RADIATION EFFECTS AND DAMAGE

The detrimental consequences of radiation are referred to as *radiation damage*. To understand the effects of radiation, one must first be familiar with the radiations and their interaction mechanisms. Further, the fundamental characteristics of the material(s) being irradiated must also be understood. A brief summary of the characteristics of radiation is first presented below. This is followed by a general discussion of the effects of radiation, along with some specific examples, with an emphasis toward the ionizing effects.

1. Ionizing Radiations

The radiations of concern here include charged particles such as electrons (beta particles), protons, alphas and fission fragment ions, and the neutral radiations including photons (gamma and X rays) and neutrons. Table I compares some key characteristics of the radiations, including charge, mass, and range in air. For a kinetic energy of 1 MeV, the electron is relativistic (0.94*c*). For the same energy, the heavier particles are slower, stopped easier and deposit their entire energy over a shorter distance. The passage of radiation through tissue is depicted in Figure 1.

Table I: Comparison of Ionizing Radiation

Characteristic	Radiation (E_K = 1 MeV)				
	Alpha (α)	Proton (p)	Beta (β) or Electron (e)	Photon (γ or X ray)	Neutron (n)
Symbol	$^{4}_{2}\alpha$ *or* He^{2+}	$^{1}_{1}p$ *or* H^{1+}	$^{0}_{-1}e$ *or* β	$^{0}_{0}\gamma$	$^{1}_{0}n$
Charge	+2	+1	–1	neutral	neutral
Ionization	Direct	Direct	Direct	Indirect	Indirect
Mass (amu)	4.001506	1.007276	0.00054858	—	1.008665
Velocity (cm/sec)	6.944×10^{8}	1.38×10^{9}	2.82×10^{10}	$c=2.998\times10^{10}$	1.38×10^{9}
Speed of Light	2.3%	4.6%	94.1%	100%	4.6%
Range in Air	0.56 cm	1.81 cm	319 cm	82,000 cm*	39,250 cm*

* range based on a 99.9% reduction

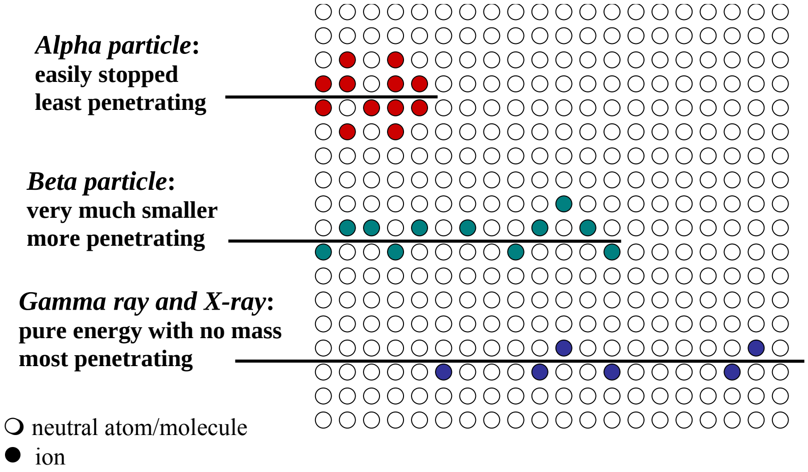


Figure 1. Radiation paths in tissue.

The behavior of charged particles (α, β, *p*) passing through matter is fundamentally different from that of the neutral radiations (*n*, γ). In particular, the charged particles strongly interact with the orbital electrons of the material through which the particles move. As such, we classify charged particles as *directly ionizing*. Further, the charged particle interactions can be subdivided into two cases based on mass: (1) heavy charged particles such as alphas and protons, and (2) light electrons (both positrons and negatrons).

