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**A Study of Behavior of Lattice Defects
by means of HVEM *In Situ* Experiments and
Real-Time X-ray Topography***

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Keywords: behavior of lattice defects, high voltage electron microscopy, in-situ experiment, X-ray topography, dislocation. direct observation of crystal growth, deformation and strength of crystalline materials

I. Introduction

It is well-known that among the properties of a crystalline material there are those which can not be discussed without knowing the behavior of lattice defects⁽¹⁾⁽²⁾. It is also well-known that dislocations, many of which exist in a crystal of practical use, have a deep relationship with crystal growth and play an important role, particularly, in plastic deformation and strength of crystalline materials. Although from the title of this paper the subject to be discussed seems to be the behavior of all types of lattice defect, the behavior of dislocations is the main subject here and the behavior of others are treated as subsidiary, since crystal growth, plastic deformation and strength are mainly discussed in this paper. Moreover, it should be mentioned in advance here that since use was made of two complementary techniques, i.e., HVEM *in situ* experiment and real-time X-ray topography, both of which enable us to observe the behavior of dislocations directly, the results obtained by these techniques are mainly described in this paper.

In the ordinary electron microscopy and X-ray topography, observations are usually made with a specimen prepared before and after the dynamic process concerned, however, in the present experiments, the aforementioned two dynamic techniques were particularly adopted for the following two reasons: (1) no speculation is needed in analyzing the sequence of events in a dynamic process, and (2) information on the speed of dislocation motion and structure changes can be obtained by observing the successive stages continuously.

However, those observations become much more useful if the results obtained with those microscopic observations are directly correlated with the stage of macroscopic behavior of the specimen. For such purposes, a new experimental technique was developed with an idea which enables one to make a direct correlation, for instance, between the behavior of dislocations and the state of the specimen represented by a point on the stress-strain curve possible by recoding both of the dislocation behavior and the stress-strain curve simultaneously.

II. High Voltage Electron Microscope and Ultra High Intensity X-ray Generator

1. High voltage electron microscope (HVEM) and its attachments for *in situ* experiments

Since 1960, HVEMs with accelerating voltage of over 500 kV have been installed in many countries. In Japan, U.S.A., England, France and some others, fundamental as well as applied researches being made, in particular, by means of *in situ* experiments in which merits of high transmissive power of high voltage electrons were successfully used, have been carried out intensively. On the occasion of the First International Conference on HVEM held Monroeville, Pa. U.S.A. in 1969, preliminary experimental results and usage of the HVEM *in situ* experiments have been reported for the first time⁽³⁾.

Since then, progress in various technological aspects, such as microscope column construction of ultra high vacuum, system for high spacial- and time-resolution recording, and image processing and simulation experiment facilities, various specimen-treatment devices for *in situ* experiments and advanced specimen preparation techniques have been made. As a result, constructions of

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HVEMs such as those for atomic resolution, and those for *in situ* experiments of high technological level, those for *in situ* studies of ion-irradiation damages have been made. More recently, at Nagoya University, a 1250kV STEM mode HVEM has been constructed, aiming improvements in various aspects such as reducing electron damages of the specimen under observation, increasing observable specimen thickness, intensifying image contrast of originally low contrast specimens, easiness of image processing, etc. For this microscope, a pioneer application of a field emission gun (FE-gun) has been made for the first time to the HVEM of over 1000 kV accelerating voltage⁽⁴⁾.

1. *In situ* specimen treatment devices

Figure 1 illustrates the usable temperature range of various types of *in situ* specimen treatment devices⁽⁵⁾ which have been developed by the author's group and by Osaka University group. Besides these, there have been

Fig. 1 Functional features of various *in situ* specimen-treatment devices. Usable temperature range extends to the shaded area when the function enclosed in brackets is excluded⁽⁵⁾.

constructed some other devices recently with advanced functions (for instance, tensile device capable of stretching a specimen at 2300 K)⁽⁶⁾. Figure 2(a) represents one example of tensile devices⁽³⁾⁽⁷⁾⁻⁽¹⁰⁾ which have been developed by our group to enable one to observe and record moving dislocations by TV-VTR system in parallel with recording the load-elongation curve of the specimen and Fig. 2(b) illustrates one of fatigue devices⁽¹¹⁾⁽¹²⁾ which is capable of making fatigue test of a foil specimen under observation by cyclic bending being induced by horizontal alternating motion of the specimen chucks. This horizontal motion is induced by converting the vertical expansion and compression of the cylindrical piezo-element with lever mechanism. For this piezo-element, plus and minus 1000 V are applied alternatively. Figure 2(c) shows schematically a specimen holder of Cu-Be plate with a center hole which insures the introduction of compression to the specimen by homogeneous bending.

