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On the local density dependence of electronic stopping of ions in solids

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We use time-dependent density functional theory to calculate the electronic stopping S_e in binary Ni-Ni atomic collisions and for a Ni projectile along channeling directions in a Ni crystal. Our results show that when S_e is reported as a function of the ground state target electronic density ρ_0 the stopping is not a single-valued function of the local density, as assumed in formalisms that date back to the origins of quantum mechanics, but shows loops, suggesting that it is inaccurate to model stopping as a dissipative force of the type $\mathbf{F} = \beta(\rho_0)\mathbf{v}$, as it is customarily done in non-adiabatic molecular dynamics simulations of ion-solid interactions. We compare our results with S_e in a uniform electron gas where the above definition for the force holds, and conclude on the validity of using jellium as a crude approximation for more realistic inhomogeneous electron gases.

Keywords: Time dependent density functional theory, binary collisions, electronic stopping power, Nickel

I. INTRODUCTION

The use of energetic ions to study properties of matter dates back to the early days of modern physics; Bohr provided the first estimates of stopping of α particles in matter even before he gave the explanation of the spectrum of hydrogen atoms [1, 2]. In the hundred years past, ion-solid interactions has become an important branch of physics covering many areas of basic and applied sciences.

There is a diversity of models to describe multi-electron atoms interacting both with uniform electron gases (jellium) and atomic targets (gas or solid) at different levels of approximation, namely, ideal degenerate gas [3], a Fermi liquid [4], a Hartree-Fock type quantum gas [5], an electronic system treated with density-functional formalism [6–9], or interacting multi-electronic atoms in gas or solid phases [10–13]. One of the first models for binary atomic collisions was proposed by Firsov in 1959 [14], valid for collision velocities less than the representative electronic velocities in the target. Firsov assumed that the energy lost by the colliding atoms is the result of electrons being exchanged between them, with the consequent change in their average velocity. His model gives the instantaneous energy loss as an integral of the unperturbed electronic density to the power of 4/3 over the area of the dividing plane between the two colliding atoms. Clearly, this expression tells that the dissipation is largest when the density overlap is greatest, that is, at the closest distance. Using the Thomas-Fermi electronic distribution, he arrived at an analytic expression in terms of the charge of the colliding atoms, their velocities, and their impact parameter, predicting with remarkable accuracy a number of cases experimentally known at the time. To be noted here is that the electronic density used in Firsov's model, the Thomas-Fermi density, corresponds to atoms unperturbed by the collision.

The importance of understanding the electronic excitation mechanisms in atomic collisions becomes apparent in cases of theoretical and practical interest, to mention just a few examples, to calculate the range of ions in solids [15, 16], in astrophysics, in fusion energy technology, and in biomedical applications. The rate of energy transfer from a projectile to a target can be cast in the form of an electronic stopping power S_e and a nuclear stopping power S_n, which are traditionally considered the two mechanisms of energy dissipation for energetic ions colliding with a target material. For more accurate information, state-resolved cross sections may be needed, requiring much more sophisticated theoretical methods, which generally provide only partial answers. For a recent review of uncertainties in theoretical atomic and molecular data, see [17].

From the perspective of materials science, one of the most accurate predictive methods today for range and damage calculations is the use of classical molecular dynamics for the nuclear motion, with the addition of dissipative terms to account for the electronic losses in both, the electron-phonon interaction and the electronic stopping regimes; these are called non-adiabatic models. A recent example of this approach, based on the Firsov model, can be found in [18]. However, the vast majority of non-adiabatic molecular dynamics (MD) simulations use a single fixed value of the friction coefficient in the electronic stopping regime, with a cutoff at low velocities to

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