A fast ion penetrating a solid creates a track of excitations. This can produce displacements seen as an etched track, a process initially used to detect energetic particles but now used to alter materials. From the seminal papers by Fleischer et al. [1] to the present [2], ‘Coulomb explosion’ and thermal spike models are treated as conflicting models for describing ion track effects. Here molecular dynamics simulations of electronic-sputtering, a surface manifestation of ion track formation, show that ‘Coulomb explosion’ produces a ‘heat’ spike so that these are early and late aspects of the same process. Therefore, differences in scaling are due to the use of incomplete spike models.

PACS numbers:61.80.Az,79.20.-m,34.50.Fa,79.20.Ap

Since Thompson and Rutherford there has been interest in the track of excitations produced by an energetic ion penetrating a solid. The fast ion excites the electron cloud producing excitons and electron-hole pairs in an insulator [6]. The electrons expelled from the track create additional excitations, then cool to the lattice, screening and finally recombining with the holes. They eventually return to the ground state by radiative or non-radiative processes. Prior to this decay an excited region with a net repulsive energy persists. Fleischer et al. [1] (hereafter FPW) proposed that this repulsion could produce displacements altering the material along the particle’s track. When accounting only for the holes the process has been called ‘Coulomb explosion’, suggested to produce amorphized tracks [6], cratering, and sputtering [6-11]. Tracks have also been observed in liquids [5], semiconductors [6] and even metals [6] when the excitation density in the track is sufficiently large.

In describing thresholds for track registration in insulators, FPW compared their ‘Coulomb explosion’ model to the often used thermal spike model for defect production [8,9]. Such a comparison is not only of historical interest but persists in the recent literature [2]. However, one model describes a mechanism for energy-input (electrostatic potential energy into kinetic energy), whereas the other describes the transport of energy out of a ‘heated’ cylindrical region. Here we present a molecular dynamics (MD) description of the response of a solid to a repulsive track. We show that at high excitation densities the primary effect is the production of a cylindrical ‘heat’ spike. Therefore, distinctions made between ‘Coulomb explosion’ and spike models can be artifacts of the approximations used.

MD simulations are carried out for a model solid with a surface because of our interest in electronically-induced sputtering, the surface manifestation of track formation in insulators [10]. As in track registration, both spike models [10] and ‘Coulomb explosion’ models [3-12] have been employed to parametrize laboratory data for electronic sputtering. In spike models the atoms in the ion track are assumed to have a radial temperature profile determined by the energy deposited per unit path length, (dE/dx). When the energy of an atom exceeds a barrier, defect production in the bulk or evaporation at the surface can occur. From the time dependent surface temperature, $T_{surf}$, the spike sputtering yield, $Y_S$, the number of atoms ejected per ion track is calculated integrating the surface flux over time and area. When a radial diffusion equation is used to determine $T_{surf}$, then $Y_S \propto (dE/dx)_{eff}$ for a fixed track radius at high (dE/dx)$_{eff}$ [3,12], where (dE/dx)$_{eff}$ is the fraction of dE/dx going into energetic non-radiative processes [3]. This has been used to describe sputtering produced by a track ‘heated’ by individual repulsive decay events [11], secondary electrons [2] and a repulsive track [3,12]. However, when conditions leading to the (dE/dx)$_{eff}$ dependence should apply, the model fails because the energy transport is not diffusive [7]. A melt and a pressure pulse control the energy transport leading to [7],

$$Y_S \approx 0.18(r_{cyl}/U)(dE/dx)_{eff}$$

(0.1)

where $U$ is the solid’s cohesive energy and the effect depth scales with $r_{cyl}$, the initial mean radius of the spike. Here we show that a repulsively-induced ‘heat’ spike is produced in an ion track over a broad range of excitation densities and the transport is not diffusive. Therefore, a correct spike model must be used.

