THE ENERGY DEPENDENCE OF RADIATION DAMAGE INDUCED IN COPPER BY LOW ENERGY H^1 AND He^4 IONS

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TITLE

The Energy Dependence of Radiation Damage Induced in Copper by Low Energy H^1 and He^4 Ions

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ABSTRACT

Bombardments of nominally 99.995% pure copper foils at 77° K have been performed using 50 to 325 keV protons and He^+ ions from the Nebraska Cockcroft-Walton accelerator. Rates of defect production as a function of energy were calculated from measured electrical resistivity changes for each type of ion. Comparison of our experimental values, corrected for low temperature annealing, with the Harrison-Seitz and Neufeld-Snyder theoretical defect production rates show that theory predicts values 10 to 12 times higher than our measurements at 50 keV for both ions. Agreement becomes better at higher energies with the discrepancy about a factor of 2 to 4 at 300 keV. The relationship between the experimental and theoretical values for the defect production rates was found to be linear. For incident protons the relationship is n(EXP.) = 0.319 (n(THE.) - 15.3) and for incident He^+ ions n(EXP.) = 0.604 (n(THE.) - 55.1), where n is the number of defects/incident particle. Various possible reasons for the disagreement between theory and experiment are discussed.

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I. INTRODUCTION

A. GENERAL

Current widespread activity in the field of radiation damage has its origin in the war years when it was realized that fission fragments have enough energy and momentum to displace atoms from their normal sites in solids and perhaps fundamentally alter the properties of these solids. For a time most investigations in this field were primarily concerned with the effects these heavy fission fragments would have on reactor materials. The emphasis has shifted in more recent times as people began to realize that experiments of a more controlled nature could be done by using lighter particles such as electrons, protons, deuterons, or alpha particles to bombard specimens at cryogenic temperatures. It is experiments of this type that have contributed greatly to the present knowledge of radiation damage phenomena.

A charged particle traversing a solid loses most of its energy by exciting electrons. In a metal the conduction electrons are loosely coupled to the lattice, however, and the excited electrons are expected to dissipate the added energy with little if any effect on the lattice structure. It is the more rare encounters with the lattice atoms of the metal that are instrumental in disrupting the structure and producing radiation damage.

A lattice atom dislodged from its normal site leaves a hole in the structure. This hole is called a vacancy. If it comes to rest between normal lattice sites it becomes an interstitial. Above certain temperatures the interstitials and vacancies are able to migrate through the metal, during which course they may annihilate by recombining or they may form vacancy or interstitial clusters. Other behavior is possible also. If impurities are present in the lattice which have introduced strain fields, they may act as trapping centers for whichever defect would help relieve the strain. Grain boundaries, dislocations, or the surface of the metal may also act as traps for the wandering defects. Such group action of the defects is normally expected when one produces the defects at a temperature below 10° K and then warms up or anneals the sample, or if the defects are produced at a higher temperature. However, group defect action has been postulated as an operative phenomenon even during their formation by heavy charged particles at low temperatures. Among such possibilities are the "thermal spike," "displacement spike" and focusing collisions. A lattice atom imparted a large energy by the energetic projectiles (called a "knock-on") may collide with lattice atoms causing them to vibrate with large amplitude as though they had been suddenly heated. No displacement occurs and the vibrational energy is given up to neighboring atoms. These momentary regions of activity, called thermal spikes (R57, R50) dissipate quickly but may cause annealing. The displacement spike (R8)[1] is produced by a knock-on at the end of its path where the mean free

1. Parenthetical numbers prefaced by letter R refer to references listed in the bibliography.

path between collisions with lattice atoms may be about equal to the average lattice spacing. The knock-on would produce a displacement at nearly every collision, producing a region of gross disturbance. Focusing collisions (R51, R31) are a sequence of collisions between lattice atoms which can transport matter and energy along close packed lines of atoms. Such collision chains could ultimately produce an interstitial defect far from the scene of the first collision.

The aim of irradiation damage experiments is to determine which of these, or perhaps other, processes are actually taking place and to gain a qualitative knowledge of them. Hopefully this knowledge will allow formulation of a defect production theory which will predict the nature and distribution of the damage induced in a solid by an energetic beam, the effect of this damage on the physical properties of the solid and the annealing kinetics of the defects as the solid is subsequently heated.

