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Dislocation motion in semiconducting crystals under the influence of electronic perturbations

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Abstract. The radiation enhanced dislocation glide effect was confirmed in h-ZnO as well as in other semiconductors, which proved further the ubiquitous nature of this effect in semiconductors. Analysis of the radiation enhanced dislocation vibrations observed in the same h-ZnO led us to a tentative model that the effect is brought about by fluctuations of the charge state of point defects which exert electrostatic forces on the charged dislocations. The hydrogen-plasma enhanced dislocation glide was studied in Ge, SiGe and GaAs in addition to the previous reports on Si. The absence of the hydrogen effect in α -dislocations in n-GaAs and in thin films of SiGe is interpreted in terms of the soliton model, the essence of which is a strong binding between kinks and hydrogens.

1. Introduction

The electronic activities of dislocations have long been one of the main subjects in the research of extended defects in semiconductors, because of the technological importance and of academic concerns on the underlying physics. One of the interesting facts is that the glide mobility of dislocations strongly depends on the electronic states of the crystals. This is usually considered to be due to the presence of some electronic band-gap states associated with the dislocations that gives rise to non-metallurgical effects. Thus, as the two aspects of dislocation behaviour, static and dynamic, are closely related in semiconductors, the study of dislocation motion under the influence of electronic perturbations has been expected to give great insights into the electronic properties of dislocations as well.

The doping effect is the oldest issue among such effects of electronic perturbations, but in the present paper, we focus on three other phenomena, the radiation enhanced dislocation glide (REDG), the radiation enhanced dislocation vibration (REDV) and the hydrogen enhanced dislocation glide (HEDG). The first effect (REDG), most intensively studied, is induced by electronic excitations such as light illumination and electron irradiation. The second effect (REDV) is not really new but little is known about its origin. The REDV effect is frequently observed during direct observation of dislocations by transmission electron microscopy (TEM); parts of dislocation segments are seen to vibrate rather irregularly. Since this effect, as shown later, is caused by the electron beam irradiation used for the TEM observation as in the REDG effect, it seems reasonable to look for some connection to the REDG effect. The last effect (HEDG) was found rather recently by Yamashita *et al* [1, 2] for Si. Since hydrogen atoms, in many cases, are known to passivate electronic active centres in semiconductors [3], we could

regard the introduction of hydrogen atoms into semiconductors as a novel type of electronic perturbation to the dislocation dynamics.

In this paper, we first go over the main points of the REDG effect together with our very recent findings on ZnO, which now draws eager attention from a technological viewpoint as a promising material of wide gap semiconductor devices. We then report some detailed study of the REDV effect in ZnO and discuss the microscopic mechanisms that are conceivable at present. Finally we introduce some new results of the HEDG effects in Ge, SiGe and GaAs and argue the microscopic mechanisms in the light of theoretical calculations performed by Ewels *et al* [4]. The relevance to the knowledge so far accumulated is also discussed.

2. Radiation enhanced dislocation glides

The main features of the REDG effect so far found may be summarized as follows.

- (a) The effect is caused by electronic excitations (light, electron irradiation) that generate electron–hole pairs.
- (b) The effect is reversible; occurring only while the electronic excitation is sustained.
- (c) The dislocation velocity increases responding to the excitations in a short time ($\ll 1$ s).
- (d) The velocity exhibits an Arrhenius-type temperature dependence characterized by a lowered activation energy and a reduced pre-exponential factor.
- (e) The activation energy is independent of the excitation intensity.
- (f) The pre-exponential factor increases in proportion to the excitation power at low intensities but saturates at high intensities.

Among possible mechanisms of the REDG effect discussed in detail in a review article [5] by one of the present authors (Maeda), the most plausible, in the authors' opinion, is the recombination enhanced defect reaction mechanism which operates at some step(s) of the fundamental glide processes controlled by the Peierls mechanism. The Peierls mechanism in covalent semiconductors consists of kink-pair formation and kink migration. So far, the present authors have concluded that at least the formation of the smallest kink pair (SKP: the smallest metastable kink pair to be formed preceding the stable double kink formation) that must be enhanced for the REDG effect to occur in the macroscopic scale.

After the review article [5] on the REDG effect was written, similar effects were found in other materials including GaN [6] and Ge [7]. As noted before, the REDG effect tends to be prominent in semiconductors having wider energy gaps. In this respect, Ge and h-GaN are at two extremes (3.4 eV in h-GaN and 0.67 eV in Ge). Although previously the REDG effect was reported to be absent in Ge, its presence was confirmed at rather high excitation intensities (several μA beam current of 1 MeV electrons). For GaN in contrast, the REDG effect had been expected because of the wide band gap, although the electronic inactivity of dislocations had been postulated from their harmless behaviour in optoelectronic properties. Nevertheless, we found that the REDG effect is present in this crystal as well [6]. Thus, the ubiquitous nature of the REDG effect is now more widely accepted.

