Energy Loss of Swift Proton Clusters in Solids*

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Energy-loss measurements in transmission experiments on carbon and gold foils with ~100-keV/nucleon H_2^+ and H_3^+ beams reveal that protons moving in spatially correlated clusters have effective charge numbers significantly larger than unity. The theory traces the origin of this effect to the coherent dynamic response of the target electrons due to the vicinage of particles in tight clusters and, in linear response approximation, predicts quantitatively the observed enhanced energy loss of proton clusters.

Consider a molecule of mass M_c and kinetic energy $E=\frac{1}{2}M_c v^2$ impinging on a solid such that the speed v exceeds $v_0=e^2/\hbar$. Within a few atomic layers, the valence electrons are stripped, at time $t=0$, leaving a cluster of spatially correlated atomic ions to penetrate into the target. During penetration, the cluster explodes: The distance between ions i and j, $R_{ij}(t)$, increases from the initial value, $R_{ij}(0)$, under the influence of Coulomb repulsion. The kinetic energy loss of the cluster per unit distance traversed in the solid, that is, the cluster stopping power $S_c(v)$, in first Born approximation can be written as $S_c(v)=Z_c^2(t)S_p(v)$, where $Z_c^2(t)$ is the square of the effective charge number of the cluster, and $S_p(v)$ is the proton stopping power. Clearly, if all R_{ij} were shorter than a characteristic small distance, r_{close}, to be determined, the cluster would act as a unified point charge with $Z_c^2(R_{ij}<r_{\mathrm{close}})=(\sum Z_i)^2$. When the $R_{ij}(t)$ become longer than a characteristic distance r_{dist}, the ions act independently such that $Z_c^2(R_{ij}>r_{\mathrm{dist}})=\sum Z_i^2$, and the stopping power of the cluster becomes equal to the sum of the stopping powers of its constituents.

This Letter reports first determinations of $Z_c^2(t)$ by energy-loss measurements in transmission through solids with H^+, H_2^+, and H_3^+ projectiles as a function of the cluster dwell time, $\tau=D/v$, in a target of thickness D. We derive $Z_c^2(t)$ from linear response theory in good agreement with experiment, and obtain estimates for r_{close} and r_{dist}. The phenomenon of dwell-time–dependent stopping powers of ion clusters in solids is important for the prediction of penetration properties. More significantly, it provides quantitative information about the interference in clusters between the wakes of electron-density fluctuations trailing charged particles.[1]

Beams of H^+, H_2^+, and H_3^+ from the New York University accelerator, of energies ranging from 60 to 300 keV, were stabilized in energy to $\lesssim 0.2\%$ through a crossed-field analyzer. The energies of the projectiles were measured in a magnetic spectrometer which was calibrated with protons by (p,γ) and (p,α) resonance reactions. The beam energies after penetration through a foil were measured with a silicon surface-barrier detector.[2] The counting statistics were adjusted to limit counting errors to less than 0.1%. Under our conditions, the influence of energy losses in nuclear collisions can be neglected.

The energy loss of protons in the detector-surface Au and Si dead layers agreed with measurements by Siffert, Forcinal, and Coche.[3] The energy losses of H_3^+, H_2^+, and H^+ projectiles were in the ratios 3.8:2.3:1 at $E_p=E/M_c=60$ keV/nucleon and 4.6:2.5:1 at $E_p=100$ keV/nucleon, rather than in the ratio 3:2:1 expected for clusters of independent protons. (After completion of our experiments, we learned of observations with qualitatively similar trends reported in an early paper by Ewing[4] and in a recent paper by Dettmann, Harrison, and Lucas.[5]) Carbon and gold foils ranging in D from 400 to 2300 Å were inserted 4 cm from the detector. The energy losses, ΔE, of the three beams were measured as a function of D at equal $E_p=E/M_c$. The distance between the target and the detector was large enough for the clusters to enter the detector as independent protons. Figure 1 shows the experimental results in the form $\Delta E(H_2^+)/2\Delta E(H^+)$ and $\Delta E(H_3^+)/3\Delta E(H^+)$.

