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Impact-parameter dependence of resonant-coherent excitation of channeled ions

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Abstract

The resonant-coherent excitation of channeled hydrogenic ions is theoretically investigated. The crystal potential averaged over the ion trajectory, together with the induced wake potential, produces mixing of its electronic states and shifting of its energy levels. This effect introduces a dependence of the resonance on the position of the ion. The periodic components of the crystal potential are responsible for the excitation. Schrödinger's equation is numerically solved to illustrate the way in which the excitation takes place. An electron promoted to an excited state is shown to evolve non-adiabatically, retaining most of its initial character while the ion travels across long distances in the channel.

The interaction of fast ions with the periodic lattice of an oriented crystal is known to produce resonant-coherent excitation (RCE) in electrons bound to the ions when one of the frequencies of the interaction potential matches the energy difference between electron states. This effect was first predicted by Okorokov in 1965 [1]. The experimental corroboration waited until thirteen years later, when Datz et al. [2–4] observed the reduction in the transmission of fixed-charge-state ions of atomic numbers ranging $Z_1 = 5$ –9, channeled through Au and Ag crystals. The RCE has been observed under both axial [2–4] and planar [5] channeling conditions.

Another way of observing the RCE consists in using projectiles of large atomic number traveling at high velocities, which can leave very thin crystals after suffering excitation without being ionized. These conditions permit to investigate the eventual emission of X-rays, whose yield has been shown to be larger near resonance conditions [6–8].

The interaction of the ion with the crystal target can be understood as being originated in two distinct sources of potential: the static crystal potential and the induced potential.

The crystal potential consists of the interaction with all electrons and nuclei in the unperturbed medium. The spa-

tial periodicity of the lattice in which those are placed is experienced by the moving ion as a periodicity in time, which enables the electron bound to it to suffer transitions of frequency corresponding to the different harmonics contributing to the potential. Experimental evidence exists that some frequencies can be suppressed due to interference effects [9].

The distortion produced in the medium by the projectile results in the emergence of an induced potential. For an infinite medium, the well-known wake potential is the part of the induced potential related to the perturbation of valence band electrons [10–12]. For an ion moving near a surface, this is the surface-wake potential [13]. The wake potential represents a net electric field responsible for the valence band contribution to the stopping of the probe. Moreover, that electric field produces mixing and splitting of excited electronic states of the projectile [14] (similar to the Stark effect).

In addition to the induced electric field, the crystal potential, when averaged over the trajectory of the channeled ion, yields a non-zero electric field, which also contributes to the Stark effect just mentioned [14].

The RCE can take place in such a way that the excited state lies in the continuum, leading to electron loss [15–17]. This process has two important consequences: firstly, the bound states can be de-populated via this effect; and secondly, it introduces a width to the electronic Stark-energy-split levels of the ion. It has been speculated that, for the case of surface channeling, the electron emitted in this way should travel with well-defined energy around

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preferential directions [17,18]. Experimental evidence of this type of electron emission is still lacking.

The RCE, particularly when the excited state lies in the continuum, can be regarded as an elastic process in the laboratory frame, similar to that taking place in low-energy electron diffraction (LEED), in which the lattice acts like a source of momentum.

This picture needs to be completed by considering the excitation of target electrons: the simultaneous promotion of the moving bound electron and a target electron can occur by absorbing energy out of the fast ion motion. These kinds of processes contribute to both excitation and ionization of the ion (and to electron capture as well to a minor extent). In this way, the collision of the bound electron with a target electron may constitute a relevant mechanism in the ionization of the projectile, as shown by Datz et al. [7]. It is interesting to point out that in regions where the target electron density is large enough, the dynamical screening of the electron–electron interaction can play a crucial role. Actually, these processes are well known in the field of charge states of low-energy ions traversing solids: they are the so-called Auger excitation and loss [19,15,16].

