CHAPTER II

A BRIEF REVIEW OF CRYSTAL GROWTH

The developments in the field of crystal growth have been mainly due to the high demand for the large and perfect single crystals in the various branches of Solid State Physics. During the last three decades a considerable progress has been made towards the understanding of the basic parameters that influence the perfection of the crystals. This chapter gives a brief review of the recent developments in the field of crystal growth. Though mention is made of the growth from the vapour phase and of the observations of spirals on metals in support of Frank's theory, stress is laid mainly on the growth of the crystals from melt. Wherever necessary the results on non-metals are also presented for the sake of continuity of the discussion.

The theory of the conventional crystal growth starts from the observations of the growth of the low index crystallographic planes of a crystal. As a matter of fact, due to the dense atomic packing of the low index planes, they exhibit a much lower surface energy than the other planes. Therefore, at absolute zero of temperature, the crystal tends to be bounded by low index planes which are as smooth as possible so as to represent the state of lowest energy. The atoms arrive at the surface continuously and some of the atoms diffuse out from the interior of the bulk of the material, creating vacancies as the temperature is raised above absolute zero. Due to this a large number of atoms will be arriving and leaving the surface of the crystal continuously and it exhibits all kinds of planes. Now, if the vapour pressure is increased above the equilibrium vapour pressure at that temperature, on an average, more atoms will arrive at the surface than leave the surface and hence the crystal grows. However, the addition of a new atom to the surface where there are no kinks or steps is a very unfavourable process because of the energy considerations. Only an atom which can reach a kink by migration over the surface before it is evaporated, does have a chance to become permanently attached to the surface. When all the kinks are filled by such a process, the growing surface becomes smooth. Further growth can proceed only by the accidental formation of an island providing kinks where the adsorbed atoms or molecules will be attached.

The critical size of such a secondary nucleus is found to be dependent on the vapour pressure p as

$$R_c = \frac{\gamma a^3}{kT \log p/p_0} \qquad (1)$$

where γ is the surface tension, a is the lattice parameter and p_0 is the equilibrium vapour pressure. This critical nucleus represents an increase of surface free energy by a factor $2\pi\gamma aR_c$ and their formation by the action of thermal fluctuations can be expected to occur at a rate proportional to $-\exp(2\pi\gamma aR_c/kT)$ which is generally a very small factor. According to this theory, below excess vapour pressure of 25% an observable growth rate cannot be expected to occur, while in actual experimental cases it is found that growth takes place at as low as 1% supersaturation.

This discrepancy was explained by Frank[1] who introduced the idea of nucleation in the presence of a dislocation. He suggested that the step formed on the surface by the presence of a screw dislocation would be a self-perpetuating one and hence the necessity of a two-dimensional nucleus does not arise. Such a step will not remain straight during the process of growth, since that would require its speed of motion to be proportional to its distance from the dislocation, whereas it will be more or less uniform. Due to this, the step lags behind at the edge and winds itself into a spiral. The growth rate is equal to the number of times per second the spiral makes a complete turn which is directly proportional to the square of the supersaturation. Many experiments have confirmed this suggestion. The shape of the spiral depends on the type and the structure of the crystal which is growing. The spirals assume a polygonal shape due to the change in growth rate with crystallographic directions. The emergence of two or more dislocations near each other can give rise to different spiral structures or even closed loops of growth steps, of which some have a configuration which generally resembles a dislocation arrangement around a Frank-Read source. This is the case when opposite dislocations are present. But, when closely situated dislocations of equal sign exist, the double pitch spirals are observed.

In 1937, prior to the introduction of the dislocation theory, Heck[2] observed the spirals on the surfaces of the crystals of paraffin. Two decades before Frank's theory Menzes and Sloat[3], Mellor[4] and few others have observed the spirals on silicon carbide. In fact no satisfactory explanation could be offered by these authors. Griffin[5] was the first to offer the experimental evidence in support of the new theory. He observed by using bright field microscopy, the line markings on the $(10\bar{1}0)$ face of beryl crystals, which, there are reason to belive, are edges of the growth layers. He also observed the patterns originating from single and double screw dislocations, which correspond to the theoretically predicted shape.

A large amount of work has been carried out by various workers which lend further support to the spiral mechanism of crystal growth. Verma[6] has given a detailed account of the various types of spirals and their significance. The development of the theory has been discussed by Frank[7]. Forty[8], in a review article, has given an account of the growth spirals observed on various crystals. The detailed study of the crystals of SiC and CdI has been made by solid state physicists.