Heavy Charged Particles

Initially, a heavy moving particle loses energy in small steps through interactions with the electrons in the material through which it passes. Once the heavy particle loses enough energy such that it no longer has sufficient energy to excite an electron, then the particle loses energy by nuclear collisions. As the particle slows, it captures electron(s) to form a neutral atom (*e.g.*, a proton becomes hydrogen, and an alpha forms helium). The heavy particles slow down almost entirely due to Coulombic interactions with the atomic electrons. Because of the large number of these interactions, the slowing down is nearly continuous. Because of their large size, ions are not easily deflected by atomic electrons and so they travel straight-line paths. In contrast with the exponential decrease of neutrons and gamma rays, the heavy particle has a well-defined range of only a few centimeters in air, even for quite energetic particles. For example, a 4 MeV alpha particle has a range of 2.3 cm in air; a 4 MeV proton has a range in air of 22 cm.

Light Charged Particles

When beta particles and electrons pass through matter, several possible processes occur, including:

1. Ionization in which the energy loss mechanism is similar to that for heavy charged particles,
2. Bremsstrahlung, which is the creation of X-rays from electron deceleration, and
3. Elastic scattering from nuclear and electronic interactions.

Scattering is more important in the case of beta particles than with heavy charged particles. This means that the path of an electron is zigzagged, and its range is greater, but its path is not well defined.

Neutral Radiations

The uncharged radiations can liberate directly ionizing particles or initiate a nuclear transformation. The fundamental interactions of neutrons are scattering and absorption, which includes both capture and fission. Neutron capture frequently produces radioactive nuclei, which in turn emit radiation(s). The three fundamental interactions by photons and their approximate energy range of interest are

1) The *photoelectric effect* (low energy: $E < 200$ keV) in which the photon transfers all of its energy to an orbital electron, which is ejected with kinetic energy equal to the incident photon energy less the binding energy (ionization energy) needed to remove the electron ($E_e = E_\gamma - BE_e$). The bottom line is that all the photon energy is absorbed into the incident material.
2) *Compton scattering* (intermediate energies: 200 keV to 1.5 MeV) which is an elastic scattering of the photon by an atomic electron, which is excited in the process. Here the photon is reduced in energy and deflected from its original direction.
3) *Pair production* (high energy: $E > 1.5$ MeV) in which the photon disappears and an electron (negatron)–positron pair is formed. Since the rest mass energy of an electron/positron is 0.511 MeV, pair production requires a photon of at least 1.02 MeV to occur. The remainder of the photon energy is received as kinetic energy by the negatron–positron pair ($KE = E_\gamma - 2\,m_e c^2$). Eventually the positron combines with an electron, and two photons (*annihilation radiation*) are produced, each having an energy of 0.511 MeV.

We have noted how the mass-less photon predominantly interacts with the atomic electrons, whereas the neutron interacts with the nucleus. Although both of these neutral radiations are classified as *indirectly ionizing*, each can cause the direct liberation of charged particles.

2. General Radiation Effects

The general types of radiation effects on materials can be categorized into

(1) *Impurity Production*, that is, transmutation of nuclei into other nuclei which themselves may be radioactive; this mechanism is caused by neutrons through fission and activation (capture). Impurities can also be deposited from the creation of hydrogen or helium when a proton or an alpha particle, respectively, becomes neutralized in the material of passage.
(2) *Atom Displacement* from their normal position in the structure of the material; displacement atoms may leave lattice vacancies and lodge in interstitial locations or cause interchange of dissimilar atoms in the lattice structure.
(3) *Ionization*, that is, the removal of electrons from atoms in the material and the formation of ion pairs in the path of the charged particles.
(4) Large *Energy Release* in a small volume, which can result in thermal heating of the material. This may be especially important in those cases where the material is a radiation shield.

Neutrons are particularly efficient at causing the first two effects above. A comparison and contrast of the radiations and their effects is presented in Table II. Consideration of both the dose and dose rate are important in assessing radiation damage as they can characterize chronic and acute exposures.

