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### The role of disclinations in two phase changes induced by temperature and pressure in crystalline solids: melting and the brittle-ductile transition

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*Letter*

**THE ROLE OF DISCLINATIONS IN TWO  
PHASE CHANGES INDUCED BY  
TEMPERATURE AND PRESSURE IN  
CRYSTALLINE SOLIDS: MELTING AND THE  
BRITTLE–DUCTILE TRANSITION**

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It has long been known that the melting temperature  $T_m$  of close-packed metals correlates well with the mono-vacancy formation energy. However, with the possible exception of the face-centered-cubic metals, there is a prior phase transition from a mechanically brittle solid phase to a ductile phase. Here the likely role of disclinations in the brittle-ductile phase change is stressed. The present picture may help to understand the brittle–ductile transition not only in crystalline materials but also in amorphous phases. The structure of such phases can probably be characterized in terms of a disordered disclination network. As examples of elemental crystalline solids, Si and graphite are finally discussed, with the melting under pressure of graphite being quantified.

*Keywords:* Disclinations; Brittle-ductile transition; Melting

## 1. INTRODUCTION

Dislocation theories of melting have a long history, and reference may be made to the early work of Kuhlmann-Wilsdorf [1], to computer simulation studies of Cotterill [2], and to the very recent investigation of Kleinert and Jiang [3].

While a dislocation is associated with a translation given by its Burgers vector [4], the less familiar defect—a disclination—involves a rotation operation. Such a defect was introduced by Volterra at the start of the 20th century in his description of a continuous solid medium, and is sometimes termed a rotation-dislocation [5]. Such a defect can be

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generated by cutting the structure along a line and adding or removing a section of solid between the two lips of the cut. It is significant to note here that in two dimensions this defected region is point-like (i.e., quasi-0-dimensional), whereas in three dimensions it is linear (i.e. topologically quasi-1-dimensional).

Turning to the higher temperature phase transition—melting—of the two that form the focal points of the present study, it has long been known to metallurgists that close-packed metals are such that the thermal energy  $k_B T_m$ , with  $k_B$  being Boltzmann's constant, associated with melting temperature  $T_m$ , correlates strongly with the mono-vacancy formation energy  $E_V$ , for example as in Ref. [6]. In turn, 'glue' models of interatomic force fields exemplified by the work of Johnson [7,8], predict that  $E_V$  correlates with shear modulus  $G$  through the relation

$$E_V = \text{constant} \times G\Omega, \quad (1)$$

where  $\Omega$  is the atomic volume. Thus, invoking the correlation already referred to between  $k_B T_m$  and  $E_V$  we can write, of course again approximately, that

$$k_B T_m = \text{constant} \times G\Omega. \quad (2)$$

To our knowledge, Eq. (2), while useful at equilibrium for close-packed metals, has not been subjected to stringent test as a function of hydrostatic pressure  $p$ ; this will be a crucial variable in what follows on the proposed relation between  $T_m(p)$  and the critical temperature say  $T_C(p)$ , of the brittle–ductile transition. In the fairly recent study of Khantha *et al.* [9] a model for the brittle-to-ductile transition (BDT) based on the Kosterlitz–Thouless [10] concept of dislocation screening has been proposed. The sharp BDTs in nearly dislocation free crystals such as Si, and the sudden bursts of plastic deformation in metal whiskers have been attributed by these authors [9] to the operation of this mechanism. This interpretation of the BDT in precracked silicon has subsequently been criticized by other authors [11–13]. Such studies have led us to propose a new point of view in which the “rotation-dislocation” or “disclination” referred to above is the crucial element in the BDT.

## 2. PROPOSAL TO USE DISCLINATIONS AS THE DEFECT TO CHARACTERIZE BDT

In parallel with the quite recent investigation of Langer and Pechenik [14], who do not, however, refer to disclinations at all, we note first that conventional (translational) dislocations cannot be the universal ingredient associated with the BDT. The reason for this is that the BDT occurs not only in crystalline solids but also in amorphous phases. Since the latter materials are basically characterized by the lack of long-range translational order, conventional dislocations do not offer a useful mode of description of such glassy materials. However, as Sadoc and Mosseri [5] write on p. 174 of their book in relation to an amorphous state, “Such a structure can probably be described in terms of a disordered disclination network.”

Thus the main idea on which the present study focuses is that the “universal ingredient” associated with a BDT is an aggregate of disclinations. All attention

below will therefore be directed towards an attempt, albeit primitive, to classify what characteristic energies associated with such a disclination aggregate can be expected to enter the energy  $k_B T_C$  associated with the BDT temperature  $T_C$  in crystals.

