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CURRENT PROBLEMS IN THE THEORY OF RADIATION DAMAGE

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CURRENT PROBLEMS IN THE THEORY OF RADIATION DAMAGE

D. K. Holmes

Abstract

The present status of the theory of radiation damage is reviewed. Emphasis is placed on the qualitative discussion of the points in the theory which appear to be most critical for a further advance in our understanding of the fundamental damage process. In particular, the need for a better knowledge of the effective interatomic potential is shown.

CURRENT PROBLEMS IN THE THEORY OF RADIATION DAMAGE* 1

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The exposure of metallic solids to high energy radiation results in the formation of relatively stable defect structures which may be removed only by appropriate annealing. The basic unit of the defect structure is the "displaced atom", i.e., an atom which has been removed from its normal lattice site and deposited at some abnormal position in the crystal. Actually, this process leads in general to two distinct defects, sometimes called a "Frenkel pair", a vacancy (the vacant lattice site) and an interstitial atom (the displaced atom at a non-lattice site). Of course, the radiation will usually disrupt the electronic structure of the solid as well as the atomic structure. In nonmetallic solids (ionic crystals, semiconductors, organic crystals) electronic changes may be semipermanent and of considerable importance (quite possibly resulting in the actual displacement of atoms as the crystal reestablishes local charge neutrality). However, in metals, with highly mobile conduction electrons, the energy deposited with the electronic system is quickly dissipated, usually leaving the crystalline structure unaltered. Consequently, in this review attention will be focused on atomic displacement processes.

The interaction mechanism by which external radiation produces a displaced atom varies, depending on the particular type of radiation.

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A further variable is the kinetic energy of the displaced atom, which may take values over a considerable range depending on the details of the interaction process. This original displaced atom is usually called the "primary displaced atom" or just the "primary"; if its energy is high, it may have important secondary effects in the solid, which will be discussed later. The most important of the types of radiation and the interactions by which they produce energetic primary displaced atoms are the following:

<u>Type of Radiation</u>	<u>Interaction with the Solid</u>
Heavy charged particles (protons, deuterons, alphas, heavier atomic nuclei, fission fragments)	Classical Rutherford scattering with the Coulomb field of the nuclei
Fast electrons	Relativistic Rutherford scattering (with the nuclei)
Fast neutrons (energies above a few hundred ev)	Elastic and inelastic scattering (nuclear interaction)
Thermal neutrons*	Capture, with subsequent nuclear recoil from gamma or beta-ray emission
Gamma rays	Photoelectric or Compton processes with the electrons of the solid which subsequently undergo relativistic Rutherford scattering with the nuclei

These interactions have been subjected to considerable investigation and may now be taken to be quite well understood. That is, for a solid exposed to a flux of each type of radiation the amount of energy deposited on the atoms (per unit volume per unit time) can be accurately calculated.

* Thermal neutrons will in most cases also cause nuclear transmutation, which will effectively introduce foreign atoms into the crystal. This effect can be kept to an essentially negligible level in most research cases and will not be discussed in this paper. Important effects can, of course, be produced by thermal neutron capture in practical cases. An example is the nuclear reaction caused in B^{10} which results in significant concentrations of He gas in steels (see the discussion by Makin).

It is at this point that currently important theoretical questions in radiation damage arise. How does an energetically moving atom dissipate its energy as it slows down in the crystalline environment?

In a general way the form of the answer to this question is well established. The primary atom will collide with other lattice atoms as it moves and will succeed, sometimes, in displacing them from their lattice sites. These "secondary atoms" may in turn displace further atoms, resulting in a "displacement cascade". Eventually all the moving atoms will come to rest (or at least find their kinetic energies reduced to the level of the energy of thermal motion in the solid) in some region about the point of creation of the original primary atom. At this time (of the order of 10^{-13} to 10^{-14} seconds after the creation of the primary) the energy originally deposited by the radiation on the primary will be found to have been mostly converted into heat spread over the domain of the displacement cascade, with a small part appearing as formation energy of the Frenkel pairs. Another small part of the original energy will be found associated with excited electrons, while a final portion will be carried away by special coherent motions of the lattice atoms. Eventually, virtually all of the energy will be dissipated as heat conducted away from the perturbed region which will return to the temperature of the whole solid with the inclusion of a non-equilibrium concentration of vacancies and interstitials, which are essentially "frozen in".

After a time of exposure to high energy radiation, a sample of metal will be found to have a number of such regions of damage which will be of a semipermanent nature and will alter, to some extent, all measurable properties of the sample. For the most part, all of this damage can be removed by heating to such a temperature that the individual defects,

vacancies and interstitials (or their clusters), have sufficient mobility to get together and annihilate each other, thus restoring the perfect lattice.

In detail, however, the present status of our understanding of the damage process is not satisfactory. Consider the problem of the number of Frenkel pairs present in the crystal after a given irradiation. Suppose the irradiation temperature is sufficiently low that there is no appreciable thermal mobility of vacancies or interstitials. The experimentally determined quantities are the total flux of radiation and the resultant change in one or more physical properties. From knowledge of radiation interactions, the amount and distribution of energy given to atoms of the solid may be correctly calculated, but it is difficult to proceed from this point to a reliable estimate of the total number of defects to be expected. The difficulty comes in the evaluation of the portion of the energy of a primary atom which is expended in a "frictional" manner, eventually appearing as heat rather than as displaced atoms. Further, it is equally difficult to work backwards from the known property change. The theory of defects in solids is not sufficiently advanced to give values of property contributions of vacancies and interstitials to better than a factor of two or three, and is mostly much poorer than that.