Table II: Radiation Damage to Materials (adapted from [1])

Radiation	Impurity Production	Atom Displacement	Ionization	Energy Release
Thermal (eV) neutron	Directly by absorption reactions (mostly thermal neutrons), also may lead to more radiations	Yes, indirectly	Indirectly	Indirectly
Fast (MeV) neutron		Multiple displacements via scattering reactions; can cause displacement of "knock-on" atoms		
Fission fragment	Become impurities themselves		These highly charged ions cause considerable ionization, and they emit β and γ	Considerable heat deposition over a very short range
Alpha	He buildup can cause pressurization problems	Yes, may cause atom displacement	Causes sizable ionization	Yes, over a very short range
Proton	H buildup can also cause pressurization	Yes	Directly	Yes, over a short range
Beta	n/a	Some displacement	Directly	Localized heat deposition
Photon (γ and X ray)	n/a	Rare displacements (via Compton effect)	Indirectly	Gamma heating over large distance

Impurity Production

In the context of this discussion, impurity production refers to radiation-caused impurities, not any impurities that may have pre-existed in a material. Impurities in a crystal constitute structural imperfections which can alter electrical and mechanical properties. As Table II indicates, electrons and photons do not directly cause impurity production. One might consider that these radiations can indirectly cause impurity production through chemical bond breakage.

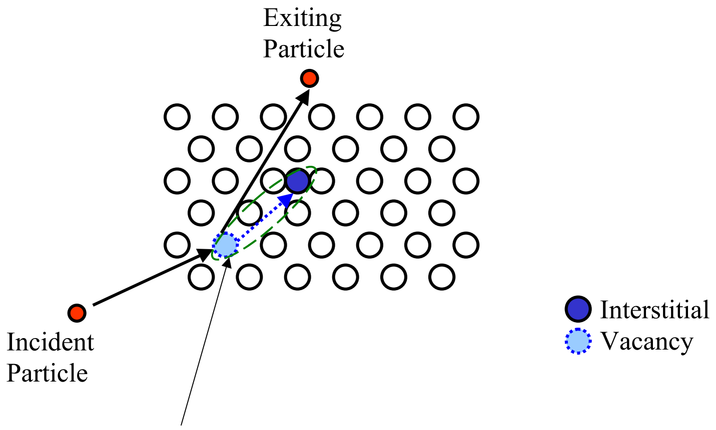
More important to impurity production are the effects of neutron and ion irradiation. Incident ions will eventually slow and capture the necessary electrons to render them neutral. As stated earlier, the protons will become hydrogen and alpha particles will become helium. In both of these cases, the neutral atom is a gas at room temperature, and hence, will exert 'pressure' on its neighboring atoms. In solids, this internal pressurization has been observed to cause swelling in the material.

Neutron and ion irradiation can also form radioactive species. Neutron capture by a nucleus does not necessarily change the chemical element present, but does change the isotope present. The new isotope may be radioactive and decay by one of several schemes, which can change the chemical element present. Of course, the decay process emits additional radiation into the material. In contrast, ion absorption by a nucleus immediately changes the chemical element present; however, the exact reaction products must be determined before a full conclusion is reached.

Atomic Displacement (Damage)

Atomic displacement can occur ballistically through kinetic energy transfer, or radiolytically by the conversion of radiation-induced excitation into atom motion (*i.e.*, recoil). As a charged particle passes through matter, the particle energy dissipates by exciting orbital electrons and by elastic collisions with the material nuclei. An elastic collision can eject an atom from its normal lattice position. The ejected atom is known as a *primary knock-on*, which, in turn, may cause a cascade of atomic displacements before eventually coming to rest. In Figure 2, the displaced atom becomes an *interstitial*, and the position the atom formerly occupied becomes a *vacancy*. Together the interstitial and vacancy are referred to as a *Frenkel pair*. Some displaced atoms can lead to secondary displacements. For example, the displaced atom may collide with and replace another atom in the material.

Displacement damage is the result of nuclear interactions, typically scattering, which cause lattice defects. Displacement damage is due cumulative long-term non-ionizing damage from the ionizing radiations. The collision between an incoming particle and a lattice atom subsequently displaces the atom from its original lattice position as shown in Figure 2. In space, the particles producing displacement damage include protons of all energies, electrons with energies above 150 keV, and neutrons.



