: Jones and Gilra (1973), X-ray topography, -15°C , range of values at single stress.
HS: Higashi and Sakai (1961), movement of tilt boundary as interpreted by Fukuda and Higashi (1973), -15°C .
FH': Fukuda and Higashi (1969), X-ray topography, approximate value at high stress and -22°C , adjusted to equivalent value at -16°C .

The broken line is a line of unit slope passing through the data of Mai.

Measurements at higher stress present difficulties, but Fukuda and Higashi (1969) have estimated from one set of topographs that the velocity at $\tau = 10^7 \text{ N m}^{-2}$ and -22°C is of the order of 10^{-5} m s^{-1} . Their value, adjusted to the higher temperature of -16°C according to Equation (2), is included in Figure 3, and can be seen to be compatible with a linear extrapolation of observations made subsequently at lower stresses. However the result is not sufficiently reliable to imply that there is no departure from the linear relation up to 10^7 N m^{-2} .

A small-angle tilt boundary consists of an array of parallel edge dislocations. Higashi and Sakai (1961) have observed the movement of such boundaries under an applied stress, and, if interaction between the component dislocations can be ignored, their observations can be taken as measurements of dislocation velocities. Their results are included in Figure 3, and are reasonably consistent with the topographic data. They observed a linear dependence of velocity on stress, and found activation energies between 0.51 and 0.72 eV depending on the angle between the boundary and the a -axis of the crystal.

As already explained, it is not possible to deduce dislocation velocities from the numerous experiments on the creep of ice because the density of mobile dislocations is not known.

However, in his review of the creep of ice, Weertman (1973) has shown that the strain-rates observed in creep are consistent with the measurements of dislocation velocities with very reasonable values for the dislocation density. By studying the stress dependence of the creep rate of pre-deformed crystals, in which the dislocation density is effectively constant, Joncich and others (1978, see also Joncich, unpublished) have shown that at -20°C and -30°C v_d is a linear function of τ .

The main part of the temperature dependence of the creep rate at a given stress is expected to come from the dislocation mobility. Values of the activation energy for creep of single crystals at temperatures above -50°C range from 0.62 to 0.78 eV (Weertman, 1973; Homer and Glen, 1978). These appear somewhat higher than values for dislocation mobility, but there is a significant variation between specimens and between authors using different techniques. It is not possible to draw firm conclusions about these differences, but a large part of the activation energy for creep must be attributed to the movement of dislocations.

The steady-state creep rate of ice has been shown to increase with hydrostatic pressure (Weertman, 1973; Jones, 1978). It is not possible at this stage to know whether this is due to a change in the mobility of dislocations or to a change in their equilibrium density appropriate to the steady-state conditions.

In principle, information about dislocation mobility can also be obtained from measurements of the internal friction of deformed crystals, but this involves problems of interpretation which go beyond the scope of this review.

Jones and Glen (1969) have shown that the deformation of ice is highly sensitive to doping with HF. Doped crystals deformed more easily and the activation energy for creep was reduced to around 0.35 to 0.40 eV. Using X-ray topography, Jones and Gilra (1973) found that HF-doped crystals had a larger dislocation density than pure crystals, and this may in part account for the softening. The change in activation energy, however, suggests that the dislocation mobility is also affected. Recent X-ray topographic measurements by Maï and others indicate that HF doping does indeed lead to an increase in dislocation velocity and to some reduction in activation energy.

All the above experiments concentrate attention on those dislocations which are observed to move, and determine an average value of the velocity of these dislocations over the time of application of the stress. It is not possible to tell whether dislocations move steadily or in jumps, but we can be sure that some dislocations are capable of moving over distances of the order of 0.1 mm at least as fast as the average values quoted.

In summary, for pure ice at -18°C and $\tau = 10^5 \text{ N m}^{-2}$, dislocations can move at a velocity of at least about $2 \times 10^{-7} \text{ m s}^{-1}$, which is equivalent to 500 Burgers vectors per second. The velocity is directly proportional to stress at low stresses and has an activation energy in the region of 0.55 to 0.60 eV.

DISLOCATION STRUCTURE

The water molecules in ice Ih are arranged in the hexagonal structure illustrated in Figure 4. The molecule at any particular site can have four possible orientations and these are occupied at random according to the Bernal-Fowler rules. It has generally been assumed that basal slip occurs between widely separated planes of molecules, i.e. on planes such as ss' or $\tau\tau'$. Slip on nn' is geometrically possible, and dislocations on this plane can dissociate into partials (Yosida and Wakahama, 1962; Glen, 1974). It has been argued that these dislocations are not likely to be mobile. However, recent experiments on Ge and Si (see, e.g. Gómez and Hirsch, 1977) and CdTe (Hall and Vander Sande, 1978), which have similar stacking to ice, indicate that glissile dislocations may dissociate and that glide therefore occurs on nn' . These observations are not directly applicable to ice, in which the bonding is very different, but they indicate that the assumption of glide on ss' or $\tau\tau'$ must be treated with

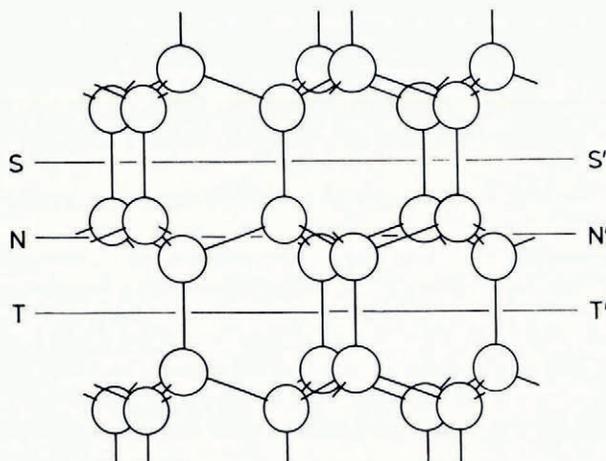


Fig. 4. The arrangement of molecules in the structure of ice Ih. The *c*-axis is vertical. Basal slip is normally assumed to occur on planes like *ss'* or *tt'*.

caution. In reviewing current theories for ice we must initially start by making the traditional assumption.