When an ion moves inside a crystal or close to its surface under channeling or surface channeling conditions, respectively, it follows a nearly classical trajectory, which is governed by the crystal potential (together with the image potential in the case of surfaces [18]), provided the ion energy is large enough to ensure a relatively negligible stopping. Moreover, since the trajectory forms a glancing angle with respect to the channel, the motion perpendicular to the planes, axis, or surface, can be decoupled from the fast motion along the remaining ‘‘parallel’’ (to the channel) direction, leading to the concept of the so-called planar potential, axial potential, or surface potential, respectively. These are nothing but the average of the crystal potential over trajectories parallel to the plane, axis, or surface, with fixed impact parameter [20–22].

The adiabatic excited states, to which the transition from the ground state can occur, change along the trajectory, because the average crystal electric field discussed above also changes. Besides, the electron excited bound states evolve non-adiabatically in that changing electric field. This effect will be discussed below.

To summarize the above remarks, the whole RCE picture has to include the following ingredients: (i) transitions connecting electronic bound states of the ion via the frequencies of the crystal potential that lie near the energy difference between those states; (ii) splitting and mixing of excited states, coming from the net electric field derived from both the induced wake and the averaged (over the ion trajectory) crystal potential; (iii) electron loss probability, part of which can be obtained as in point (i) by considering that the bound electron state resonates, via harmonics of the crystal potential, with the continuum; and (iv) the ion trajectory and the dynamical evolution of different bound

states. A substantial non-coherent contribution to points (i) and (iii) is expected to come from Auger excitation (de-excitation) and loss, respectively.

In what follows, we will focus on a hydrogenic ion of nuclear charge Z_1 moving near or inside a crystal under surface or planar channeling conditions, respectively. The evolution of the originally bound electron state $|\Phi\rangle$ will be described by the one-electron Schrödinger equation

$$(H_0 + V)|\Phi\rangle = i\partial_t|\Phi\rangle, \quad (1)$$

where H_0 is the electronic Hamiltonian of the free projectile and V represents the interaction with the solid, that is, the crystal and induced potentials. The electron wave function will be expressed in terms of stationary solutions of H_0 according to

$$|\Phi\rangle = e^{-i\epsilon_0 t} a_0 |0\rangle + \sum_{\alpha > 0} e^{-i\epsilon_\alpha t} a_\alpha |\alpha\rangle + \sum_k e^{-i\epsilon_k t} a_k |k\rangle, \quad (2)$$

where $|0\rangle$ is the electron ground state (i.e., $|1s\rangle$), $|\alpha\rangle$ denotes an excited bound state, and $|k\rangle$ represents a free state of momentum k . We will solve Eq. (1) in the projectile rest frame.

As mentioned above, the potential V can be split as

$$V = V_C + V_1,$$

where V_C is the crystal potential and V_1 denotes the induced potential. The latter will be approximated by the wake potential created by the projectile and acting on the bound electron, i.e.,

$$V_1 = -(Z_1 - 1/2)\phi_{\text{wake}}(\mathbf{r}),$$

where $\phi_{\text{wake}}(\mathbf{r})$ represents the wake induced by a unit charge at the position \mathbf{r} relative to the nucleus of the ion, Z_1 is the atomic number of the ion, and the $1/2$ term stands for the electron self-interaction. In the case of surface channeling, V_1 may have a smooth dependence on the time, introduced through the surface–atom separation.

The periodic crystal potential experienced by the bound electron can be written

$$V_C(\mathbf{r}, t) = \sum_G V_{G,z+z_0} e^{iG(\mathbf{R}_0 + \mathbf{R} + \mathbf{v}t)},$$

where $\mathbf{R}_0 = (0, y_0)$, \mathbf{v} is the velocity of the ion, which will be taken parallel to the x -axis, z_0 stands for its initial impact parameter with respect to the surface/atomic plane under consideration, the z direction is taken normal to the surface/plane, and the sum is extended over reciprocal lattice vectors. Upper-case vectors will be reserved for components parallel to the surface/plane from now on. In particular, $\mathbf{r} = (\mathbf{R}, z)$.