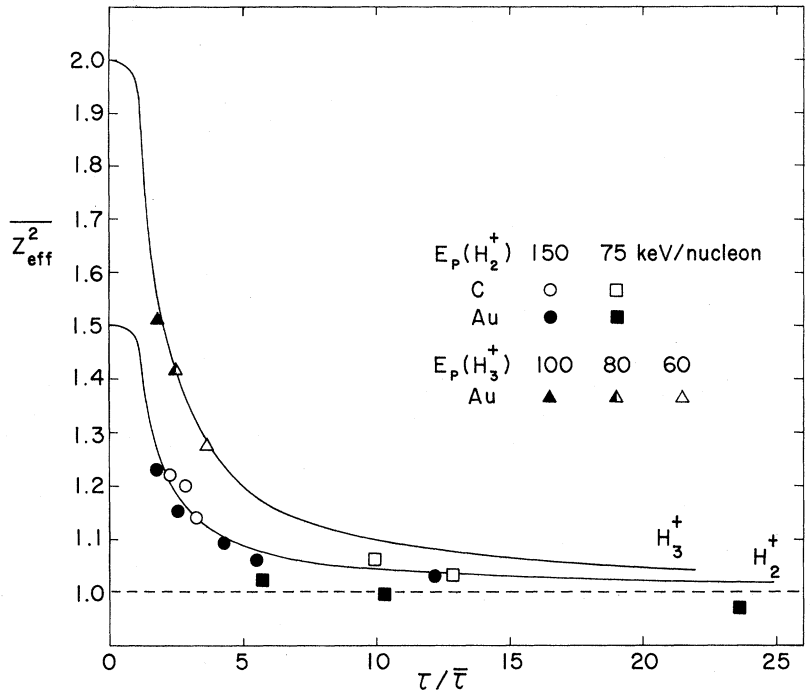

FIG. 1 Measured energy-loss ratios $\Delta E(H_2^+)/2\Delta E(H^+)$ and $\Delta E(H_3^+)/3\Delta E(H^+)$ in various foils as a function of the dwell time in the foil, in units of $\bar{\tau}$. Experimental uncertainties are ±5% for the H_2^+ points and ±10% for H_3^+. The values of $\bar{\tau}$ for H_2^+ are calculated as described in the text following Eq. (6); the $\bar{\tau}$ for H_3^+ at 60, 80, and 100 keV/amu in Au were estimated to be 3×10^2, 3.8×10^2, and 4.7×10^2 a.u., respectively.

Each point represents the average of three or four independent measurements which were reproducible within ± 5 and 10%, respectively. The three measurements with H_3^+ cover our available energy range. The abscissa gives τ in a reduced scale suggested by the theory developed presently for the example of a diatomic cluster applicable to H_2^+.

Let two point charges, Z_1e of mass M_1 and Z_2e of mass M_2, proceed with velocity $\vec{v}$ parallel with the z axis of a cylindrical coordinate system (z, ρ) in a medium characterized by the dielectric function $\epsilon(k, \omega)$. The particles are separated by a vector $\vec{R}$ from 1 to 2, with components $(\vec{d}, \vec{b})$ in the z and ρ directions, respectively. Starting from an initial distance $R(0) \equiv R_0$, the separation has grown under the influence of the force $Z_1Z_2e^2/R^2$ to $R(t)$ at the time

$$t = t_0\{\xi^{1/2}(\xi-1)^{1/2} + \ln[\xi^{1/2} + (\xi-1)^{1/2}]\},$$

where $t_0 \equiv (\mu R_0^3/Z_1Z_2e^2)^{1/2}$ and $\xi \equiv R/R_0$, with μ the reduced mass. This function is displayed as the dashed curve in Fig. 2.[6]

In linear response theory, the particles set up the scalar electric potential

$$\varphi(\vec{r}, t) = \frac{e}{2\pi^2 v}\int d^2\kappa \int d\omega \frac{e^{i[\vec{\kappa}\cdot\vec{\rho} + (z/v - t)\omega]}(Z_1 + Z_2 e^{-i(\vec{\kappa}\cdot\vec{b} + \omega/vd)})}{(\kappa^2 + \omega^2/v^2)\epsilon(k, \omega)} \tag{1}$$

where $k^2 = \kappa^2 + \omega^2/v^2$. The cluster stopping power,

$$S_c = Z_1 e\,\partial\varphi/\partial z\,|_{\vec{r}=\vec{v}t} + Z_2 e\,\partial\varphi/\partial z\,|_{\vec{r}=\vec{v}t+\vec{R}(t)},$$

becomes

$$S_c = \frac{2e^2}{\pi v^2}\int_0^\infty \kappa\, d\kappa \int_0^\infty \frac{\omega d\omega}{(\kappa^2 + \omega^2/v^2)} \mathrm{Im}\left(-\frac{1}{\epsilon(k, \omega)}\right)[Z_1^2 + Z_2^2 + 2Z_1Z_2J_0(\kappa b)\cos(\omega d/v)], \tag{2}$$