2. Dynamic observation and continuous recording

For the continuous recording of moving EM images, a movie camera was used in the past, however, the recording speed of it is low even under a strong electron beam and it has many inconveniences such as invisibility of recording images during recording, impossibility of inspecting focusing condition during recording and necessity of development of the film to inspect the proper exposure etc.

To eliminate these inconveniences, we considered the usability of TV-VTR system and pioneer application of it to HVEM *in situ* experiments was made for the first time already in 1967⁽¹⁴⁾. The recording speed of moving EM images is increased dramatically (60 fields/s, 30 frames/s) under reduced electron illumination and moreover, recording of the moving image can be made simultaneously with observing the image on monitor TV screen. Increase of the brightness of the image and adjustment of the focus can be made easily under reduced specimen illumination (about two orders of magnitude less incident electron density) also. As regards a TV image tube, there are two types, namely, direct type and indirect type⁽¹⁵⁾⁻⁽¹⁷⁾. As for the former type, imaging electron beam is received directly with the target plate of the vidicon type TV tube

Fig. 2 Tensile device capable of recording moving dislocations by TV and VTR system and load-elongation curve of the specimen by x-y recorder simultaneously during observation (a), fatigue test device capable of measuring strain amplitude in carrying out fatigue test by alternating tension and compression (b) and special specimen holder (c)⁽¹¹⁾⁻⁽¹³⁾.

in vacuum to form the image on the TV screen, and in the latter case, the optical image formed at first on a semi-transparent fluorescent screen is transferred to the target plate of an ordinary TV camera with tandem lenses or fiber plate to form the image on TV screen. Taking into account the high gain and low lag, we decided to use the latter type and as an image tube, the image orthicon tube was chosen because of its nice performance of recording a fast moving image without appreciable lag⁽¹⁶⁾⁽¹⁷⁾. Figure 3 represents schematically the complete assembly of our instrument for HVEM *in situ* experiments in which the aforementioned tensile device and TV-VTR system are indicated⁽¹⁸⁾⁽¹⁹⁾.

2. Ultra high power X-ray generator of rotating anode type

Since the intensity of X-ray scattering by matter is much weaker than that of electron beam, ultra high intensity X-ray source is necessitated at all rates for carrying out real time X-ray topography observation. At first, use of X-rays being generated by SOR came into mind, however, taking account of the fact that the time available is very limited for one research group to carry out a systematic study of crystal growth, deformation, etc., we decided to construct an ultra high intensity X-ray generator of rotating anode type. Figure 4 indicates schematically the out-line of ultra high intensity X-ray generator of 90kW-class (60 kV $\times$ 1500 mA) which was developed by our Nagoya University group with the co-operation of Rigaku-Denki Co. in Japan. Figure 5(a) illustrates a newly constructed tensile device set on a large goniometer stage for real-time X-ray topography apparatus. This tensile device enables us to carry out tensile deformation of a specimen at a temperature up to 1200 K in vacuum or in any inert gas environment⁽²⁰⁾⁻⁽²²⁾.

Figure 5(b) indicates a schematic sketch of a specimen-heating device which was designed and constructed by

Fig. 4 Overview of ultra high power X-ray generator of rotating anode type.

the author's group to carry out direct observations of crystal growth from the melt and also to anneal the crystal. This enables one to heat the specimen up to 1300 K in vacuum and in any inert gas atmosphere by using a graphite heater⁽²³⁾⁽²⁴⁾. Figure 6 indicates schematically the complete assembly of the real-time X-ray topography apparatus which includes X-ray goniometer with the aforementioned specimen heating stage, TV-VTR recording system and monitor TV⁽²⁴⁾. A PbO vidicon tube has been used so far for this TV camera, however, in the future, use of a SATICON tube will be considered to obtain higher speed recording.