In this review, attention will be concentrated on the production of defects rather than on their property contributions. The aim is to reveal the sticky points in the theory which are currently being investigated in order to allow a much more accurate understanding of the formation and structure of a defect cascade.

ELASTIC COLLISIONS BETWEEN ATOMS

The basic process in the formation of the defect cascade is the interaction of a moving atom with the atoms of its lattice environment. One well-established and important interaction leads to elastic collisions, i.e., in one extreme approximation the moving atom may be regarded as an elastic ball traveling through an open collection of identical balls, every so often bumping into one of them, thus not only changing its trajectory but also transferring some of its kinetic energy to the struck ball (atom), which in turn becomes a moving ball. In truth, of course, the atoms do not interact as elastic spheres but have some relative potential energy of interaction which increases as the atoms come closer together, as schematically shown in Fig. 1. Thus, in the energy range of interest here, the interaction potential is positive, so the atoms repel each other. If this potential were known, because of the relatively large mass of the atoms, the entire history of the movement of the atom through the lattice, involving many such binary collisions, could be exactly solved by classical mechanics. And if such elastic interactions were the only physical processes occurring, it would be possible to calculate exactly the expected total number of displaced atoms in the cascade.

Of course, this implies knowing the potential over a large energy range, in general, all the way from the energy of the primary, possibly hundreds of kev, down to values near that of the binding energy per atom of a solid, some few ev. In fact, the low energy end is especially critical for the calculation, as may be seen by approximating the elastic collisions as an equal sharing of energy between the two atoms. Then a primary of energy E would, on the first collision, yield a primary and a

secondary each of energy $E/2$; the next collisions of each of these would result in four moving atoms, each of energy $E/4$, and so on, until the n -th collision stage is reached, at which there are $2n$ moving atoms each having energy $E/2n$. This process terminates when the kinetic energy is too low to allow an atom to leave its lattice site. Under the most favorable circumstances an atom must push through its neighboring atoms at a distance of something like half the normal lattice spacing in order to leave its lattice site permanently. The situation is depicted schematically in Fig. 2, which shows the moving atom climbing over a potential saddle-point half-way between two atoms, which, at the energies involved, do not have time to move apart to lower the effective barrier. The energy an atom of a lattice would require to squeeze through the shell of neighboring atoms in the easiest direction is called the "threshold energy". It is characteristic of each solid and is believed to have values in the range of 10-40 ev. In any case, it is clear that if a moving atom strikes a lattice atom and shares its energy so that each has energy below the threshold energy, then further displacement-type motion is impossible, and the two atoms will come to rest. This gives rise to an interstitial atom, since there are two atoms for one lattice site. This gives a basis for an elementary estimate of the number of displaced atoms to be expected from one primary of energy E . When a moving atom has an energy just below $2E_d$, it is no longer able to produce other displaced atoms. If an energy just above E_d is transferred to a lattice, the original atom will be left with an energy below E_d and will fall into the empty lattice site (replacement). On the basis of the very simple "equal-sharing" model at the n -th collision stage, there are 2^n displaced atoms; let these each have energy

$2E_d$, so that no further displacements are produced. Then the total number of displaced atoms in the cascade may be approximated as

$$n_d = 2^n = \frac{E}{2E_d} \quad (1)$$

Improvements of this simple model are based on two criticisms:

(1) The scattering law is more complicated than a single equal sharing of energy.

(2) The apparent potential well is much more complicated than implied in the derivation of Eq. (1), in which it is effectively assumed that the energy E_d is a sharp bounding energy between escape and non-escape of the struck atom.

Consider the first point. On the basis of an interatomic potential such as shown in Fig. 1, the kinetic energy (T) transferred to the struck atom (by a moving atom of energy E) has the general type of distribution, $K(E, T)$, as shown in Fig. 3 (equal mass atoms assumed). Thus, low energy transfers are more prevalent, which leads to nonproductive dissipation of energy. For example, on the sharp threshold model all of the energy transferred below E_d would be lost without yielding an additional displacement as the struck atom bounces around inside its potential well, eventually giving up its extra energy to its neighbors. The collisions in which small energy transfers occur are those in which the colliding atoms do not approach each other closely, thus being affected only by the values of the interatomic potential at larger separations. Since in the formation of a large cascade very many binary collisions take place, and for each there is a good chance of a nonproductive, glancing encounter, it appears that precise knowledge of the potential in the range of 10 ev to 100 ev is essential for evaluation of the expected number of displacements.