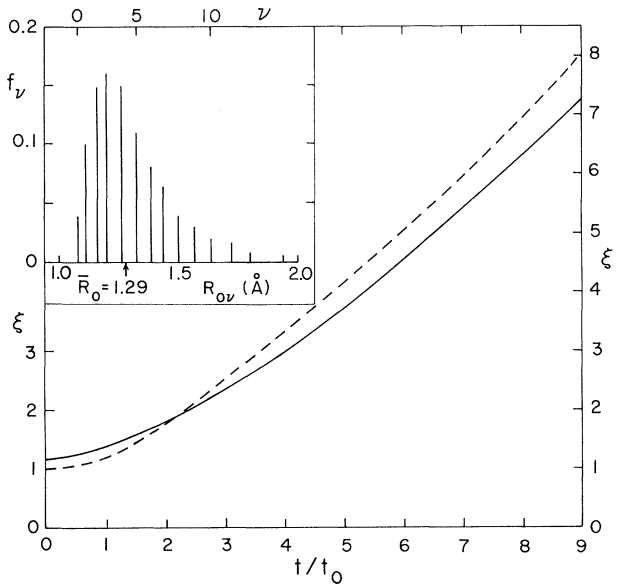

FIG. 2. Relations between intermolecular distances and time for exploding proton pairs. The dashed curve represents the function $\xi(t/t_0)$ given in the text preceding Eq. (1). The solid curve shows the same function averaged over the distribution of internuclear distances in H_2^+ ion beams displayed in the inset, as discussed in connection with Eq. (6).

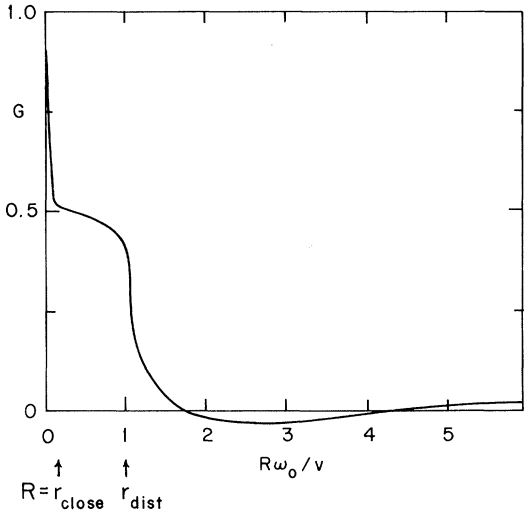

FIG. 3. The directionally averaged interference function versus the reduced interparticle distance, with $r_{close}=\hbar/2mv$ and $r_{dist}=v/\omega_0$.

since $\dot{\vec{R}} \ll \vec{v}$ in the present context. The last term describes the interference caused by the vicinage of the projectiles, with J_0 the zero-order Bessel function of the first kind. At our particle velocities, or under channeling conditions, the target core electrons are ineffective; the stopping power from interactions with the valence electrons dominates, to which Bethe's theory applies.[7] If the valence electrons can sustain well-defined plasmons of energy ω_0, where $\omega_0=(4\pi e^2 n_0/m)^{1/2}$, one can set $\mathrm{Im}(-1/\epsilon)\simeq(\pi/2)(\omega_0^2/\omega_k)\delta(\omega-\omega_k)$, with $\omega_k^2\simeq\omega_0^2+k^4/4m^2$, and Eq. (2) becomes

$$S_c=\frac{e^2\omega_0^2}{v^2}L\left[Z_1^2+Z_2^2+2Z_1Z_2 g\frac{\vec{R}\omega_0}{v}\right]. \tag{3}$$

In the limit $v\gg v_0$, the forefactor with the stopping number

$$L=\int_0^\infty \frac{\kappa\, d\kappa}{\kappa^2+\omega_k^2/v^2}=\ln\frac{2mv^2}{\hbar\omega_0} \tag{4}$$

is equal to the Bethe formula for S_p. We have averaged the interference function $g(\vec{R}\omega_0/v)$ over all cluster orientations with the result $G(R\omega_0/v)$ shown in Fig. 3. When $R<r_{close}=\hbar/2mv$, G is of the order unity and $Z_c^2=(Z_1+Z_2)^2$. When $r_{close}<R<r_{dist}=v/\omega_0$, G has values close to $\frac{1}{2}$ and varies only slowly with R so that $Z_c^2\simeq\frac{1}{2}[(Z_1^2+Z_2^2)+(Z_1+Z_2)^2]$. When $R>r_{dist}$, G drops sharply to small values and oscillates with decreasing amplitude about zero, giving $Z_c^2=Z_1^2+Z_2^2$ as for independent point particles. The oscillations signify the interference between the two wakes of electron-density fluctuations trailing particles[1] separated by more than the screening length v/ω_0. In terms of r_{close} and r_{dist} so obtained, one retrieves Eq. (4) in Bohr's formulation $L=\ln(r_{dist}/r_{close})$.