III. Plastic Deformation and the Behavior of Dislocations

1. Introduction

It is well-known that a star player in crystal deformation is a dislocation and that elemental process of crystal deformation is a glide motion and multiplication of dislocations. Since plastic deformation is essentially a dynamic phenomenon, time-dependent macroscopic as well as microscopic change of physical quantities will occur as the progression of deformation. Macroscopic physical quantities which will be usually subjected to measurements are the shape and dimensions of the specimen and also the deformation resistance of the specimen. The microscopic origin which produces such a change of the macroscopic quantities is the motion of dislocations. The change of density and distribution of dislocations occurs in the specimen as deformation proceeds so that the change will occur in dislocation interaction and also in free path and velocity of dislocation motion. Then, in the meanwhile, the state of the specimen becomes such that since the behavior of individual dislocations does not directly reflect the macroscopic behavior of the specimen, the collective motion of dislocations represents the macroscopic behavior instead. Even so, when we want to discuss and understand the mechanical properties like deformation and strength of a crystal, a final goal of understanding will not be obtained without knowing

Fig. 3 Schematic representation of a complete assembly for carrying out HVEM *in situ* experiments⁽¹²⁾.

Fig. 5 Schematic representation of high temperature tensile apparatus set on a large goniometer stage for real-time X-ray topography (a) and specimen heating device for direct observation of crystal growth from the melt (b)⁽²³⁾⁽²⁴⁾.

Fig. 6 Schematic drawing of a complete assembly of apparatuses for real-time X-ray topography⁽²⁴⁾.

the fundamental properties of individual dislocations. Therefore, it may be said that such approach is

necessitated in the first place and then with the progress of deformation a wealth of knowledge pertaining to the collective motion of dislocations is requested to reach the goal. From such a point of view, detailed studies on the behavior of individual dislocations near the yield point of each crystal are made and then we go into further study of the behavior of dislocations in the progressed stage of deformation.

2. Yielding of a crystal and behavior of dislocations near yield point

There are two fundamental equations which correlate the behavior of dislocations with macroscopic crystalline deformation⁽²⁵⁾⁽²⁶⁾. One of those is a fundamental equation which expresses the relation between a macroscopic strain of the crystal ε and mean displacement of dislocations $\langle d \rangle$, that is,

$$\varepsilon = b \cdot N \cdot \langle d \rangle. \quad (1)$$

Another fundamental equation which correlates mean velocity of moving dislocations $\langle v \rangle$ with macroscopic strain rate of the crystal $\dot{\varepsilon}$ is:

$$\dot{\varepsilon} = d\varepsilon/dt = b \cdot N \cdot \langle v \rangle, \quad (2)$$

where b denotes a size of Burgers vector, N the density of mobile dislocations. In these two equations, N is described as if it is independent to time with the progression of deformation stage. However, it is unlikely in general. N is usually a function of ε and $\dot{\varepsilon}$ and $\langle v \rangle$ is dependent

on the applied stress τ . In practice, N and $\langle v \rangle$ change in accordance with progression of deformation, so that in case we like to describe the macroscopic deformation and deformation rate of a crystal in terms of the aforementioned type of the motion of dislocations quantitatively, it is necessitated to know the functional forms of the change of N and $\langle v \rangle$ of each crystal with time.

By the way, now let us consider first the yielding phenomenon according to the view of Gilman and Johnston, that is, from the point of view of interrelation between a specimen and a testing machine⁽²⁵⁾. In case a crystalline specimen is deformed by Instron-type hard beam machine, the crosshead is displaced at a fixed rate, and since at the very beginning of deformation where the density of mobile dislocations is low, it is hard for the specimen to deform correspondingly to the displacement of the crosshead, the increase of applied stress is needed to make further deformation of the specimen because of the fact that the increase of deformation resistance of the whole linear system occurs. As the deformation proceeds further, the density of mobile dislocations increases due to multiplication of dislocations and at the same time the velocity of moving dislocations increases also due to the applied stress increase, resulting in the increase of deformability of the specimen. In this way, the deformability of the specimen crystal increases and at the particular point where the microscopic strain rate of the specimen due to this deformability-increase exceeds the crosshead speed, that is the applied macroscopic strain rate, the accumulated elastic strain in the specimen is suddenly released and the yielding phenomenon will occur due to the sudden decrease of the applied stress. At the instant when the yielding phenomenon begins, the macroscopic strain rate, which is given by the machine, and the deformation speed of the specimen being induced by the motion of dislocations given by the equation (2) will be balanced. If the functional forms of N and $\langle v \rangle$ for a given crystalline specimen, are known in advance, the relation between the applied stress acting to the specimen (τ) and the strain of the specimen (ϵ) can be calculated in principle using eqs. (1) and (2). In other

words, the yield stress for a crystal can be calculated with using the characteristic functions of N and $\langle v \rangle$ for the crystal. It may be said now that the yield stress can be obtained from the $N \cdot v$ characteristic of a crystal without carrying out experiments.

