

## ELECTRON DENSITY FLUCTUATIONS INDUCED BY ION CLUSTERS IN CONDENSED MATTER

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Valence electron density fluctuations induced around a trailing ion by a leading ion of a swift dicluster moving through condensed matter depend on the orientation of the dicluster axis with respect to the beam axis. These density fluctuations may appear experimentally in the angle dependence of the knock-on collision electrons emitted under bombardment of molecular ions. We present linear-response theory calculations of the density fluctuations at the position of the trailing ion, as a function of the orientation. Second-order perturbation theory calculations of the dependence on the angle of the scattering probabilities of target valence electrons by a two-center Coulomb field moving with different velocities are presented. The applicability of linear theories as the charges and velocities of diclusters are changed is discussed.

### 1. Introduction

Since the pioneering work of Ritchie and coworkers [1], who first gave an explicit expression for the wake potential induced by coherent electron displacements due to the passage through condensed matter of swift charged particles, much theoretical [2,4] and experimental [5-9] work has been performed. Most of the experimental work is related to measurements of the electric field strength and the wake potential, quantities that are obtained from certain averages of the induced electron density fluctuations.

When molecular ions penetrate solid targets, valence electron density fluctuations induced around the trailing ion by the leading ion depend on the orientation,  $\theta$ , of the molecular axis with respect to the beam axis. These fluctuations may appear experimentally in the dependence on  $\theta$  of the knock-on collision electrons emitted under bombardment by molecular ions. In particular, measurements of the knock-on collision electrons emitted at  $0^\circ$  under  $N_2^+$  molecular ion bombardment as a function of the orientation ( $\theta$ ) of the molecular ions have been recently performed by Yamazaki [10].

In this paper we first present linear-response theory calculations, in the plasmon-pole approximation, of the density fluctuations induced by the leading ion of the dicluster at the position of the trailing ion, as a function of  $\theta$ . Next we go beyond first order and present the results of a second-order quantum-mechanical calculation of the  $\theta$  dependence of the scattering probabilities of target valence electrons by a two-center Coulomb field.

### 2. Density fluctuations

The electron density fluctuations  $\delta n(\rho, z)$  in a homogeneous isotropic medium induced by a swift point charge  $Z$  having constant velocity  $v$  are related to the wake potential by the Poisson equation and given, in

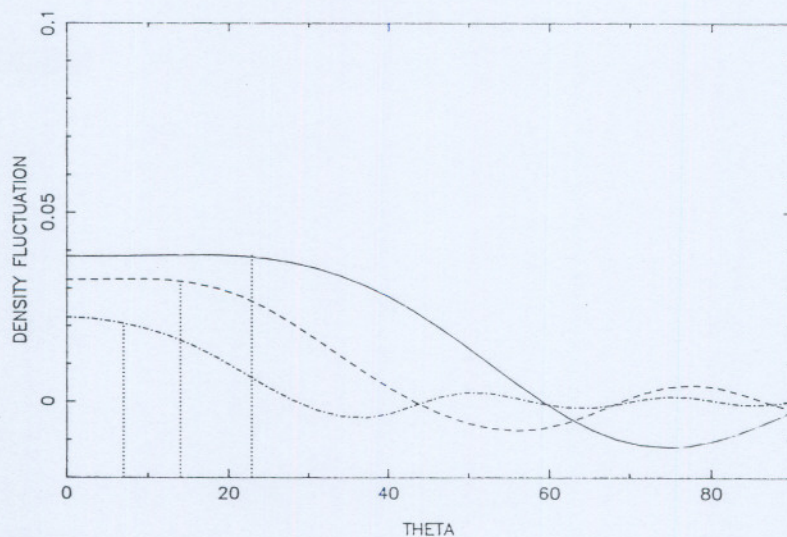


Fig. 1. Induced density fluctuations at the position of the trailing ion versus the orientation of ion diclusters moving with velocity  $v = 2.5$  a.u. (solid curve),  $v = 4.0$  a.u. (dashed curve) and  $v = 8.0$  a.u. (dashed-dotted curve). The medium is characterized by a density parameter  $r_s = 1.53a_0$ . Also shown are the corresponding  $\theta \approx \sin^{-1}(\beta/v)$  angles (vertical dotted lines).

the plasmon-pole approximation, by eq. (26) of ref. [3].  $\rho$  and  $z$  are the cylindrical coordinates relative to the position of the point charge  $Z$ .