That the potential well in which a regular lattice atom is situated is more complicated than can be represented by the sharp threshold model is apparent from Fig. 2. Clearly the probability that the atom will escape through the restraining wall of neighboring atoms is very low if the atom happens to be struck in such a way as to head directly toward another atom. It must be also taken into account that insofar as the total production of displaced atoms is concerned it does not matter if the struck atom itself escapes from its lattice site or if a neighboring atom is caused to escape from its lattice site. The latter could occur if the originally moving atom were to suffer a nearly head-on collision with a neighbor; the original atom would then fall back into its old position (equal masses) or perhaps into the lattice position vacated by the second atom which would then become the displaced atom. Since atoms tend to occur in rows in crystal lattices, it is possible to envisage a chain of such events leading to the final appearance of the displaced atom far from the point of the original deposition of energy.

A useful concept in radiation damage theory is that of the "displacement probability function". Exact knowledge of the interatomic potential (especially, again, in the large separation, low energy range) along with considerable effort in the classical mechanics of the movements of the struck atom and its neighbors would permit an averaging over all possible initial directions of motion of the atom to find the probability, $p(T)$, that a (permanently) displaced atom will eventually result from an initial kinetic energy transfer, T . This might appear schematically as shown by the solid curve of Fig. 4, in which the extreme approximation of the sharp threshold is shown as the dashed curve. If this function is

assumed to be known along with the scattering law, $K(E,T)$, also obtained from the potential, an integral equation may be set up for the average number of displaced atoms to be expected in the cascade resulting from a primary of energy E . The reasoning on which the equation is based is that the expected number of displacements $n(E)$ for a primary of energy E is equal to the number of displacements in the two cascades which will result from the primary and secondary atoms after the first binary collision, when the energies will be $E - T$ and T , respectively. To complete the calculation it is only necessary to average over the distribution of T and incorporate the probability that each of the atoms emerging from the collision is able to escape from the potential well. The resultant equation is, then (for the equal mass case),

$$n(E) = \int_0^E n(E - T)K(E,T)p(E - T)dT + \int_0^E n(T)K(E,T)p(T)dT. \quad (2)$$

(Eqn. 2 neglects for high E the case in which both atoms fail to be displaced.)

For the relatively crude approximations

$$1) \text{ Hard sphere scattering, } K(E,T) = \frac{1}{E} \text{ (all transferred energies equally probable)} \quad (3)$$

$$2) \text{ Sharp threshold displacement probability, } p(T) = \begin{cases} 0 & T < E_d \\ 1 & T \geq E_d \end{cases} \quad (4)$$

this equation has the simple solution of Eq. (1) for energies, E , above $2E_d$.

Thus the formalism of counting defects is completed and everything depends on a proper knowledge of the interatomic potential for elastic scattering. Correct calculation of this function involves treating with quantum mechanical rigor the dynamic behavior of two interpenetrating atomic

systems at internuclear separations well below that of any molecular binding. This problem is at present far beyond our capabilities. However, a great deal of effort is being expended in approximation procedures, mostly based on the Thomas-Fermi approach with refinements. The most useful calculations at present utilize either these approximate potentials or mathematically convenient forms which match the approximate potentials fairly well, to obtain $p(T)$ and $K(E,T)$ so that approximate values of $n(E)$ may be obtained. The general conclusion at present is that the number of displacements is not so sensitive to the potential as might have been expected. Specifically, if the $p(T)$ curve rises from zero to unity over not too large an interval of T , the precise functional form does not influence $n(E)$ greatly. However, if the displacement probability remains significantly below unity (say, 0.80-0.90) for a few hundred ev after the initial rise, the effect on $n(E)$ is too large to be neglected. Secondly, the exact form of $K(E,T)$, within certain limits, does not affect $n(E)$ greatly. This may be seen most easily by a change of variables in Eq. (2), casting it into the form

$$n(E) = \int_0^E n(T)p(T)dT \left\{ K(E, E - T) + K(E, T) \right\} . \quad (5)$$

Here the scattering law appears in a symmetrical way with respect to high and low energy, as shown in Fig. 5. This corresponds physically to the indistinguishability of the two atoms emerging from a binary collision. Thus dissipative losses by energy transfers below E_d are, to some extent, balanced by an equal probability for maintaining high energies, above $E - E_d$.

All in all, it is now believed that for many cases the estimate given in Eq. (1) is not so bad, and that the principal deficiencies in our knowledge of radiation damage arise mostly in areas other than that of elastic collisions between atoms.

INELASTIC LOSSES

One of the vital areas in which present-day knowledge is most inadequate is that of the loss of energy of an atom or ion moving through a solid due to excitation and ionization of the electrons of the solid. In the preceding discussion it was assumed, tacitly, that all of the energy of the primary atom was shared, as kinetic energy, with other atoms of the crystal. In this approximation the electrons play a role only in screening the Coulomb charge of the nucleus to provide the physical origin of the interatomic potential. However, it is also true that atoms or ions moving through solids and colliding with lattice atoms may interact directly with the electrons, tearing them away from their parent atoms, or exciting them to higher energy levels of the solid. Such excited electrons are of insufficient energy to produce further displaced atoms, so their energy is dissipated as heat (very quickly in a metal). Thus the moving atom is continuously being slowed down by inelastic losses to the electron system in a way which does not act to produce displaced atoms.