3. Multiplication and mobility of dislocations

Figure 7 represents the curve obtained by plotting the density of moving dislocations N_m versus the normalized applied stress τ/τ_y (the applied stress τ is normalized by dividing it with the yield stress τ_y)⁽¹²⁾. The accuracy of this measurement of moving dislocation density is low as compared with that obtained by etch-pit method and others which provide an average value in a wide area, but this obtained curve indicates clearly the occurrence of an abrupt increase of the number of moving dislocations near the yield point and also no appreciable temperature dependence of N_m versus τ/τ_y relation within the present experimental temperature difference.

Figure 8 illustrates the mobility data obtained for the first time by observing directly moving dislocations (bold lines)⁽¹⁰⁾⁽²⁷⁾⁻⁽²⁹⁾. The velocities of edge and screw component can be distinguished by this direct observation of moving dislocations. Since in the range of the present experiments, the relation $\langle v \rangle$ versus $\langle v_0 \rangle$ can be described in the form of $\langle v \rangle / \langle v_0 \rangle = (\tau/D)^m$, the mobility parameter m and D for pure Fe, Fe-3%Si, Mo and β CuZn are calculated by using the aforementioned relation and tabulated as in Table 1. As regards multiplication mechanisms or sources of dislocations, Fig. 9(a) illustrates sequential pictures of a single-ended F-R source in operation as an example of dislocation sources observed by HVEM *in situ* experiments and Fig. 9(b) illustrates schematical drawing of various multiplication mechanisms observed frequently. One can recognize that many other mechanisms of multiplication can be seen besides F-R source and cross slip mechanisms.

4. Behavior of dislocations in the process of work-hardening and fracture

With further deformation after yielding, the density of dislocations increases further and the interaction of dislocations with forest dislocations and other obstacles

Fig. 7 Density of moving dislocations as a function of applied stress in Fe⁽¹²⁾.

Table 1 Mobility parameter m and D of a dislocation (RT: room temperature, s: screw component, e: edge component).

Specimen	m	D/Pa
Mo (RT s)	4.8	6.8×10^7
Mo (150 K s)	7.4	5.3×10^8
Mo (150 K e)	14.2	1.7×10^8
Fe (RT s)	3.5	2.0×10^8
Fe (150 K s)	3.5	1.4×10^9
Fe (150 K e)	3.1	9.0×10^8
Fe-3%Si (RT e)	34.5	1.6×10^8
β -CuZn (550 K e)	1.3	5.2×10^{10}

[†] In $\langle v \rangle / \langle v_0 \rangle = (\tau/D)^m$, the mobility parameters m and D are dependent on the kind and physical state of materials. If $\langle v_0 \rangle$ is assumed to be 10^{-2} m/s, the value of D corresponds to the stress value necessary for moving a dislocation at the speed of 10^{-2} m·s⁻¹.

Fig. 8 Dislocation velocity versus resolved shear stress relations obtained by observing moving dislocations under applied stress directly. Bold lines represent the data of the present experiments^{(10),(27)-(29)}.

Fig. 9 Successive pictures of operating Frank-Read Source (a) and schematic drawing of others indicating various multiplication mechanisms observed frequently (b)⁽³⁰⁾⁽³¹⁾.

becomes frequent. The formation of jogs due to the moving dislocations with forest dislocations, which is one of the elementary process of work-hardening⁽¹⁹⁾⁽³⁰⁾⁽³¹⁾, the formation of cell walls of dislocations and the fractionization of cell structures were studied *in situ*⁽³⁰⁾⁽³¹⁾, however, one example indicating the interaction of dislocations with internally oxidized particles in the Cu-Al matrix is illustrated here in Fig. 10⁽³⁰⁾. It can be observed in a series of sequential pictures that motion of dislocations which encountered with oxide particles on the slip plane, was hindered upon meeting the particle. Then, they bowed out and finally overpassed the particles at a certain critical angle 2θ .