Fig. 1 shows plots of  $\delta n(R \sin \theta, -R \cos \theta)/Z$ , that is, the electron density fluctuation induced at the trailing ion position by the leading ion when a dicluster transverses the target, as a function of the dicluster polar angle with respect to the beam direction, for different constant velocities.  $R$  is the distance between the pair of ions of the dicluster, which is assumed to be constant and equal to  $2.2a_0$  ( $a_0 = 0.529 \text{ \AA}$ ). It can be seen from this figure, that the density fluctuation of target valence electrons around the trailing ion is prominent when it is in the conical region behind the particle with half-angle of opening  $\theta \sim \sin^{-1}(\beta/v)$ , which defines the envelope of the collective wake [3], and this fact may appear experimentally in the  $\theta$ -dependence of the knock-on collision electrons emitted under bombardment of molecular ions.

If we are interested in electrons associated with high momentum transfers, the scattering processes are mainly determined by the short strong range of the Coulomb potential; however, no detailed nonlinear theory exists at present. In the next section we present some theoretical work based on second-order perturbation theory, discuss its validity comparing with the very scarce experimental data, and suggest new experiments.

### 3. Quantum analysis

We consider the process of noninteracting electrons being scattered by a two-center Coulomb field moving with velocity  $v_0$ , each center having a charge equal to  $Z$ , in the coordinate system in which the two centers, separated by a fixed distance  $R$ , are at rest.

If the electron is initially in the state  $|i\rangle$ , the probability per unit time of finding it in the state  $|f\rangle$  after the perturbation caused by the ion and represented by  $H'$  is given to second order, in atomic units, where  $|e| = \hbar = m = 1$ , by [11]

$$\gamma_{fi} = 2\pi \left| H'_{fi} + \sum_n \frac{H'_{fn} H'_{ni}}{\omega_{in}} \right|^2 \delta(E_f - E_i), \quad (1)$$

where  $H'_{fi}$ ,  $H'_{fn}$  and  $H'_{ni}$  represent matrix elements of the perturbation,

$$\omega_{in} = E_i - E_n, \tag{2}$$

and  $E_i$ ,  $E_f$  and  $E_n$  are the eigenvalues of the unperturbed Hamiltonian corresponding to  $|i\rangle$ ,  $|f\rangle$  and the  $|n\rangle$  intermediate states, respectively.

The initial and final states are assumed to be plane waves, and the perturbation Hamiltonian

$$H' = V(\mathbf{r}) + V(\mathbf{r} + \mathbf{R}), \tag{3}$$

where  $V(\mathbf{r})$  is a Yukawa potential, that is, we approximate the wake by a spherically symmetric form of the total potential that electrons in the medium feel when a point charge  $Z$  having a constant velocity  $v_0$  passes through the medium:

$$V(\mathbf{r}) = \frac{Z e^{-\alpha r}}{r} \quad \left( \alpha = \frac{\omega_p}{v_0} \right), \tag{4}$$

which should be a good approximation for transitions to electronic states of high momentum. Then we obtain

$$H'_{fi} = \langle f | H' | i \rangle = \frac{Z}{2\pi^2} \frac{1 + e^{-i(\mathbf{k}_i - \mathbf{k}_f) \cdot \mathbf{R}}}{\alpha^2 + |\mathbf{k}_i - \mathbf{k}_f|^2}, \tag{5}$$

and

$$H'_{fn} H'_{ni} = \langle f | H' | n \rangle \langle n | H' | i \rangle = \frac{Z^2}{4\pi^4} \frac{1 + e^{-i(\mathbf{k}_i - \mathbf{k}_f) \cdot \mathbf{R}} + e^{-i(\mathbf{k}_i - \mathbf{k}_n) \cdot \mathbf{R}} + e^{-i(\mathbf{k}_n - \mathbf{k}_f) \cdot \mathbf{R}}}{(\alpha^2 + |\mathbf{k}_n - \mathbf{k}_f|^2)(\alpha^2 + |\mathbf{k}_i - \mathbf{k}_n|^2)}, \tag{6}$$

