

Coherent resonant excitation of fast Ne^{9+} ions in a crystal: a computer experiment

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A method has been developed for simulating the internal dynamics of hydrogen-like ions as they pass through crystals. Quantitative agreement has been reached with the results of recent experiments on the coherent resonant excitation of Ne^{9+} ions in a gold crystal.

Coherent resonant excitation of fast hydrogen-like ions in a crystal (the Okorokov effect¹) arises when a harmonic of the periodic perturbing field of the crystal coincides (in the c.m. frame of the ion) with the frequency of a transition between two levels of the ion:

$$\omega_n = \frac{2\pi\gamma\beta cn}{a} = \omega_{ik}. \quad (1)$$

In the derivation of (1) it is assumed that the ion is moving along a crystallographic axis; γ and βc are the Lorentz factor and velocity of the ion; and a is the distance between atoms of the axis. Although coherent resonant excitation has been under experimental study for a fairly long time now (see the review²), we do not yet have a definitive quantitative theory. The basic difficulty is in dealing correctly with the simultaneous effects of a periodic perturbation with the set of frequencies in (1), the continuous potential of the axes, and the wakefield potential,³ which lead to a splitting and mixing of the degenerate electronic levels of the ion in the crystal; inelastic factors which lead to a loss of the electron; and the changes in the amplitudes and frequencies of the perturbation along the complex trajectory of the ion in the crystal (the slowing of the ion due to the ionizational energy loss is to be taken into account). These difficulties were pointed out in Refs. 4 and 5, where attempts were undertaken to find a qualitative interpretation of experimental data on the basis of a two-level model (or a model with more than two levels⁵) in the resonance approximation, with rectilinear ion trajectories in the crystals.

We have now developed a new approach to this problem, which is based on a numerical (computer) simulation of ion trajectories in a crystal and a calculation on the quantum dynamics of an ion on the trajectory which is found. Here are the basic principles of the method.

1. The crystal potential is written in the form

$$V(\mathbf{R}) = U(\mathbf{R}_\perp) + W(\mathbf{R}_\perp, z); \quad (2)$$

$$U(\mathbf{R}_\perp) = \sum_{\mathbf{k}_\perp, k_\parallel=0} V_{\mathbf{k}} e^{i\mathbf{k}_\perp \cdot \mathbf{R}_\perp} S(\mathbf{k}_\perp, k_\parallel = 0); \quad (3)$$

$$W(\mathbf{R}_\perp, z) = \sum_{\mathbf{k}_\perp, k_\parallel \neq 0} V_{\mathbf{k}} e^{i\mathbf{k}_\perp \mathbf{R}_\perp} S(\mathbf{k}_\perp, k_\parallel) e^{i\mathbf{k}_\parallel z}; \quad (4)$$

$$\mathbf{k} = \{\mathbf{k}_\perp, k_\parallel\} = \left\{ \frac{2\pi l}{a_x}, \frac{2\pi m}{a_y}, \frac{2\pi n}{a_z} \right\}.$$

Here $V_{\mathbf{k}}$ is a Fourier component of the crystal potential, $S(\mathbf{k}_\perp, k_\parallel)$ is a structure factor, and the Z axis runs along some selected crystallographic axes. If the ion enters the crystal at a small angle from the crystallographic axes, its trajectory in the crystal is determined primarily by potential (3), which is the continuous potential of the axes. The trajectory is found in this case by the standard multistring model, in which the multiple scattering and energy loss are taken into account.

2. For the ion trajectory $\mathbf{R}(t) = \mathbf{R}_\perp(t) + \beta_\parallel ct$ which is found (\mathbf{R}_\perp is the displacement transverse with respect to the axes), the Schrödinger equation (or the von Neumann equation for the density matrix), which describes the internal dynamics of the hydrogen-like ion, is solved numerically. In the c.m. frame of the ion, scalar and vector potentials arise. These potentials depend on the transverse coordinates and on the time t' :

$$V'(\mathbf{R}') = \gamma \left\{ U(\mathbf{R}'_\perp) + \sum_{\mathbf{n} \neq 0} U_{\mathbf{n}}(\mathbf{R}_\perp) \exp \left[i \frac{2\pi n \gamma z'}{a} + i \omega_{\mathbf{n}} t' \right] \right\}; \quad (5)$$

$$\mathbf{A}'(\mathbf{R}') = \vec{\beta}_\parallel \mathbf{V}'(\mathbf{R}'), \quad U_{\mathbf{n}}(\mathbf{R}'_\perp) = \sum_{\mathbf{k}_\perp} V_{\mathbf{k}} e^{i\mathbf{k}_\perp \mathbf{R}'_\perp} S(\mathbf{k}). \quad (6)$$