As the progress of deformation after cell formation, the cell fractionization reaches a certain limit size ($\sim 0.1 \mu\text{m}$) and then a crack is initiated usually at the edge of the thin film specimen. Fracture will occur by the crack propagation. It is worth noticing here that a large number of dislocations are emitted from the crack tip⁽⁸⁾⁻⁽¹⁰⁾⁽³³⁾ during crack propagation and the emitted dislocations immediately move away from the crack tip, creating a dislocation-free zone⁽³²⁾. Besides, in the case of a fatigue test of

Al the flip-flop motion and shuttling motion of dislocations could be observed actually under the alternating stress for the first time by using the fatigue test device developed by the author's group⁽³⁴⁾⁽³⁵⁾. It was also succeeded to observe directly the formation of so-called ladder structure (wall structure) of dislocations during the fatigue process of Cu (cf. Fig. 11)⁽³⁶⁾. In zone-refined Nb single crystals, one of the mechanisms indicating anomalous slip could be observed by the *in situ* experiments⁽³⁰⁾.

IV. Behavior of Dislocations Associated with Temperature Change and High Energy Particle Irradiation (Application of High Resolution Techniques to *In Situ* Experiments)

1. Temperature dependence of stacking fault energy (γ)

It becomes now possible to observe a very sharp image of dislocations by the development of high resolution

Fig. 10 Successive pictures indicating the interaction between moving dislocations and distributed internally oxidized oxide particles in a Cu-Al alloy⁽³⁰⁾.

Fig. 11 Successive pictures indicating formation mechanisms of ladder structure of dislocations observed directly during fatigue test⁽³⁶⁾.

techniques such as multiple beam imaging and weak beam method^{(36)–(38)}. Owing to the development of those, determination of stacking fault energy (γ) can be made with precision by observing directly the separation of partials of the extended dislocation in fcc metals without applying the former node method. The determination of temperature dependence of γ , which was a long pending question, has been carried out successfully by the weak beam method. Figure 12 illustrates the intrinsic temperature dependence of γ obtained with the aforementioned method on pure Cu, Cu–Al alloys, pure Ag, Ag–Al, Ag–Sn and Ag–In alloys^{(39)–(42)}. In both of Cu alloy and Ag alloy systems investigated, γ decreases with the increase of temperature for both pure Cu and Ag, and according as the increase of content of higher valent solute atoms the slope of the γ versus temperature curves changes. At a certain amount of solute content, the sign of the slope changes from minus to plus. Figure 13 indicates the curve obtained between $d\gamma/dT$ versus e/a and it is likely to have a single curve for both Cu and Ag alloy systems, having a point of inflection at $e/a=1.14$. This indicates a good accordance with the theoretical result by Tisone⁽⁴³⁾. Using a similar method the experimental evidence on the

presence of so-called Suzuki effect (chemical interaction between a stacking fault and solute atoms in fcc alloys) has been obtained by Saka *et al.*⁽⁴⁴⁾

2. Effects of high energy particle irradiation to the behavior of dislocations

In case a high energy particle with an energy higher than a certain threshold value is irradiated onto a crystal, the influence of the irradiation to the behavior of dislocations can be seen due to the formation of point defects and others by irradiation. Besides the influence of irradiation onto crystal growth, phase transformation and glide deformation^{(45)–(46)}, the irradiation effects to the followings have also been studied, i.e., (a) the mechanism of climb of extended dislocations (cf. Fig. 14)⁽⁴⁷⁾, (b) conservative motion of extended jogs along the extended dislocation (c) the formation mechanism of a helical dislocation⁽⁴⁸⁾ from an extended dislocation, (cf. Fig. 15)^{(49)–(50)} (d) an experimental evidence of irradiation-induced solute segregation⁽⁴⁸⁾ and (e) the formation of blister-like defects in Al by electron irradiation at a high temperature in an environmental chamber containing air⁽³⁷⁾. These are mentioned only by indicating related figures or reference articles due to limited pages given to this paper.

Fig. 12 Intrinsic temperature dependence of stacking-fault energy γ in pure Ag, Ag–Al and Ag–In alloys^{(39)–(42)}.

Fig. 13 Relation between $(d\gamma/dT)$ and (e/a) in Cu and Ag alloys⁽⁴³⁾.

