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High-voltage electron microscope studies of low-temperature radiation damage in silicon

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Abstract. Thin foils of high-purity silicon were investigated in a high-voltage electron microscope at temperatures T between 15 and 60 K at beam voltages between 400 kV and 650 kV (ie, at voltages high enough for producing atomic displacements). The following results were obtained: between 10 K and 35 K we observed the formation of point defect clusters of so far unknown type and with diameters d of about 50 Å. These clusters are formed in p-type material at a significantly lower electron dose than in n-type material. At $T > 35$ K the number of clusters decreases and their sizes grow. At the same time the spatial distribution of the clusters becomes inhomogeneous, the major part being localized near the upper foil surface. In cases where the defect nature could be analysed in more detail, all of them were found to be of interstitial type.

In a high-voltage electron microscope (HVEM) the energy of the imaging electrons is high enough to displace crystal atoms from their lattice sites, thus producing vacancies and interstitials. If these point defects agglomerate to point defect clusters of diameters $d \gtrsim 25$ Å, they become visible on the microscopic images. This technique for the study of radiation damage has so far been mainly applied to metals (Wilkens and Urban 1974). The present paper reports preliminary results obtained on high-purity n- and p-type silicon using a Hitachi HU 650 electron microscope at voltages up to 650 kV. Previous HVEM irradiation experiments have been performed at temperatures above 70 K (Thomas 1968, for a review see Nelson 1972). In the present work silicon was irradiated for the first time at temperatures down to about 10 K in a HVEM using the cooling stage described by Heide and Urban (1972).

The specification of the specimens are: {111} orientation; oxygen content $\approx 10^{15}$ atoms/cm^3, carbon content $\approx 10^{16}$ atoms/cm^3, room-temperature resistivity $\rho = 50\ \Omega$ cm in the case of n-type material (phosphorus-doped) or $\rho = 10\ \Omega$ cm for the p-type material (boron-doped). A thin layer of aluminium (about 100 Å thick) was evaporated onto the surface of the specimen to avoid accumulation of electrical charge in the sample during irradiation.

In order to obtain a high current density during irradiation the electron beam was focused to an illuminated area of about 2 μm diameter on the specimen. After a sufficient time of irradiation point defect clusters became visible within the area of the illuminated spot. Figure 1 shows as an example a specimen of n-type silicon irradiated at 35 K up to a dose of about 5×10^{22} electrons/cm^2. The micrograph was taken using the so-called weak-beam technique (ie, by dark-field microscopy using a weakly excited Bragg

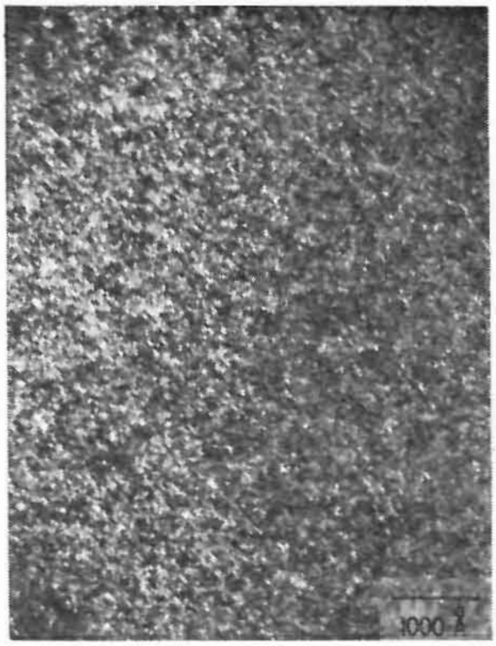
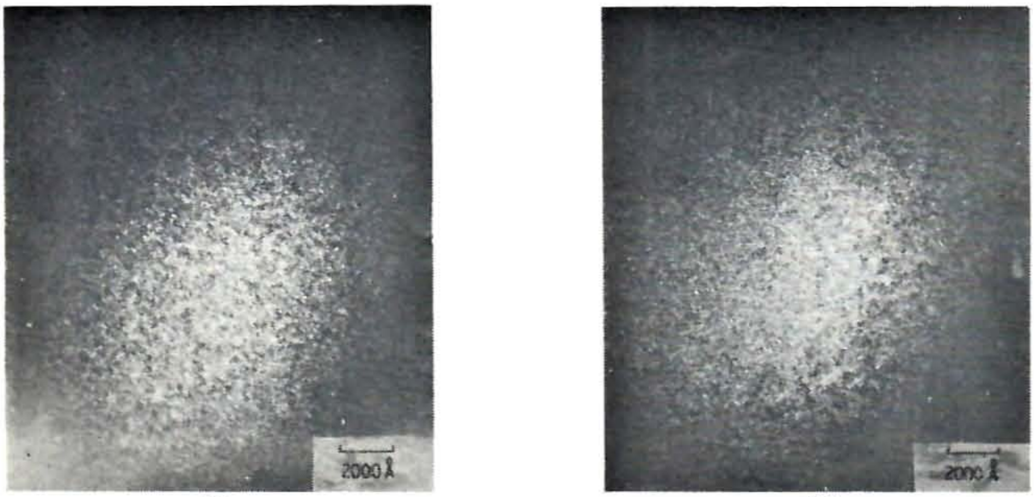


Figure 1. Weak-beam micrograph of 50 Ω cm n-type silicon irradiated with about 50×10^{22} electrons/cm² at 35 K.

reflection). The weak-beam technique has the great advantage of high resolution, but under the present circumstances does not allow one to obtain information on the nature of the defects. For a more detailed description the reader is referred to Häussermann *et al* (1973), and Goringe and Ray (1974). The conventional bright- or dark-field microscopy, which is often used to obtain information on the type of clusters is unfortunately not applicable in the present situation. The reason is that the small size and the small mean distances between the clusters prevent the observation of normal black–white contrast.