For simplicity, the excited states will be restricted to those of the L shell, so that we shall work in the reduced basis of five atomic states ($1s, 2s, 2p$). This allows us to replace the energy ϵ_α by $\epsilon_1 = -\frac{1}{8}Z_1^2$ for $\alpha > 0$.

The transitions to the continuum and to higher excited states introduce a width to the energy of the reduced basis [14]. This permits to write, from Eqs. (1) and (2),

$$\dot{a}_\alpha = -i \sum_\beta \left(V_{\alpha\beta} - \frac{i}{2} \Gamma_{\alpha\beta} \right) a_\beta - i \tilde{V}_{\alpha 0} e^{i(\epsilon_1 - \epsilon_0 - G_0 v)t} a_0, \quad (3a)$$

$$\dot{a}_0 = -i \sum_\beta \tilde{V}_{0\beta} e^{-i(\epsilon_1 - \epsilon_0 - G_0 v)t} a_\beta - i \left(V_{00} - \frac{i}{2} \Gamma_{00} \right) a_0, \quad (3b)$$

where a dot represents the time derivative. The matrix elements

$$V_{\alpha\beta} = \langle \alpha | V_1 | \beta \rangle + \sum_{Gv=0} e^{iGR_0} \langle \alpha | V_{G, z+z_0} e^{iGR} | \beta \rangle, \quad (4)$$

incorporate the mixing of excited states through the induced and crystal potentials. The average over a large time interval Δt such that $Gv\Delta t$ is either zero or $\gg 1$, but small in terms of the variation of the projectile impact parameter along the trajectory z_0 , has been performed to