We simulate the evolution of a track of repulsive energy that might be produced by a fast incident ion. Since sputtering [1] and crater formation in both amorphous and crystalline, atomic or molecular solids can be scaled over a broad range of material properties, we use a model solid made of atoms interacting via Lennard-Jones (LJ) potentials. We use parameters for condensed gas solids, but the results scale with the energy and length parameters. Also, we showed earlier that scaling was maintained using more complex potentials [13]. Interactions occur between all atoms within a cut-off radius $r_{cut} \approx 2.5l$, where $l$ is the mean atom spacing. The (001) layer spacing in the fcc lattice is $l_s \approx 0.8l$. MD simulations focusing...
on damage have been made for repulsive energy between a few charges in LiF [18], a distribution of holes in Si [19], and using an assumed velocity distribution [20]. Here we follow the conversion of a track of repulsive energy into atomic motion in a solid.

On electronically exciting a condensed gas solid, the track of excitations can produce large sputtering yields at high $dE/dx$, a process relevant to icy outer solar system bodies [18]. Ejection is presumed to be due to the net repulsive energy, but a quantitative description is lacking. At high ionization densities the net repulsion can persist if the holes have low mobility and are not sufficiently screened during the time displacements could occur. Even when holes are neutralized, the excited atoms have overlapping charged clouds which act repulsively at high $dE/dx$ [16]. Here we describe the net repulsion between ‘excited’ atoms in a track using $V = (ε^2/\rho) \exp(-r/\alpha)$, where $\alpha$ is an average screening constant [21]. We do not distinguish between closely spaced, partially screened holes or overlapping excited species, since in both cases the electrons screen the interactions between ‘excited’ neighbors [16,17]. The interactions of ‘excited’ atoms with unexcited neighbors was left unchanged; including a weak polarization had a small effect. For energy conservation, a larger cut-off, $r_{cut} = 7\alpha$, is used for interactions between ‘excited’ species.

The material is ‘excited’ by instantaneously changing the potentials between atoms in the track. The resulting velocities and positions of all particles are then followed [14]. This is done for a number of excitation densities and screening constants. In a second set of calculations the excitations were quenched (recombination). This was done statistically so the average number of excitations in the track decayed exponentially, $\exp(-t/\tau)$ where $\tau$ is the quenching (neutralization) time. The number of excitations per unit path length, $dN/dx$, is related to $dE/dx$ for fast ions [1] and the mean radius of the distributions, $r_{cout}$, depends on the speed of the incident ion. For given track parameters $[dN/dx, r_{cout} (here \sim l_s)]$ excited species are chosen randomly. On excitation, atoms are sputtered if they cross a plane $2r_{cut}$ above the ‘top’, here the (001) surface. The number of atoms ejected in each run is called the yield, which is related to the track of damage in FPW but is more easily measured. Sample depth was chosen to be at least twice $r_{cout}$. Different boundary conditions did not change the average yields significantly. The sample size ($3 \times 10^4 - 3 \times 10^5$) and simulation times ($15 - 80$ ps) were adjusted to $a$ and $dN/dx$. Extending times by tens of picoseconds or doubling the thickness did not change the average yield. The distribution in the yield is broad, especially for small $l_s(dN/dx)$ where sputtering occurs when two excitations are produced close together [3]. Results, averaged over $\sim 10 - 200$ excitation distributions, are given in Fig. 1a as a function of charge density, $l_s(dN/dx)$, for 3 values of the screening constant $\alpha$. The yield is seen to be quadratic in $dN/dx$ for each $\alpha$ and increases non-linearly with $a$; lines $Y = 14. \ln[1.3(\alpha/r_{cout})] l_s(dN/dx)^2$ give a good fit [3]. The yields as a function of neutralization time, $\tau$, for a fixed $(dN/dx)$ and $\alpha$ are given in Fig. 1b scaled by the fit in Fig 1a. Because neutralization is treated stochastically, the size of the yield is affected even for relatively large $\tau$ and only becomes independent of the neutralization rate for $\tau \gtrsim 10\tau_D$, with $\tau_D$ the Debye period for the lattice ($\sim 0.5$ ps) [17]. However, the yield is still seen to be nearly quadratic in $(dN/dx)$ for the full range of $\tau$ with an additional weak dependence on excitation density.