In metal crystals, Amelinckx[9] and others[10] obtained direct experimental proof for the screw dislocation theory on chemically deposited gold. However, due to their large step heights the dislocations are anomalous for there is no evidence for the existence of such large dislocations in metals. Forty[11] has observed growth spirals on hexagonal metals, magnesium, cadmium and zinc grown from their vapour in vacuo or in inert atmosphere. They are observed on the facets which might have been deposited on the cooler parts of the tube which contains the stock metal. These facets were true to the crystallographic orientation within a few seconds of an arc. Faint growth spirals which from their low visibility could be inferred as being monomolecular in height were observed on the crystals of magnesium and cadmium. Also growth hills with step lines and closed loops without a terrace at the centre were observed. In each of these crystals the evidence for the presence of two or more screw dislocations was not available. A large number of loops with a speck of material at the centre and not the growth spirals has been observed by Forty. He suggested that this could be due to the growth by two dimensional nucleation catalysed by the crystalline overgrowth of zinc oxide. If this was the case, the magnesium crystals should also have grown by the catalysis of magnesium oxide. Such an observation was not made in the case of magnesium and his suggestion cannot be considered conclusive. Dawson and Anderson[12] explain this by suggesting that the growth hills are produced by pairs of dislocations of opposite sign which have moved together to form a closed inner loop. However, this should not happen so completely in every case.

The cubic and octahedral faces of silver crystals grown from vapour phase have been studied by Forty and Frank[13] and they reported the growth spirals on these faces. They were the first to observe screw dislocations of unit Burgers' vector in cubic crystals. A major criticism of the dislocation theory of crystal growth was that spirals have been observed only on thin plates with layered structure, for example, cadmium and crystals of complex structure, such as beryl. The growth spirals have also been observed on platinum crystals grown from $PtCl_4$[14]. Steinberg[15] observed the growth spirals on titanium crystals. By using a microscope fitted with an infrared tube, Dash[16] has shown excellent closed loop patterns not only on the surface but also inside the crystals of silicon. The study of the growth of lead nitrate crystals from solution and the effect of local deformation during the process of growth have been done by Williams[17,18]. She has observed that the growth centres can be initiated at the deformed regions and these can also overtake the existing ones, if they are strong enough.

Henschke[19] has studied the growth of the microblisters of copper films sputtered on a glass substrate and observed closed loops, partly centric and partly eccentric with large step heights, of the order of a micron each. This large step height is explained by considering a double alternative active growth mechanism analogous to the Frank-Read sources without assuming the existence of the dislocations having large Burgers' vector.

The study of the effect of supersaturation on the form of spirals on paratoludine and other organic crystals was made by Dukova[20]. He observed polygonal spiral layer, rounded spiral layer, disintegration of the spiral layer to concentric loops and layers of dendritic form from low to high supersaturation.

Testuya et al.[21], observed three-fold symmetrical pyramids developed on the (111) surface of germanium crystals due to the deposition of germanium vapour produced by decomposing GeI_2. They argued that these are formed at the sites of dislocations due to the rapid growth which is suggested by Frank's theory. Their further suggestion was that an obliquely inclined edge dislocation can also give rise to a perpetuating step.

Lang[22] has given another explanation for getting large step heights of the spirals and closed loops. He has shown that a periodic bunching of advancing or retarding atomic sheets occurs on the crystal surface inclined at a small angle to the densely packed atomic plane and causes observable steps to appear. Such steps form concentric rings generally. But, in the early stages a simple mistake in the step formation can produce a continuous spiral step rather than ring. In such cases the screw dislocation is not required for the production of the spiral.

Spirals and loop patterns have been observed by Pandya and Balasubramanian[23] on the facets of zinc crystals grown from the melt. The spirals showed low step heights while the loops had a step height of about one micron. The large step heights were explained by assuming a double spiral mechanism as used by Henschke[19] on microblisters of copper films sputtered on to a glass substrate. They showed that these patterns are not due to the growth from melt but they are due to growth from vapour. Pandya and Thattey[24] have reported the growth spirals on cadmium crystals with large step heights. They suggested that the large step height is either due to the initial formation of multiple dislocations during nucleation or due to the different rates of advance of successive layers piled up on the top of each growing step. These growth spirals are shown to develop during growth from the vapour phase. Thattey[25] has also observed the closed loop patterns on cadmium single crystals. This has been attributed to the existence of two opposite types of screw dislocations.