The H_2^+ molecules from an ion source impinging on a solid are in one of the nineteen vibrational states $\nu=0,1,\ldots,18$, with probability f_ν and initial distance $R_{0\nu}$ shown in Fig. 2.[8,9] The lowest value is $R_{00}=1.06$ Å, and the mean $\bar{R}_0=1.29$ Å. After time t, the internuclear distance has grown from $R_\nu(0)\equiv R_{0\nu}$ to $R_\nu(t)$. We make contact with the experiment by averaging G over time up to τ,

$$\bar{G}(\tau)=\frac{1}{\tau}\sum f_\nu\int_0^\tau G\frac{R_\nu(t)\omega_0}{v}dt \simeq\begin{cases}\frac{1}{2} & \text{for } \tau<\bar{\tau},\\ \frac{1}{2}\bar{\tau}/\tau & \text{for } \tau>\bar{\tau},\end{cases} \tag{5}$$

in terms of $\bar{\tau}\equiv\sum f_\nu\tau_\nu$, where $\tau_\nu=\tau(\xi_\nu=v/R_{0\nu}\omega_0)$ is the time required for $R_\nu(t)$ to become equal to $r_{dist}=v/\omega_0$. The vicinage function $\zeta^2(\tau)\equiv Z_c^2(\tau)/\sum Z_i^2$ measures the enhancement of the energy loss of clusters in the electron gas. For diatomic clusters it becomes

$$\zeta^2(\tau)=[Z_1^2+Z_2^2+2Z_1Z_2\bar{G}(\tau)]/(Z_1^2+Z_2^2). \tag{6}$$

In the approximation Eq. (5), it is a universal function of $\tau/\bar{\tau}$, as shown in Fig. 1 by the curve for the (homonuclear) diproton cluster in the form $\bar{Z}_{\rm eff}^2 = Z_c^2(\tau)/2$. We calculated $\bar{\tau}$ of H_2^+ for the distribution shown in Fig. 2. The result is displayed as the solid curve in the same figure. Given v/ω_0 and $R_{00} = 1.06$ Å, the abscissa value corresponding to $\xi = v/R_{00}\omega_0$ is $\bar{\tau}/t_0$, where $t_0 = 85.7$ a.u. $= 2.07\times10^{-15}$ sec. Actually, $\bar{\tau}$ is insensitive to the detailed distribution of vibrational states, and values of adequate accuracy can be obtained simply from the dashed curve by using $\bar{R}_0$ for the initial distance. Similar but more approximate calculations lead to the curve shown in Fig. 1 for H_3^+ as $\bar{Z}_{\rm eff}^2 = Z_c^2(\tau)/3$.

The representative response frequency, $\omega_{\rm core}$, of the electron cores of target atoms of atomic number Z_T is typically so high, of the order $Z_T\mathfrak{R}$, where $\hbar\mathfrak{R} = 13.6$ eV, that $\omega_{\rm core} > v/R_{0v}$. Then only the valence electrons contribute to the vicinage effect, and the ratio of the cluster stopping power S_c to the sum of the stopping powers S_i of its components becomes

$$S_c/\sum S_i \simeq 1 + (S_p^{\rm val}/S_p)[\xi^2(\tau) - 1], \qquad (7)$$

where $S_p^{\rm val}/S_p$ is the fraction of the proton stopping power of the target due to its valence electrons. In our low-velocity regime, where $S_p^{\rm val} \simeq S_p$, we measure $\xi^2(\tau)$ directly and find it in agreement with the theory, Eq. (6), as shown in Fig. 1.

One may, in conclusion, restate the theoretical results in an approximate but succinct manner by invoking the equipartition rule of stopping powers[10,11] according to which $S = S_{\rm close} + S_{\rm dist}$ is composed of contributions $S_{\rm close}$ from close collisions and $S_{\rm dist}$ from distant resonant collisions in approximately equal parts, $S_{\rm close} \simeq S_{\rm dist} \simeq S/2$. Any cluster of ions acts as a united charge, and $S_c = (\sum Z_i)^2 S_p$ when $R_{ij} < r_{\rm close} = \hbar/2mv$, a condition never found in molecules. When $r_{\rm close} < R_{ij} < r_{\rm dist} = v/\omega_0$, the ions act as independent particles with regard to close collisions and as a united charge with regard to distant collisions, and $S_c \simeq \frac{1}{2}[\sum Z_i^2 + (\sum Z_i)^2]S_p$. When $R_{ij} > r_{\rm dist}$, the ions act as independent particles in all collisions, and $S_c = \sum Z_i^2 S_p$. Our measurements of the effect of ion proximity in di-proton and tri-proton clusters follow the transition from the former to the latter condition as the cluster dwell time in the target increases. In this sense, the results present the first experimental confirmation of the equipartition rule of stopping powers.

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