In MD simulations details of the energy transfer and sputtering can be extracted. At very low excitation densities ($Y \lesssim 1$) the probability of neighbors being excited near the surface leads to ejection. At the highest excitation densities ($Y \gtrsim 5$), the repulsive energy is transferred to neighbors in times $\sim 0.2\tau_D$ producing a cylindrically heated region. Comparing the evolution of the resulting radial temperature profiles with our earlier simulations of ‘heat’ spikes, the energy transport is found to be similar. That is, we find for the repulsively produced heat spike transport is not diffusive [14], a melt and a pressure pulse are formed and these control the energy transport. Seiberling et al. [12] suggested that a repulsive
track could produce a heat spike but used the standard diffusive model to obtaining a yield dependence very different from that calculated here.

![Diagram](image)

**FIG. 2.** Energy distribution of the ejecta for \( l_a dJ/dx = 2 \), \( a = 1.13 l_s \) and \( r_{coul} = l_s \), for \( \tau = \tau_D \) (a) and \( \tau = 20 \tau_D \) (b). The dotted line shows the energy spectrum from a cylindrical spike, as in ref. [14], with \( (dE/dx)_{cyl} \approx 45U/l_s \) and \( r_{cyl} \approx 2.6 l_s \). For the “Coulomb” spike produced in (b), \( (dE/dx)_{coul} \approx 38U/l_s \) and \( r_{coul} \approx 2.6 l_s \), while the spike in (b) has a 30% lower \( (dE/dx)_{eff} \). Peaks are prompt ejecta, with position determined by the potential energy between neighbors and the surface binding energy.

From the energy distributions for the ejecta at high excitation density two regions can be seen in Fig. 2. At large ejecta energies, \( E > U \), the spectra has peaks due to prompt ejection from the upper surface layers of the initial track. This accounts for \( \approx 20\% \) of the ejecta at large \( dJ/dx \) but dominates at very small \( dJ/dx \). On the other hand, the principal component of the ejecta in Fig. 2 exhibits an energy distribution like that found in our studies of ejection from a narrow cylindrical heat spike shown as the dashed line [17]. That is, there is a broad, quasi-thermal distribution at low ejecta energies, \( E < U \) which gives way to a non-Maxwellian, \( \sim E^{-2} \) dependence at \( E > U \). The latter dependence is characteristic of low energy cascades in a solid [4] but differs from thermal spike model predictions. Surprisingly, although the ejection process changes in going from the lowest to the highest \( dJ/dx \), there is no dramatic change in the dependence of the yield on \( dJ/dx \) in Fig. 1. This is due to the nearly linear dependence of spike yield on \( (dE/dx)_{eff} \) discussed below.

From the above, the primary effect of the repulsive decay at the high excitation densities is the production of a ‘heat’ spike. This is also shown quantitatively using the MD calculation to determine the repulsive energy driving atomic motion in times short compared to desorption times. At \( \sim 0.2 \tau_D \) the energy density deposited via repulsion is \( (dE/dx)_{rep} \approx 0.15 e^2 \), for \( a = 1.13 l_s \). This energy is localized in a cylindrical region of radius \( r_{rep} \approx 2.6 l_s \) for our values of \( r_{coul} \). Using \( (dE/dx)_{eff} = (dE/dx)_{rep} \) and \( r_{coul} = r_{rep} \) as initial conditions in the expression for the yield from a cylindrical ‘heat’ spike in Eq. 1 gives a yield that has the same dependence on \( dJ/dx \) and a size within 10% of those in Fig. 1a. Therefore, if a repulsive region is sustained, a ‘heat spike’ is formed at high \( dJ/dx \) which determines the subsequent energy transport, sputtering and displacements. Therefore, ‘Coulomb explosion’ and spikes are the early and late aspects of the same process, so that contradictions for effects occurring later than \( \sim 0.2 \tau_D \) are due to incorrect descriptions of the ‘heat’ spike. Below we examine under what conditions a ‘Coulomb’ track is sustained long enough to produce a ‘heat’ spike.