Generally speaking, excitation and ionization losses become more important with increasing velocity of motion of the atom. In fact, it has been customarily assumed, in the absence of more precise calculations, that a certain energy, E_I (ionization threshold), could be appropriately chosen for a given atom in a given crystal, such that for energies of motion below E_I elastic atomic collisions were the dominant mode of energy

loss, while above E_I inelastic losses to the electrons were predominant. To the extent that such a crude model is acceptable, Eq. (1) for the number of displaced atoms in a cascade is modified to read

$$n(E) = \begin{cases} \frac{E}{2E_d} & E < E_I \\ \frac{E_I}{2E_d} & E \geq E_I \end{cases} \quad (6)$$

Thus, for energies of motion greater than the ionization threshold, the atom suffers nonproductive inelastic losses until its energy reaches the value E_I , when it immediately enters the elastic range. In terms of measured damage from a given irradiation, this implies that as the energy of the bombarding particles is raised (thus, in general, raising the average energy transferred to the primary atoms), the damage increases essentially linearly for a while, but then levels off, with little more damage produced, per incident particle, when the average transferred energy goes above E_I . Values chosen for E_I tend to be in the range of many kev, e.g., for a copper atom moving in copper, E_I is usually taken as about 60,000 ev.

The theoretical portion of the task of obtaining a more precise knowledge of ionization losses seems at present to be the most formidable in radiation damage theory. In the high energy range ($E > E_I$) the moving atom is very likely to be itself charged due to the loss of one or more electrons, so that a dual problem is to be faced. First, what is the charge state of the moving atom as a function of velocity? The second problem is the calculation of the effects on a many-electron atom when an electrically charged particle passes by at such a high velocity that the atomic system does not have time to adjust adiabatically to the rapidly changing field.

Although this is a very old problem in quantum mechanics, it has never been treated to a satisfactory approximation. These questions are under active investigation at the present time. As an example of the approximations available, Fig. 6 presents (schematically) the results of Lindhard and Thomsen.² The two contributions to the energy lost by the moving atom per unit distance, $(-dE/dx)$ (given in Fig. 6 in relative units), (1) due to elastic collisions, (2) due to ionization losses, are shown separately in their dependence on the square root of the energy of the moving atom (the energy is also given in relative units). The square root is chosen for the abscissa because these investigators find that $(-dE/dx)_{\text{ionization}}$ is proportional to the velocity of the moving atom. If these results were to be applied to the rough "ionization threshold model", the quantity E_I would be taken at approximately the crossing point of the two curves.

The theoretical difficulties in the low energy range are equally great. Here the problem also has two parts: (1) the continuation of the general ionization loss curve into the low energy region, for which Fig. 6 gives one approximation, (2) the treatment of large-deflection "elastic" collisions. Attention to the latter problem has arisen from recent experimental work which shows that, even at such low energies that only slight inelastic losses are suffered by an atom moving through the solid along an only slightly deflected course, still on the rare occasions of a close encounter with a lattice atom (a small fraction of the atomic spacing) the binary collision is not wholly elastic. Thus one or the other of the two atoms emerges from the collision having lost an electron or more, or having electrons in excited atomic states. This problem has scarcely been attacked at all, theoretically, and the difficulties to be expected are

emphasized by the fact that the most recent experimental results indicate that the inelastic losses associated with "hard" collisions tend to be characteristic, with values not obviously related to atomic level spacings.

LATTICE EFFECTS

Much of the work that has been done to date on defect cascades has neglected the lattice geometry of the solid. The tendency has been to treat the collision sequences statistically, only requiring that the average density of atoms be that of the solid. We have already discussed the importance of the local lattice arrangement in determining the nature of the potential well in which a lattice atom finds itself. It has been emphasized that there is an important consequence of this structure in the low energy range in the possibility of "replacement collisions". Suppose the atoms of a crystal could be labeled so that after a defect cascade has been created by an energetic primary it would be possible to determine not only how many lattice sites were vacant and how many atoms were in interstitial positions, but also how many lattice atoms were at different lattice positions than before the cascade. How would the number of such replacements compare with the number of structural defects? This theoretical problem depends very much on the details of the lattice structure (and the potential!) in the immediate region of a collision and its solution has two important consequences. Even in a monatomic lattice, where replacements are unobservable as such, each replacement collision represents an energy loss from the cascade which is not productive of observable defects. In an alloy, the number of replacement collisions plays a large role in the local disordering resulting from the primary atoms.

The possibility was mentioned earlier of a more drastic effect of lattice structure on the replacement process. If the momentum of a struck atom happens to be directed along a row of lattice atoms, a "replacement chain" may result, in which atoms replace each other down the row, finally resulting in the permanent ejection of a lattice atom from its lattice site at the end of the chain. This is but one aspect of a more general, low-energy, lattice effect of "collision chains" along lattice rows, especially close-packed rows. The most dramatic of such events is the "focusing sequence". If a lattice atom is directed sufficiently nearly parallel to the axis of a row of atoms and with sufficiently low energy, it transfers nearly all of its momentum to the neighboring atom (and falls back into its own lattice site); in such a way that the neighboring atom finds itself moving more nearly parallel to the atomic row than was the original atom. So the momentum is transferred down the row, with, of course, some energy loss at each collision, but with the momentum being focused more and more nearly along the line of centers of the atoms as the collision sequence proceeds, as shown in Fig. 7. Such a chain might, for example, begin with an energy of 100 ev, which might in the normal unfolding of a cascade yield three or four Frenkel pairs. If the conditions are right for a focusing sequence, however, this same 100 ev is dissipated by some 100 elastic collisions down the close-packed row, yielding no Frenkel pairs.