The most striking feature of figure 1, which is also representative of the damage visible at irradiation temperatures below 35 K, is the homogeneous distribution of the clusters within the irradiated area. This is especially well seen in figure 2 (figure 1 is an enlarged part of figure 2(*b*)). Since the electron beam has a nearly gaussian-shaped intensity distribution, the dose in the outer parts of the irradiated area of figure 2 is much lower than in its centre. The clusters are relatively small, their mean diameters are about 50 Å. From a comparison of figures 2(*a*) and 2(*b*) the change in the damage as a function of dose can be clearly seen. Above a critical dose the clusters no longer grow, the damage reaches a saturation. Only in the outer parts of the irradiated area, where the dose was too small to reach saturation, did further irradiation produce new clusters. Therefore, we conclude that the damage in the saturated area is almost independent of the flux density of the electron beam. This might be regarded as an indication that the diffusion coefficient related to the point defects forming the clusters is a function of the flux density of the electron beam.

Figure 3 shows the damage obtained after irradiation at 40 K to a dose of about 5×10^{22} electrons/cm². The striking difference between the damage at 40 K and at lower temperatures can be seen clearly. The total number of clusters is considerably smaller, and the distribution is nonuniform. The average cluster size is larger than at lower



(*a*) (*b*)

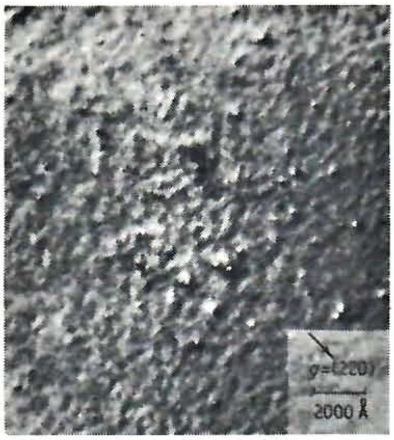
Figure 2. Dose dependence of the damage structure in 50 Ω cm n-type silicon irradiated at 35 K. (*a*) Dose ~ 3×10^{22} electrons/cm^2. (*b*) Dose ~ 5×10^{22} electrons/cm^2.

temperatures. A number of bigger clusters have been formed. Further experiments have shown that this damage structure occurs between about 35 and 45 K.

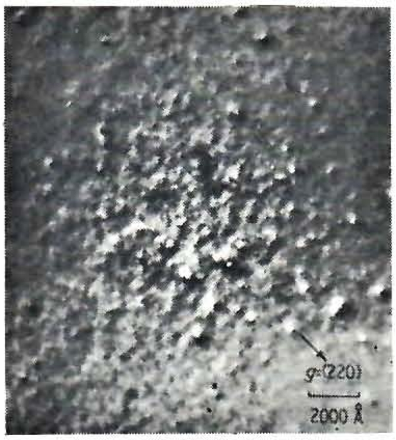
Figure 4 shows the damage structure after irradiation up to a dose of about 5×10^{22} electrons/cm^2 at a temperature of 60 K. (It should be noted that the big white spots in figure 4 and the big black–white contrasts in figure 5 have not been introduced by irradiation, but originate from sample preparation.) Only a small number of clusters have been produced, which in dynamical dark-field images exhibit black–white contrast.

Figure 3. Weak-beam micrograph of 50 Ω cm n-type silicon irradiated with about 5×10^{22} electrons/cm^2 at 40 K.

Figure 4. Weak-beam micrograph of 50 Ω cm n-type silicon irradiated with about 5×10^{22} electrons/cm^2 at 60 K. The big white spots are not due to irradiation but to sample preparation.



(a)



(b)

Figure 5. Dynamical dark-field micrographs of 50 Ω cm n-type silicon irradiated with approximately 5×10^{22} electrons/cm^2 at 60 K. The big black–white contrasts are not due to irradiation but to sample preparation. (a) Immediately after irradiation at 60 K, (b) after warming to room temperature.

Figure 5 shows the same area using dynamical dark-field imaging conditions. Figure 5(a) has been taken at the irradiation temperature, figure 5(b) after warming up to room temperature. Two essential features may be noticed: firstly, all black–white vectors (ie, the vectors pointing from the black to the white part of the contrast dots) lie in almost the same direction. Secondly, warming up the specimen causes no significant changes in the damage. This is also true for the damage obtained at all other temperatures.