Most recently, we found the REDG effect in h-ZnO also. As the results are related to the REDV effect described in the next section, let us describe some details of the experimental procedure. The samples were prepared from reagent-grade (99.9%) hexagonal monocrystalline ZnO powder (Soekawa Rikagaku Co., Ltd) that were crushed in an agate mortar to introduce fresh dislocations and simultaneously obtain platelets thin enough for TEM observations. The samples mounted on a grid mesh holder were examined in a JEM-2010F operated at 200 kV.

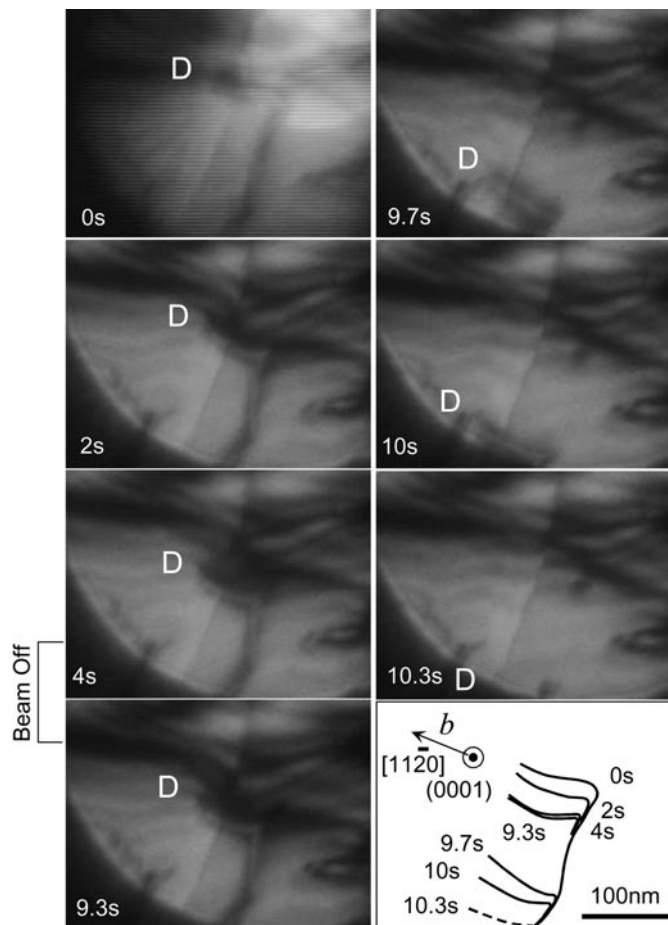


Figure 1. Sequential TEM images showing the REDG effect observed in a basal screw segment (marked by D) in h-ZnO. The test temperature and the accelerating voltage are room temperature and 200 kV, respectively.

The dislocation motion was recorded by videotapes. Since no external stress was applied, the stress driving the dislocation motion is considered to be stresses unintentionally built into the crystals when the specimens were prepared by crushing.

The TEM micrographs in figure 1 demonstrate the dislocation glide motion on the basal plane. As shown in figure 2(a), the interruption of the electron beam irradiation causes an immediate halt of the dislocation glide and on resuming the irradiation the dislocation restarts its glide exactly at the same position as before the intermission, exhibiting a similar behaviour as observed in other materials (GaAs [8, 9], ZnS [10], SiC [11–13] and GaN [6]) that show the REDG effect. The standard, gb contrast analysis indicates that the dislocation segments that exhibited the REDG effect are screw dislocations on the basal (0001) plane (figure 2(a)) and edge dislocations on a pyramidal (presumably $\{1\bar{1}01\}$) plane (figure 2(b)), both of a type (the Burgers vector being of $a/3\langle 11\bar{2}0 \rangle$ type, the same as that of the basal dislocations). No definite proof of the REDG effect in the prismatic $a/3\langle 11\bar{2}0 \rangle/\{1\bar{1}00\}$ system was found.

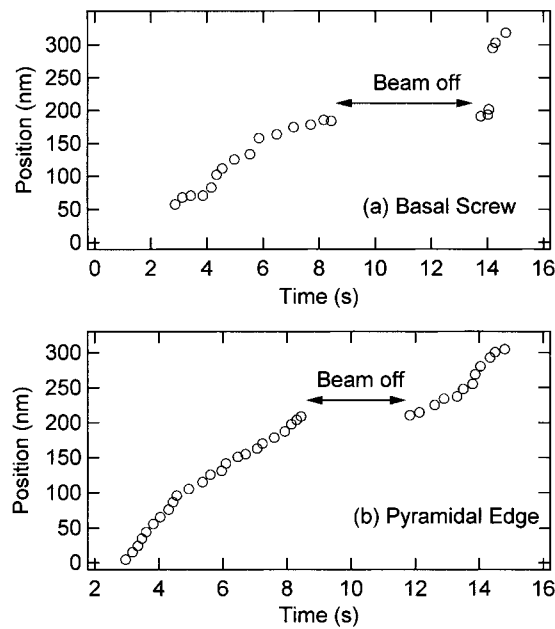


Figure 2. The effect of beam interruption on the dislocation motion, (a) for a basal screw dislocation and (b) for a pyramidal edge dislocation. Note the intermission of beam irradiation completely freezes the dislocation motion.