The above discussion presents the uptodate data on the spirals and closed loops observed on the crystals. In brief it can be stated that the only possible mechanism of the growth of the crystals from the vapour and dilute solution under low supersaturation is spiral growth mechanism. Impurities can also provide suitable steps for the growth of the crystals as observed by Forty on zinc. Also the macroscopic spirals can be formed without screw dislocation. This shows that one must carry out careful analysis of the spirals and closed loops before drawing any specific conclusions.

A good amount of information has been obtained on the mechanism of crystal growth from the melt during the last three decades. Chalmers and his co-workers have carried out much work on metals while on semiconductors the work has been carried out by major electrical concerns in the United States of America and Great Britain. U.S.S.R. and other countries have also taken keen interest in the study of the crystal growth from the melt. Actually the process of growth is through an atom by atom addition to the lattice as in the case of vapour phase growth but the observations in the recent years tend to suggest that the presence of screw dislocations is not a prerequisite to the growth of single crystals from melt. This conclusion has been drawn from the results of Dash[26], Hillig and Turnbull[27], Chalmers and Maritus[28], etc. Dash[26] has successfully grown single crystals of silicon having large portions free from dislocations. Large single crystals of some metals have also been grown without observable dislocations. Theoretical calculations for the growth rates of crystals by screw dislocations were carried out by Hillig and Turnbull[27] and showed that the results do not fit in the case of some metals like tin but agree for organic substances. Chalmers and Maritus[28] have proposed a step-like interface which provides permanent re-entrant steps which are not propagated by dislocation. The evidence for the existence of such steps have been obtained. Hence one can conclude that nucleation at the re-entrant edge or a screw dislocation is not important for the growth of metal crystals from melt.

The basic condition to be satisfied for the production of single crystals from the melt is that a progressive freezing of the liquid takes place in a controlled manner. In other words the solid-liquid interface moves in a controlled manner. The growth rate of the crystal is the rate at which this solid-liquid interface moves in a direction perpendicular to the interface. The latent heat is continuously evolved during freezing, which must be removed by conduction along the solid charge in order that crystal may grow. This means a temperature gradient must exist in the solid. More frequently, a temperature gradient in the liquid with the temperature increasing away from the interface exists, in which case the heat evolved is conducted through the interface down to the solid. The heat transfer equation can be given as

$$K_s A_s \left(\frac{dT}{dx}\right)_s - K_1 A_1 \left(\frac{dT}{dx}\right)_1 = AL\rho \frac{dx}{dt} \qquad (2)$$

where s = suffix for solid, 1 = suffix for liquid,

K = thermal conductivity, T = temperature,

A = cross-sectional area of the charge,
ρ = density of the metal,
L = latent heat of metal and
x = co-ordinates of the interface measured along the charge.

If G is the temperature gradient, R, the rate of growth, and since $A_s = A_l = A$, equation (2) can be written as

$$K_s G_s - K_l G_l = RL\rho \tag{3}$$

From the equation (3), it can be seen that the factors controlling the growth of crystals from the melt are the growth rate and temperature gradient.

Most of the investigations in the field of crystal growth from melt were aimed at (1) the study of the morphology of the interface and the effect of the various growth parameters on it, (2) the study of the substructures observed on the crystals grown from the melt, (3) the study of the imperfections, their formation and distribution in the crystals and (4) the study of the preferred orientation and the influence of the various parameters responsible for this property of the crystals. Generally the growth of single crystal from melt is influenced mainly by the rate of growth, the temperature gradient along the solid and impurities. A large number of reviews has been published on the crystal growth from the melt. Holden's article[29] clearly shows that our knowledge about conditions governing the successful growth of metal single crystals is very often contradictory. Maritus[30] also gave a review article on the growth from the melt. The various problems were well-discussed at the international conference on growth and perfection of crystals[31]. A review on the mechanisms of growth of metal single crystals from the melt is given by Hurle[32] where he explains the morphology of the solid-liquid interface, preferred orientation, lattice defects produced during growth, etc., in detail. The latest literature available on the mechanism of the growth, not only from the melt but also from the solution, vapour and recrystallization is that by Gilman[33].