Several methods have been used to detect the presence of irradiation produced defects in metals. Among these are measurement of lattice parameter changes, length changes, and increase in the stored energy of the lattice, but the most common method has been measurement of resistivity increase caused by the defects which interrupt the periodic nature of the lattice.

B. THIS EXPERIMENT

In this experiment we have bombarded copper foils with 50 to 325 keV protons and helium ions. The induced damage was detected

by measuring the increase of electrical resistance of the copper sample. Irradiation and electrical measurements of the sample were carried out with the copper held at a temperature of about 77° K. Detection of the resistance increments was by a unique bridge method, wherein we compared the resistance of a portion of the sample struck by the beam with another portion shielded from the beam. The major objective of these measurements is to determine the rate at which defects are produced as a function of beam energy and to compare this with predictions of the defect production theories. As we shall see, the defect production rates were found to increase monotonically with kinetic energy for both kinds of incident particles, but the observed production rates were always lower than those predicted by theory. When the production rates found for both types of incident ions were corrected for annealing during the irradiation and for the effect of the incident particle remaining in the copper as a scattering center, it was found that the damage production rate for the He^+ ions was about 4 to 6 times the rate for H^+ ions.

II. THEORY OF DEFECT PRODUCTION

A. GENERAL OUTLINE

An energetic particle penetrating a solid substance whose atoms are arranged in a periodic structure usually produces radiation damage by disrupting the periodic structure in a two stage process. The primary stage consists of the direct interaction of the radiation with the lattice by means of displacement of electrons (ionization), displacement of atoms from their lattice sites (the effect that most concerns us), excitation of both atoms and electrons without displacement, and the transmutation of nuclei (R18). Assuming that a lattice atom has been displaced from its normal site because of an interaction with one of the high speed particles, it will in general be given more energy than it needs to merely escape its lattice site. It is called a primary knock-on and can in turn collide with other lattice atoms, displacing them from lattice sites. These collide with and displace others, etc., until no further displacements are energetically possible. This sequence of events initiated by a primary knock-on comprises the secondary stage of the radiation damage process.

Let us assume that we bombard a metal foil with a proton or helium ion beam having uniform flux φ ions/cm^2-sec. over the face of the specimen. In our experiment the beam will come to rest in the foil since the beam energies available to us do not penetrate

our thinnest foils. The number of primary displacements induced by an incident particle of energy E in traversing a distance dx in the foil is given by

$$dn_p = N \sigma_d(E)\, dx$$

where N is the number of atoms per unit volume in the stopping substance and $\sigma_d(E)$ is the cross section per atom for collisions that produce displacements. Assume each primary knock-on produces on the average ν secondary displacements where ν also includes the primary knock-on. ν depends on the energy of the primary knock-on and when averaged over the knock-on energy spectrum we obtain the value $\bar{\nu}$ which is the average number of displacements produced by a beam ion if it undergoes a collision. The total number of displacements produced by the beam particle of energy E is then

$$dn = N \sigma_d(E)\, \bar{\nu}(E)\, dx$$

as it traverses thickness dx. To obtain the number of displacements as a result of the total number of interactions as the primary and secondaries come to rest we integrate the above equation

$$n(E) = N \int_0^E \frac{\sigma_d(E)\, \bar{\nu}(E)\, dE}{-\frac{dE}{dx}} \qquad (1)$$

making use of the stopping power -dE/dx.

Our concern in this section is to arrive at estimates of $\bar{\nu}$, σ_d and dE/dx in order to evaluate this integral.

Turning our attention to the primary process, we can state immediately that transmutation of nuclei will not occur for the particles and energies used in this experiment. The excitation and displacement of electrons will be very important only in that the major portion of a primary particle's energy will be given up to electrons, as will presently be seen. The permanent displacement of atoms is the mechanism that will ultimately lead to measurable radiation damage effects.