![Diagram](image)

**FIG. 3.** The change in the total energy in the track of interacting holes and electrons vs. time for ‘excited’ electrons with \( l_s dJ/dx = 1 \), and a dielectric constant \( \epsilon = 1 + 6 \epsilon_o \). A fraction of the energy will be converted to atomic motion. No scattering or inelastic energy loss (solid), no inelastic energy loss but scattering \( \lambda_s = l \) (dashed), for \( \lambda_{in} = l \) and \( \Delta \epsilon_{in} = 0.03 \) eV (dotted), for \( \lambda_{in} = l \) and \( \Delta \epsilon_{in} = 0.1 \) eV (dash-dotted). Since neutralization quenches the energy of a spike, we simulated the neutralization and screening of holes by free electrons. That is, we make a separate simulation in which we track the cooling of the ‘excited’ electrons in the field of the track of positive charges. The lattice is like that in the MD simulations and we choose appropriate mean-free paths for elastic, \( \lambda_s \), or inelastic, \( \lambda_{in} \), scattering, both assumed to be isotropic [22]. A time dependent \( a \) or an average \( a \) and \( \tau \) can be determined from such a simulation. Instead, we give in Fig. 3 the instantaneous total potential energy density in the track vs. time. This is the energy available for repulsive heating of the lattice. The interactions between the positive charges in the track and the electrons are Coulombic beyond an atomic radius, \( a_o \). Inside \( a_o \), the potentials gradually become flat giving a binding energy of \( \sim 8.6 \) eV. We examined a number of potential forms inside \( a_o \) and initial electron energies and the trends are the same. Electrons are ‘excited’ by receiving, on the average, 15 eV, roughly the energy dissipated to the lattice (W-value minus ionization energy [23]). This does not treat the very fast electrons which would further slow neutralization.

For only elastic scattering, the total energy in the track
in Fig. 3 is sustained with a very large effective a. Examining the radial dependence of the electron density, a fraction of the electrons cool rapidly by electron-electron collisions partially neutralizing the track. This is di-electronic recombination, also called Auger recombination. The remainder of the electron cloud, now ‘hotter’ but more fully screened, expands to a larger average radius. Varying λ affects the electron density distribution, whereas including an inelastic energy loss, Δε_{in}, decreases the potential energy in the track. This also heats the lattice, but here we are interested in the repulsive energy. For a Δε_{in} and λ_{in} roughly corresponding to electrons in a polymer or in ice (dotted curve) [22], it is seen in Fig. 3 that most of the initial track energy remains during the time it takes to produce a ‘heat spike’ repulsively, \( \sim 0.2\tau_D \). This would correspond to an intermediate \( \tau \) and relatively large a in Figs. 1b and 2a. Therefore, for a reasonable inelastic energy loss, screening is not sufficient to quench the repulsive production of a ‘heat spike’. Increasing Δε_{in} (dot-dashed curve) or decreasing λ_{in} leads to more rapid but still incomplete quenching over the relevant time. Quantities for specific measurements need to be used and the role of excited states in the track must be included.

In this paper we close the circle on one aspect of an old [1] but topical [2] problem, the effect on a solid of a track of excitations produced by a fast penetrating ion. A MD simulation of repulsive explosion in a track of excitations has been carried out and the effect of neutralization has been examined. This was done for narrow screened Coulomb spikes in a condensed-gas solid to constrain the simulation size. However, we showed earlier that the yields scale with U and \( (dE/dx)_{eff} \). Whereas at low excitation densities ejection can occur if neighboring ‘excited’ species are formed near the surface [3,4], at the higher excitation densities the repulsive energy in the track produces a ‘heat’ spike. Therefore, ‘Coulomb explosion’ and spike models are the early and late aspects of the repulsive decay process. The spike formed by the repulsive heating of the lattice can lead to displacements and ejection (electronic sputtering), examined here. Because the yield is roughly linear in \( (dE/dx)_{eff} \) at high excitation densities and \( (dE/dx)_{eff} \) is proportional to \( (dJ/dx)^2 \), the yield from a repulsive track is seen (Fig. 1a) to vary smoothly, having the same quadratic dependence on dJ/dx in going from low to high excitation densities although the character of the ejection process changes. At high excitation densities the yield can now be estimated by substituting the repulsive energy deposition into the new, useful expression for spike sputtering in Eq. [1].