where  $\mathbf{k}_i$ ,  $\mathbf{k}_f$  and  $\mathbf{k}_n$  represent the wave vectors of  $|i\rangle$ ,  $|f\rangle$  and  $|n\rangle$  states, respectively. Substituting eqs. (2), (5) and (6) in eq. (1) and integrating over final states, the probability per unit time and unit solid angle of the transition to one of a group of final states that have nearly the same  $\mathbf{k}_f$ , is

$$\frac{d\gamma}{d\Omega} = \frac{Z^2 k_i}{2\pi^3} \left| \frac{1 + e^{-i(\mathbf{k}_i - \mathbf{k}_f) \cdot \mathbf{R}}}{\alpha^2 + |\mathbf{k}_i - \mathbf{k}_f|^2} + \frac{Z}{\pi^2} (I_1 + I_2) \right|^2, \tag{7}$$

if  $\mathbf{k}_f = \mathbf{k}_i$ , where

$$I_1 = \int \frac{d\mathbf{q}}{k_i^2 - q^2} \frac{1 + e^{-i(\mathbf{k}_i - \mathbf{k}_f) \cdot \mathbf{R}}}{(\alpha^2 + |\mathbf{q} - \mathbf{k}_f|^2)(\alpha^2 + |\mathbf{k}_i - \mathbf{q}|^2)} \tag{8}$$

and

$$I_2 = \int \frac{d\mathbf{q}}{k_i^2 - q^2} \frac{e^{-i(\mathbf{k}_i - \mathbf{q}) \cdot \mathbf{R}} + e^{-i(\mathbf{q} - \mathbf{k}_f) \cdot \mathbf{R}}}{(\alpha^2 + |\mathbf{q} - \mathbf{k}_f|^2)(\alpha^2 + |\mathbf{k}_i - \mathbf{q}|^2)}. \tag{9}$$

To obtain the scattering probability per unit time and unit solid angle of any target valence electron to be emitted after the perturbation with a wave vector  $\mathbf{k}_f$ , it is necessary to perform the average of eq. (7) over all allowed initial states. In particular, if electrons emitted in a velocity region of  $\lambda_1 v_0 \leq v_f \leq \lambda_2 v_0$ , at  $0^\circ$  with respect to the beam direction of bombarding diclusters, are considered, the only contributions to eq. (7) come from initial states whose wave vector  $\mathbf{k}$ , in the dicluster rest system, satisfies

$$\lambda_1 k_0 \leq |\mathbf{k} - \mathbf{k}_0| \leq \lambda_2 k_0, \tag{10}$$

where  $\mathbf{k}_0$  is the wave vector of the bombarding dicluster. Then, by taking  $\mathbf{k}_f = k_i \mathbf{k}_0 / k_0$ , we obtain the following average scattering probability per unit time and unit solid angle:

$$\left( \frac{d\gamma}{d\Omega} \right)_{av} = \frac{3Z^2}{8\pi^4 k_F^3} \int_{\lambda_1 k_0}^{\lambda_2 k_0} dk_i k_i^3 \int_{k'}^1 d\mu_i \int_0^{2\pi} d\phi_i |f(k_i, \mu_i, \phi_i)|^2, \tag{11}$$

with  $k' = (k_i^2 + k_0^2 - k_F^2)/(2k_i k_0)$  and where

$$f(k_i, \mu_i, \phi_i) = f_1(k_i, \mu_i, \phi_i) + \frac{Z}{\pi^2} [f_{21}(k_i, \mu_i, \phi_i) + f_{22}(k_i, \mu_i, \phi_i)], \quad (12)$$

with

$$f_1(k_i, \mu_i, \phi_i) = \frac{1 + e^{-i\zeta}}{\alpha^2 + 2k_i^2(1 + \mu_i)}, \quad (13)$$