Fig. 14 Chern-Hirsch-Saka model⁽⁴⁷⁾ for climb of a dissociated dislocation in low γ fcc crystals, indicating the change of shape by irradiation of 1000 kV electrons at about 400 K. A indicates a dissociated dislocation of Burgers vector b before irradiation, B-D the same dislocation loop after irradiation. Sub-loops are created on two partial dislocations and climb motion can be seen according to the aforementioned model by contrast experiment (changing g , C-D).

V. Crystal Growth from the Melt

1. Morphology of solid-liquid interface and Jackson's theory

Clarification of the mechanism of crystal growth from

the melt is very important not only from the scientific view but also from the view of growing highly perfect crystals in industrial circle. Many experimental as well as theoretical studies have been carried out so far for such reasons⁽⁵¹⁾⁻⁽⁵⁵⁾. As compared with our understanding of crystal growth from vapor, solution and solid, however, it has not yet been clarified so completely even now due to our poor knowledge on the structure of the melt and also on the morphology and atomic structure of solid-liquid interface.

From the kinematical theory of crystal growth from the melt⁽⁵¹⁾, the growth rate of a crystal or the moving speed of the solid-liquid interface u (m/s) can be given as a function of ΔT , as in the following three cases, where $\Delta T = T_m - T$, a measure of supercooling, and T_m is the melting point;

(a) growth due to the uniform movement of the interface; $u = A \cdot \Delta T$,

(b) screw dislocation aided growth: $u = A(\Delta T)^2$, and

(c) lateral growth due to two-dimensional nucleus formation; $u = Ae^{-B/\Delta T}$ where A is a constant.

These cases correspond to the following three growth mechanisms, respectively:

(A) continuous growth on the atomistically rough interface (non-faceted growth),

(B) growth aided by a screw dislocation, and

(C) faceted growth on the atomistically flat interface.

The experimental studies have been made to get evidences for these cases mainly by carrying out optical microscope observation of crystals which are transparent to visual light and the observed results on the interface were used to confront with the corresponding cases⁽⁵⁶⁾⁽⁵⁷⁾. In the case of metallic crystals which are not transparent with visual light, the growth mechanism of crystal from the melt is considered to be usually due to the atomistically rough surface growth (A) or screw dislocation aided growth (B), although there was no direct evidence.

On the other hand, there have been two well-known theories on the solid-liquid interface morphology, being based on the two types of models, i.e. continuous interface model by Cahn⁽⁵⁸⁾ and discontinuous interface model by Jackson⁽⁵³⁾. Indirect evidence obtained so far seems to be favorable to the latter model. Anyway, in the case of metallic materials there have been no direct evidences due to the difficulty of observing the interface atomistically and even macroscopically during the crystallization *in situ*.

To clarify the mechanism of crystal growth from the melt, it should be the most powerful way to observe atomic structure as well as macroscopic configuration of the growing interface and also the effect of dislocations directly and continuously during crystal growth. However, as mentioned above, satisfactory results can not be expected by use of optical microscope for metallic crystals. From such reason, two new techniques were chosen and adopted, namely, high resolution *in situ* HVEM and real time X-ray topography. These are the most powerful techniques available at present to obtain

Fig. 15 *In situ* sequence of formation of a helix; a-e, Shape change indicated b-c-d corresponds to (b)-(c) of the schematic model by Cherns *et al.* Complete contraction indicated by C_1/C_2 in e indicates the halfway of transition to regular helical dislocation⁽⁴⁹⁾⁽⁵⁰⁾.

the information on atomic and macroscopic configuration of the solid-liquid interface during crystal growth, although the development of new attachments and powerful X-ray generator were needed in advance to carry out such *in situ* experiments (Fig. 2-Fig. 5). Firstly, as the specimens for carrying out dynamic observations of macroscopic configuration of the interface, superhigh purity Al of 99.998–99.999% in purity and Ga of 99.9999% in purity were chosen and dynamic observations of the interface during melting and solidification process of Al and Ga were made by means of real-time X-ray topography⁽²³⁾⁽²⁴⁾⁽⁵⁹⁾. The reason why Al and Ga were chosen are as follows;

(a) according to Jackson's theory, continuous growth on the atomistically rough interface which will extend along the isothermal plane can be expected in the case of Al which has the value of α parameter^{†(24)} less than 2 in Jackson's theory, and (b) in the case of Ga which has α larger than 2 for its (010) face and less than 2 for other crystallographic faces, faceted growth can be expected to occur on atomistically flat (010) plane by lateral growth mechanism and non-faceted growth can be expected to occur on other planes by growth surface growth (continuous growth) along isothermal planes according to the

[†] $\alpha = (L/kT_m)(\eta_0/v)$, where L : latent heat of melting, k : Boltzmann constant, v : coordination number and η_0 : number of the nearest neighbor atoms on the habit plane concerned.