In the process of the crystal growth, three basic techniques or some modifications thereof have been adopted. They are Czochralski's method, Bridgman's method and Chalmers' method. In the first method a seed crystal is used which is immersed in the molten metal which is just above its melting point. The seed is pulled up vertically at the rate at which the crystal solidifies, while remaining in contact with the liquid. For maintaining a uniform temperature gradient, the crystal or the crucible is rotated about its vertical axis. This method is widely used in growing the germanium and silicon single crystals. In recent years, it has been applied for the production of bismuth single crystals[34,35]. In Bridgman's method, the metal is put in a capsule having a capillary end and the capsule is then lowered vertically in an externally imposed temperature gradient. The metal solidifies from the bottom upwards. The rate of freezing is determined by the rate of heat flow from the solid, which depends on the rate of travel of the capsule through the gradient. The third method, namely Chalmers' method, is the modified form of the Bridgman's, in which the container is made to travel horizontally. Whenever necessary, the seed crystals can be used. This technique has an advantage over the Bridgman's technique that the upper surface of both solid and liquid can be observed.

The understanding of the morphology of the solid-liquid interface is the key to the solution of the problem of crystal growth. The morphology of the interface depends on the thermal pattern, solute distribution and the crystallographic character of the specimen. Depending on the degree of supercooling, the interface develops a variety of microstructures. In the absence of the supercooling the interface is either planar or step-like which will be obtained only with very high purity metal. If there is constitutional supercooling the interface develops initially a pox-like structure, which are small projections and give rise to the cellular structure. When the supercooling is large the cellular structure breaks up into dendritic structure. Using decanting technique the interface structure is studied in detail.

On the decanted interface of oriented single crystals and bicrystals of lead $\{111\}$ or $\{001\}$ planes parallel or nearly parallel to the interface have been observed by Elbaum and Chalmers[36]. When these planes coincided with the interface the steps became large and wide terraces were observed, but when they were not parallel to the interface no such structure was observed. By varying the growth conditions no systematic variation was observed. Rosenberg and Tiller[37] also observed similar results who named it the platelet structure. Elbaum and Chalmers[36] have given the explanation for this phenomenon in terms of the creation and migration of steps when the interface is slightly tilted to the close-packed planes. Jackson[38] attributes this to the formation of faceted growth of the low index planes when the interface is parallel to the isotherm.

A detailed study of the cellular structure on tin has been made first by Rutter and Chalmers[39]. After this the study of this structure has been made in a number of metals which includes lead, aluminium, silver, copper and zinc. An observation of corrugation on the surface of the growing crystal interested many workers. To study whether this corrugation was a surface feature or a body feature, the interface was exposed by decanting. On examining the surface, it has been found that the interface consisted of a number of hexagonal cells. Subsequent cutting of the crystal with a chemical saw showed that this structure remains the same inside the crystal. As a matter of fact, this corrugation was also observed by Goss and Weintroub[40] but they called them as "fine lines". The corrugations and cells are two perspectives of a columnar structure of the crystal growing under conditions of supercooling caused by the rejection of solute at the interface. The cell boundaries are the places of concentration of the solute. Thus a growing crystal under the influence of supercooling can be pictured as a close stack of pencils, where ends of the pencils represent the cellular interface and the axes of the pencils represent the growth axis. The nature, size and direction of the cells and corrugations have been studied under different conditions of growth rate, temperature gradient, impurity content and the crystallographic nature of the interface. It has been found that the cellular structure can be suppressed by a steep temperature gradient and a slow growth rate.

A dendritic interface is observed at a fast growth rate or a large impurity content. The transition from a smooth interface to that of a cellular one is always characterised by the appearance of a pox-like structure[41-44]. The observations of Hulme[43] on zinc single crystals grown in a horizontal boat under the conditions of constitutional supercooling show that the shape of the cell boundaries depends markedly on the orientation of the crystals.

Balasubramanian[45] has grown zinc single crystals by Bridgman's method and observed that the size and direction of the cell boundaries which can be revealed by etching depend on the crystal orientation. He, by employing a slow rate of growth and very high purity zinc, showed that the development of the cellular structure did not take place abruptly but gradually. His results agree with the theoretical predictions regarding the requirement of an incubation distance in the process of growth. The cellular structure has also been observed on cadmium by Thattey[46]. His study includes the dependence of the cell structure on the growth rate and purity of the material.

Rutter and Chalmers[39] have developed a qualitative theory for the formation of the cellular structure, which is outlined as follows. The increase in the impurity content near the interface, forms a liquid layer adjacent to the interface, which is below its equilibrium liquid state temperature and hence it is an unstable state. A spontaneous nucleation is initiated in this zone which results in the formation of a projection in the interface which is initially plane. The impurities from the tips diffuse to the sides. This helps the tips to solidify at a higher temperature than the interface temperature and the solute at a lower temperature than that at the interface. This increase in temperature at the tips produces the favourable thermal conditions for latent heat flow from the projection to the surrounding liquid and solid, promoting the projection to grow. These conditions for the growth of the projections are maintained only if they do not cross a certain length, the temperature increasing away from the interface. As a consequence of the above factors there results, during the non-equilibrium solidification, an array of projections in the initially smooth interface.