The simplest orientations of basal dislocations are the screw orientation with the dislocation line parallel to the $\langle 11\bar{2}0 \rangle$ Burgers vector and the 60° orientation with the line at 60° to the Burgers vector. Figure 5(a) shows the structure of a 60° dislocation drawn on the assumption that molecules near the core conform as far as is possible to the normal crystalline arrangement in ice. There are inevitably severe distortions of bond lengths and bond angles close to the core. The precise nature of these distortions is not known, and the figure is meant only to indicate the linkages between molecules, not their actual positions. As this dislocation has an edge component there is of course a dangling bond. This is on molecule *c* in the figure, and is in this case occupied by a proton. Dangling bonds along the length of the core may be randomly with or without protons, though there may be some imbalance resulting in the dislocation being charged. The nature of charged dislocations in ice has been considered by Whitworth (1975, section 15.5). A dislocation can carry its charge with it as it glides, leading in principle to a flow of charge during deformation or to a movement of dislocations in an electric field. Itagaki (1970, 1978) claims to have observed such phenomena, but Joncich (unpublished) could not detect any effect of this kind.

The model of the core represented by Figure 5(a) maximizes the number of hydrogen-bond linkages between molecules. It assumes that the free energy is minimized in the arrangement shown rather than with, say, three dangling bonds on molecules *b*, *c*, and *c'*. This being so the position of the core has a definite location at the symbol \perp just below molecule *c*. If the dislocation moves to the left it will do so by a jump in which bond bd' is broken and *c* becomes joined to *d'*. Such a jump can occur in one $\{1\bar{1}00\}$ plane at a time, and the dislocation will then advance by the propagation of a kink along its length. Figure 5(b) shows a kink on a 60° dislocation at the point β , with the molecules labelled to correspond to Figure 5(a); the jump just described involves the movement of the kink from β to γ .

Figure 6 illustrates the arrangement of water molecules around the core of a screw dislocation, again on the assumption that the molecules link together as far as possible in the normal crystalline arrangement. There are distortions of about 10% in some bond lengths and around 15° in some bond angles. The position of the core is definitely located between two rows of bonds crossing the slip plane, and movement of the dislocation involves the propagation of kinks along its length.

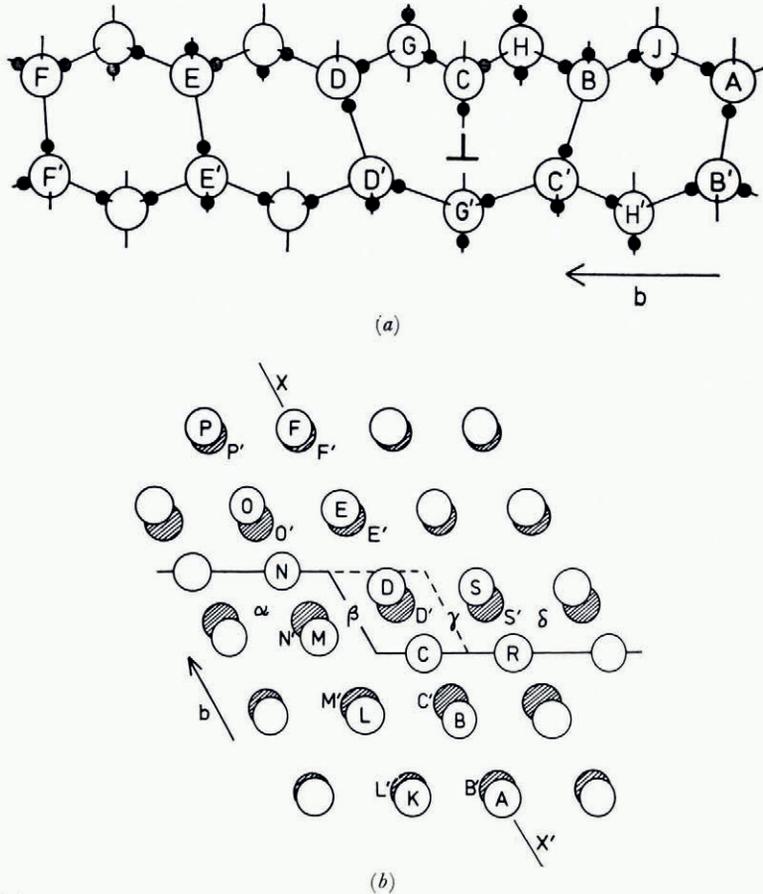


Fig. 5. A 60° dislocation lying on a (0001) plane in ice. Diagram (a) is a section in the $(1\bar{1}00)$ plane which includes the Burgers vector \mathbf{b} . The large circles represent the oxygen atoms, and the dots a possible set of positions of the protons. Diagram (b) is a projection on to the glide plane of a section of the dislocation (solid line) including a kink at β . The plane xx' is that of diagram (a) and the atoms are similarly labelled. Open circles represent molecules lying immediately above the slip plane and shaded circles those immediately below it. (Reproduced by permission from Whitworth and others, 1976.)