A Frenkel pair consists of a vacancy and an interstitial atom.

Figure 2. Displacement damage.

A single incident particle can cause a cascade of collisions to occur to a portion of the affected material (*e.g.*, Si) lattice atoms. These collisions are produced by both incident "heavy" particles (*p*, *n*, ions) and secondary particles. Defects (vacancies, interstitials, Frenkel pairs, dislocations) are produced along the tracks of the secondary particles and in clusters at the end of these tracks as shown in Figure 3.

Displacement damage can be quantified using the non-ionizing energy loss (NIEL). The NIEL is energy lost to non-ionizing events per unit length, MeV/cm or MeV·cm^2/g. The NIEL is based upon fact that displacement damage effects are proportional to the non-ionizing particle's energy loss and the nuclear recoils produced. A *displacement damage dose* can be computed from $D_d = \text{NIEL}\,\Phi$.

The production of vacancies and interstitials involves a transfer of particle kinetic energy to potential energy stored in the crystal lattice. Both vacancies and interstitials, especially the latter, are mobile at sufficiently high temperature and annealing facilitates their recombination. At the higher temperature, the vibration of the atoms in the lattice increases, thereby providing a mechanism by which an interstitial atom can migrate to a vacancy; and hence, fix both defects.

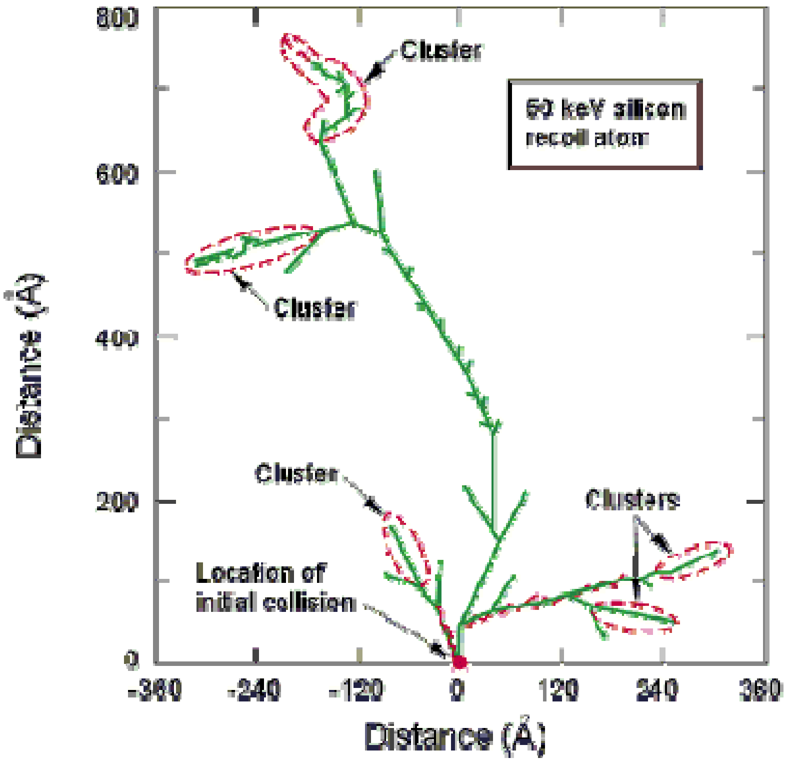


Figure 3. Displacement cascade damage from movement of silicon atom after primary collision [2].

Ionization

Ionization is the process of removing or adding an electron to a neutral atom, thereby creating an ion. The term is also often used in connection with the removal of an electron from a partially ionized atom. A closely related process is *excitation*, in which the energy level of an electron is raised; however, excitation occurs at an energy less than that required for ionization. Table I shows that those radiations which are charged (α, β, p) can directly ionize matter; however, those radiations which are neutral (n, γ) are said to indirectly cause ionization.