3. Radiation enhanced dislocation vibrations

Dislocations in ZnO were quite frequently vibrating with visible amplitudes during TEM observations. The anomalous vibrations are evidently caused by the irradiation of the TEM electron beam. Similar vibrations have been reported also for CdS [14], CdTe [15] and SiC [12] and hence we believe that this effect is rather common in semiconducting materials.

The experimental set-up is the same as in the study of the REDG effect. Since the average position of the dislocations did not change much as in the REDG effect, it was possible to conduct more systematic studies on the same dislocation segments. Figure 3 demonstrates the typical vibrations observed, in which the dislocation vibrates not necessarily on the whole segment between pinning points but is seen as if it were being 'whipped' by some external force. Detailed analysis of real time images of the dislocation vibrations recorded in videotapes revealed that

- (a) The REDV effect has been observed in basal dislocations and pyramidal dislocations, all of *a* type. We had no definite experience that the prismatic dislocations vibrate.
- (b) The fluctuation amplitude saturates as the radiation intensity increases (figure 4(a)).
- (c) Elevating temperature only slightly enhances the vibrations (figure 4(b)).
- (d) It takes some incubation time for the vibrations to start. After then the amplitude grows with irradiation time (figure 5).
- (e) Prolonged irradiation suppresses the vibrations.
- (f) As typically demonstrated in figure 5, the vibrations of dislocation segments are rather asymmetric: the fluctuations towards one side are more short lived.
- (g) The vibrations of adjacent dislocations are sometimes correlated; edge segments of different signs on the same dislocation line (figure 6) move in the same direction concomitantly.

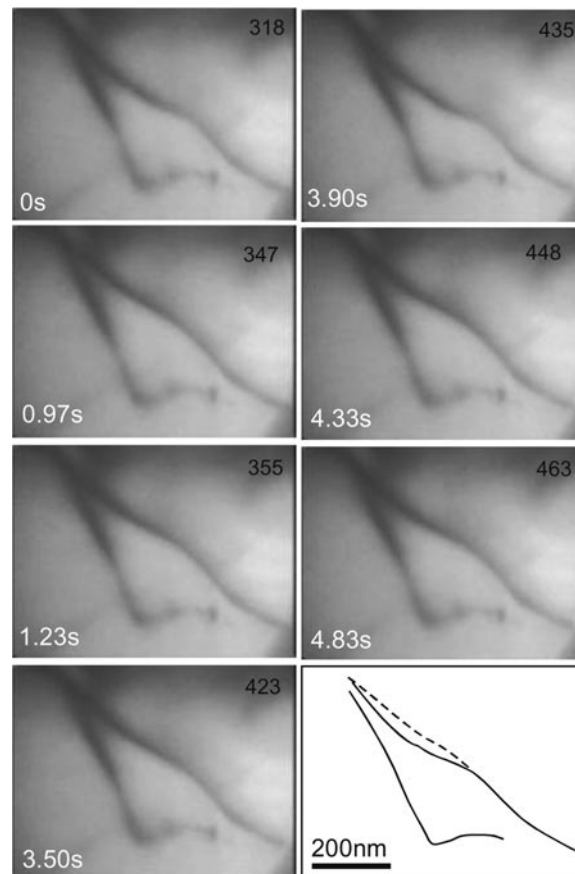


Figure 3. Sequential TEM images showing typical dislocation vibrations induced by the electron beam irradiation used for the TEM observations. The sketch at the bottom right indicates the dislocation line shape at the maximum amplitude.

- (h) Some of the dislocation segments that have been vibrating started to glide over a long distance once they escaped from the pinning points.

The last fact (h) above strongly suggests that the glide mobility was actually enhanced in all vibrating segments. Obviously, the REDG effect alone cannot explain the REDV effect since the REDG effect does not provide any straightforward reasoning for the dislocation motion in alternating directions. The possible heating of the specimen does not account for the REDV effect either, because the intensity dependence (b) shown in figure 4(a) exhibits a saturation, incompatible with the growth of the effect with the temperature (c) shown in figure 4(b).

In contrast to the REDG effect that starts very quickly after the commencement of electron beam irradiation (see figures 2(a) and (b)), the vibrations start rather gradually (figure 5). The displacement damage threshold in ZnO is known to be 310 keV [16], well above the 200 keV used for the TEM observations. The fact (e) above, however, may indicate an occurrence of subthreshold damage [16], though not visible in the TEM images, leading to pinning down of the dislocation segments. Therefore, the incubation time mentioned in (d) above may correspond to the time necessary for a sufficient accumulation of the point defects for the REDV effect to become detectable.