The collision chain may also be a "defocusing sequence". The energy is so high that the directions of motion of the atoms tend to be necessarily directed away from the atomic row, leading relatively quickly to an atom's being displaced to one side of the row. Thus a relatively widely separated (for such low energies) Frenkel pair is produced.

The question for theory is the extent to which collision chains play a role in the development of cascades. How much energy is dissipated by focusing, how many widely separated Frenkel pairs are formed, and to what extent is the volume affected by the cascade process increased by long-range focusing sequences? The two essential steps in answering these questions are (1) the development of a reliable interatomic potential, not only for atomic interactions down the chain but also for interactions of the collision chain atoms with neighboring rows of atoms, and (2) the use of a calculational model which incorporates the essentials of the lattice structure. Such calculations are being carried out extensively using large-capacity, high-speed computers by many groups of investigators at the present time. All of them, however, must rely on approximate potentials.

There is also an important high energy lattice effect, called "channeling". It may happen during the development of a cascade that an atom finds itself in motion down a relatively open channel in the lattice, bounded by close-packed rows of lattice atoms. Then, as illustrated in Fig. 8, the moving atom (with an energy of hundreds or thousands of ev) makes glancing collisions with atoms of the channel walls, which keep it moving fairly closely along the channel axis with small losses of energy. Thus an atom may be led as much as thousands of lattice distances away from the region of the cascade to end up finally as an interstitial atom. In the process its original energy has been dissipated in very small quantities transferred to hundreds of atoms of the lattice without the production of defects. Just as in the case of focusing chains, the combination of a precise knowledge of the interatomic potential and the use

of a correct lattice model are needed to provide theoretical answers to the questions of the probability that a moving atom will become channeled by elastic collisions and of the expected energy loss per unit distance traveled.

Some of the most interesting experiments in this area at present are demonstrating that atoms, once channeled, have a smaller interaction with the solid than otherwise. This is shown by the observation of lower ionization losses (electron density low in the channel) and a lower incidence of nuclear reactions (channel axis far from all nuclei). These observations are made possible by causing the moving ions to be incident upon a single crystal from the outside. By choosing a crystallographic surface to which open channels are normal, a far higher percentage of channeled ions may be achieved than would be the case for primary atoms which were ejected from lattice positions by radiation.

Two additional points on the lattice effects are:

(1) Both focusing and channeling have possibly important implications for the sputtering of atoms from the surfaces of solids. In the experimental work, "spot patterns" of emitted atoms are observed which are related to low-index crystallographic directions in the target. To differentiate the roles of these two lattice effects it is important to investigate the energies of the sputtered atoms, low energy (100 ev or so) implying focusing chains but high energies (1000 ev or more) implying channeling events.

(b) Both focusing and channeling depend upon lattice regularity. Thus imperfections and impurities in the crystal may interrupt the long-range travel of energy or mass. This leads to the possibility of detecting

these effects by doping or working metals and observing consequent changes in the production and annealing of radiation damage.

DETAILED STRUCTURE OF THE DAMAGED REGION

Using computing machines it is possible to follow the full history in an atomic lattice of a displacement cascade developing from an original, energetic primary atom. The calculational model must, of course, assume an interatomic potential for elastic scattering and a form for ionization losses, and the fact that these are poorly known represents a severe limitation on the value of the resulting picture of a damaged region. Further, no actual calculation to date has allowed for all of the special effects at all energies, replacement, focusing, channeling, and ionization losses. However, the work which has been done allows an overall description of a defect cascade which would be expected to be at least qualitatively correct.

The three most vital questions are:

- (1) How many defects are in the final, stable damaged region?
- (2) What is the spatial extent of this region?
- (3) What has happened to the energy of the primary atom?

The answer to the first two questions depends directly on the energy of the primary. A more energetic primary produces more defects and spreads them out over a larger volume of the crystal. Since primaries of a great range of energies are of practical interest and are also found in experimental studies, it is impossible to apply one damaged region description to all important situations. A primary in copper created by a one-Mev electron will have perhaps 50 ev of energy. It will travel a short way from its point of creation, give rise to perhaps one other displaced atom,

and the cascade will be over. On the other hand, a 200,000 ev primary in copper from fast neutron irradiation will travel perhaps 100 Å before it begins to produce secondaries efficiently, and the resultant cascade will include something like 1500 Frenkel pairs spread over a region of about 200 Å in diameter. There is the further point that in applying our knowledge of damaged regions to actual irradiations of solids it must be remembered that for most types of radiation primaries are produced with a significantly broad range of energies so that there may be no really typical damaged region. In this section attention will be concentrated on damaged zones arising from primaries having some tens of kev of energy as providing some insight into the structure of a relatively heavily damaged region.