B. ENERGY LOSS TO ELECTRONS

When the moving atom has a velocity greater than the velocity of a Fermi electron in the target, the collisions with electrons are quite likely to leave them in excited states, thereby taking energy from the projectile, whereas if the projectile moves slower than the above-mentioned velocity, collisions with the electrons leave them for the most part unexcited and the main energy loss in this region is due to collisions with lattice atoms (R18, R43, R28). In estimating the energy E_i, at which we assume a sharp transition from electronic excitation to atomic excitation takes place, we follow the estimates of Seitz (R49) and Kinchin and Pease (R28). Seitz suggested that, in insulators

$$E_i = \frac{1}{8}\left(\frac{m_1}{m}\right) I \qquad (2)$$

where m_1 is the mass of the moving ion, m is the electronic mass, and I is the lowest electronic excitation energy, given by the low

energy limit of the main optical absorption band. In rough analogy to this, Kinchin and Pease give for metals:

$$E_i = \frac{1}{16}\left(\frac{m_1}{m}\right) E_f \tag{3}$$

where E_f is the Fermi energy. This relation is admittedly quite rough, yet it is reasonable because when the moving ion velocity is much lower than that of the electrons at or near the top of the Fermi level very few electrons can be excited. Using the value of E_f = 7.04 eV for copper (R29), we get E_i = 810 eV for protons, 3200 eV for helium ions, and 50 keV for copper ions as projectiles. As Kinchin and Pease state, these are energy limits below which ionization losses may be neglected. More recently, Lindhard et al. (R32, R33, R34) have treated the sharing of energy dissipation between electronic and atomic motion in some detail. As an example one may calculate from their curves the energy at which the electronic cross section per atom, S_e, is equal to the nuclear stopping cross section, S_n. For copper ions traversing a copper target this energy is calculated to be about 500 keV.

C. DISPLACEMENT OF PRIMARY ATOMS

The displacement of atoms in our target material takes place because of electrostatic forces between the projectile and the atom. Bohr (R7) found a potential of the form

$$V(r) = \frac{Z_1 Z_2 e^2}{r} e^{-\frac{r}{a}} \tag{4}$$

to be reasonably accurate in describing such interactions. $Z_1 e$ is the charge on the moving ion, $Z_2 e$ is the charge on the stationary or lattice atom, r is the separation of the two nuclei, and a is called the screening radius. For very small separations, r, this potential approximates the coulomb interaction between nuclei, while for separations of magnitude a the potential decreases faster, because of the screening effect of the orbital electrons. The screening distance is approximately given by

$$a = \frac{a_o}{\left(Z_1^{2/3} + Z_2^{2/3}\right)^{1/2}} \tag{5}$$

where a_o is the radius of the first Bohr orbit in hydrogen (5.292×10^{-9} cm). $a = 1.637 \times 10^{-9}$ cm for protons on copper and $a = 1.594 \times 10^{-9}$ cm for helium ions on copper.

The classical Rutherford differential scattering cross section is given by

$$d\sigma = \frac{\pi b^2}{4} \cos\frac{\theta}{2} \csc^3\frac{\theta}{2}\, d\theta \tag{6}$$

which is valid for coulomb interactions. Θ is the angle of deflection in the center of mass coordinate system of the incoming particle and b is the distance of closest approach for two charges of $Z_1 e$ and $Z_2 e$ having the same sign when particle 1 has laboratory energy E. These quantities are connected by the following relationship:

$$\frac{Z_1 Z_2 e^2}{b} = \frac{\mu E}{m_1} = \frac{\mu v^2}{2} \tag{7}$$

where v is the lab velocity of particle 1, m_1 its mass, m_2 the mass of particle 2 and μ is the reduced mass $m_1 m_2/(m_1+m_2)$. Assuming elastic collisions (no energy given up in excitation), it is easily shown that the kinetic energy transferred to the stationary particle is

$$T = T_m \sin^2\left(\frac{\theta}{2}\right) \tag{8}$$

where

$$T_m = \frac{4 m_1 m_2}{(m_1 + m_2)^2} E. \tag{9}$$

T_m is the maximum energy that can be transferred, as in a head on collision. Using (8) we may rewrite (6) to give the differential cross section for a transferral of kinetic energy in the range between T and T+dT:

$$d\sigma = \frac{\pi b^2}{4} T_m \frac{dT}{T^2} \tag{10}$$

Bohr (R7) shows that the above formula is quite adequate provided

$$\lambda\!\!\!^{-} \ll b \quad \text{or equivalently,} \quad E \ll \frac{2 Z_1^2 Z_2^2 e^4 m_1}{\hbar^2} \tag{11}$$

where $\lambda\!\!\!^{-} = \hbar/\mu v$ is the de Broglie wavelength divided by 2π. If condition (11) holds, it is possible to construct the quantum

mechanical wave packets which follow the classical orbits assumed in the derivation of (6). Using (11) one sees that if the incident particles are protons and the target is copper we require $E \ll 84$ MeV to use the classical picture and if the particles are helium ions $E \ll 1.34$ BeV. Since the maximum energy used in the present experiment is 325 keV, the above condition is adequately fulfilled.