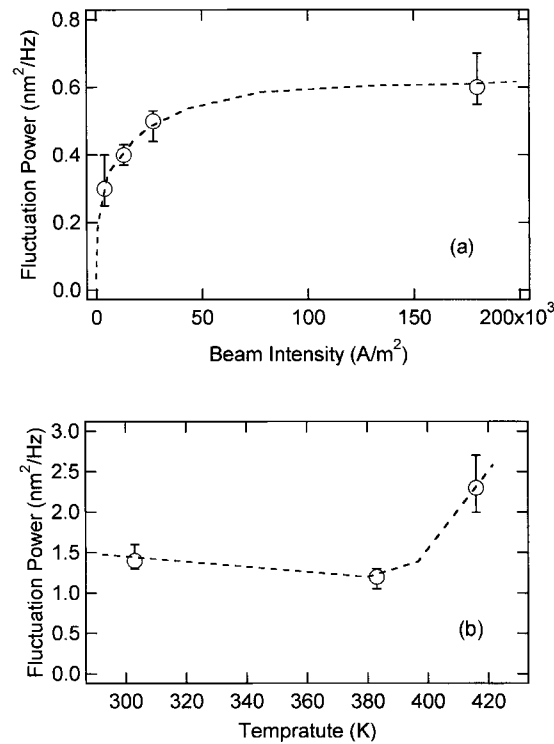


Figure 4. The intensity dependence (a) at room temperature and the temperature dependence (b) of the REDV effect studied for a pyramidal dislocation. The ordinates indicate the fluctuation power (displacement amplitude squared per unit width of frequency) at 3 Hz, which was chosen as a representative frequency in observed $1/\omega^2$ type spectra (not shown) where ω is angular frequency.

ZnO is one of the typical materials which exhibit the photoplastic effect (PPE) [17], a reversible increase of flow stress upon illumination of the crystal with a supra-band-gap light. This effect, different from the REDG effect in this sense, is considered to be caused by a change in the interaction between point defects and dislocations. Although not conclusive, the PPE possibly arises from a change in the charge state of the point defects or the dislocations due to light illumination leading to a change in electrostatic interaction between the point centres and the somehow charged dislocations [18, 19]. A charged centre located at a distance r from a charged dislocation can exert an effective shear stress

$$\tau^* = \frac{\rho e}{4\pi \epsilon b r^2} = \frac{f e^2}{4\pi \epsilon a b r^2}$$

on the dislocation. Here e is the electronic charge, ρ the dislocation charge density per unit length, b the magnitude of the Burger vector, a the separation of chargeable sites along the dislocation, f the occupancy of the chargeable sites and ϵ the static dielectric constant of the crystal. If we take tentative values of $a \sim b$ and $f \sim 0.5$ [20], the effective stress is ~ 1 MPa for a centre at a distance $r \sim 100b$ from the dislocation. This stress may be insufficient to cause dislocation motion but if multiple centres are involved, the stress can exceed a level sufficient for dislocation motion provided that the dislocation segment is mobile enough. The entity that changes its charge state can be the point defect or the dislocation. The strong correlated vibrations (g) seen in figure 6 are consistent with the former case if we assume that the edge segments of opposite signs are charged in different polarities and the point defect is situated in between.

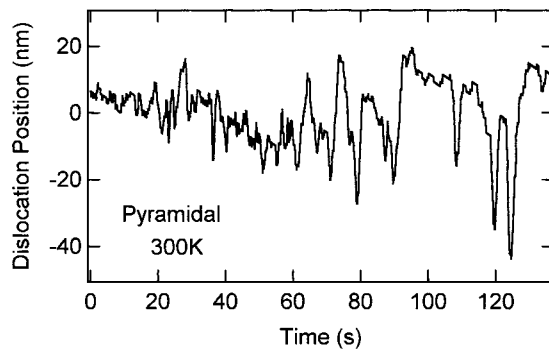


Figure 5. The growth of dislocation vibrations with time.