Another important point we have investigated is the spatial distribution of the clusters. The observation that all black–white vectors point in roughly the same direction indicates that all the clusters are located at roughly the same depths, with respect to the foil surface. This interpretation is fully confirmed by stereo-micrographs. In particular, stereo-micrographs of specimens irradiated at 40 K show very clearly that most clusters are localized in a layer, some 100 Å thick, close to the upper foil surface (ie, the surface through which the electron beam enters). A small number of clusters are found in the bulk of the crystal, randomly distributed with respect to their depth. This result was obtained in three different specimens in both n-type and p-type silicon. If the irradiation temperature is increased, no clusters are seen in the bulk, and all of the visible damage is concentrated in a layer near the upper foil surface. Indications of a similar effect were mentioned by Thomas (1968) and Matthews and Ashby (1972) after room-temperature irradiation in n- and p-material. The depth distribution at temperatures below 35 K is not yet known, since the high density and the smallness of the defects do not permit an evaluation of stereo-micrographs.

After determining the depth location of the defects, it is possible to find out whether they are of the interstitial or vacancy type by analysing the black–white contrast (for the methods employed see, for example, Wilkens *et al* 1973). This method has been successfully applied for damage patterns obtained above 35 K. Without any exceptions all analysed black–white contrasts were found to originate from interstitial-type defects, presumably dislocation loops on {111} planes. It has not been possible to analyse the nature of the defects produced at $T \lesssim 35$ K, because they neither give rise to black–white contrast nor is their depth position known.

Irradiation of p-type silicon leads to quite similar damage structures and spatial distributions of the clusters as in n-type material. However, in order to obtain the same defect density by irradiation at 20 K as in n-material, a dose of about two or three times smaller than in n-type material is necessary. A further interesting observation which was made in p-material, but most likely is also true in n-material, is the dependence of the damage on the beam voltage. If the voltage is lowered from 650 to 600 kV the damage structure remains the same. However, the number of clusters produced at a given dose at 600 kV is much lower than at 650 kV. This is demonstrated in figure 6. Both 'spots' are obtained with a fully focused electron beam and are therefore directly comparable. (The apparent difference in brightness of the irradiated areas in figures 6(*a*)

(*a*) (*b*)

Figure 6. Weak-beam micrographs of the damage structure in 10 Ω cm p-type silicon at 20 K. The big white and dark spots are not due to irradiation but to sample preparation. (*a*) $E_{\text{electrons}} = 600$ keV, dose $\sim 5 \times 10^{22}$ electrons/cm^2, (*b*) $E_{\text{electrons}} = 650$ keV, dose $\sim 3{\cdot}6 \times 10^{22}$ electrons/cm^2.

and (*b*) is due to photographic processing.) This voltage dependence is unexpected since the threshold voltage belonging to a threshold energy of 13 eV (Corbett *et al* 1972) is about 140 kV.

Figure 6 also shows that the damage structure is similar to that in n-material (see figure 2).

Firstly, we will discuss the dose and voltage dependence. The doses required for obtaining observable damage are unexpectedly high compared with metals. From this and the unexpected voltage dependence we conclude that only a small fraction of the defects created by the electron beam participate in the clustering process. Presumably most of the Frenkel pairs created by the electron beam recombine spontaneously or under the influence of the electron beam. Consequently, only Frenkel pairs whose interstitials come to rest far away from their vacancies, or defects trapped by impurities may participate in the clustering process. In this context it should be mentioned that in germanium which we have irradiated with 650 kV electrons in similar experiments as in silicon no observable defects could be found.

The distinct white dots observed in the weak-beam micrographs in the temperature region down to 10 K have to be attributed to small well defined point-defect clusters. In order to obtain such clusters an athermal migration of point defects must have taken place. (From contrast arguments we can exclude the so-called Lück–Sizmann effect (Lück and Sizmann 1964), ie, clustering due to statistical fluctuations of the point-defect density at sufficiently high doses, as the main clustering mechanism.) Among the athermal diffusion mechanism, which could give rise to the observed damage pattern are the radiation-induced diffusion according to Urban and Seeger (1974) (see also Urban 1974) or the ionization-enhanced athermal diffusion as suggested by Bourgoin and Corbett (1972, 1974). We will not discuss any one mechanism in detail, because a distinction between these mechanisms, based on the damage observed, has not yet been possible.

The observation that clustering occurs faster in p-type silicon than in n-type silicon suggests that electronic properties play a role in the athermal diffusion mechanism, but this does not yet allow the identification of a particular mechanism.

Whereas the homogeneous distribution of the clusters below 35 K indicates that the diffusion coefficient may be related to the flux density of the electron beam (this is true for all athermal diffusion mechanisms), the change in the damage structure above 35 K might be attributed to a thermal activated migration of point defects. In this temperature range the nucleation may well be heterogeneous. This may be the explanation for the localization of the clusters near the upper foil surface. It is conceivable that the required concentration of impurities have been 'shot in' from the oxide layer or from surface contaminations by the interaction with the electron beam. Such a mechanism would also explain the high doses required to obtain visible damage.

Acknowledgments

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