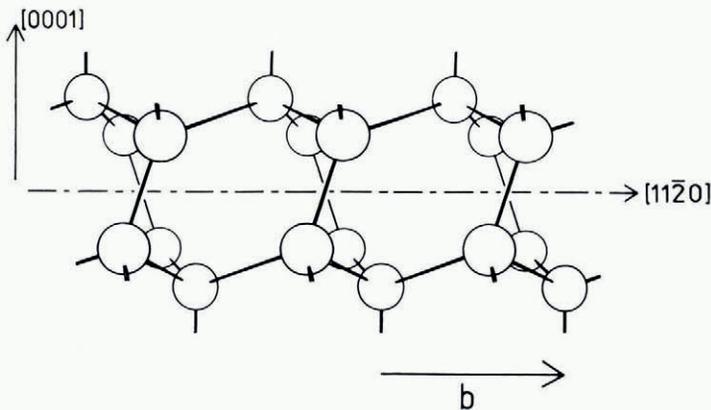


Fig. 6. The arrangement of water molecules around a screw dislocation lying along the $[11\bar{2}0]$ axis indicated by the broken line.

The structures just described have been built up by filling the hole containing the question mark in Figure 1 with molecules linked according to the normal crystalline arrangement as far as is possible considering the constraints imposed by the boundary conditions on the hole. We will refer to this as the *crystalline-core model*.

The distortions in the core will inevitably increase the energy of the system as compared with a perfect lattice. It is possible that the free energy could be lowered by allowing some bonds to break completely with a consequent gain in entropy due to the disorder of the molecular positions. This leads to the idea of filling the hole in Figure 1 with a liquid-like arrangement of molecules. Nye (1973) has discussed this possibility using a macroscopic argument in which the radius of the liquid core is determined by a balance between the elastic energy of the solid and the surface energy of the ice-water interface. His calculation predicts a core radius somewhat less than the Burgers vector. This indicates that a large core of liquid is not a realistic possibility, but the macroscopic model is clearly not applicable to a core of radius of the order of one atomic spacing. We must take into account the possibility that a few molecules close to the core are disordered, and we will refer to such an arrangement as the *non-crystalline-core model*. Little theoretical attention has been given to the properties of such a model. It is unlikely that a non-crystalline core would be stable at very low temperatures, so that if the core is non-crystalline near the melting point, a transition between the two types of core would be expected at some lower temperature.

THEORIES OF DISLOCATION MOBILITY

A dislocation-free crystal is very strong. The introduction of a dislocation enables slip to occur easily. Theories of dislocation mobility have to account for the finite stress which is in fact required to move the dislocation at a given speed. Various processes may be involved simultaneously or consecutively in limiting the motion. The actual stress needed will be a combination of the stresses required for the separate processes, and must be at least as large as the largest of these components. As we will see, the problem in ice is not to account for the stress needed but to explain how the dislocations manage to move as easily as they do.

In many materials where dislocation movement is basically easy, the dislocation velocity is in practice limited by obstacles such as impurities (in solution or as precipitates), intersection with other dislocations, or the trailing of jogs. Ice is a very pure material yet the movement of dislocations is comparatively slow. Moreover the motion of the isolated dislocations studied in X-ray topographs does not appear to be limited by obstacles, and we are led to the conclusion that the processes limiting dislocation movement are properties of the ice itself. Nevertheless we may have to accept that obstacles of one kind or another are preventing the movement of the many dislocations which are observed not to move at low stresses.

We will now consider various limiting processes which may be applicable in ice, and will confine attention at first to those based on the crystalline model of the core.

Kink formation

In a material where there are definite bonds across the slip plane, as is the case in ice, the movement of a dislocation is normally thought of in terms of the formation of kinks and their subsequent movement along the dislocation line as illustrated in Figure 5(b). For very small displacements, as studied in internal friction experiments for example, it may be sufficient merely to move pre-existing kinks, but for large displacements such as we are considering, fresh kinks will inevitably have to be generated. To do this it is normally envisaged that a short segment of a straight dislocation is thermally activated forward into an adjacent stable position so forming a pair of opposite kinks. This will require energy U_k , which may depend on the separation l of the kinks, and an amount of work of order $\tau b^2 l$ will be done by the stress. Under conditions appropriate to ice $\tau b^2 l$ will be much less than kT , and the stress merely

applies a small bias to the random formation of kinks. This model has been applied to germanium, for which the dislocation velocity conforms to the predicted equation:

$$v_d = A\tau^m \exp(-U/kT), \quad (3)$$

with m in the region of 1 to 1.5, and U assumed to be related to U_k (Alexander and Haasen, 1968). The form of this equation is similar to that of the empirical Equation (2) for ice, but this in itself does not justify the model.

Kink movement limited by proton disorder

It has been pointed out by Glen (1968) that the disorder of the protons on the hydrogen bonds presents a special kind of obstacle to the glide of dislocations in ice. If in Figure 5(a) the dislocation were to glide to the right, atom c would have to link to c' and the resulting bond cc' would then have two protons on it. Thus the glide would generate a D-defect which requires energy, but there is not sufficient energy available from the applied stress to do this. In general any attempt to slip one plane over another in ice will generate Bjerrum defects on about half of the bonds intersected. An attempt to avoid the problem by moving the proton from c' to b in Figure 5(a) merely generates an ion-state at b and is of little help. Glen therefore suggested that the dislocation can only move forward when the bonds are correctly oriented, and that the rate of movement is limited by the need to wait for such reorientations to occur. Reorientation occurs by the same movement of electrical point defects as gives rise to dielectric loss (Fletcher, 1970; Hobbs, 1974), the average time interval between reorientations of a given bond τ_b being twice the relaxation time τ_{diel} of the main audio-frequency Debye dispersion.

