Nuclear Instruments and Methods in Physics Research B 317 (2013) 56–60

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Dynamic screening and energy loss of antiprotons colliding with excited Al clusters

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ARTICLE INFO

Article history:
Received 15 September 2012
Received in revised form 18 January 2013
Available online 13 February 2013

Keywords:
Energy loss
TDDFT
Metal cluster

ABSTRACT

We use time-dependent density functional theory to calculate the energy loss of an antiproton colliding with a small Al cluster previously excited. The velocity of the antiproton is such that non-linear effects in the electronic response of the Al cluster are relevant. We obtain that an antiproton penetrating an excited cluster transfers less energy to the cluster than an antiproton penetrating a ground state cluster. We quantify this difference and analyze it in terms of the cluster excitation spectrum.

1. Introduction

A charged particle moving across a metallic target is able to create electronic excitations in the medium at the expense of its own kinetic energy. Research on this phenomenon has been broad in condensed matter physics and materials science because of its relevance in various fundamental and applied topics, such as radiation damage, medical physics, and ion sputtering.

A key point in the theoretical analysis of the slowing down of charged particles in metals is the intensity of the perturbation that the moving particle introduces in the medium. For a particle of charge Q moving with a velocity v in a standard metal, the perturbation strength can be roughly characterized by the Sommerfeld parameter $\eta = Q/v$ [1]. If such ratio is small, $\eta \ll 1$, linear theory is naturally applied and accurate results for the particle energy loss are found. If $\eta \gg 1$ and v is much smaller than the typical velocities of the electrons in the medium, various non-perturbative methodologies have been successfully applied [2]. In between these two cases, in the regime of intermediate velocities, accurate descriptions of the energy loss process are much more involved because quasistatic or perturbative approximations break down even for unit-charge projectiles. Only recently calculations based on time-dependent density functional theory (TDDFT) [3–10] have shown its potential to close this gap.

The achievements of TDDFT in the non-linear description of electronic excitations pave the way to answer new and challenging questions in the field. In the traditional description of energy loss processes, the target is always considered as initially in its ground state. However, the energy lost by a travelling charge in a metallic medium should be affected by the electronic state in which the target is. Otherwise said, if electronic excitations have been already created in the system, the electronic response to the incident perturbation, and consequently the energy loss, will be different. In this work we try to quantify this difference for the particular case of a point charge crossing metallic clusters of a few Å size.

The experimental investigation of the excitation and ionization of neutral metal clusters by collision with positively or negatively charged particles has been intensive. In particular, the ionization of metal clusters by low energy singly and multiply charged ions and protons [11,12] and the ionization of neutral metal clusters by slow electrons [13–16] have been studied. Description of such processes from the theoretical point of view is incomplete and requires further investigation. In our work we study the collision of an Al cluster with a slow negative point charge (an antiproton). The choice of the antiproton as a projectile allows us to avoid complications related to the electron capture by the cluster if the projectile is an electron and the electron capture by the projectile if the latter is an ion. Our goal is to identify the distinct effects that arise in the dynamic screening and the projectile energy loss when the metallic target has been previously excited by a preceding projectile. In spite of the fact that our model is simplified, the results of our study can contribute to the understanding of the fundamentals of the dynamical processes during collision of charged particles with metallic clusters.

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http://dx.doi.org/10.1016/j.nimb.2013.01.053

We perform an explicit time propagation of the electronic state of the system using TDDFT and evaluate the energy lost by the charge when crossing ground-state clusters. We compare this quantity with the amount of energy lost when the projectile crosses a cluster excited from a previous collision. We show that the difference is appreciable and give the explanation of this change as a consequence of the excited state of the cluster as well as of the emission of electronic charge from the excited cluster.

Non-linear effects in the excitation of metal clusters have been previously analyzed with TDDFT. In particular, electron dynamics in clusters under intense laser fields are an active hot topic of research [17] because of the possibilities offered to explore and control ultrafast processes. The resonance energy of collective excitations in these systems has been shown to depend on the intensity of the perturbation [18]. Here we focus on a different type of external perturbation, namely, that derived from a point Coulomb charge crossing the system. We will show, nevertheless, that similar shifts in the position of the plasmon peaks are found.

Hartree atomic units ($e = \hbar = m_e = 1$) will be used throughout this work unless otherwise stated.

2. Methodology

Let us first define the system under study: We will focus on a negative point charge (an antiproton) crossing a metal cluster. Electron dynamics in metallic systems typically lie in the femtosecond and subfemtosecond time scales [19,20]. For this reason and for the kind of processes that we study, we consider the cluster ion cores as frozen. We further simplify the problem and use the spherical jellium model (JM) to represent the cluster. In the JM, the ions are substituted by an homogeneous background of positive charge with density $n_0^+(\mathbf{r}) = n_0(r_s)\Theta(R_{cl} - r)$. Here R_{cl} is the radius of the cluster, $\Theta(x)$ is the Heaviside step-function and $n_0(r_s)$ is the constant bulk density, which depends only on the Wigner–Seitz radius r_s ($1/n_0 = 4\pi r_s^3/3$) [21]. The number of electrons in a neutral cluster is $N = (R_{cl}/r_s)^3$.

The ground state electronic density of the cluster $n(\mathbf{r})$ is obtained using the Kohn–Sham (KS) scheme [22] of density functional theory (DFT) [23]. The ground state KS wave functions $\varphi_i(\mathbf{r})$ are expanded in the spherical harmonics basis set [24].