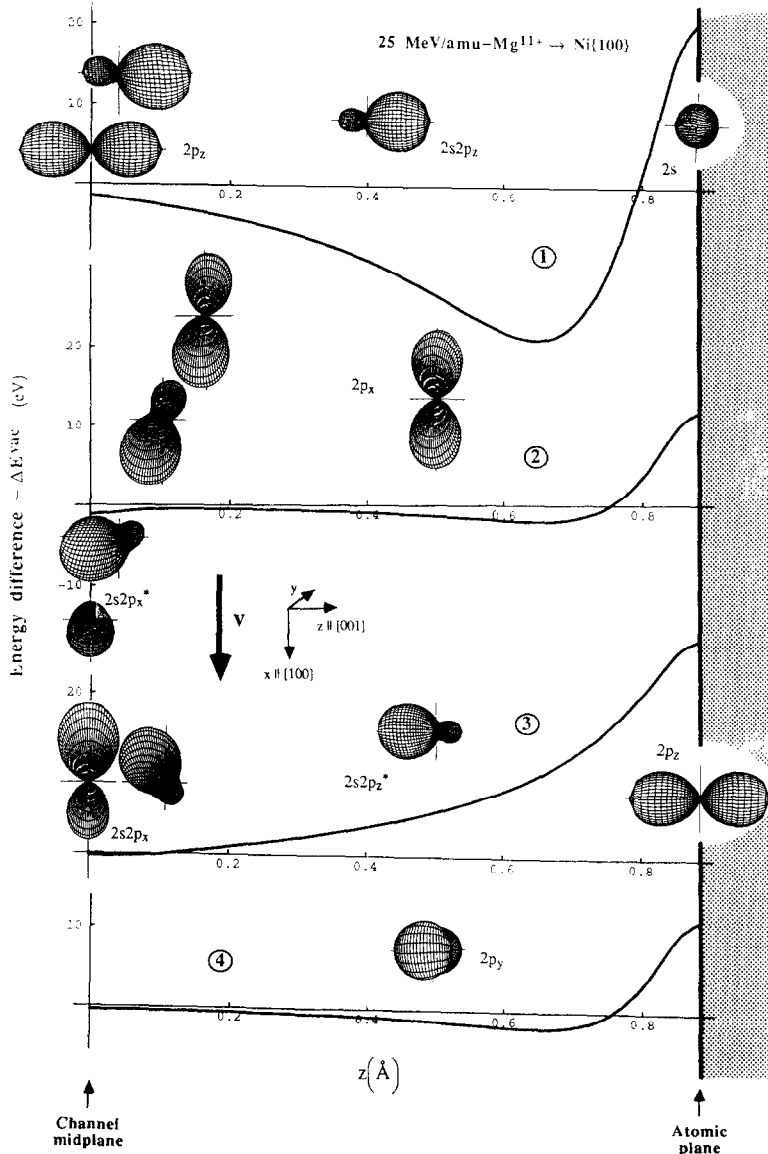


Fig. 1. Transition energies between the $|1s\rangle$ state and the hybrid states of the L shell of a 25 MeV/amu Mg^{11+} ion channeled in the (001) planar channel of a Ni crystal as a function of distance from the channel mid-plane. The energy differences are given relative to the vacuum value $3Z_1^2/8$. The shape of the electronic cloud in the different excited states is shown for various positions in the channel by means of the accompanying insets. They represent the radial integral of the squared electron wave function. The ion velocity runs from top to bottom.

obtain Eq. (4); these two conditions can be simultaneously accomplished for ion trajectories that are grazing with respect to the channel. The RCE is accounted for by matrix elements connecting the state $|1s\rangle$ with those of the L shell:

$$\tilde{V}_{\alpha 0} = \langle \alpha | V_{-G_0, z+z_0} e^{-iG_0 R} | 0 \rangle$$

and $\tilde{V}_{0\alpha} = \tilde{V}_{\alpha 0}^*$, where G_0 is such that $|\epsilon_1 - \epsilon_0 - G_0|$ is small. Notice that a factor $e^{-iG_0 R_0}$ has been disregarded, since it only contributes like an overall phase multiplying a_α in Eq. (3a). Finally, $\Gamma_{\alpha\beta}$ represent the effect of the transitions to the continuum. These non-Hermitian matrix elements include Auger transitions and RCE to the continuum, or resonant-coherent loss (RCL). The former deserves a more detailed analysis beyond the scope of the present work. The theory of these transitions is formally equivalent to the existing development for low-energy ions in solids [16]. The RCL contribution for excited states can be shown to be

$$\begin{aligned} \Gamma_{\alpha\beta}^{\text{RCL}} &= 2\pi \sum_k \sum_G \langle \alpha | V_{G, z+z_0} e^{iGR} | k \rangle \\ &\times \langle k | V_{-G, z+z_0} e^{-iGR} | \beta \rangle \delta(\epsilon_k - \epsilon_1 - Gv), \\ &\alpha, \beta > 0, \end{aligned} \quad (5a)$$

$$\begin{aligned} \Gamma_{00}^{\text{RCL}} &= 2\pi \sum_k \sum_G \langle 0 | V_{G, z+z_0} \rangle e^{iGR} | k \rangle \\ &\times \langle k | V_{-G, z+z_0} e^{-iGR} | 0 \rangle \delta(\epsilon_k - \epsilon_0 - Gv). \end{aligned} \quad (5b)$$

It is important to realize that only some vectors G contribute significantly to Eqs. (5). The reason for this is that the form factors of the sort $\langle \alpha | V_{G, z+z_0} e^{iGR} | k \rangle$ take very small values unless $(k_x, k_y) \approx G$, and this condition is consistent with the conservation of energy, expressed through the δ function, only for certain vectors G .