As already explained the dislocation may be expected to move by the propagation of kinks. The velocity v_k of a kink along a dislocation under an applied stress τ has been calculated by Whitworth and others (1976) and, using a different method of analysis, by Frost and others (1976). An important feature of the problem is that, at the stresses typically used, the work done by the stress in moving the kink through one atomic spacing, $(\sqrt{3}/2)\tau b^3$, is much less than kT . This implies that kinks will move along the dislocation in a random fashion, with the stress merely producing a bias to this motion. The theories further assume that kinks move easily amongst the sites which, for a particular configuration of bonds, are accessible to them. If they do not, the velocity would be limited by kink movement to a value less than that being calculated on the basis of proton disorder. The mathematics can most easily be analysed in the limit $\tau b^3 \ll kT$, and the velocity in lattice constants per second is found to be

$$\frac{v_k}{b} = \alpha \frac{5}{6} \frac{\sqrt{3} b^3}{2\tau_b kT} \tau, \quad (4)$$

where α is a constant between 3 and 6 depending on the orientation of the dislocation and whether or not dangling bonds are subject to reorientation by the defects. The factor of $\frac{5}{6}$ is the correlation correction introduced by Frost and others (1976). Equation (4) predicts a linear dependence of v_k on τ at small τ . Neither version of the theory is restricted to this limit; at high stresses ($>10^7 \text{ N m}^{-2}$) they predict a deviation from linearity with v_k tending to a constant value where maximum advantage is taken of every relevant bond reversal. These models assume throughout that the stress does not directly influence τ_b , and the limit is a consequence of this assumption.

Perez and others (1975) have also attempted to calculate the velocity of kinks as limited by proton disorder. In their calculation the stress is introduced through a factor in the kink jump rate of $\exp[-(E_M \pm \frac{1}{2}\tau b^3)/kT]$, where E_M is the activation energy of movement of a Bjerrum defect. This implies that the kink moves because the stress modifies the rate of reorientation of critical bonds. This is contrary to the basic concept of the model that the

bonds reorient in a random fashion and that the only effect of the stress is to influence the movement of kinks within the sites available to them. Their equation for v_k can be expressed in a form similar to Equation (4), from which it differs only by a numerical factor of about 4.

The only significant parameter that determines v_k in Equation (4) is τ_b , which, on the usual interpretation of the properties of ice, can be simply deduced from experiments on dielectric relaxation. At -18°C , $\tau_b = 2\tau_{\text{die1}} = 2.36 \times 10^{-4}$ s (see Hobbs, 1974). For $\tau = 10^5 \text{ N m}^{-2}$ and a 60° dislocation with reorientation by Bjerrum defects, this gives $v_k/b = 48 \text{ s}^{-1}$. The velocity of a dislocation v_d involves the number of kinks n_k per unit length, and is given by

$$v_d/b = (\sqrt{3}/2)n_kv_k. \quad (5)$$

Therefore, at the above temperature and stress, $v_d/b = 42n_kb \text{ s}^{-1}$. For the kink model to be meaningful there must be much less than one kink per atomic plane along the dislocation, and thus $n_kb \ll 1$. The model therefore predicts $v_d/b \ll 42 \text{ s}^{-1}$, which is much smaller than the value of order 500 s^{-1} observed experimentally.

A complete theory based on kink movement should include the process of kink nucleation outlined in the previous section and should incorporate the fact that the bonds must be correctly oriented throughout the length of the segment of dislocation thrown forward to generate a pair of kinks. The theory should also take account of the possibility that kinks may not move between the sites available to them as easily as is assumed in the models of Whitworth and others (1976) and Frost and others (1976). Such considerations can only lead to a reduction of v_d below the upper limit deduced from Equations (4) and (5) with $n_kb = 1$. It is clear that models based on kink movement with τ_b determined by τ_{die1} cannot account for dislocation movement at the rates in fact observed.

Perez and others (1975) have presented a theory in which they combine their model of kink migration with a calculation of n_k , but their calculation involves an unconvincing mixture of equations related to rate processes and those applicable to conditions of thermodynamic equilibrium. In particular the factor 2^{-n} , which arises from the need to match correctly the n bonds along a segment thrown forward to nucleate a pair of kinks, has been included in the thermal equilibrium equation for the kink concentration. As already explained the premises on which such models are based cannot lead to results in agreement with experiment. Contrary to what is implied in their paper the models of Perez and others (1975) do not satisfactorily account for the results shown in Figure 3.

Movement of flexible dislocations limited by proton disorder

Faced with the problem that models involving kink migration with bond reorientation at a rate determined by τ_{die1} cannot account for the observed dislocation velocities, Whitworth and others (1976) developed a model at the opposite extreme. This is the flexible dislocation model in which the kink energy and line tension are taken to be so small that the dislocation can move easily over all positions permitted by the orientations of the bonds across its slip plane. The model is illustrated in Figure 7 in which the solid line represents a 60° dislocation with its slip plane in the plane of the paper. The circles with dots represent dangling bonds on the dislocation and the other circles represent bonds cutting the slip plane. Bonds that are oriented to match the corresponding dangling bond are shown as open circles, and bonds that are mismatched are shown as solid circles. The dislocation is free to move over the area bounded by the broken lines, and it is assumed that it will indeed move over the sites within this area on a time scale short compared with τ_b . If bond F is reversed the segment can move forward into new positions, and if bond B is reversed while the dislocation is in front of it the dislocation will be trapped in a forward position. The stresses we are considering do not push the dislocation hard up against the mismatched bonds, but they bias the randomly