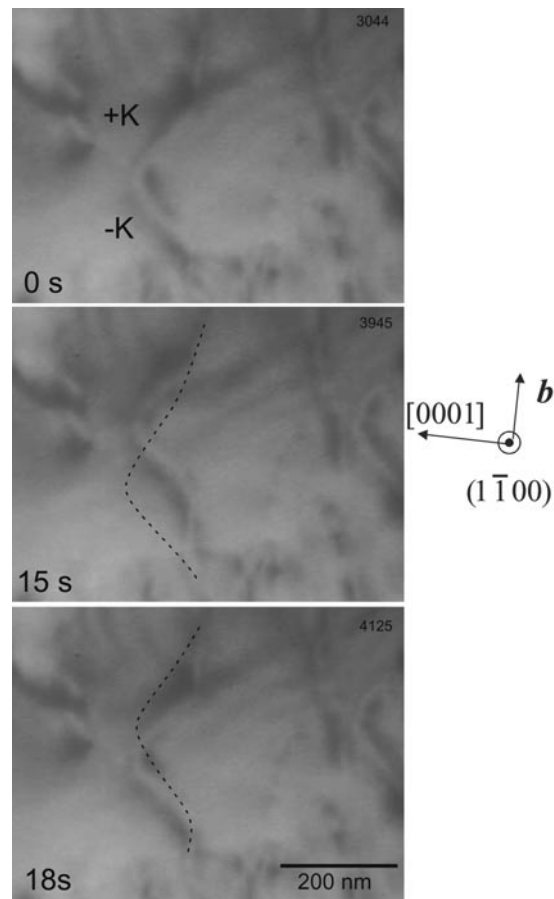


Figure 6. Sequential TEM images showing correlated vibrations of kinks of edge character with opposite signs. The kinks +K and -K occasionally moved together in the same directions. The broken curves indicate the dislocation positions in the previous frames.

The expected charge alternation with time is schematically illustrated in figure 7. The alternation rate and hence the vibration frequency should be increased in proportion to the minority carrier injection rate at low excitation levels, and at high excitation levels it should

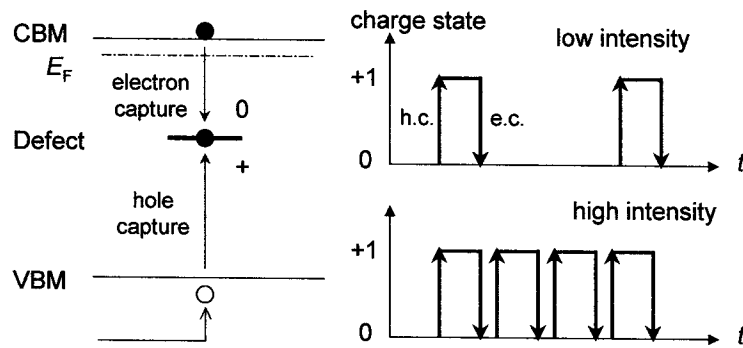


Figure 7. Temporal change in the charge state of a defect centre that gives rise to a band-gap state. Although in this illustration we suppose a donor-type defect in an n-type crystal, the qualitative feature is generally the same in different situations. The abbreviations e.c. and h.c. indicate electron capture and hole capture, respectively. Note the similarity in the dislocation positions in figure 5.

saturate to a value determined by the majority carrier density. This is consistent with the saturation behaviour (b) above and the saturation behaviour of the PPE in ZnO [21]. The temporal charge alternation illustrated in figure 7 also accounts well for the characteristic asymmetry in the wave form of the vibrational displacement (figure 5).

Thus, the REDV effect is considered to be brought about by a cooperation between the REDG effect and the fluctuations of the electrostatic interaction between the dislocations and point defects. When the dislocations are not charged, the REDV effect would not occur, which may be the reason why the REDV is absent in the prismatic system. In fact, the prismatic slip systems in ZnO [17] and CdS [22] do not exhibit the PPE and this was explained by the fact that the prismatic dislocations in wurtzite CdS are uncharged or only weakly charged [23]. However, as shown in the last section, prismatic dislocations in ZnO may not exhibit the REDG effect, which might account for the absence of the REDV effect in this slip system. Nevertheless, we should point out that the REDG effect is not a necessary condition for the REDV effect; the REDV effect could occur as long as the dislocations are mobile (thermally for example).

It was reported [20] that screw dislocations in II–VI compounds are generally uncharged. If the above interpretation is correct, the dislocation charges responsible for the vibrations should originate from the edge parts on the screw segments. In fact, the vibrations typically observed in globally screw orientation are characterized by a ‘whip’ motion, where the vibrations are achieved by the propagation of giant kinks of edge character.

4. Hydrogen enhanced dislocation glides

The experiments by Yamashita *et al* [1, 2] demonstrated that, prior to dislocation velocity measurements in Si, the exposure of the samples to hydrogen plasma results in a pronounced enhancement of the dislocation velocity. The main features of the HEDG effect in Si may be summarized as follows.

- (a) The pre-hydrogenation treatment before velocity measurements rather than a hydrogen environment during stress loading is essential for the effect to occur.
- (b) There is an upper limit in the pre-hydrogenation temperature above which the HEDG effect becomes weakened. When the pre-hydrogenation temperature is above the upper limit, the HEDG effect disappears sharply above a critical temperature (referred to as the dropping effect).