First consider the dimensions of the region containing the bulk of the defects produced in the cascade. Figure 9 shows the results of calculations of the range of a copper primary in copper, i.e., the average distance from the starting point to the final rest position. The calculation is based on elastic collisions only, with no lattice effects included, and assuming a particular form for the interatomic potential. The results show reasonable agreement with measured ranges of ions in solids. Thus it may be taken as a fair estimate that the damaged region has a spatial extent of approximately the values given in Fig. 9. Of course, some of the defects produced will lie outside this region--in special cases, such as highly channeled or focused events, far outside the region. With this reservation, however, it might be expected that the greater fraction of defects will be inside this region.

For an estimate of the number of vacancy-interstitial pairs produced, the simple formula of Eq. (1) should not be too bad. Over the same

energy region as in Fig. 9, again for a copper primary in copper, this would give the result of Fig. 10. An average value over this energy range for the atomic fraction of Frenkel pairs may be seen to be about 0.005.

Virtually all of the energy of the primary appears as heat in the lattice within a time of the order of 10^{-13} sec after creation of the primary. If all of it is contained within a region of the size suggested in Fig. 9, the instantaneous temperature would be of the order of 1200°K . At such a high temperature the defects created would be expected to be mobile and to begin to diffuse toward annihilation. However, it seems at present very questionable that this "thermal spike" persists long enough to allow much free migration. Even on the basis of thermal conductivity, the temperature of the damaged region would have fallen over to the lattice temperature in something like 10^{-11} sec. However, such a calculation is not really valid, for there is insufficient time (and space) for the establishment of phonon equilibrium. It seems likely that rapidly moving phonons, focusing events and channeling events carry energy sufficiently far away, sufficiently rapidly that there is little thermal annealing due to the thermal spike. One reservation must be made, however. Particularly unstable defect configurations which require only one or two lattice jumps to anneal may be partly destroyed by the thermal spike. These will be discussed later.

Lattice calculations show that there are interesting effects to be expected in the more detailed structure of the final defect cascade. For example, it seems that, as indicated in Fig. 11, the vacancies and interstitials tend to be segregated to some extent due to the mechanics of the problem. The primary and secondaries knock atoms far away from their

lattice sites, leaving a cloud of vacancies behind, inside a "shell" of interstitials. Of course, this is only a tendency and the actual situation will be that of overlapping of two partially separated distributions. Nevertheless, this trend may have important consequences.

One of these has to do with "self-annealing". As the cascade process, i.e., the dynamic motion, comes to a halt, it may happen that an interstitial atom comes essentially to rest near a vacancy. It is to be expected that, since a vacancy-interstitial pair represents an excited state of the lattice, there would be a tendency for the pair to annihilate. This would appear as an attractive force between the two. Thus if the two are sufficiently closely spaced, it is to be expected that mechanical forces will cause them to annihilate even in the absence of thermally activated motion. Processes similar to that shown in Fig. 12 may be envisaged. Recent calculations indicate that unstable defect configurations might reduce the estimated total number of defects by as much as a factor of two. However, this is a very uncertain quantity since no reliable theory, at all, exists of the range and size of the attractive force between vacancies and interstitials. It may be said, however, that the tendency over recent years, in interpreting experimental results, is to assume a greater and greater effectiveness of this force over large distances.

DEFECT CONFIGURATION AND MOTION

Because of the difficulties met in trying to correlate irradiation-produced changes in physical properties with defect production, it is of great importance to utilize the information available from annealing studies. Suppose, for example, we imagine an ideal example in which

irradiation at a low temperature, such as 4°K , produces a distribution throughout the solid of an equal number of vacancies and interstitials which are unable to move at any appreciable rate. If the solid is now warmed up, a temperature will first be reached at which one of the two (usually presumed to be the interstitial atom) will diffuse at an appreciable rate. Some moving interstitials will annihilate vacancies and the radiation-produced property change will begin to reduce. With further temperature rise this process completes itself and for a very simple model we may imagine that those interstitials which have not disappeared by annihilation find temporary trapping sites in the lattice, such as at impurity atoms or dislocation lines. The vacancies, which remain frozen in, begin to move with a further rise in temperature, and in our simplified model the radiation damage is completely annealed out when the temperature is high enough so that all vacancies have had time enough to move as far as needed to reach annihilation points (trapped interstitials). Schematically, then, the radiation-induced property change would look as shown in Fig. 13. The derivative curve (b) should reveal single processes taking place at temperatures T_I and T_V which would correspond (in some way depending on the kinetics of the process) to E_I and E_V , the activation energies for motion of the interstitials and vacancies, respectively. Such information, along with the kinetics of the processes which can be studied by isothermal anneals in the appropriate ranges, and along with the percentage recovery associated with each type of defect, should give valuable clues to the basic nature of radiation damage.

In fact, however, it has not been possible to unravel the annealing data in any case so far studied in terms of underlying defect motion. At

least, it has not been possible to do so with sufficient finality as to convince everyone of the correctness of the analysis. A schematic but realistic plot of the annealing of copper analogous to 13(b) illustrates the complications (Fig. 14).^{*} The unexpected features needing explanation are:

- (1) the multiplicity of processes,
- (2) the presence of broad peaks,
- (3) ranges (such as shown between 40°K and 250°K) where continuous annealing seems to occur.