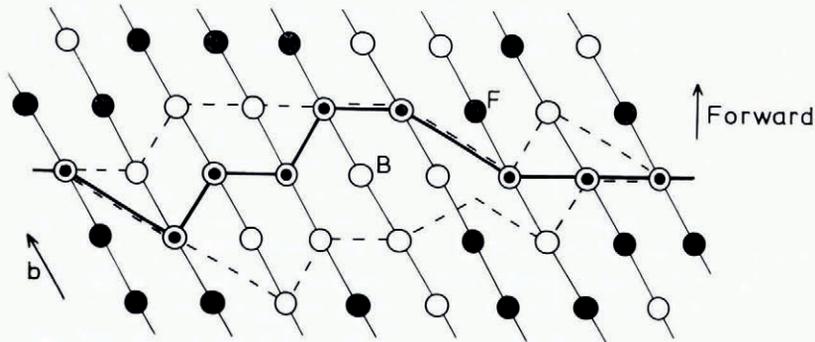


Fig. 7. Diagram illustrating movement of nominally 60° flexible dislocation on (0001) glide plane. The circles represent bonds crossing the slip plane, and to simplify the diagram the distortions apparent in Figure 5(b) have been eliminated here. The dislocation is in the position shown by the solid line and circles with dots represent dangling bonds. Bonds shown by open circles are matched to the corresponding dangling bond; bonds shown by solid circles are mismatched. For the arrangement of protons shown, the dislocation is free to move over positions within the area bounded by the broken lines.

moving segment towards the forward part of its range. In the presence of a stress the dislocation will gradually migrate forward amongst the array of continually reversing bonds.

The theory of this model is more difficult than that of the relatively precisely defined kink model. A range of approximations and assumptions have to be made, leading eventually to the dislocation velocity

$$\frac{v_d}{b} = 1.18 \frac{\sqrt{3}b^3}{2\tau_b kT} \tau, \quad (6)$$

for a 60° dislocation with reorientation of bonds by Bjerrum defects. For a temperature of -18°C , at which $\tau_b = 2.36 \times 10^{-4}$ s, and for $\tau = 10^5$ N m $^{-2}$ this equation predicts $v_d/b = 11$ s $^{-1}$. This is considerably less than the experimental value of 500 s $^{-1}$. Even if maximum advantage is taken of the approximations and uncertainties in the model the theory still predicts a velocity which is an order of magnitude too small.

The corresponding calculation is very difficult to formulate for a screw dislocation, but the physical processes are essentially the same and it is hard to envisage any way in which screw dislocations could move appreciably faster than 60° dislocations.

Anelastic-loss model

Ice exhibits anelastic relaxation which arises because a stress can produce a slight ordering of the orientation of the water molecules. As a dislocation moves, every nearby part of the lattice will be subjected to a stress cycle which will result in a dissipation of energy and therefore in a force retarding the movement of the dislocation. Weertman (1963, 1973) has drawn attention to the importance of this effect and proposed it as the process which ultimately limits dislocation mobility in ice.

The theory has been developed by Eshelby (1961) for a screw dislocation of width ζ moving in an isotropic anelastic continuum for which the relaxation time is τ_r and the logarithmic decrement at the loss maximum is δ_{\max} . In the limit $v_d \ll 2\zeta/\tau_r$, the force on the dislocation is a linear function of the velocity. Assuming this to be the only process limiting the movement,

$$\frac{v_d}{b} = \frac{4\pi^2}{G\tau_r\delta_{\max}} \left(\frac{\zeta}{b}\right)^2 \tau, \quad (7)$$

where G is the shear modulus. This theory inevitably involves some approximation concerning the region close to the axis of the dislocation. However, the process is essentially one arising from the behaviour of material outside the core, and the details of these approximations and of the core structure are therefore not of fundamental importance.

Experiments on internal friction in pure ice by Schiller (1958) and Kuroiwa (1964) show a clear Debye peak with a relaxation time close to that observed for dielectric relaxation. This has therefore been attributed to the anelastic loss due to stress-induced ordering. Onsager and Runnels (1969) have shown theoretically that the mechanical relaxation time $\tau_r = \frac{2}{3}\tau_{\text{dieI}}$, and using the already quoted values of τ_{dieI} we will therefore take $\tau_r = 8 \times 10^{-5}$ s at -18°C . The strength of the relaxation depends on the orientation of the stress within the crystal, but a typical mean value for the peak logarithmic decrement δ_{max} would be 0.025. G is also anisotropic with a mean value of about 3.5×10^9 N m $^{-2}$. Taking $\zeta = b$, and substituting the above values and a stress $\tau = 10^5$ N m $^{-2}$ into Equation (7) gives $v_d/b \approx 560$ s $^{-1}$. This is close to the experimental value.

The dominant contribution to the temperature dependence of v_d will be due to τ_r , and the model therefore shares with the Glen model the feature that v_d will vary with temperature in the same way as τ_{dieI}^{-1} . The activation energy for dielectric relaxation in "pure" ice lies in the range 0.57 to 0.64 eV (Hobbs, 1974; Camplin and Glen, 1973), which is consistent with the value found by Maï (1976) for dislocation movement. The limited evidence available shows that both v_d and τ_{dieI}^{-1} are reduced by doping with HF, but the effect of doping on v_d is much less than that on τ_{dieI} (paper in preparation by C. Maï, J. Perez, J. Tatibouët, and R. Vassoille, quoted by Perez and others, 1978, p. 371).