An adiabatic basis of states can be defined such that it diagonalizes the matrix given in Eq. (4) for each impact parameter. This will remove (completely almost anywhere) the degeneracy of the L shell. Fig. 1 shows the difference between ground and excited states energy for 25 MeV/amu Mg^{11+} ions channeled in the (001) planar channel of a Ni crystal. The wake potential has been calculated using the random-phase approximation dielectric function [23]. The crystal potential was derived from the ZBL inter-atomic potential [24,25]. The shape of the electronic cloud in each state is indicated in the insets. Notice that state 1 is oriented towards the channel wall, and consequently, it can be more easily populated by RCE [8].

The electron transition energy of resonance ΔE_{res} depends on the orientation of the trajectory through $G_0 v$ (see Eq. (3a)). For an ion moving parallel to a [001] surface or planar channel of an fcc crystal, like that considered in Fig. 1, one has

$$\frac{\Delta E_{\text{res}} a}{2\pi v \gamma} = k \cos \varphi + l \sin \varphi, \quad (6)$$

where $\gamma = 1/\sqrt{1-c^2/v^2}$ accounts for the relativistic Lorentz contraction of the lattice constant a , φ is the angle

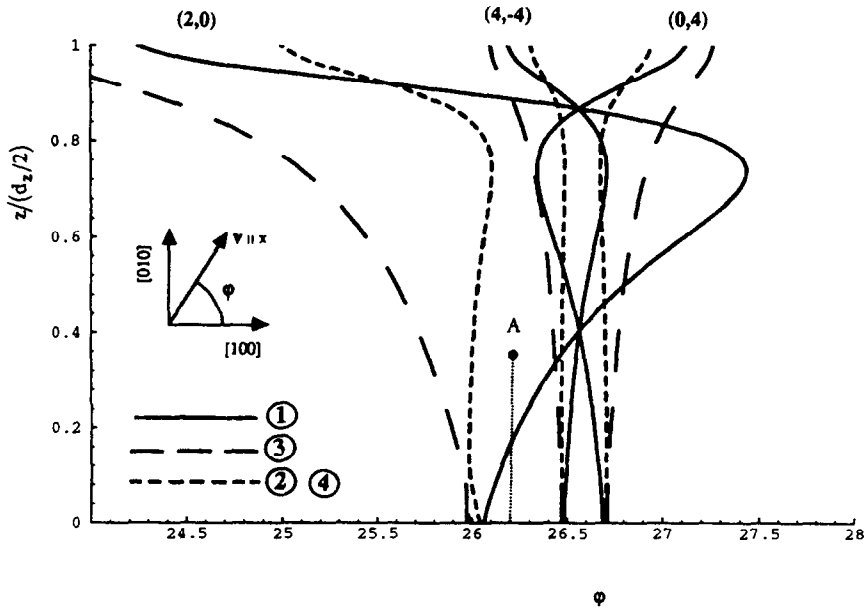


Fig. 2. Distance from the channel mid-plane at which the resonance condition is fulfilled for the (2, 0), (4, -4), and (0, 4) harmonics (see Eq. (6)) as a function of the angle φ between the ion velocity vector and the [100] direction, under the same conditions as in Fig. 1. d_z represents the channel width.