Ionizing radiation tends to be increasingly damaging in the following order of molecular formation (largely due to the ability of ionization to disrupt the bonds):[1]

1. metallic bond (*least damaged*)
2. ionic bond
3. covalent bond (*most damaged*)

Since biological tissue is characterized by substantial covalent bonding, it is generally more susceptible to radiation damage than metallic-bonded structural components.

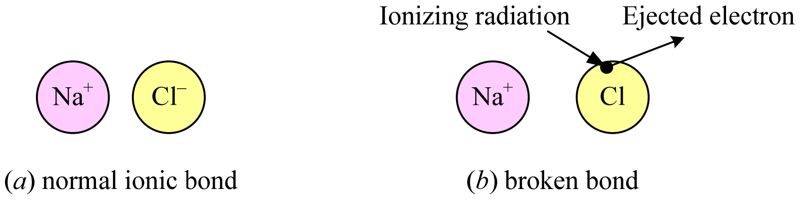
Metallic bonds

Metallic bonding consists of positive ions with free valence electrons, which hold the ions together. Ionizing radiation increases the kinetic energy of the electrons or excites the electron to a higher energy level, but they shortly return to their normal energy level. In either case, there is no permanent damage from ionization, only temporary internal heat production. *Note*: this is not to say that metals are not damaged by ionizing radiation as we examined earlier the fact that displacement damage can occur in materials.

Ionic bonds

Ionic bonds are weaker than metallic bonds from an ionizing radiation point of view. Recall that for an ionic bond, electrons are transferred from one element to another element; hence the ionic compound (*e.g.*, NaCl) is composed of positive (cation) and negative (anion) ions which attract one another. Overall, the electrostatic attractive and repulsive forces between the ions lead to well-ordered, three-dimensional arrangements of the ions in crystalline substances. Radiation causes only temporary ionization of the lattice atoms, which soon become neutral. Discoloration (*e.g.*, in glass) may occur due to free electrons being trapped at lattice perfections.

Illustrative Example: Let us use salt (NaCl) as an example. Sodium transfers its loosely bound valence electron to chlorine, such that Na^+Cl^- is formed, as shown in Figure 4(*a*). Ionization of the sodium cation would result in a doubly charged Na ion, and the Na^{2+} would continue to be attracted to the Cl^- anion. Conversely, ionization of the Cl^- anion would result in the chlorine atom returning to a neutral state, and the Na^+ cation is no longer attracted to the chlorine atom, and hence, the ionic bond is broken, as illustrated in Figure 4(*b*).



(*a*) normal ionic bond (*b*) broken bond

Figure 4. Ionic bond of salt before and after irradiation.

Covalent bonds

For a covalent bond such as found in water (H_2O), the outer electrons in a molecule are no longer uniquely associated with a particular atom, but rather are shared between all the atoms in the molecule. Covalent compounds (*e.g.*, H_2O, CO_2, C, Si, and Ge) share electrons among the joined atoms, with the binding force being the attraction of each atom to the jointly held electrons. Covalent bonds are typical of the gases, liquids and organic materials. The resultant covalent molecules do not attract one another to any degree. Covalent bond energies are in the low eV range. Radiation of sufficient energy to overcome the covalent bond can permanently separate the molecule into its constituent atoms or radicals. Hence, the chemical composition of the material is fundamentally changed.

Energy Deposition

All of the radiations cause energy (and charge) deposition within the absorbing material through the ionization process. In water and organics, most of the absorbed ionization energy breaks chemical bonds. In metals, almost all of the absorbed energy from ionization appears as heat. It is the kinetic energy deposition that generally manifests itself as thermal heating of the material. The corresponding temperature rise can change a number of material properties. For direct heating by radiation, the product of the absorbed dose rate ($\dot{D}$) and the material density (ρ) gives the volumetric heat generation rate (*i.e.*, the energy deposition rate per unit volume)

$$\dot{Q} = \dot{D}\,\rho \tag{1}$$

3. Specific Radiation Effects

Radiation can cause changes to a variety of material properties including chemical, electrical, magnetic, mechanical, optical, and so on. In this section, we look at two examples of these effects. Materials such as insulators, dielectrics, plastics, lubricants and rubber are but a few of the materials that are ionization sensitive.