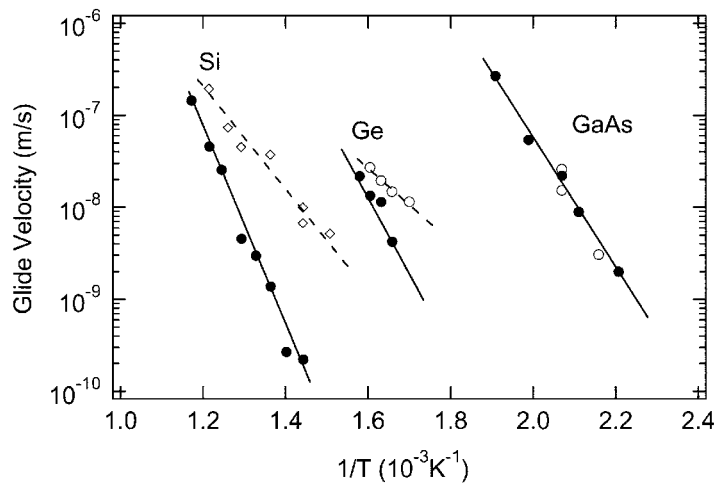


Figure 8. The dislocation velocity and temperature relations for three different materials, hydrogen-free (filled symbols) and pre-hydrogenated (open symbols). For the pre-hydrogenation conditions, see the text.

- (c) The velocity exhibits an Arrhenius-type temperature dependence characterized by a lowered activation energy and a reduced pre-exponential factor.

After the first observations in Si, the authors attempted similar experiments on some other materials such as Ge, SiGe and GaAs. The Ge samples were prepared from a p-type (111) wafer doped with In to a concentration of $3 \times 10^{14} \text{ cm}^{-3}$, and the GaAs samples from an n-type (111) wafer doped with Si to a concentration of $\sim 10^{18} \text{ cm}^{-3}$. The $\text{Si}_{1-x}\text{Ge}_x$ samples ($x = 0.08$, undoped) were thin monocrystalline 485 nm thick films epitaxially grown on Si(100) substrates. The samples containing dislocation sources introduced by scratching were exposed to a hydrogen plasma generated by a radio frequency electromagnetic wave source (13.56 MHz, 50 W). The condition of the pre-hydrogenation depended on the material (at 533 K for 45 min in Ge, for example), but was varied (623 K to 683 K for 3 h in GaAs) especially when the effect was not observed. The samples were then bent in vacuum with a given load (producing a resolved shear stress of 30 MPa for Ge, 228 MPa for SiGe and 20 MPa for GaAs) at an appropriate temperature for a prescribed duration. Finally the samples were etched to determine the displacement of the dislocations from the dislocation sources. For more experimental detail, the readers should refer to a separate paper [24].

Figure 8 compiles the new results together with the previous results in Si. The results of the SiGe alloy are not included to avoid complications. It is evident that 60° dislocations in Ge crystals exhibit the HEDG effect just as 60° dislocations do in Si. The apparent activation energy was found to decrease from a value of 1.7 eV in hydrogen-free samples to 1.1 eV in pre-hydrogenated ones. In contrast to Ge, however, the HEDG effect was observed neither for α -dislocations in n-GaAs nor for (both 60° and screw) dislocations in $\text{Si}_{0.92}\text{Ge}_{0.08}$ films. It should be noted that dislocations in the hydrogen-free $\text{Si}_{0.92}\text{Ge}_{0.08}$ crystals of such low Ge content exhibit a dislocation mobility very close to that of bulk crystals of pure Si. This means that the absence of the HEDG effect in the SiGe films may indicate the absence of the effect in pure Si when the crystal thickness is small.

As proposed in our previous paper [2], we could conceive of several mechanisms for the HEDG effects. One is the hydrogen-catalytic effect, which enhances the kink migration (referred to as the kink migration enhancement model). Another is the hydrogen termination

of dangling bonds associated with the kinks, which effectively reduces the kink formation energy (referred to as the kink formation enhancement model). In accord with these ideas, Ewels and his coworkers [4] carried out first-principles calculations of the formation energies of kinks and their motional barriers for various cases, for kinks free of hydrogen and hydrogen-attached kinks. Heggge and Jones [25] claimed previously that the smallest kink pairs (SKPs) are most easily generated at soliton sites where the bond reconstruction gets out of phase leaving a dangling bond. According to recent calculations [4], a hydrogen atom terminates the dangling bond at the soliton to lower the electronic energy associated with the dangling bond. More interestingly the soliton–hydrogen complexes thus formed can serve as ‘spontaneous’ nucleation sites for the smallest kink pair formation with a ‘negative’ formation energy. The dropping effect mentioned above (b) suggests the presence of some defects that could trap hydrogen atoms with a binding energy. Analysis of the experimental drop in the dislocation velocity against temperature in hydrogenated Si indicates that the binding energy is as large as 3.6 eV [2]. Although qualitatively the soliton can be a candidate for such a hydrogen-trapping centre, the calculated binding energy, 2.56 eV [4], is not so large. Nevertheless, as the calculations are done for 90° partials, and not for 30° partials that are considered to govern the total dislocation motion, the quantitative disagreement might not be serious. From these considerations, we could conceive of the following scenario:

- (1) Hydrogen atoms from the plasma incorporated into the crystals are stored at some sites along the dislocation line with a large binding energy.
- (2) Hydrogen atoms supplied from the deposit assist to form soliton–hydrogen complexes by virtue of the effectively lowered soliton formation energy.
- (3) Thus, the solitons are more abundantly formed than in hydrogen-free crystals.
- (4) The SKP formation takes place spontaneously at the soliton–hydrogen complex.
- (5) As a result, the dislocation line advances with a larger velocity.