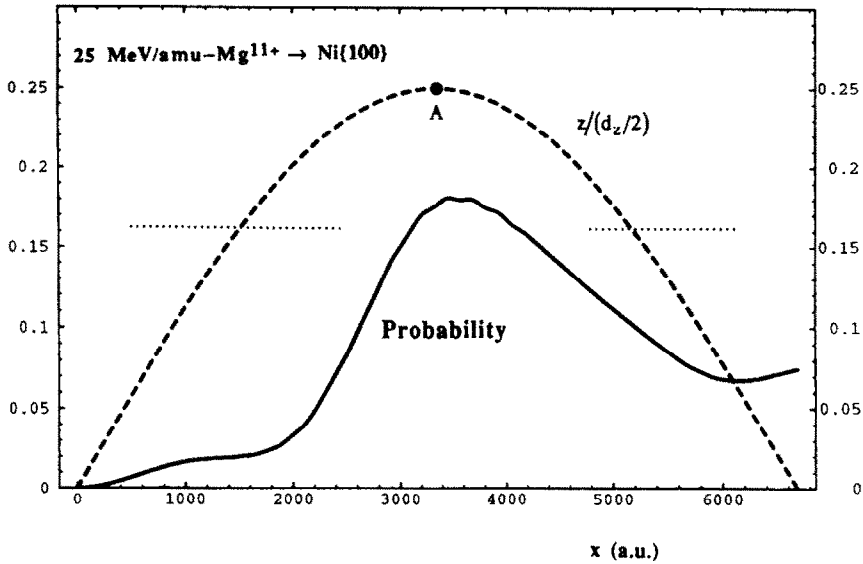


Fig. 3. Under the same conditions as in Fig. 1, the probability of occupancy of the L shell (solid curve) is shown along the trajectory (dashed curve). The latter forms an angle of $\varphi = 26.2^\circ$ with respect to the [100] direction. The projectile is prepared in the $|1s\rangle$ state at $x = 0$. The trajectory lies along the dotted vertical line in Fig. 2. The dotted horizontal bars indicate the impact parameter of the resonance.

between the ion velocity and the [100] direction, and (k, l) are integers such that $k + l$ is an even number. Combining Eq. (6) with the data displayed in Fig. 1 one obtains a dependence of the impact parameter at which the resonance condition (6) is fulfilled on the angle φ . This is shown in Fig. 2 for various harmonics (k, l) . Notice that by means of tuning the angle φ one can select the excita-

tion to a given state via a given harmonic. That is the case for trajectories corresponding to the dotted vertical line shown in the figure, which cross a $(2, 0)$ resonance with state 1 (see Fig. 1). We have analyzed those trajectories in Figs. 3 and 4.

Schrödinger's Eq. (3a) has been solved numerically for a trajectory corresponding to the dashed line shown in Fig.

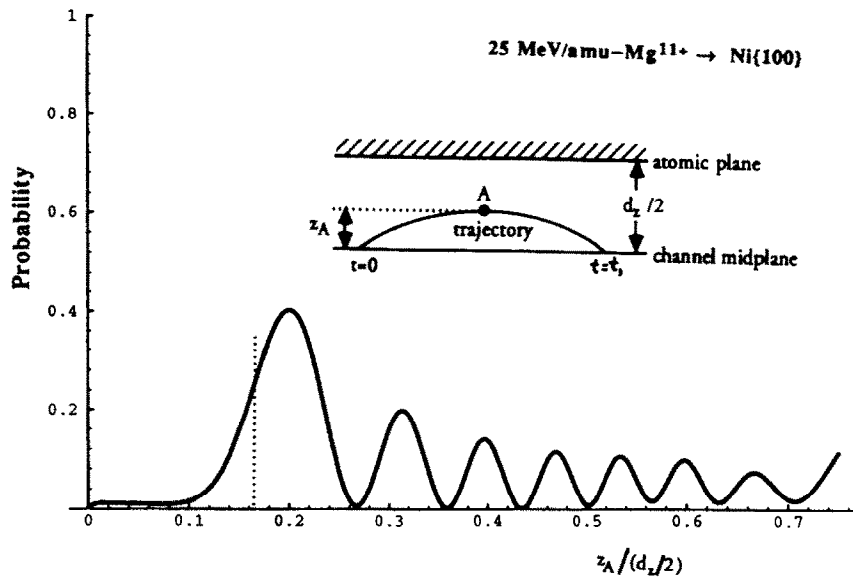


Fig. 4. Probability that an electron prepared in the ground state at time $t = 0$ is in the L shell at time $t = t_1$ due to RCE for different impact parameters z_A , under the same conditions as in Fig. 1. The ion trajectory is schematically shown in the inset. The angle with respect to the [100] direction has been taken $\varphi = 26.2^\circ$. The trajectory runs across the dotted line shown in Fig. 2. A dotted line shows here the distance at which the state labeled 1 in Fig. 1 is at resonance with the $(2, 0)$ harmonic (see Eq. (6)). d_z represents the channel width.