A large supercooling, either thermal or constitutional, causes the interface to become dendritic one. Dendrites are common features in a crystal grown from the melt or from the solution. Weinberg and Chalmers[47,48] and others have made a detailed study on dendrites. Transition from cellular structure to dendritic structure has been studied by Tiller et al.[49,50] On germanium, the growth of strip dendrites has been studied by Billig and Holmes[51-53] and others. Maritus[30], Jackson[38] and others have given detailed accounts of the growth of dendrites. Yet the morphology of dendrites is not completely understood and the available information on secondary dendritic arms is very little.

Another substructure of metals is striations or lineage structure. This substructure has been studied in detail by Chalmers and others[54-59]. These striations are body phenomena and are parallel to the crystal axis or, at fast rates occur generally along certain close-packed directions. They divide the crystals into different regions differing in orientations by 0.25° to 5°. The lineage boundaries are low angle boundaries consisting of number of dislocations. Chalmers and Teghtsoonian[55] have proposed that the boundaries are formed by the condensation of vacancies at, or behind, the interface. Chalmers[60] has reviewed the vacancy condensation theory, but it is open to criticism.

The last two decades have witnessed a remarkable advancement in the investigations on the preferred orientations of the crystals during growth. Goss and Weintroub[40] have made a detailed study of this property on metallic single crystals having low melting point. They found that for metals cadmium, tin and bismuth, the preferred orientation was that for which the direction of maximum thermal conductivity coincided with the axis of the specimen. The preferred orientations at fast rates and at slow rates are different. To explain the preferred orientation, Tammann[61] used the theory of the anisotropy of growth. But this theory fails in the case of cubic crystals which are isotropic. Platelet growth system and the growth direction of dendrites are also the cases of preferred orientation. Rosenberg and Tiller[37] have shown that in the case of crystals of columnar casting the orientation is directly related to the morphology of the interface. Tiller[62] has explained how platelets having orientation different from others are suppressed by the process of competitive growth of more than one grains. However, there exists a disagreement between the experimental results of Rosenberg and Tiller[37] and those of Walton and Chalmers[63]. This conflict must be resolved first before drawing any definite conclusions on preferred orientation in cubic crystals. Very little work has been done on metals with other symmetry. Recently, in the case of cadmium crystals, Grinberg[64] has observed that the rate of travel of the mould in a fixed temperature gradient has a pronounced effect on the orientation of the basal plane with respect to the crystal axis. Balasubramanian[45] has reported that the results, in the case of zinc crystals grown by Bridgman's method, are in agreement with those of Grinberg. In the case of bismuth single crystals grown by Bridgman's technique, Shah[65] has shown that the results are in complete agreement with Tammann's theory.

A study on the perfection and the factors controlling the perfection of 99.999% purity and OFHC copper crystals has been made by Young and Savage[66] with the crystals grown by Bridgman's method. They observed that the dislocation densities in these crystals were low. They also found that the orientation of unseeded crystals of both purities was random; but in the case of high purity copper a correlation was found between perfection and growth direction. The growth axis of the crystal is away from the [111] axis.

The origin of the dislocations in as-grown crystals is not yet fully understood. Theory of vacancy condensations can be ruled out as has already been explained. Dash[26] has proposed that the propagation of dislocations from the seed, multiplication of dislocations by thermal shock and the generation due to poor epitaxy between the seed and the crystal are the various possible means for generating as-grown dislocations. A mechanism of the production of dislocations at platelet and cell structures was proposed by Tiller[67]. According to this mechanism, the dislocations are produced due to the microseggregation of impurities which produce local changes in the lattice parameter and this may produce as many as 10^5 to 10^6 lines/cm^2.

In the case of cellular interface this value may be as high as 10^7 lines/cm^2.

The review presented above is neither comprehensive nor exhaustive. The situation of the crystal growth theory at present can be summarised as follows:

Under the conditions of low supersaturation screw dislocations facilitate the growth of crystals from vapour or from dilute solution. For crystals growing from the melt, nucleation at the re-entrant corner of a screw dislocation is not necessary. The solid-liquid interface topography is governed mainly by solute distribution, thermal conditions and crystallographic features of the growing crystals. Also the orientation of the crystal is influenced by the same factors. The aspects of preferred orientation and the production of dislocations when the crystal grows have not yet been satisfactorily solved.

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