For nonrelativistic ions ($\vec{\beta} \ll 1$), the effect of the magnetic field is negligible, but we add the wakefield (polarization) potential $\vec{\phi}_{\text{wake}}(\mathbf{R}')$, whose form depends on the velocity.³ If the electron coordinates \mathbf{r} are reckoned from \mathbf{R}' , then the perturbation acting on the electron of the ion is found from (5), in which we replace \mathbf{R}' by $(\mathbf{R}' + \mathbf{r})$. We seek a solution of the Schrödinger equation with a perturbation of this type in the form

$$\Psi(t) = \sum_m C_m(t) \Psi_m(\mathbf{r}) \exp(i\epsilon_m t / \hbar),$$

where $\Psi_m(t)$ and ϵ_m are the wave functions and energies of the unperturbed states of the ion before it enters the crystal. Substitution into the time-dependent Schrödinger equation results in a system of coupled equations for $C_m(t)$ with the initial condition $C_m(0) = \delta_{m0}$. The corresponding transition matrix elements are calculated by expanding the potential $\Phi = \phi_{\text{wake}}(\mathbf{R}' + \mathbf{r}) + V'(\mathbf{R}' + \mathbf{r})$ in powers of \vec{v} and by retaining terms of second order (i.e., dipole and quadrupole transitions are taken into account). We thus use the resonance approximation for the case in which one of the harmonics of perturbation (5), $\hbar\omega_n$, is close to one of the transition energies, i.e., for the case of coherent resonant excitation proper. For the $1S \rightarrow 2S$, $2P$ transition, it is sufficient to retain the $1S$ ground level and four degenerate excited levels ($2S$, $2P_0$, $2P_{\pm 1}$) in the system of equations for $C_m(t)$, for ions with relatively small Z ($\alpha Z \ll 1$). As a result, we find a system

$$i\hbar\dot{C}_0 = \sum_{l=1}^4 C_l M_{0l}^n \exp\{i(\omega_{l0} - \omega_n)t\} + M_{00}^0 C_0 - \frac{i\hbar\Gamma_0}{2} C_0; \quad (8)$$

$$i\hbar\dot{C}_i = \sum_{l=1}^4 M_{il}^0 C_l + M_{i0}^n \exp\{-i(\omega_{i0} - \omega_n)t\} C_0 - \frac{i\hbar\Gamma_i}{2} C_i, \quad \omega_{ml} = (\epsilon_m - \epsilon_l)/\hbar, \quad (9)$$

to which we have added the decay constants Γ_i . These constants describe the loss of the electron by the ion as it moves through the crystal and a possible radiative decay of the excited levels. In particular, for the case of Ne^{9+} ions, radiative decay over a distance of 2000 Å in a crystal (as in the experiments of Ref. 6) can be ignored, and Γ_i can be calculated from the cross sections for electron-impact ionization of the ions (in the c.m. frame of the ion). The local electron density in the channel and the cross sections for photoionization by nonresonant harmonics of the potential are to be taken into account. In Eqs. (8) and (9), the transition matrix elements are

$$M_{ik}^n = \langle I \frac{\partial \Phi_n}{\partial \mathbf{r}_\perp} \mathbf{r}_\perp + \sum_{j,l} \frac{\partial^2 \Phi_n}{\partial x_{j\perp} \partial x_{l\perp}} x_{j\perp} x_{l\perp} | K \rangle,$$

where Φ_n means that only the n th harmonic from the Fourier expansion for Φ is included in the matrix element.

3. A fundamental distinction between the scheme for integrating system (7) and the approach of using rectilinear-trajectory approximations^{3,5} is that there are changes in M_{ik}^n and Γ_i upon the transition to the next integration step (the next fragment of the trajectory) on which the motion is assumed to be rectilinear. On each occasion, one calculates the matrix elements M_{ik}^n and M_{i0}^n , which depend on the position of the ion in the crystal channel and which determine the new set of eigenfunctions and new energy eigenvalues of the electron in the ion (a Stark effect which depends on the coordinates of the ion). In addition, one calculates the new value of the perturbation frequency ω_n , which changes because of the decrease in the ion energy due to ionization loss. One furthermore calculates new decay constants Γ_i , because the probability for electron-impact ionization depends on the local electron density. This procedure makes it possible to follow the dynamics of the excitation and loss of the electron along each trajectory of the ion in the crystal, for various values of the initial energy, the entrance coordinate, and the angular distribution at the entrance. It is also possible to calculate the probability that the ion will be in any of the states $1S, 2S, 2P_0, 2P_{\pm 1}$ at the exit. This capability is particularly important for describing the spectral-angular and polarization properties of the emission of excited ions.