We would like to point out that our picture can be posed in terms of dislocations first developing below  $T_C$  and above  $T_C$  dissociating to disclinations. To this end we consider two-dimensional graphite (for which a comprehensive characterization of different possible disclinations has recently been made [15,16]). Initially dislocations can form via a local transformation from the undefected lattice as pairs with opposite Burgers vectors, as in Figs. 1(a) and (b) for the example of (two-dimensional) graphite. Then they can move apart as the result of further local transformations, as in Figs. 1(c) and (d). Each dislocation can be viewed as a positive-curvature disclination (say as

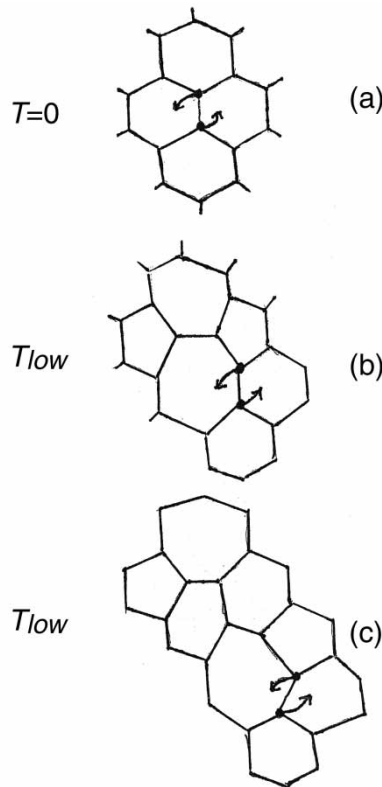


FIGURE 1 Schematic representation of dislocations and disclinations in crystalline graphite. Here (a) and (b) illustrate formation via a local transformation from the perfect lattice of dislocation pairs with opposite Burgers vectors. Next (c) and (d) illustrate movement apart of the dislocation pair formed in (b) all as a result of further local transformations. Each dislocation can be regarded to be comprised from a (pentagonal) positive-curvature disclination and a (heptagonal) negative-curvature disclination. If the brittle-to-ductile transition exists in crystalline graphite, it is proposed that such disclination pairs as in (e) dissociate into well separated disclinations of opposed curvature, and net Burgers vector as indicated there. This process is not just a simple unadorned separation of the opposed disclinations since such a separated pair has a different (larger) Burgers vector as indicated in (f). The separation of disclination pairs then entails the generation of additional dislocations, such as shown in (g), where the Burgers vector of the well separated disclination pair when added to the Burgers vectors for the two additional dislocations (consisting of neighbouring pentagonal and heptagonal rings) gives a net Burgers vector matching that in (e). Hence (g) is obtainable from (e) via local transformations, and separation of opposed-curvature disclination pairs generates additional dislocations.

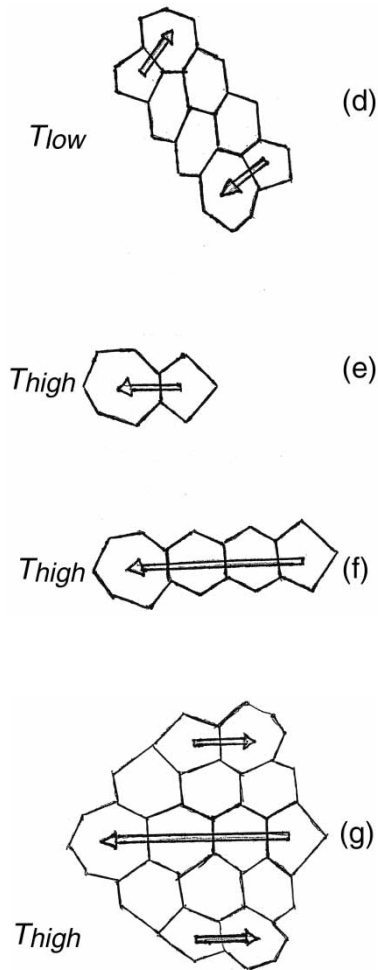


FIGURE 1 Continued.

formed by a pentagonal ring in graphite) and a negative-curvature dislocation (say as formed by a heptagonal ring), so that the overall curvature of the dislocation is 0. At  $T_C$  then such disclination pairs are imagined to dissociate into well separated disclinations of opposite curvature but notably this process necessarily entails the generation of additional dislocations. For example, in Fig. 1(e) a dislocation with the pentagonal and heptagonal rings immediately adjacent is shown, while in 1(f) a dislocation with the pentagonal and heptagonal rings more well separated is depicted, and it is noted that this pair has a different overall Burgers vector, so that this latter dislocation (of (e)) cannot (in isolation) result from the earlier dislocation (of (f)) through local transformations. However, Fig. 1(g) shows the disclination pair with two additional dislocations whose Burgers vectors together combine with that of the separated disclination pair, so as to give a net Burgers vector the same as that for the unseparated pair, as in Fig. 1(e).

