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Theoretical Treatment of Ion Stopping in Complex Systems

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The sort of materials treated by theoretical methods in physics has changed radically in recent years, from small molecules and biomolecules, as well as perfect solids and gasses at zero temperature, to much more complex systems such as plasmas [1], warm dense matter [2] and complicated biomolecular structures [3]. For example, plasmas are almost always mostly ionized, depending on target temperature being much higher than ionization potentials, with electrons, atoms, and ions composing a quasi-neutral mixture, while biomolecules, generally neutral systems, can be large and complicated molecules. It is frequently desired to pump energy into these systems for reasons ranging from creating an operable fusion reactor to perform ion based radiation treatment of tumors. Such energy deposition is often done by focusing a fast ion beam on the target, where collisions, perhaps the result of complicated motions, depending on whatever $\vec{E}$ and $\vec{B}$ fields are present, result in conversion of some of the kinetic energy of the fast ion to kinetic and electronic energy of the components of the target system. Understanding of the details of the deposition of energy by a swift ion in materials is thus important to the understanding of many processes, from radiation treatment of tumors to interaction of molecules in space with the solar wind.

It is thus desirable that energy deposition by swift ions in complex modern systems be understood more deeply.

In all such cases, projectile kinetic energy is converted to target electronic energy on collision. The quantity describing such energy transfer is the energy deposited by the projectile per unit length of the trajectory, known as the stopping power, $-dE(v)/dx$, of the target, which depends on the projectile velocity, given in units of the Bohr velocity, v_0, and the scatterer target density, n. To facilitate comparison among different target systems, the stopping power is frequently normalized with respect to scatterer density, to produce what is referred to as the stopping cross-section of the target $S(v)$:

$$S(v) = -\frac{1}{n}\frac{dE(v)}{dx} \tag{1}$$

The stopping cross section with appropriate constants removed, is referred to in Bethe-like theories [4], as the stopping number, $L(v)$:

$$S(v) = \frac{4\pi e^4 Z_1^2 Z_2}{mv^2} L(v) \tag{2}$$

Here Z_1 and Z_2 are the projectile charge and target electronic charge, respectively. The stopping number is further expanded in powers of the projectile charge,

$$L(v) = \sum_{i=0} Z_1^i L_i \approx L_0 + Z_1 L_1 + Z_1^2 L_2 \tag{3}$$

yielding

$$S(v) = \frac{4\pi e^4 Z_1^2 Z_2}{mv^2} \sum_{i=0} Z_1^i L_i(v) \tag{4}$$

The stopping number, $L(v)$, is normally written using derived quantities which, using the Bethe [5], Lindhard [6,7], and Bloch [8] forms for L_0, L_1 and L_2, respectively, yields

$$L(v) = \ln\frac{2mv^2}{I_0} - C_1 v e^{-C_2 v} + Z_1 \cdot \frac{3\pi e^2 I_0}{2\hbar m v^3} \ln\frac{2mv^2}{I_0} - Z_1^2 \frac{1.202}{v^2} \tag{5}$$

for the stopping number. The dominant term here is the L_0, or Bethe, term $\left(\ln\frac{2mv^2}{I_0} - C_1 v e^{-C_2 v}\right)$, and calculations of the stopping power of a system are often approximations using only this term. If high accuracy is needed, this is not reasonable, as although the higher terms are small, they are not negligible.

Of the quantities in Equation 5, the critical quantity is the mean excitation energy of the target, I_0, which is defined [9] as the first energy weighted moment of the target dipole oscillator strength distribution (DODS):

$$\ln I_0 = \frac{\int \frac{df}{dE} \ln E \, dE}{\int \frac{df}{dE} dE} \tag{6}$$

The mean excitation energy describes how easily a target can absorb kinetic energy from the projectile, primarily as electronic (including ionization) and vibrational (including full and partial fragmentation) excitation.

The mean excitation energy is characteristic of the target only, and has no dependence on the properties of the projectile ion. (One should note parenthetically that if the target is in an excited electronic state before the collision, the projectile might absorb energy from the target [1].) As the mean excitation energy enters the stopping power expression through the logarithm and in atomic units, small changes in mean excitation energies do not produce major changes in the stropping cross section [10].

The question then arises as to how the mean excitation energies of the current complex systems now being studied should be calculated.

Perhaps the most æsthetically pleasing way is simply to calculate I_0 directly from Equation 6. Thus, the dipole oscillator strength distribution and excitation energies for the target must be calculated [11]. For atoms and small molecules, this is not a problem using presently available methods. However, different theoretical methods and basis sets used in such calculations can produce somewhat different results, as do estimation methods such as Bloch's [5] estimate:

$$I_0 = 10 Z_1 \; eV \tag{7}$$

of atomic mean excitation energies. If the target is a molecule, geometry, orientation with respect to the projectile beam and state of aggregation can be important.

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As an example, consider water. A variety of *ab initio* and semi-empirical calculations of I_0 give values differing by only some 6% around a mean of ~74 eV over the range of calculational schemes employed [12]. As noted above, since I_0 enters the calculation of stopping power only as the natural logarithm, the difference in computed stopping power will be very small regardless of which of the computed I_0´ is used.

The question then arises as how to determine the mean excitation energy for a particular complex target of the type mentioned above. There are several presently available possibilities.

In principle, one need calculate the DOSD for the complex mixture of the target, and then apply Equation 6 to obtain I_0 for the mixture. However, this is, as yet, not possible.

Another way the theoretical mean excitation energy for a complex, bulk system can be determined from a theoretical dielectric response of the bulk system [13]. In this approach, the mean excitation energy can be obtained from:

$$\ln I_0 = \frac{\int_0^\infty E \ln E \, \mathrm{Im}\left[-1/\varepsilon(E,0)\right] dE}{\int_0^\infty E \, \mathrm{Im}\left[-1/\varepsilon(E,0)\right] dE} \tag{8}$$

using, for example, a Drude-like dielectric function of the form

$$\varepsilon(E,q) = 1 + f_j(q) E_p^2 \left\{ E_j^2(q) - E^2 - iE\gamma_j(q) \right\}^{-1} \tag{9}$$

However, using such an approach requires determination of the dielectric function of the complex sample which determination is not easier than determination of the first energy weighted moment of the target DODS of the sample.

An approach frequently used to describe the mean excitation energy of a mixture of components in a target is related to the Bragg Rule [14], which calculates the stopping of a mixture as a density fraction weighted sum of the stopping of the components.

$$S(v) = \sum_{i=components} S_i(v) \tag{10}$$

In terms of the mean excitation energy, this gives a mean excitation energy of

$$\ln I_0 = \frac{1}{N_e} \sum_{i=components} \omega_i \ln I_0^i \tag{11}$$

Where ω_i is the number of electrons in component i, and N_e is the weighted number of electrons summed over the components of the target.

This formulation assumes that the components are not interacting, or that the interactions are small. Such an assumption is clearly not true for many complex targets such as plasmas.

Were one to use a Bragg-like scheme for the stopping power of a complex target, the mean excitation energy of neutral atoms and molecules, of free electrons, and of ions would need be known. Although the literature has many references to methods and values for calculating accurate mean excitation energies or stopping powers for atoms and free electrons [15,16], methods for calculation of accurate mean excitation energies for ions are nearly unknown.

Thus, the conclusion is that there appear to be two major projects that need be completed before energy absorption from swift ions in complicated target systems can be accurately described and understood:

1. New methods and programs need be developed which can deal with ion stopping in dense, strongly coupled systems.

2. To do so will require development of methods for accurate calculation of mean excitation energies of target ions.

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