At higher excitation densities than those studied here, the more distributed ‘heat’ spike produced by the electron cooling to the lattice might also be important [3]. Earlier applications of a ‘heat’ spike for describing track formation or electronic sputtering founded on the use of a spike model in which the energy transport was described incorrectly. That is, the transport is not diffusive [4] and the local kinetic energy distribution is not Maxwellian (Fig. 2). Calculations should now focus on accurately describing the interactions in an ion track for times \( \lesssim 0.2\tau_D \) including electrons, holes and excitons, as the latter also contribute to the net repulsive energy.

**ACKNOWLEDGMENTS**

We thank M. Liu for the neutralization calculations, H. Urbassek, R. Vidal, and R. Baragiola for comments, the NSF Astronomy and Chemistry Divisions for support.

---

[1] R. L. Fleischer, P. B. Price, and R. M. Walker, J. App. Phys. 36, 3645 (1965); R. L. Fleischer et al., Phys. Rev. 156, 353 (1967). (FPW).
[2] C. Trautmann, S. Klaumunzer and H. Trinkaus, Phys. Rev. Lett. 85, 3648 (2000).
[3] R. E. Johnson and W. L. Brown, Nucl. Instrum. Meth. 170, 103 (1982).
[4] P. K. Haff, Appl. Phys. Lett. 29, 443 (1976).
[5] W. M. Bartzac, L. D. A. Siebels, and A. Hummel, J. Phys. Chem. A 101, 8135 (1997).
[6] T. Schenkel et al., Phys. Rev. Lett. 81, 2590 (1998).
[7] Z. G. Wang et al., J. of Phys. Condens. Matter 6, 6733 (1994).
[8] M. Toulemonde et al., Nucl. Instrum. and Meth. B 166-167, 903 (2000).
[9] H. Trinkaus and A. Y. Ryazanov, Phys. Rev. Let. 74, 5072 (1995).
[10] R. E. Johnson, Rev. Modern Phys. 68, 305 (1996).
[11] R. E. Johnson, M. Pospieszalska, and W. L. Brown, Phys. Rev. B 44, 7263 (1991).
[12] L. E. Seiberling, J. E. Griffith, and T. A. Tombrello, Rad. Eff. 52, 201 (1980).
[13] M. Jakas, Rad. Effs. and Deffs. in Solids 152, 157 (2000).
[14] E. M. Bringa, R. E. Johnson and M. Jakas, Phys. Rev. B 60, 15107 (1999).
[15] C. Watson and T. Tombrello, Rad. Eff. 89, 263 (1985).
[16] P. Stampfl and K. H. Bennemann, Appl. Phys. A 60, 191 (1996).
[17] E. M. Bringa and R. Johnson, Nucl. Instr. and Meth. B 143, 513 (1998), ibid 152, 267 (1999).
[18] R. E. Walkup and Ph. Avouris, Phys. Rev. Lett. 56, 524 (1986).
[19] M. Hedström and H-P. Cheng, Phys. Rev. B 62, 2751 (2000).
[20] S. A. Fedotov et al., Nucl. Instr. and Meth. B 118, 724 (1996).
[21] A. F. Lishchitz and N. R. Arista, Phys. Rev. A 57, 200 (1998).
[22] L. Sanche, T. D. Mäerk, and Y. Hatano, in IAEA-TECDOC-799, IAEA, Vienna, 1995, p. 277.