Such constructions can also be made for silicon, as shown in Fig. 2, the details being given in the caption. It is seen that again dissociation of a dislocation into (oppositely

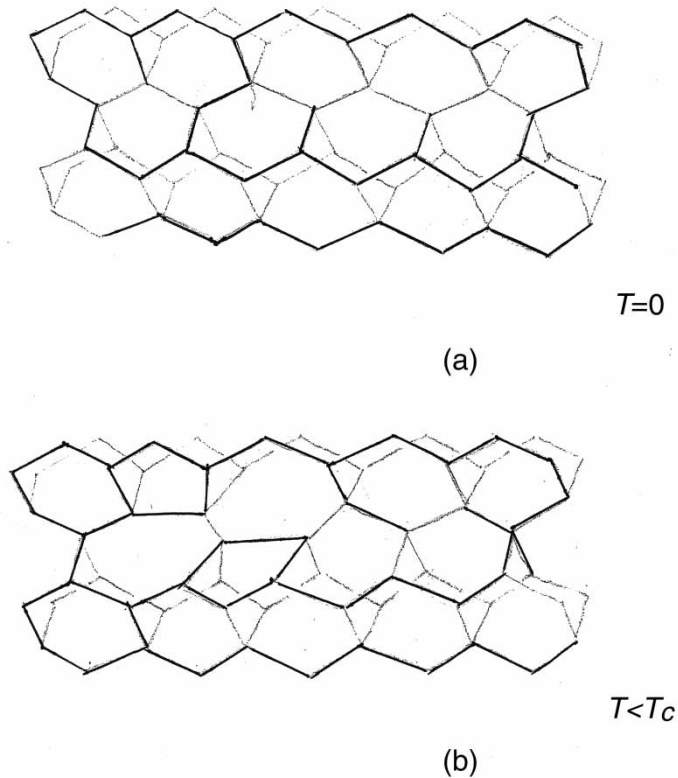


FIGURE 2\* A parallel schematic representation of dislocations and disclinations in crystalline silicon. Here (a) and (b) illustrate formation via a local transformation from the perfect lattice of dislocation pairs with opposite Burgers vectors. Next (c) and (d) illustrate movement apart of the dislocation pair formed in (b) all as a result of further local transformations. In (e) a lone dislocation consisting of pentagonal and heptagonal positive- and negative-curvature disclinations is shown, while in (f) there is shown a dislocation with unadorned positive- and negative-curvature disclinations more well separated and with a consequent different Burgers vector. In (g) a similarly well separated positive- and negative-curvature disclination pair occurs now adorned with two dislocations (of neighboring pentagon-heptagon pairs) such that the overall Burgers vector is the same as in (e). Again separation of opposed-curvature disclination pairs generates additional dislocations.

curved) disclinations generates a number of simple dislocations proportional to the separation between the disclinations. For crystalline Si the existence of the BDT is well established experimentally [9].

To conclude this letter we want to make a number of comments, the first of which is to emphasize a possible analogy to an inter-relationship between  $T_C$  and  $T_m$ . This analogy concerns the plastic phase of solid nitrogen, which then melts at  $T_m$ . Here the plastic phase is characterized by orientational disorder of the  $N_2$  molecules on the sites of a crystal lattice. Characterizing the possible orientations by a single activation energy  $U$  say, Tozzini *et al.* [17] have related the melting temperature  $T_m(p)$  of

\*Because of space limitations, only Figs. 2 (a) and (b) are reproduced. The reader requiring Figs. 2(c)–(g) is welcome to contact DJK.

solid nitrogen at applied hydrostatic pressure  $p$  to the phase boundary ( $\equiv T_C(p)$ ) of the plastic phase again as a function of  $p$ . We feel that in crystalline Si, using an analogous theory with an activation energy related to disclination networks, a similar inter-relationship should exist between  $T_m(p)$  and critical temperature  $T_C(p)$  for the BDT. Further work, both experiment and theory, on crystalline Si under pressure, would seem fruitful in clarifying the concepts put forward in this letter.

The second comment concerns graphite again. The phase diagram of  $C$  at high  $p$  and  $T$  has been considered by us [18–20] and also in relation to the measurement by Togaya [21] of the melting line  $T_m(p)$  of graphite. He discovered that  $T_m$  increased from 4650 K at 1.4 GPa to a maximum of 4790 at 5.6 GPa and subsequently decreased to 4640 K at 9.4 GPa: i.e.,  $T_m$  is practically independent of  $p$ . In the disclination picture proposed here, we conclude that linear disclination networks scarcely are changed in their interaction energies in the above range of pressures. If we use the ‘rule of thumb’ mentioned by Khantha *et al.* [9] that  $T_C \sim T_m/2$  at atmospheric pressure, there should be a BDT in graphite at  $\sim 2200$  K. Its pressure dependence, if indeed such a BDT exists, should then be investigated experimentally, as it would help either to confirm, or to indicate possible extension of, the concepts proposed here.

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