Mechanical Effects

The effects of radiation on the mechanical (and metallurgical) properties include changes to strength and ductility. Mechanical properties are directly related to microstructural characteristics of a given material.

In general, nuclear radiation tends to destroy the well-defined lattice structure of crystalline materials. These imperfections ultimately alter the basic material properties such as hardness, ductility, etc. The radiation damage is primarily due to point defects being created in the crystalline structure. As an example, if a fast neutron causes displacement damage, then the ordered structure of the material has been weakened and the material properties changed due to the irregularities and vacancies. In a process known as *amorphization* or *metamictization*, radiation may convert crystalline material to an amorphous structure.

Microstructural change then affects macroscopic properties. Consider a neutron-caused atomic displacement in a crystalline solid. The radiation has therefore caused point defects in the lattice. Various mechanical properties are affected by the way in which one plane of atoms slips over the adjacent plane. The dislocations inhibit *slip* processes, that is, more energy is required to initiate slipping. Consequently, the material resistance to penetration (hardness) and the stress required to initiate failure (strength) increase, but there is a concomitant decrease in the energy needed for failure by fracture (toughness) and permanent strain (ductility).

Metals: Metals represent an appropriate material for which to examine mechanical effects of radiation. The changes produced by radiation are comparatively small in metals. The relaxation time for self-annealing in metals is short. The radiation effects are similar to those produced by cold working; specifically, the hardness and the creep rate are increased, and the electrical and thermal conductivities are decreased. In addition, ordered alloys become disordered. Metals with their shared valence electrons are affected very little by ionization.

Neutron damage to metals is more pronounced for higher energy neutrons. Under fast neutron irradiation, all steels experience radiation-induced hardening and embrittlement. Steels bombarded by fast neutrons also experience swelling (volume increase) and radiation-induced creep. Typically, radiation causes the hardness and strength to increase with a concurrent decrease in the ductility and toughness. Annealing will soften and toughen materials.

Steels used in nuclear reactors are exposed to significant neutron fluence. Irradiation damage is sometimes quantified in terms of the number of displacements per atom (dpa). Klueh states that typical displacement rates in steel are 0.03, 30 and 60 dpa/yr, respectively, for light-water, fast and fusion reactor applications [3]. It is phenomenal to consider that each steel atom being used in a fast reactor is displaced 30 times in a single year!

Electrical Effects [4]

Radiation can change electrical properties such as the conductivity of a material. Primary radiation damage mechanisms from indirectly ionizing radiation (neutrons and photons) are of two types: atomic displacements resulting in lattice defects, and changes at the molecular level. The former is the key damage mechanism in metals, the latter in nonmetals. A combination of both mechanisms is important for electrical components such as semiconductors and insulators. We separate our discussion of electrical properties into those on metallic conductors and those on semiconductors and insulators.

Metallic conductors: First, we examine radiation effects on metallic conductors. We have already seen that ionization does little to the metals, rather atomic displacement is the radiation damage mechanism of importance. The conductivity (σ) and resistivity (ρ) of a metal can be expressed as

$$\sigma = \frac{1}{\rho} = q\, n\, \mu \tag{2}$$

where q is the fundamental charge unit, n is the number of charge carriers and μ is the carrier mobility. The mobility is the ratio of the drift velocity and electric field $\left(\bar{v}/E\right)$.

Valence electrons travel through a metal as standing waves. The introduction of a point defect (imperfection) in the material can cause electrons to be deflected. Such a disordered structure has a shorter mean free path for electron movement. The electron drift velocity is correspondingly reduced which subsequently causes a decrease in the carrier mobility. As can be seen from Eq. (2), a decrease in μ causes conductivity of metals to decrease (and resistivity to increase). Hence, any radiation that introduces irregularities into the crystal structure of a metal causes an increase in the electrical resistivity since the lattice imperfections decrease the mobility of the charge carriers (electrons). Again, ionization does little to change the conductivity of a metallic conductor.