Equation (7) was derived in the limit $v_d \ll 2\zeta/\tau_r$, which implies that the stress cycle imposed on the lattice by a moving dislocation is everywhere slow compared with τ_r . Assuming Equation (7) to be valid and $\zeta = b$, this condition is equivalent to

$$\tau \ll \frac{G\delta_{\text{max}}}{2\pi^2},$$

the right-hand side being approximately 4×10^6 N m $^{-2}$ and independent of temperature. Examination of Eshelby's theory shows that a linear relation between v_d and τ may be expected up to about one-fifth of this limit, and all the reliable measurements in Figures 2 and 3 lie in this linear range. The theory is consistent with the behaviour observed at lower temperatures. Although the deviations from linearity predicted are in the same sense as those found by Maï above -10°C (Fig. 2), the simple model cannot account for their occurrence at such low stresses, or at values of the stress that depend on temperature.

The theory further predicts that the stress passes through a maximum value of about 1.4×10^6 N m $^{-2}$ at $v_d \approx 2.6\zeta/\tau_r$. This arises because dislocations moving much faster than this produce a stress fluctuation in the lattice lasting for a time short compared with τ_r , and this cannot produce so great an ordering of the protons as longer pulses. It is worth looking experimentally for evidence of dislocations moving exceptionally easily under very high stresses ($> 2 \times 10^6$ N m $^{-2}$).

DISCUSSION OF CRYSTALLINE-CORE MODEL

We have seen that if we adopt a crystalline model of the core the disorder of the protons appears to present a serious obstacle to dislocation motion. Even in the extreme case of a fully flexible dislocation, the stress required to move it is 10 to 100 times larger than that found experimentally, and on a crystalline-core model we would not expect to be able to ignore line tension and kink energies so readily. It is therefore necessary to consider carefully what possibilities there may be of overcoming this difficulty.

The theories involve the mean time τ_b between reorientations either of the dangling bond or of adjacent bonds across the slip plane such as bc' in Figure 5(a). The process of reorientation is the same as that which gives rise to dielectric relaxation, and in the numerical calculations τ_b has been taken as $2\tau_{die1}$. The value of τ_{die1} is well-established from experiments on dielectric relaxation and is consistent with results on anelastic relaxation and n.m.r. Itagaki (1978) has however reported that the main Debye relaxation does not occur in dislocation-free ice; if substantiated this throws into doubt the whole of the normally accepted model of the electrical properties of ice, and consequently the argument to be developed here. Our justification for using in Equation (4) or (6) a value of τ_b appropriate to bonds far removed from dislocations is that even near the core it is not possible to reorient a single bond in isolation without generating electrical point defects. The orientation of every bond is uniquely determined by the orientations of all the neighbouring bonds, and a bond can only be reversed as part of a chain of reversals along the path of a Bjerrum defect or ion. Dangling bonds are only reversed when a defect is created or destroyed at the site. The rate at which a given bond is visited by a reorienting defect is therefore determined by the rate at which such defects move about in the crystal as a whole. This argument breaks down if defects are attracted towards dislocations so that their concentration on nearby sites is greater than the thermal equilibrium value elsewhere, and this possibility was pointed out when the model was proposed by Glen (1968).

One source of attraction is that dislocations may become charged, as has already been mentioned, due to too many or too few of the dangling bonds having protons on them. A charged dislocation will be surrounded by a Debye-Hückel screening cloud of defects of the opposite sign, and it is these screening defects which could speed up bond reorientation near the core. Whitworth and others (1976) proposed this as a mechanism which might just possibly bring the predictions of the flexible dislocation model into agreement with experiment, but further quantitative analysis shows that this is not so. The potential difference between sites near the core and distant parts of the crystal can be calculated by simple modification of theories developed for ionic crystals (Whitworth, 1968, 1972). As the screening is influenced by the polarization of the lattice itself, the theory involves the permittivity of the material. For ice the static permittivity is appropriate because the dislocations are stationary over times much longer than τ_{die1} , and this permittivity is exceptionally large. In consequence, whatever other adjustable parameters are fed into the calculation, it is impossible to generate a potential near the core of more than about 0.02 V. This would at most double the concentration of defects and halve τ_b . (This does not of course prevent the dislocations from being charged and exhibiting other phenomena related directly to their charge rather than to the concentrations of defects in the surrounding charge cloud.)

Another possible cause of attraction of Bjerrum defects or ions to dislocations is their elastic interaction with the surrounding strain fields. The effects of this interaction on the numbers of defects passing along bonds close to the core is very difficult to analyse because the interaction is anisotropic and defects may follow paths involving a sequence of favourably and unfavourably oriented bonds. The order of magnitude of the interaction energy of a suitably oriented defect at a site a distance r from a screw dislocation is

$$E \approx \frac{\epsilon G b^4}{2\pi r}, \quad (8)$$

where ϵ is the tetragonal distortion of the defect. There may also be a similar interaction arising from the dilation around the defect in a region of hydrostatic stress. With ϵ taken rather arbitrarily as 0.1 and $r = b$, Equation (8) gives $E \approx 0.03$ eV. The interaction energy would have to be two to three times larger than this to bring even the flexible dislocation model into agreement with experiment. The interaction merits further analysis, but it seems unlikely that it will solve the problem of how dislocations move so easily.

Ice will contain vacancies, possibly at concentrations as high as 1 in 10^6 (Mogensen and Eldrup, 1978). If the site c' in Figure 5(a) were vacant there would be no particular difficulty in moving the dislocation one step to the right. The vacancy would then become "invested" with a D-defect (i.e. there are three protons pointing into it (see Glen, 1974)). It would still be mobile but would continue to move as an invested vacancy until an appropriate Bjerrum defect was created or destroyed at the site. Vacancies could in principle speed up the movement of kinks by allowing a violation of the Bernal-Fowler rules to be stored and released by the appropriate electrical defect at a later time. However, to be of any use in this process the vacancy must actually be present at the instant that the kink passes the mismatched bond concerned.