4. In experiments on coherent resonant excitation, the most common approach is to measure the charge states of the ion beam as it leaves the crystal as a function of the initial energy of the ions. Since the radii of excited states of the ion are about an order of magnitude larger than the radius of the ground state, the ionization probability is higher. Consequently, at ion energies near the resonant energy (when excited levels are being filled), the relative number of completely stripped ions increases.² The relative number of hydrogen-like ions, which is measured experimentally, is described in

our model by the quantity (E_{ion} is the initial energy of the ion)

$$R(E_{\text{ion}}) = \left\langle \sum_{n=0,1,2,3,4} |C_n(E_{\text{ion}})|^2 \right\rangle, \quad (10)$$

where the angle brackets mean an average over all possible ion trajectories in the

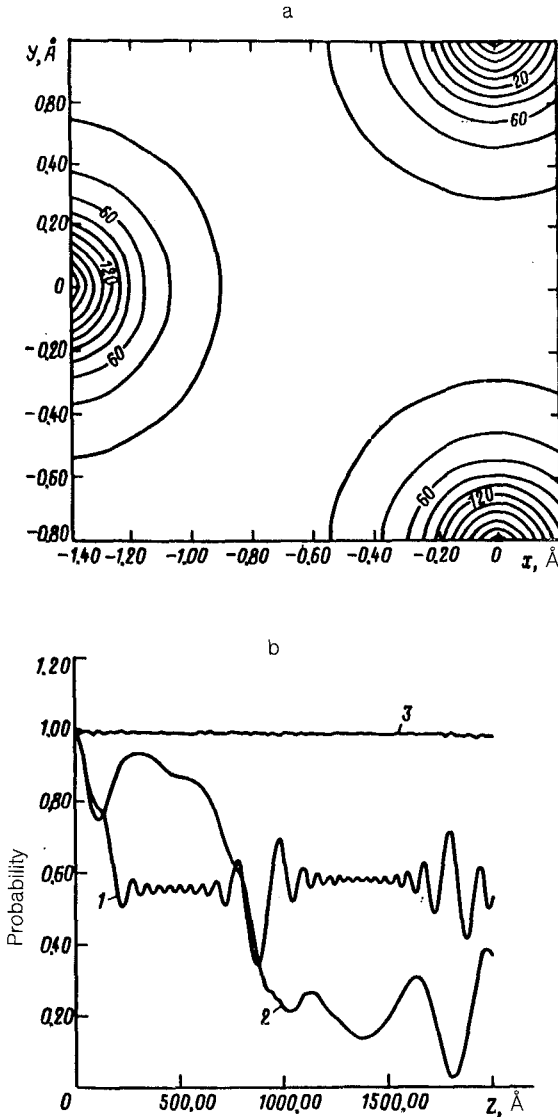


FIG. 1. a: Map of the potential $U(\mathbf{R}_1)$ for Au $\langle 111 \rangle$ and a typical trajectory of a Ne^{9+} ion in the transverse plane for an initial energy $E_{\text{ion}}/E_{\text{res}} \sim 1$. The entrance angle is ~ 0.1 of the Lindhard angle. The crystal thickness is 2000 \AA , and the temperature is $T = 293 \text{ K}$. b: The probability for the ion to remain in the $1S$ state as a function of the penetration depth in the crystal, Z , for several initial energies: $E_{\text{ion}}/E_{\text{res}} = 0.99, 1.00, \text{ and } 1.01$ (curves 1-3, respectively).