Semiconductors and Insulators: To investigate the effects of radiation on semiconductor and insulator electrical properties, we begin our discussion by recalling the basic electronic difference between metals, and semiconductors and insulators.

The outermost electrons, which are not associated with any particular atom, have energy levels that fall into two bands: a *valence* (filled) band and a *conduction* band. In all materials, a region of forbidden energies separates these two bands. In an insulator, electrons occupy all the energy levels in the valence band and the conduction band is empty. However, in a conductor, not only is the valence band full, but the conduction band also has electron(s). Electrons in the conduction band are relatively free to move throughout the material under the influence of an electric field, and create an electric current.

The forbidden energy gap between the valence and conduction bands is small in semiconductors whereas the gap is large for insulators. Thermal agitation of valence electrons in semiconductors is sufficient to allow the electrons to jump the gap and move into the conduction band.

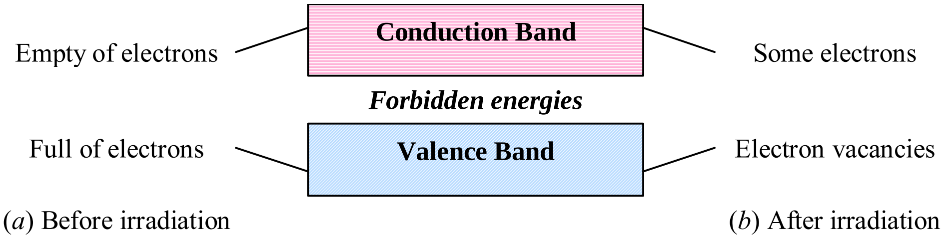


Figure 5. Electron population in an insulator.

Consider the passage of radiation through some material. Ionizing radiations can carry more substantial excitation energy than thermal agitation, such that large numbers of valence electrons in both semiconductors and insulators can be readily excited to the conduction band. The radiation produces a large number of excited atoms along its track. After the traversal of the radiation, vacancies may be left in the valence band and there will be some electrons in the conduction band (see Figure 5). Since the direct recombination of the conduction band electron with the vacancy is a highly forbidden process, the electron is free to drift through the material. Thus, this irradiation event has created *electron-hole pairs*—an electron in the conduction band and a hole in the valence band.

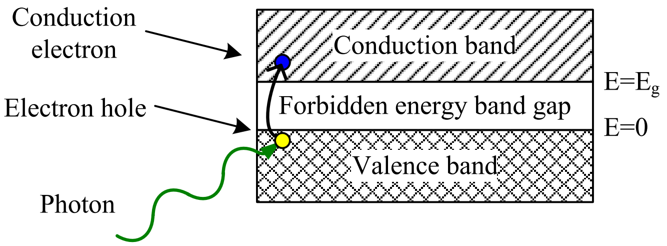


Figure 6. Photoconduction in which a photon raises the energy of a valence electron to the conduction band and leave behind a hole in the valence band.

Photoconduction occurs when an electron is excited across the forbidden energy gap between the valence and conduction bands, as depicted in Figure 6. Radiation of sufficient energy (for example, photons in the ultraviolet-to-gamma range) can increase the number of intrinsic carriers (electrons and holes) by several orders of magnitude. Since photoconduction increases intrinsic carrier concentration, both electrons and holes, the resistivity of semiconductors is decreased according to

$$\sigma = \frac{1}{\rho} = q\,(n_n\,\mu_n + n_p\,\mu_p) \qquad (3)$$

where the subscript *n* denotes the negative electron carriers and *p* signifies the positive hole carriers. The excitation of valence electrons into the conduction band can significantly increase the electrical conductivity of both insulators and semiconductors, this phenomena is known as *radiation-induced conductivity*. Recombination occurs when the electron drops from the conduction band back down to the valence band. The first-order time constant for recombination to occur is known as the *relaxation time*.