A number of interesting hypotheses along with some supporting calculations have been proposed toward the understanding of these results.

a) Close Pairs. As suggested above, Frenkel pairs probably have a significant attraction to annihilation in the lattice. Sufficiently close pairs which have survived the thermal spike and are mechanically stable at the irradiation temperature could have a very low activation energy for recombination. Further, there could be many different activation energies of pairs, varying with the separation of the two defects. However, the spectrum of such energies would be expected to be discrete because of the lattice nature of the crystal. Such pair recombination processes would be expected to show first order kinetics; this has been found to be true and activation energies of the order of 0.1 ev measured for the group of peaks in the 15-50°K range (in copper). Clearly one future job for theory, and one on which little progress has been made to

* No one experiment has ever shown this entire range with just exactly these details.

date, is that of calculating the elastic attractive force between defects, and the possible stable configurations.

b) Metastable Form for Defects. As the specific example of this idea it has been proposed that during irradiation a special form of the interstitial atom called the "crowdion" may be created (see Fig. 15(c)). In this form the extra atom is crowded into a close-packed row of atoms which relaxes somewhat so that the whole configuration includes 10 or so atoms. This is not supposed to be the lowest energy form of the interstitial, but might be unable to convert spontaneously into a lower energy form at the temperature of irradiation. As the temperature is raised, this crowdion may become able to move along the close-packed row until it reaches a defective portion (vacancy, impurity atom or dislocation line), at which it converts or is trapped. Those which reach vacancies annihilate and thus produce recovery. At a higher temperature the more "normal" interstitial becomes able to move and produces further recovery. In one model (for copper) most of the recovery around 40°K is attributed to crowdions,* moving with an activation energy of about one ev, while the recovery around 250°K is attributed to motion of the normal interstitial moving with an activation energy of about 0.6 ev. This more normal form is thought to be a "split" or "dumbbell" interstitial (Fig. 15(a)), i.e., two atoms are just on either side of a lattice position such that the line between them is oriented in a 100 direction (in the fcc lattice). (The concept of a simple interstitial atom has not been abandoned; some models of annealing in copper rely on the motion of the "body-centered"

*The crowdion form may conceivably exist in a close pair arrangement with a vacancy, just as well as any other form of the interstitial.

interstitial (see Fig. 15(b)), just an extra atom at the most open, non-lattice, site in the fcc lattice.)

This reveals another area for theory to tackle, that of the lowest energy form of the interstitial (and of the vacancy, though this at present is thought to be fairly simple). Along with this must come calculations of the energy of motion of each type and of the possibility of metastability. Then comes modification of the close pair configurations and energies depending on which type of interstitial is being considered. It must even be allowed that occurrence in close proximity to a vacancy may cause the interstitial to choose another configuration as energetically more favorable than that taken in the otherwise perfect lattice.

c) Clustering of Defects. One of the most fruitful suggestions is that the individual defects, vacancies and interstitials, may have an attractive force for their own kind, as well as for their opposites. If this is true, another temporary resting place for defects in motion is in clusters of similar defects. It is not, at first, evident that two interstitials or two vacancies should have a binding energy holding them together as the lowest order clusters, called the di-interstitial and the divacancy, respectively. However, it seems very likely, at present, that such is the case, the forces involved being short range and associated with the lattice structure of the crystal. Further, the possibility must be considered that these defects may be able to move by thermal activation without decomposing into single defects. Then the possibilities for basic processes in annealing become greatly increased. For example, if a locally high density of single interstitials is created by low temperature irradiation, upon subsequent heating a significant fraction of these interstitials may combine to form di-interstitials. At an even higher temperature

the di-interstitials may become mobile or may dissociate and produce another annealing stage as they annihilate at vacancies. It is quite possible that non-negligible concentrations of both double defects are formed under certain irradiation conditions or by accidental creation of very close pairs of like defects, or by mechanical or thermal instability with respect to combination of somewhat more distant pairs.

The various possible clusters, including tri-interstitials, tri-vacancies, etc., are under theoretical investigation at the present time, using, again, approximate interatomic potentials and large computing machines. The goal is to find the lowest energy form of such defects in the lattice, to find the binding energy (if there is one), and to calculate the mode of motion and the corresponding activation energy. For example, Johnson and Brown (in one particular model) have found that a di-interstitial consisting of two 100-split interstitials on neighboring lattice sites is stable in copper with a dissociation energy of 0.6 ev and an energy of migration of 0.26 ev. As another example, calculations have suggested that the divacancy moves with a lower activation energy than the single vacancy. A further suggestion is that an intermediate (excited) state of the trivacancy (in fcc) may be of the lattice form of a tetrahedron of four empty lattice sites with one atom at the center of the tetrahedron.

CONCLUSION

The mode of attack which must be used in the theory of radiation damage now seems clear. There must be a fundamental effort to understand the physics of the basic processes, elastic scattering of atoms and inelastic losses by ionization and excitation. This depends on the correct

calculation of an interatomic potential, whether two-body or many-body, whether central or non-central. Upon the basis of this knowledge large-scale calculational models of the lattice may be constructed in which (probably using high-capacity computers) the true course of development of a defect cascade may be studied. This would ultimately lead to an evaluation of the role played by focusing and channeling and by the non-productive, low-energy-transfer elastic collisions and replacements. It would also allow for the calculation of the lattice configuration and of the formation and migration energies of point defects and their clusters. In this way the complexities underlying observed radiation damage production and annealing may be resolved.