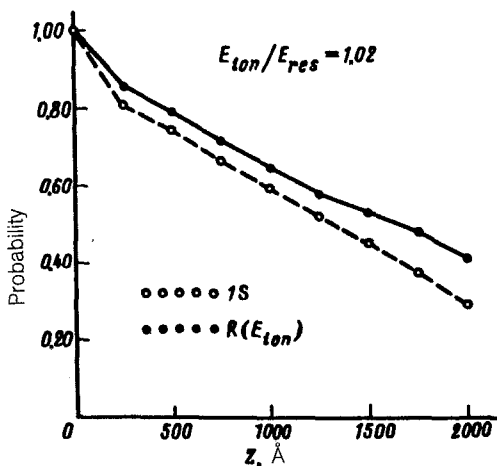


FIG. 2. Population of the 1S state and value of $R(E_{ion})$ as functions of the penetration depth in the crystal, Z , for $E_{ion}/E_{res} = 1.02$ after an averaging over 10^3 trajectories.

crystal or, equivalently, an average over the initial points, the entrance angle, and the spread in initial energy.

The procedure described above has been used to interpret the experiments of Ref. 6 on the coherent resonant excitation of Ne^{9+} ions in a gold $\langle 111 \rangle$ crystal $0.2 \mu m$ thick. Figure 1a shows a potential map for the $\langle 111 \rangle$ direction in Au, along with the projection of a typical trajectory onto the plane perpendicular to the $\langle 111 \rangle$ direction for an ion energy $E_{ion} = 87.7$ MeV near the $n = 6$ resonance. Figure 1b shows the probability for the ion to remain in its ground state, $|C_0(z)|^2$, along the given trajectory for three values of the energy: $E_{ion}/E_{res} < 1$, $E_{ion}/E_{res} = 1$, $E_{ion}/E_{res} > 1$. Clearly, there is resonance, and the energy loss is having an effect (a passage through a reso-

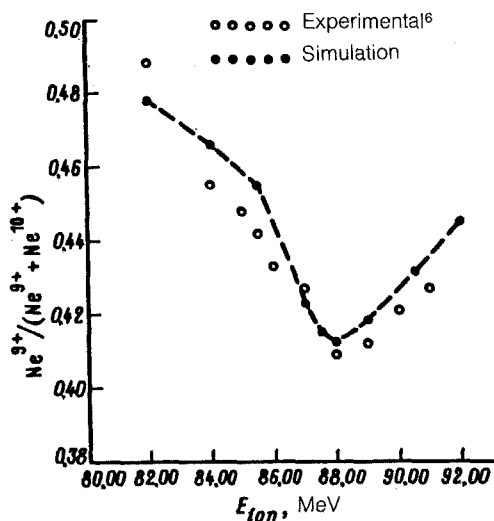


FIG. 3. Ratio of the yields of the relative numbers of Ne^{9+} ions in the 1S, 2S, and 2P states to the total number of ions as a function of the initial energy of the ion beam, E_{ion} , along with experimental data. The statistical base is 10^3 particles; the angular spread is ~ 0.1 of the Lindhard angle; and the energy spread is $\Delta E/E \sim 0.001$.

nance inside the crystal). Figure 2 shows the behavior $R(E_{\text{ion}})$ and the population of the $1S$ state versus the penetration depth after an averaging over trajectories for $E_{\text{ion}}/E_{\text{res}} = 1.02$. It follows that the $1S$ state is filled predominantly during coherent resonant excitation. The yield of photons beyond the crystal due to the radiative decay of excited states is therefore small. Finally, Fig. 3 shows the final results of the numerical simulation: a plot of $R(E_{\text{ion}})$ versus the ion energy E_{ion} , along with experimental data from Ref. 6. The fairly good agreement between the calculated and experimental data allows us to conclude that the initial physical principles of the model are correct. The calculations also reveal that the size of the dip in $R(E_{\text{ion}})$ at $E_{\text{ion}} = E_{\text{res}}$ depends very strongly on the strength of the wakefield potential, and the width of the resonance depends strongly on the decay constants of the excited states, particularly that of the $2P_0$ state. In this particular calculation, we selected the values of these constants on the basis of the average electron density over the crystal. A more rigorous calculation would include a direct calculation of the decay constants from the states deformed by the crystal field, with allowance for the local electron density. The breakdown of the resonance approximation at $E_{\text{ion}}/E_{\text{res}} \gtrsim 1$ should also be taken into account.

In summary, the model developed here makes it possible to follow the dynamics of transitions to excited states and the formation of the charge states of hydrogen-like ions over the crystal thickness. It thus becomes possible to select optimum experimental conditions for reaching a better understanding of the physics of coherent resonant excitation. Relativistic effects (for ions with $\alpha Z < 1$ and $\gamma > 1$) can be incorporated in this method easily.

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