Impurities in a crystal constitute structural imperfections which create trapping sites for holes and electrons that are diffusing. Damage also arises in insulators and semiconductors as a result of atomic displacements, whereby point defects serve as charge-carrier donors and traps. Transient effects in these materials are analogous to molecular effects. Irradiation creates secondary-electron charge carriers, and thus affects electrical properties. The longer relaxation time in semiconductors as compared to metals leads to damage that is more permanent. Semiconductors, such as germanium, can have their conductivity altered by irradiation.

Example:
A beam of 5 MeV protons with current density 10^5 /cm^2·sec is incident to the face of a silicon wafer. Find the number of ion pairs formed per unit volume, and the fraction of Si atoms experiencing ionization.
Solution:
The range of 5 MeV protons in air is

$$R_{air} = \left(\frac{E_p}{9.3}\right)^{1.8} = \left(\frac{5}{9.3}\right)^{1.8} = 0.3272\text{ m}$$

Using the Bragg-Kleeman rule, their range in silicon (*i.e.*, the penetration depth into the Si wafer) is

$$R_{\mathrm{Si}} = 3.2\times10^{-4}\,\frac{\sqrt{M_{\mathrm{Si}}}}{\rho_{\mathrm{Si}}}\,R_{air} = 3.2\times10^{-4}\,\frac{\sqrt{28.0855}}{2.3296}\,(0.3272\,\mathrm{m}) = 0.238\,\mathrm{mm}$$

From Table III, silicon requires 3.6 eV to create an ion pair, therefore the number of ion pairs produced is

$$IP = \frac{5\times10^{6}\ \mathrm{eV/proton}}{3.6\ \mathrm{eV/i.p.}}\left(10^{5}\,\frac{\mathrm{protons}}{\mathrm{cm}^{2}\cdot\mathrm{sec}}\right)\frac{1}{0.0238\,\mathrm{cm}} = 5.84\times10^{12}\,\frac{\mathrm{ion\ pairs}}{\mathrm{cm}^{3}\cdot\mathrm{sec}}$$

The number density of silicon is

$$N_{\mathrm{Si}} = \frac{(2.3296\,\mathrm{g/cm}^{3})(6.022\times10^{23}\,\mathrm{atoms/mole})}{(28.0855\,\mathrm{g/mole})} = 4.995\times10^{22}\,\frac{\mathrm{atoms}}{\mathrm{cm}^{3}}$$

So, the fraction of atoms experiencing ionization per second is

$$\frac{IP}{N_{\mathrm{Si}}} = \frac{5.84\times10^{12}}{4.995\times10^{22}} = 1.2\times10^{-10}$$

Table III: Properties of intrinsic germanium, silicon, gallium arsenide, silicon dioxide, silicon nitride, and aluminum oxide at 27°C [5]

Material	Ge	Si	GaAs	SiO_2	Si_3N_4	Al_2O_3
Type	Semi-conductor	Semi-conductor	Semi-conductor	Insulator	Insulator	Insulator
Atomic/Molecular Weight	72.6	28.09	144.63	60.08	140.27	101.96
Density (g/cm^3)	5.33	2.33	5.32	2.27	3.44	3.97
Electron-hole pair generation energy (eV)	2.8	3.6	4.8	17.	10.8	19.1
Dielectric constant ε	16.3	11.9	12.5	3.9	7.5	9.4
Breakdown field (V/µm)	8	30	35	600	900	48
Displacement damage threshold energy (eV)	27.5	25	9.9			
Carrier generation constant (g_0) 10^{13} ehp/cm^3·rad(·)	11.9	4.05	6.92	0.81		
Resistivity (Ω·cm)	50.	2.3×10^{5}	7×10^{7}	$>10^{16}$	10^{7}	$>10^{16}$

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