Since the scattering potential is not the coulomb interaction due to two bare nuclei, but rather is probably more like that given by (4), deviations from Rutherford scattering may be expected whenever the exponential factor assumes importance. The major part of the scattering is done inside the screening distance, provided

$b \ll a$ or equivalently $$E \gg 2E_R \frac{m_1}{\mu} z_1 z_2 (z_1^{2/3} + z_2^{2/3})^{1/2} \tag{12}$$

where $E_R = e^2/2a_o$ is the Rydberg energy for hydrogen (13.6 eV). For protons on copper this requires $E \gg 2.59$ keV, for H_e ions on copper $E \gg 5.57$ keV, and for copper on copper $E \gg 199$ keV. Let us denote this limiting energy by E_H. When the energies are much less than E_H the screening is very strong and the atoms are assumed to scatter as if they were classical hard spheres. (R50) Although the beam energies used in this experiment are always 50 keV or greater, the above limits will be violated because the beam will not penetrate the copper foil thicknesses used and will reach zero kinetic energy inside the foil. For purposes of calculation in this paper we will use the Rutherford cross section for energies above E_H and a hard sphere cross section below E_H.

An atom can be permanently removed from its normal lattice site only if it receives some minimum energy E_d, called the threshold displacement energy. Seitz (R49) first estimated the magnitude of this energy to be about 25 eV. Eggen and Laubenstein (R20) measured this threshold energy in copper by bombarding with high energy electrons and noting at what electron energy the electrical resistivity of the copper began to increase. Their measured threshold electron energy corresponded to an E_d of 25.0 ± 1.0 eV. We shall use this value of E_d in this paper although other workers have measured E_d values somewhat lower (R11, R4).

We assume that if a lattice atom receives an energy greater than E_d it will be displaced. An incident particle of energy E will produce displacements, then, whenever it transmits an energy in the range E_d to T_m. The total cross section for producing displacements is gotten by integrating (10) as follows:

$$\sigma_d = \frac{\pi b^2}{4} T_m \int_{E_d}^{T_m} \frac{dT}{T^2}$$

$$= \frac{\pi b^2}{4} \left(\frac{T_m - E_d}{E_d} \right)$$

which becomes

$$\sigma_d = \frac{4\pi E_R^2 a_o^2 Z_1^2 Z_2^2}{E_d} \frac{m_1}{m_2} \left(\frac{E - \frac{1}{M} E_d}{E^2} \right) \qquad (13)$$

where M is defined by the equation

$$T_m = ME \qquad \text{or} \qquad M = \frac{4 m_1 m_2}{(m_1 + m_2)^2}$$

Note that E_d/M represents the lowest energy an energetic particle may have and still transfer energy E_d to a lattice atom in a

head on collision. For protons on copper

$$\sigma_d = 34.6 \times 10^{-18} \left(\frac{E - 0.41}{E^2}\right) \text{ cm}^2$$

for E in keV, and for helium ions on copper assuming the helium ions have a charge of 1 e,

$$\sigma_d = 138.6 \times 10^{-18} \left(\frac{E - 0.112}{E^2}\right) \text{ cm}^2$$

The differential cross section for energy transfer when hard sphere collisions are taking place may be shown to be (R18)

$$d\sigma_H = \frac{\pi a^2}{T_m} dT \tag{14}$$

where a is the diameter of the effective hard sphere and is approximately equal to the screening distance as given by (6) (R18). We shall use the above way of defining the hard sphere diameter although there are other ways which are energy dependent, such as the distance of closest approach or distance at which dr/dt = 0 in an encounter where the potential of equation (4) is used. Note that all energy transfers from E = 0 up to E = T_m are equally probable, and that the scattering is isotropic. Integrating (14) to get the total displacement cross section for transferral of energy between T_m and E_d we find

$$\sigma_{Hd} = \pi a^2 \left(\frac{E - E_d/M}{E}\right) \tag{15}$$

For protons on copper $\sigma_{Hd} = 8.419 \times 10^{-18} \left((E - 0.41)/E\right)$ cm^2 and $\sigma_{Hd} = 7.979 \times 10^{-18} \left((E - 0.112)/E\right)$ cm^2 for helium on copper.