This picture is consistent with the predicted kink pair formation model and appears to work quite well.

However, there is an embarrassing fact that should be viewed seriously. As mentioned before, the HEDG effect is absent (or very weak) in thin films of SiGe. At present, we have two hypothetical explanations for this fact. One is that hydrogen in a concentration sufficient for the HEDG effect to occur cannot dissolve into heteroepitaxial thin films stressed in lateral compression. Another is that the HEDG effect is absent in crystals of small sizes, as suggested already. The latter hypothesis is not an *ad hoc* idea but based on our previous study of dislocation glide in heteroepitaxial thin films of SiGe on Si substrates [26]. Based on the experimental fact that the glide activation energy in SiGe of low Ge contents is invariable, independent of the film thickness which determines the length of the dislocations whose velocity was measured, we proposed that some point obstacles, that are present only on dislocation lines of length longer than a critical value, block the kink migration along the dislocation line but this blocking activity diminishes when another kink of the opposite sign arrives at the blocking site [27]. If we adopt this hypothesis, the absence of the HEDG effect in thin films should be ascribed to the absence of the point obstacles on short segments of dislocations in such films.

The problem that has remained unsolved in the point obstacle model is on the microscopic origin of the ‘point obstacles’ which must be intrinsic in nature. One conceivable model for the intrinsic ‘point obstacles’ is the transformation of two kinks into a single superkink (superkink model). Our recent high-resolution electron microscopy (HREM) study [28], in which partial dislocation line shapes were imaged in a plan-view geometry with atomic resolution, showed that some of the kinks on 30° partials combine to form superkinks of a

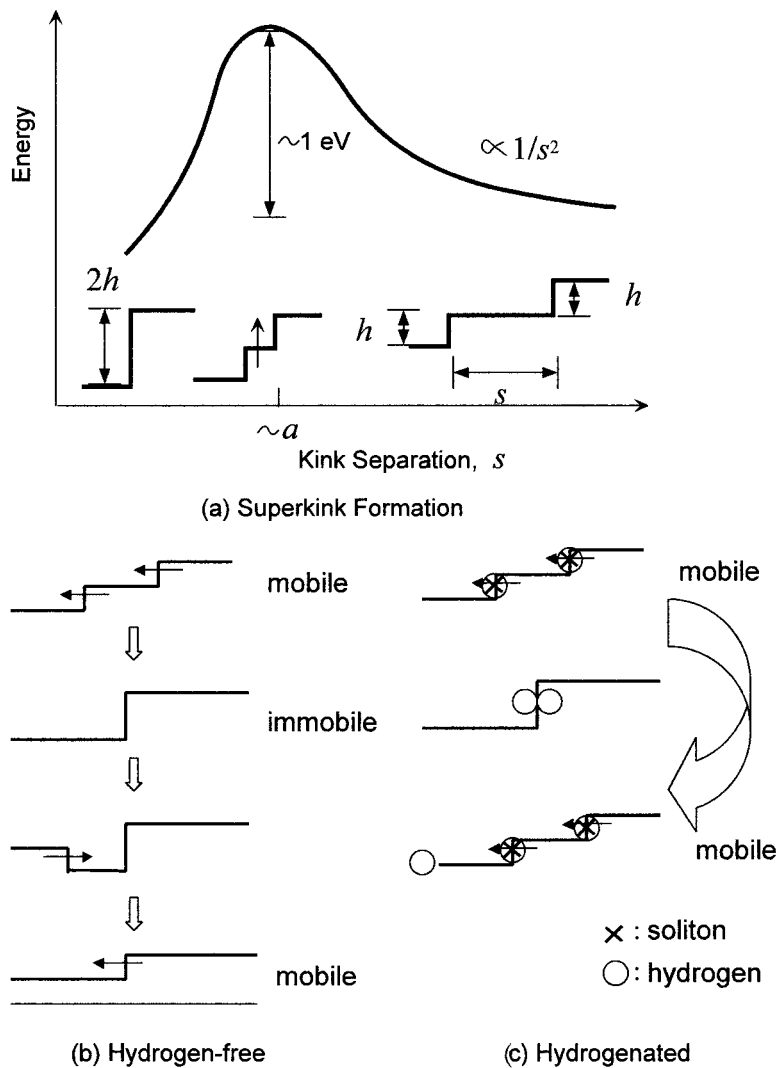


Figure 9. Superkink model: (a) the energy barrier for superkink formation, (b) the superkink formation effectively blocking kink migration and (c) hydrogen atoms suppressing (circumventing) the blocking effect because breaking the complexes with solitons costs energy.