Let the fractional concentration of vacancies be f_v and let the average time for which a vacancy remains on a particular site be τ_v . If $\tau_v > \tau_b$ then a fraction f_v of mismatched bonds will be associated with vacancies and so be ineffective barriers to kinks; as f_v is small this is clearly unimportant. It is more probable that $\tau_v \ll \tau_b$, in which case, during the interval τ_b for which a kink is confined to a segment of dislocation terminated by a particular mismatched bond, that bond will have a vacancy at one end for a fraction f_v of the time. If the kink is to get past, it must do so during this short interval of time. If this is to happen often the random movement of the kink within the segment available to it must occur on a time scale less than τ_v and consequently very much less than τ_b . This seems improbable, but the possible role of vacancies in determining dislocation mobility is a problem that deserves further study.

Throughout the discussion it has been assumed that dislocations glide on the planes ss' or tt' of Figure 4. Glide on nn' involves the breaking and reforming of more bonds. The molecules at the core are free to rotate about the c -axis, but this does not allow them sufficient freedom to reorient in a way that avoids the need for correct matching of bonds prior to glide.

The conclusion of this discussion is that no mechanism has yet been devised which will allow a dislocation with a crystalline core to move through the disorder of the protons at the stresses observed experimentally. The Bernal-Fowler rules impose constraints on the disorder which it is very difficult to avoid by processes involving purely local changes. As soon as we start to consider processes involving the simultaneous rearrangement of several molecules we leave the territory of the crystalline-core model and introduce features characteristic of a non-crystalline core.

DISCUSSION OF NON-CRYSTALLINE-CORE MODEL

If a crystal of ice is sheared on the basal plane ss' in Figure 4 about 50% of the bonds will subsequently fail to match up correctly. However, if shear occurs between planes ss' and tt' and the two layers of molecules between these planes are then reorganized, it is possible to produce matching of most of the bonds, though there will be a very few cases where matching cannot be achieved without reorientation of bonds outside these layers. In terms of dislocation movement this means that if there is sufficient freedom of rearrangement of molecules near the core the difficulty over matching bonds can be largely overcome. Such co-operative rearrangements are impossible on the crystalline-core model, but become simple if the core is disordered and the molecules within it are sufficiently mobile. The important feature is not the disorder itself but the assumption that this disorder allows molecules to move more freely than is possible in crystalline ice where the Bernal-Fowler rules can only be violated at well-defined point defects. A rigid glassy structure as in "amorphous ice" would be of no value here. The core needs to have characteristics approaching those of a fluid, though this does not imply that its properties closely resemble normal liquid water.

An important question concerns the minimum size of such a core which will allow adequate rearrangement. Consideration of simple examples shows that it is insufficient to have a core consisting only of rows c , g , and h in Figure 5(a); the constraints imposed by the bonds on the

boundary of the core do not allow molecules to be fitted back as the dislocation moves on. A molecule such as c' cannot usefully be included in the core without G' or H' because the orientation in which it fits back is entirely determined by the bonds below it. Arguments of this kind lead to a probable minimum core involving the eight rows of molecules shown missing in Figure 8. The core could be larger than this, but if much larger would tend to violate the already explained conclusion of Nye that a large core of macroscopic liquid water would be energetically unfavourable.

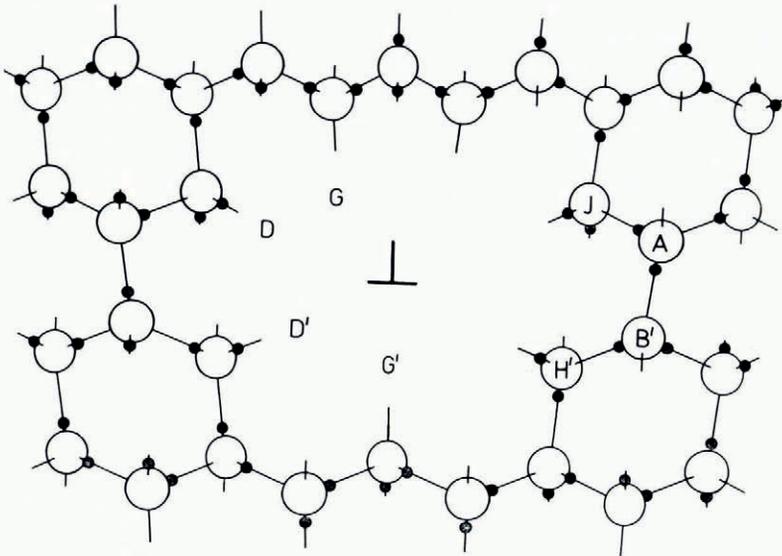


Fig. 8. Diagram similar to Figure 5(a) showing arrangement in $(1\bar{1}00)$ plane of molecules around a 60° dislocation with a non-crystalline core incorporating 8 lattice sites. The disordered molecules in the core are not shown.

The movement of a non-crystalline core avoids all difficulties concerning kink movement and kink nucleation. As the dislocation in Figure 8 moves to the right, the bonds ahead of it will become increasingly distorted and the four rows of molecules J, A, B', and H' will break away from the regular lattice into the core. This process does not involve a change in free energy, and we should think in terms of a dynamic equilibrium with molecules continually moving back and forth from the crystalline wall to the disordered core. Four rows of molecules must then re-attach themselves to the boundary at sites G, D, D', and G'; it is at this point that the question of bond matching arises. In general this will not present much problem, but situations can arise where proper matching is impossible. It will then be necessary to wait for an electrical point defect to produce a suitable reorientation somewhere on the boundary, but this will not delay the dislocation motion by anything like as much as the need for reorientations in the crystalline-core model. Nye (1973) has likened this process to Tyndall's theory of the flow of ice by melting and regelation.