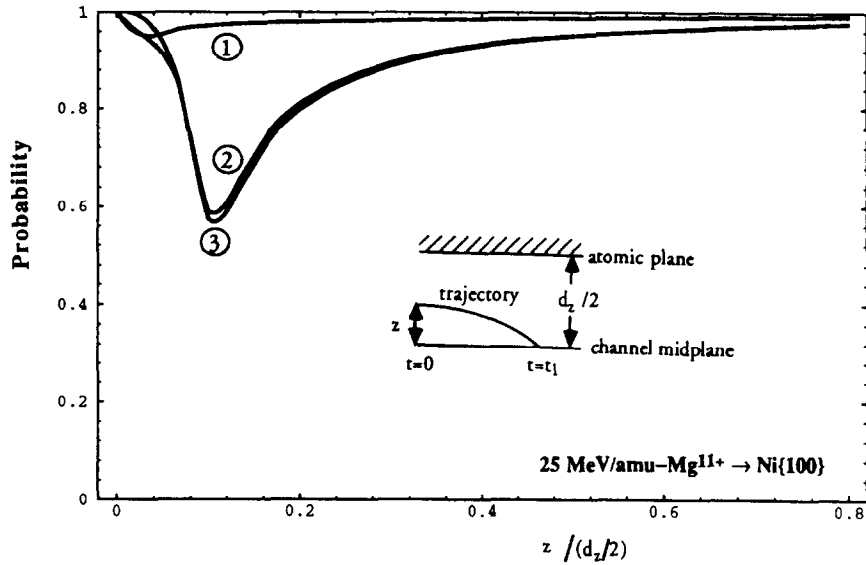


Fig. 5. Probability that an electron prepared in the state i (numbers in circles in the plot) at time $t = 0$ stays in the same state at time $t = t_1$, as a function of the initial impact parameter z . The velocity normal to the channel is zero at $t = 0$.

3. The Auger and RCL processes have been neglected. The ion trajectory was in turn calculated using the ZBL inter-atomic potential. The electron was placed in the ground state at $x = 0$. The solid line shows the probability of finding the electron in the excited states as a function of the distance across the direction parallel to the channel, x . The impact parameter of resonance, obtained from Fig. 2 has been shown by dotted horizontal bars. Notice that the RCE occurs after the ion has crossed the resonance. In the way back to the channel mid-plane, the resonant-coherent interaction acts in the sense of reducing the population of excited states (resonant-coherent de-excitation).

This reduction can be complete for certain impact parameters z_A , as shown in Fig. 4, where the occupancy of the L shell at the end of the trajectory shown in the inset is shown for an ion initially prepared in the ground state. Each point of the curve corresponds to a different trajectory. The oscillations observed in the figure are familiar in the study of potential-curve crossing of low-energy ions [26].

In order to test the adiabaticity of the evolution of an excited electron, we have solved Schrödinger's equation for the trajectory shown in the inset of Fig. 5. Each of the curves shown in the figure corresponds to an initial excited state at time $t = 0$. The figure shows the probability that the electron remains in the same excited state at time $t = t_1$. The projection of the wave function at that time over the adiabatically evolved initial state would result in a considerable loss of probability. Thus, the excited states evolve non-adiabatically and they retain their initial character to a large extent.

In summary, the RCE of channeled ions depends on the impact parameter with respect to the atomic plane under

consideration. Moreover, the evolution of an excited electron is non-adiabatic in the sense that it retains most of its initial character after excitation. A further effort has to be made in order to incorporate the effect of Auger and RCL processes. Work in this direction is in progress.

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