Jackson's theory. It was thought exciting to testify whether or not the Jackson's theory is applicable for the growth of these crystals. In addition, using InSb single crystals which have the α parameter larger than 2, high resolution observations of the atomic configuration of the interface have been carried out by means of high resolution *in situ* HVEM at 523 K.

Other subjects such as (1) behavior and role of dislocations intersecting with the interface, (2) dislocation generation near the growing interface, (3) the effect of sudden change of growing condition such as growth rate onto dislocation generation, (4) morphology of the solid-liquid interface in the dendritic growth of alloy crystals like Al-Mg alloys, (5) the effects of growth rate, growth direction and alloy composition to dendrite morphology, and (6) the effect of cooling rate to the change of microscopic structure of as-grown crystals after solidification etc, have been made by means of real time X-ray topography to obtain new information on these subjects through direct dynamic observations⁽⁶⁰⁾.

2. Results of observation on solid-liquid interface and the mechanism of crystal growth

(1) Principal results of observation in the case of Al⁽²⁴⁾

(a) In both of melting and solidification, the solid-liquid interface is always observed along isothermal

Fig. 16 A series of X-ray topographs (TV images) taken after the sudden change of growth rate from $20 \mu\text{m/s}$ to $40 \mu\text{m/s}$ in the growth process. Dislocation generation due to sudden change of growth rate can be observed⁽²⁴⁾.

plane.

(b) Even if there are dislocations intersecting with the interface during growth process, appreciable change could not be observed for the further growth.

In Fig. 16, it can be seen that many dislocations were generated in the solidified region near the interface by the sudden change of growth rate from 20 to $42 \mu\text{m}$ (black thick lines intersecting with the interface are dislocations).

(2) Principal results of observation in the case of Ga⁽³⁴⁾⁽⁵⁹⁾

(a) In the melting process, the interface appeared always along the isothermal plane similar to the case of Al.

(b) In the growth process, not only along the (010) plane of α larger than 2 but also along other planes of α

Fig. 17 A series of X-ray topographs (TV images) showing the growth of a Ga single crystal. Growth rate was about $3 \mu\text{m/s}$. Facets of (110) and $(1\bar{1}\bar{1})$ planes at the solid-liquid interface are indicated by arrows⁽²⁴⁾⁽⁵⁹⁾.

smaller than 2, facets could be observed (cf. Fig. 17) which could not be expected from the Jackson's theory. From these experimental facts, microscopic structure of the interface and the growth mechanism in the cases of Al and Ga are understood as in the following:

Firstly, in Al, (A) the microscopic structure of the interface in the growth process can be expected to be atomistically rough, (B) the growth mechanism operating should be the continuous growth mechanism.

Secondarily, in Ga, (A) in the growth process, the microscopic structure of the interface can be expected to be atomistically flat, (B) the lateral growth occurs and the rate of formation of two dimensional nucleus should be nearly the same as the migration speed of atoms along the interface, and (C) since it has been found that there is an experimental evidence which is not consistent with the Jackson's theory, it should be considered to correct or at least modify his theory. In Jackson's model, the number of the first nearest neighbor atoms is taken into consideration to calculate the α parameter. However, if the second nearest neighbor atoms, which have atomic distance very close to that of the nearest neighbors (the difference is only 0.007 Å) are also taken into account, it is found that modified Jackson's type of theory can be used to interpret the observed results.

Figure 18 provides very important result indicating the atomic configuration of the solid-liquid interface of InSb observed directly for the first time in the world⁽⁶¹⁾. For TEM observations, a foil specimen of InSb was made by electro-polishing and it was observed at 523 K *in situ* by high resolution HVEM. Since the In-Sb alloy system assumes eutectic, evaporation of Sb occurs from InSb

when heated in vacuum and a partial melt can be seen in the meanwhile due to compositional change of the specimen. And then, the edge of melt-zone, i.e., the solid-liquid interface was observed. The lattice fringes correspond to the projection of $(\bar{1}\bar{1}1)$ layers perpendicular to this print and the direction of the fringe lines is parallel to $[\bar{1}10]$ on the (111) planes. In this figure, atomistically flat solid-liquid interface can be seen clearly. The reason why the melt appeared at 523 K is due to the fact that the composition of the InSb specimen became In rich because of the selective evaporation of Sb at 523 K in the EM vacuum. This experimental result is consistent with the prediction of Jackson's theory which predicts the appearance of atomistically flat interface in the case of α larger than 2.