No calculation has yet been published of the stress required to move a non-crystalline dislocation core through the lattice, but an attempt at this difficult problem is to be presented at this conference (Perez and others, 1978). In addition to this effect all dislocations must also experience the drag arising from the anelastic loss which occurs in regions of the lattice outside the core. As already explained this model predicts a stress for dislocation movement of the same order as that found experimentally, and the effect could therefore be the limiting process as proposed by Weertman (1973). The mechanism predicts a dependence on temperature and

HF-doping similar to that of τ_{die1} . The deviation from a linear relation between stress and v_d is expected to be in the direction found experimentally (Fig. 2), but the theory does not account for its occurring at such low stress. The anelastic-loss model has to be combined with the as yet unknown stress required to move the core. If the drag on the core is small, anelastic loss becomes the limiting process, and with refinements could well account for the observations on pure ice. If however the anelastic loss is reduced by doping with HF, then the drag on the core could eventually become rate limiting. This may account for the already mentioned observation that HF doping does not increase v_d by as much as it reduces τ_{die1} .

The non-crystalline-core model can apply to screw as well as 60° dislocations. The core would be of similar size and would include at least all the molecules shown in Figure 6.

A dislocation may be electrically charged even if it has a non-crystalline core and no obvious dangling bonds. It is merely necessary that there is an imbalance between the numbers of bonds with and without protons pointing into the core region. A screw dislocation may also be charged in this way, although this would not have been possible if it had had a crystalline core and no dangling bonds. As a dislocation glides it will carry its charge with it, the imbalance in numbers of bonds pointing into the core being perpetuated in the layers that recrystallize behind the dislocation.

The core may be a source or sink of vacancies or interstitials. There is no necessity that in thermal equilibrium the density of molecules in the core should correspond exactly to the number of missing lattice sites. Similarly the core could trap impurity molecules and carry them along with the dislocation. Such modifications to the contents of the core may influence the mobility of the dislocation.

A non-crystalline core to a dislocation is not a feature commonly encountered in the theory of other materials. It is interesting to speculate on whether it arises in ice primarily because interest is concentrated on temperatures comparatively near to the melting point. One would expect on thermodynamic grounds that the core would become crystalline at a sufficiently low temperature and that this would lead to an important change in mechanical properties.

CONCLUSIONS

This review has moved towards the conclusion that dislocations in ice have a small non-crystalline core within which molecules are free to reorient easily. The anelastic loss due to the stress-induced ordering of protons in the surrounding lattice plays an important, and in pure ice a probably dominant, role in limiting the dislocation mobility. The rate-determining process is the reorientation of bonds, which explains why the dependence of dislocation mobility on temperature and HF-doping is related to that for dielectric relaxation.

These are dangerous conclusions! It is most unsatisfactory to adopt a model by a process of elimination, and to believe in it only because the calculations required to disprove it have not yet been carried out. We know enough about the Glen model to know that it cannot apply under the range of conditions so far studied, but we have not yet eliminated the possible role played by vacancies. We do not know enough about the non-crystalline core to be reasonably satisfied that it can allow dislocation movement at the rates observed.

Further theoretical work in this field should give priority to the following:

1. An analysis of the nature, size, and dynamics of a non-crystalline core. This is a very difficult model to formulate.
2. The application of the theory of stress-induced ordering to the movement of dislocations for the particular case of the structure of ice. The assumption of a non-crystalline core should greatly ease the difficulties arising in this model in the non-linear region near the axis.
3. A study of the effect of vacancies on the Glen model.

Experiments could usefully look for the following things:

1. The correlation between measurements of v_d and τ_{die} on identical specimens over a wider range of temperature and doping.
2. The possibility that dislocations can be made to move easily at velocities above those corresponding to the peak of the anelastic loss (i.e. $v_d/b > 2/\tau_r$).
3. Is there a change in mechanical properties at a temperature at which the core becomes crystalline?

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DISCUSSION

J. PEREZ: Among the different possibilities of structure of dislocation cores you did not mention the dissociation effect; what do you think about it and about its consequence on the anelastic-loss model?

R. W. WHITWORTH: A dislocation can dissociate if it glides on the plane NN' in Figure 4. This is not the usually assumed glide plane, but, as indicated in the written paper, by analogy with recent results on Ge and Si it may be relevant to consider it. The partial dislocations would still be subject to the limitations of the Glen and anelastic-loss models, so we cannot overcome the difficulty of moving dislocations in this way.

T. HONDOH: Generally, it is difficult to move pre-existing dislocations but it is easy to move fresh dislocations. How do you explain this difference?

WHITWORTH: There are many ways of pinning a dislocation, for example by impurities, jogs, or other dislocations. Presumably fresh dislocations are less hampered by such obstacles than ones which have been aged in the crystal. There is no difficulty in slowing dislocations down. Our problem is to explain how a free dislocation can move as fast as is observed.

HONDOH: I think there are two different approaches to interpreting mobility of dislocations, the one is that made by you and others, and another is to solve the equation of motion of a dislocation (of course, in a continuum body) supposing some distributions of obstacles, such as the theory by Dr Ninomiya in Tokyo University. Do you have any idea how to combine these two different approaches?

WHITWORTH: The two approaches are not alternatives. They must both be taken into consideration and the actual stress needed to move the dislocation will be a combination of the stresses for the separate processes. This will not be an average but some kind of summation, and we can only decide how to do the summation when the models have been formulated in more detail.

A. HIGASHI: With respect to the amorphous or "liquid" structure of the core, have you considered the thermodynamic processes associated with its movement?

WHITWORTH: No, we have not developed a quantitative model, but Dr J. Perez will report some work of this kind in his paper later in this symposium.