The evolution of the electronic density in the cluster in response to the field of the moving charge is calculated using TDDFT [25]. We propagate the ground state wave functions $\varphi_i(\mathbf{r}, 0) = \varphi_i(\mathbf{r})$ solving time-dependent KS equations:

$$\mathrm{i}\frac{\partial}{\partial t}\varphi_i(\mathbf{r}, t) = \left\{-\frac{1}{2}\nabla^2 + V_{\mathrm{eff}}(\mathbf{r}, t)\right\}\varphi_i(\mathbf{r}, t). \quad (1)$$

The effective potential includes four terms $V_{\mathrm{eff}}(\mathbf{r}, t) = V_{\mathrm{ext}}(\mathbf{r}, t) + V_H(\mathbf{r}, t) + V_{\mathrm{xc}}(\mathbf{r}, t) + V_p(\mathbf{r}, t)$, where V_{ext} is the external potential created by the positive background. V_H is the Hartree potential created by the electronic density. V_{xc} is the exchange–correlation potential, calculated with the standard adiabatic local density approximation (ALDA) with the Perdew–Zunger parametrization of Ceperley–Alder exchange and correlation potential [26]. Finally, $V_p(\mathbf{r}, t) = -\frac{Q_p}{\sqrt{(z_p(t)-z)^2+\rho^2}}\Theta(t)$ is the potential created by the antiproton and acting on the valence electrons of the cluster. We use cylindrical coordinates (ρ, z) in the time-dependent calculations, which are more appropriate since the problem has axial symmetry. The origin of coordinates is located at the center of the cluster. The antiproton is represented by a negative point charge ($Q_p = -1$) which moves with constant velocity υ along the z-axis. At time $t = 0$, the antiproton is located at a distance from the cluster (50 a.u.) far enough to avoid a significant interaction between the projectile and the target. The time propagation of the electron wave function is performed using the time-stepping algorithm: $\varphi_i(\rho, z, t + dt) = e^{-iH_i dt}\varphi_i(\rho, z, t)$. The split operator approximation is then used to separate the potential and kinetic energy terms in the $e^{-iH_i dt}$ time propagator. The action of the kinetic energy operator is calculated using Crank–Nicolson propagation scheme. A detailed description of the numerical procedure can be found in Refs. [27–29].

From the time-dependent KS orbitals we obtain the time-evolving electronic density of the excited cluster $n(\rho, z, t) = \sum_{i\in occ}|\varphi_i(\rho, z, t)|^2$. The force acting on the moving antiproton along the z-axis is obtained from the time-dependent electronic density and includes the effect of the positive background:

$$F_z(t) = 2\pi\int \rho d\rho dz \frac{n(\rho, z, t) - n_0^+(\rho, z)}{\left[(z_{\bar{p}}(t) - z)^2 + \rho^2\right]^{3/2}}[z_{\bar{p}}(t) - z]. \quad (2)$$

The energy loss is then obtained from the integral:

$$E_{\mathrm{loss}} = -\upsilon\int_0^\infty F_z(t)dt. \quad (3)$$

We will study the energy loss in two different motion cycles. In the first cycle, the antiproton moves towards the cluster with a constant velocity υ, crosses it following a symmetry axis through the cluster center, and eventually moves away until it reaches a turning point arbitrarily defined. The turning point is at a distance from the cluster far enough not to have any residual interaction. The cluster is then left in an excited state. The electronic energy transferred to the cluster during the collision is calculated. In the second cycle, the antiproton turns back from the turning point and starts to approach the excited cluster with the same constant velocity υ. In the second crossing of the cluster, the latter now in an excited state, energy is again transferred to the cluster. We calculate the energy lost in this second cycle and compare the obtained value with that of the first cycle.

3. Results and discussion

We have chosen a small Al ($r_s = 2.07$) cluster with $N = 18$ electrons and with radius $R_{\mathrm{cl}} = 5.43$ a.u. (≈0.29 nm). In all the calculations shown in this article, the projectile velocity is $\upsilon = 0.5$ a.u. The ALDA–TDDFT method used here predicts very well the energy loss of antiprotons in Al targets. The method gives very good agreement with measurements in Al bulk for antiproton velocities up to 1.8 a.u. Above this velocity the excitations of the inner shells in Al start to contribute to the energy loss and results deviate from the experimental ones [4].

The antiproton starts its motion at time $t = 0$ from the position $z_0 = -50$ a.u. After the first collision, the projectile continues to move until $t = 1000$ a.u. At this time the second cycle starts and the antiproton takes the way back to collide again with the cluster. We call τ to the time interval between both collisions. In both cycles we calculate the force $F_z(t)$ experienced by the projectile due to the interaction with the cluster through Eq. 2. From $F_z(t)$ we obtain the value of the energy lost by the antiproton E_{loss} by means of Eq. 3. In addition, we consider three other different time spots for the second cycle to start: 1003.5, 1005, and 1010 a.u. The purpose of using different time delays is to check the sensitivity of the final result to the dynamics followed by the electron density in the cluster excited state. With our choice of time delays, the antiproton reaches the excited cluster respectively $\Delta\tau = 7$, 10 and 20 a.u. of time later than in the reference calculation. Depending on the value of $\Delta\tau$, the antiproton will start to cross the surface of the excited cluster meeting a minimum or a maximum in the electronic density oscillations, or an intermediate state. The density oscillations will be discussed later. The results for the energy loss are summarized in Table 1.