References

1. For more detailed reviews of this field and references to original research papers, see the following:
 - (a) The Interaction of Radiation with Solids, ed. by Strumane, Nihoul, Gevers, and Amelinckx (North Holland Pub. Co., Amsterdam, 1964).
 - (b) D. S. Billington and J. H. Crawford, Jr., Radiation Damage in Solids (Princeton University Press, 1961).
 - (c) G. J. Dienes and G. H. Vineyard, Radiation Effects in Solids (Interscience, 1957).
 - (d) Proceedings of the XVIIIth Summer School "Enrico Fermi", Radiation Damage in Solids, Ispra, Italy, 1960 (Academic Press, 1962).
 - (e) Proceedings of a Symposium on "Radiation Damage in Solids", Venice, Italy, 1962 (IAEA, Vienna, 1962) (especially in this volume the article by A. Seeger and also that by R. O. Simmons, J. S. Koehler and R. W. Balluffi).
 - (f) Proceedings of the International Conference on Crystal Lattice Defects, J. Phys. Soc. Japan, Vol. 18, Suppl. I-III (1963).
2. J. Lindhard and P. V. Thomsen, p. 65 in Radiation Damage in Solids (IAEA, Vienna, 1962).
3. J. R. Beeler, Jr., and D. G. Besco, p. 43 in Proceedings of a Symposium on "Radiation Damage in Solids", Venice, Italy, 1962 (IAEA, Vienna, 1962).

Figure Captions

- Fig. 1 The Interatomic Potential (Schematic).
- Fig. 2 Escape of a Lattice Atom from Its Potential Well (Schematic).
- Fig. 3 The Differential Cross Section for Energy Transfer (Schematic).
- Fig. 4 The Displacement Probability as a Function of the Kinetic Energy Transferred to a Lattice Atom.
- Fig. 5 Kernel of the Integral Equation for $n(E)$ (Schematic).
- Fig. 6 Schematic Representation of the Energy Loss per Unit Distance ($-dE/dx$) of a Moving Ion (after Lindhard and Thomson²).
- Fig. 7 Focussing Collision Sequence. The Arrows Show Successive Directions of Motion along a Close-Packed Row of Atoms.
- Fig. 8 Channeling Path of an Atom Channeled between Two Close-Packed Rows of Lattice Atoms. Points Indicate Sites of Binary Elastic Collisions.
- Fig. 9 Average Distance of Penetration of a Moving Copper Atom into Copper.
- Fig. 10 Approximate Dependence of the Number of Frenkel Pairs on the Primary Energy.
- Fig. 11 The Spatial Disposition of Vacancies and Interstitials Resulting from an Energetic Primary (Planar Calculation of Beeler and Besco³).
- Fig. 12 Schematic Representation of a Possible Mode of Mechanical Collapse of a Frenkel Pair.

Fig. 13 Idealized Annealing Pattern for Interstitials and Vacancies.
(a) Warm-up Annealing, (b) Derivative of (a).

Fig. 14 The Pattern (Schematic) of the Annealing Behavior Actually
Observed.

Fig. 15 Some Possible Interstitial Configurations in a Face Centered
Cubic Lattice. (a) 100-Split Interstitial (Dumbbell), (b) Body
Centered Interstitial, (c) 110 Crowdion Interstitial.

The Interatomic Potential (Schematic).

Escape of a Lattice Atom from Its Potential Well (Schematic).

Fig. 2

The Differential Cross Section for Energy Transfer
(Schematic).

The Displacement Probability as a Function of the Kinetic Energy Transferred to a Lattice Atom.

Fig. 4

Kernel of the Integral Equation for $n(E)$ (Schematic).

Fig. 5

Schematic Representation of the Energy Loss per Unit Distance $\left(-\frac{dE}{dx}\right)$ of a Moving Ion (After Lindhard and Thomson²).

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Focussing Collision Sequence. The Arrows Show Successive Directions of Motion Along a Close-Packed Row of Atoms.

Fig. 7

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Channeling Path of an Atom Channeled Between Two Close-Packed Rows of Lattice Atoms. Points Indicate Sites of Binary Elastic Collisions.

Fig. 8

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Average Distance of Penetration of a Moving Copper Atom into Copper.

Fig. 9

Approximate Dependence of the Number of Frenkel Pairs on the Primary Energy.

The Spatial Disposition of Vacancies and Interstitials Resulting from an Energetic Primary (Planar Calculation of Beeler and Besco³).

Schematic Representation of a Possible Mode of Mechanical Collapse of a Frenkel Pair.

Idealized Annealing Pattern for Interstitials and Vacancies. (a) Warm-up Annealing, (b) Derivative of (a).

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The Pattern (Schematic) of the Annealing Behavior
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Fig. 14

Some Possible Interstitial Configurations in a Face Centered Cubic Lattice
 (a) 100-Split Interstitial (Dumbbell) (b) Body-Centered Interstitial
 (c) 110 Crowdion Interstitial.