double height. This fact is not surprising if we consider that two kinks of the same sign on approaching each other closely could lower their total energy by merging to a single superkink. Although there must be an energy barrier for the formation of a superkink due to the kink–kink repulsion (figure 9(a)), the barrier height should be as moderate as ~ 1 eV in Si. Surmounting this barrier height thermally would produce the ‘point obstacles’ the separation of which is experimentally estimated to be $\sim 1 \mu\text{m}$. If the superkinks are assumed to be far less mobile than normal kinks of a single height, the transformation of two kinks into a single superkink will act as an apparent ‘point obstacle’ against the kink migration. As illustrated in figure 9(b), the arrival of another normal kink of the opposite sign at the superkink resumes the mobility of the kinks so that the ‘point obstacle’ will effectively disappear.

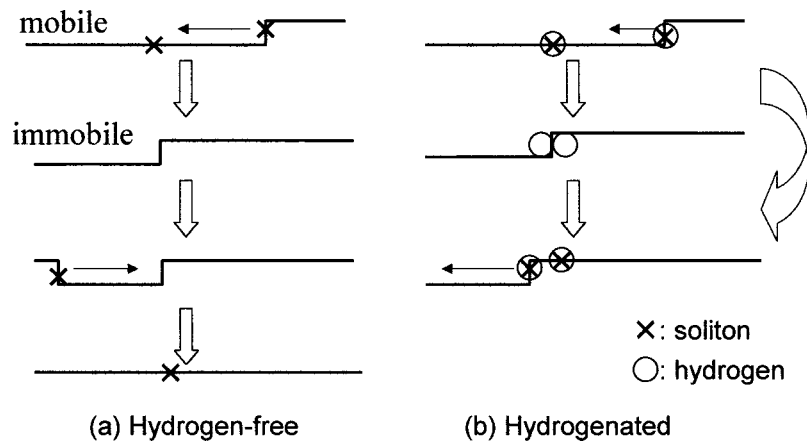


Figure 10. Soliton annihilation model: (a) when a kink–soliton complex encounters a soliton, the kink transforms to a kink without a soliton, which is immobile and hence blocked. Once another kink–soliton complex arrives at the blocked kink, the blocking state disappears. (b) When hydrogenated, the blocking effect is suppressed (circumvented) to avoid the energy cost for breaking kink–soliton complexes.

An alternative model for the intrinsic ‘point obstacles’ is the transformation of the kink structure (soliton-pair annihilation model). Let us imagine a situation as illustrated in figure 10(a) where a kink–soliton complex (a Hirsch kink with a dangling bond) encounters a soliton on the dislocation line. Once the soliton pair recombines to annihilate, the kink without a soliton loses its mobility [29]. The immobilized kink disappears, however, when another soliton–kink complex arrives at the former kink.

When hydrogen atoms are present, the calculations by Ewels *et al* show that the soliton–kink complex favours the form acting as a trimer with a hydrogen atom. In the framework of the first superkink model (figure 9(c)), the cause of the HEDG effect is ascribed to the suppression of the superkink formation, because if the solitons are annihilated on the merge of two soliton–kink–hydrogen trimers (in this case we have to assume soliton–kink complexes), two hydrogen atoms must be released from the solitons, which costs an excess energy corresponding to twice the soliton–hydrogen binding energy (~ 5 eV). Therefore, the blocking mechanism will not operate. The cause of the HEDG effect in the second soliton-pair annihilation model (figure 10(b)) is ascribed to the suppression (circumvention) of the soliton-pair annihilation reaction for the same reason as above on encounter of a soliton–kink–hydrogen trimer with an approaching soliton–kink complex.

Both models assume the presence of the solitons as a requisite concept and are therefore applicable only to elemental crystals (Si and Ge). Since the first-principles calculations of Öberg *et al* [30] showed that α -dislocations in GaAs are only weakly reconstructed, we could account for the absence of the HEDG effect in α -dislocations in GaAs as due to the absence of solitons necessary for the above mechanisms to operate.

The similarity in the temperature dependence of the HEDG effect and that of the REDG effect tempts us to consider that both share a common mechanism. Especially in Si, the reduced activation energies are close to each other (1.2 eV in HEDG and 1.2 ± 0.1 eV in REDG). However, the activation energy in Ge is 1.1 eV in the HEDG effect [24] while it is 0.7 eV in the REDG effect [7], differing more than the experimental error. This implies that the electronic excitations in the REDG effect affect not only the SKP formation but also some other processes such as kink migration. Whether or not the ‘point obstacles’ to the kink

motion are involved in these processes is not yet answered and is still the subject of future studies.

Acknowledgments

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