3. Generation of dislocations in growing crystal

As regards the generation of dislocations in the growth process of crystalline matters, the following facts were found by the present experiments. (a) It was found newly in a series of experiments on Al that a dislocation free region of about 1 mm thick was observed for the first time near the advancing solid-liquid interface during crystal growth with the temperature gradient of 10^3 K/m and at the growth speed of $9 \mu\text{m/s}$. This band-like region could be observed constantly near the advancing interface during crystal growth and in the crystal region behind this dislocation free region the generation of dislocations could be observed after the advancement of interface (cf. Fig. 19). This is an important new finding on the introduction of dislocations in as-grown crystals. (b) As was illustrated before in Fig. 16, when the growth

Fig. 18 High resolution electron micrograph of solid-liquid interface taken at 523 K. The lattice fringes correspond to the $(\bar{1}\bar{1}1)$ plane perpendicular to the point. The transition from solid to liquid is very sharp when the interface is parallel to $(\bar{1}\bar{1}1)$ ⁽⁶¹⁾.

Fig. 19 A series of X-ray topographs showing growth process of a Al single crystal (99.999% pure). Dislocation free zone along solid-liquid interface and generation of dislocations with the advancement of the interface behind the dislocation free zone⁽²⁴⁾⁽⁶²⁾.

rate was changed suddenly, a group of dislocations were introduced after a short latent time. (c) In the case of Ga, whose solidification temperature is close to room temperature, very slow cooling ($\sim 10^{-3}$ K/s) can be made rather easily near the solidification temperature. Taking this advantage of easy keeping of slow cooling, a big dislocation free single crystal of Ga (which shows expansion at solidification) were obtained by using a highly perfect seed crystal of Ga.

As regards the mechanism of intrinsic generation of dislocations in growth process of pure materials, two types of view were presented⁽⁶³⁾⁽⁶⁴⁾, namely, one being based on the segregation of super-saturated atomic vacancies and another being based on an atomic structure inherited from the melt. Taking into account of the aforementioned experimental results of (a) to (c) and these two ways of thinking, the origin of dislocations introduced in unidirectionally solidified crystals of Al and Ga in the present experiments is considered to be due to the segregation of super-saturated atomic vacancies. Since Al and Ga of very high purity were employed in the present experiments, it is unlikely to consider a role of impurities in generating dislocations. As regards the results obtained in the experiments of solid-liquid interface during dendritic growth of Al-Mg alloys⁽²⁴⁾, a high rate reduction of the density of dislocations by thermal-cyclic annealing after solidification (in Cu, Ag, Fe-3%Si, Si, etc.)⁽²⁴⁾⁽⁶⁵⁾, the generation of dislocations in dislocation free Si by annealing, the behavior of dislocations in Si at high temperatures⁽²²⁾, etc., only references are quoted at the end of this paper.

VI. Future of *In Situ* Experiments and Applied Research

For further applications of *in situ* experiments by

HVEM and real-time X-ray topography, it is desirable to have technological developments in (a) high speed and high resolution recording, (b) collection of three-dimensional information on static as well as dynamic phenomena. Use of an F-E gun is recommendable as a bright electron source for electron microscopy and construction of X-ray generator of higher power than our 90 kW is desirable for real-time X-ray topography to carry out dynamic observations of high speed phenomena. Development of TV image tube of higher gain and less lag is also desirable for recording fast changing phenomena. Use of high vision TV for EM observations and SATICON tube for real-time X-ray observation will provide recording of images with high speed and high resolution. Moreover, the scanning by two alternately inclined electron beams in STEM mode operation may realize the dynamic stereoscopic observation and provide more information on the generation and behavior of lattice defects⁽⁶⁶⁾.

Recently, the technology of electron beam holography has been developed in Japan and as its application, such experiments pertaining to (a) observation of magnetic flux in motion, (b) biomedical polymer and (c) three-dimensional structure of matters have been going on. The author do hope very original studies will be made by using those advanced technologies.

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