$$f_{21}(k_i, \mu_i, \phi_i) = (1 + e^{-i\zeta}) \int_0^\infty dq \frac{q^2}{k_i^2 - q^2} \int_{-1}^1 \int_0^{2\pi} d\mu d\phi \frac{1}{A} \quad (14)$$

and

$$f_{22}(k_i, \mu_i, \phi_i) = \int_0^\infty dq \frac{q^2}{k_i^2 - q^2} \int_{-1}^1 \int_0^{2\pi} d\mu d\phi \frac{e^{-i\zeta_1} + e^{-i\zeta_2}}{A}. \quad (15)$$

In these expressions,

$$A = (\alpha^2 + q^2 + k_i^2)^2 - 2k_i q (\alpha^2 + q^2 + k_i^2)(\mu + xk) + 4k_i^2 q^2 \mu xk,$$

$$xk = -\mu \mu_i \sqrt{(1 - \mu_i^2)(1 - \mu^2)} (\cos \phi \cos \phi_i + \sin \phi \sin \phi_i),$$

$$\zeta = k_i R [\sqrt{1 - \mu_i^2} \cos \phi_i \sin \theta - (1 + \mu_i) \cos \theta],$$

$$\zeta_1 = R [(k_i \sqrt{1 - \mu_i^2} \cos \phi_i - q \sqrt{1 - \mu^2} \cos \phi) \sin \theta - (q\mu + k_i \mu_i) \cos \theta]$$

and

$$\zeta_2 = R [q \sqrt{1 - \mu^2} \cos \phi \sin \theta - (k_i - q\mu) \cos \theta].$$

$k_F$  is the Fermi momentum ( $k_F = 1.92/r_s$ ), and  $r_s$  is the mean distance between electrons, i.e.  $n = 3/(4\pi r_s^3)$ .

If the target valence electrons are considered to have zero velocity in the laboratory system, we have

$$\left( \frac{d\gamma}{d\Omega} \right)_{av} = \frac{3Z^2 k_0^2}{8\pi^3 k_F^3} \left[ \frac{(k_F^2 - k_0^2)(\lambda_2^2 - \lambda_1^2)}{2} + \frac{2k_0^2(\lambda_2^3 - \lambda_1^3)}{3} - \frac{k_0^2(\lambda_2^4 - \lambda_1^4)}{4} \right] |f(k_0, 1, \phi_i)|^2, \quad (16)$$

where  $f(k_0, 1, \phi_i)$  is given by eq. (12).

We first considered ion diclusters moving with various velocities, each ion having a charge equal to  $Z = 1$ , and calculated the scattering probabilities per unit time and unit solid angle for electrons to be emitted in a velocity region of  $1.95v_0 \leq v_f \leq 2.15v_0$  at  $0^\circ$  with respect to the beam. Figs. 2a–c exhibit plots of the scattering probabilities computed from eq. (16), as a function of  $\theta$ , when only the first-order term is considered and when both first- and second-order terms are included in our calculations, for the velocities given in the figure caption. Notice that as the velocity increases, the second-order term becomes comparatively smaller, as we expected, since the perturbation also becomes smaller, and more fluctuations occur in the dependence of the scattering probabilities on the orientation of bombarding molecular ions,  $\theta$ .

In order to compare our results with the experimental data [10], next we have considered 4.5 MeV  $N_2^+$  molecular ions ( $v = 2.5$  a.u. and  $R = 2.2$  a.u.) bombarding a carbon foil target ( $r_s = 1.53a_0$ ) and calculated the scattering probabilities per unit time and unit solid angle for electrons to be emitted in a velocity region of  $1.95v_0 \leq v_f \leq 2.15v_0$  at  $0^\circ$  with respect to the beam. Fig. 3 shows the results obtained from eqs. (11) and (16), plotted as a function of  $\theta$ , assuming that the effective charge,  $Z$ , of the ions of the dicluster is given by the following expression [12]:

$$Z = Z_0 [1 - \exp(-vZ_0^{-2/3})], \quad (17)$$

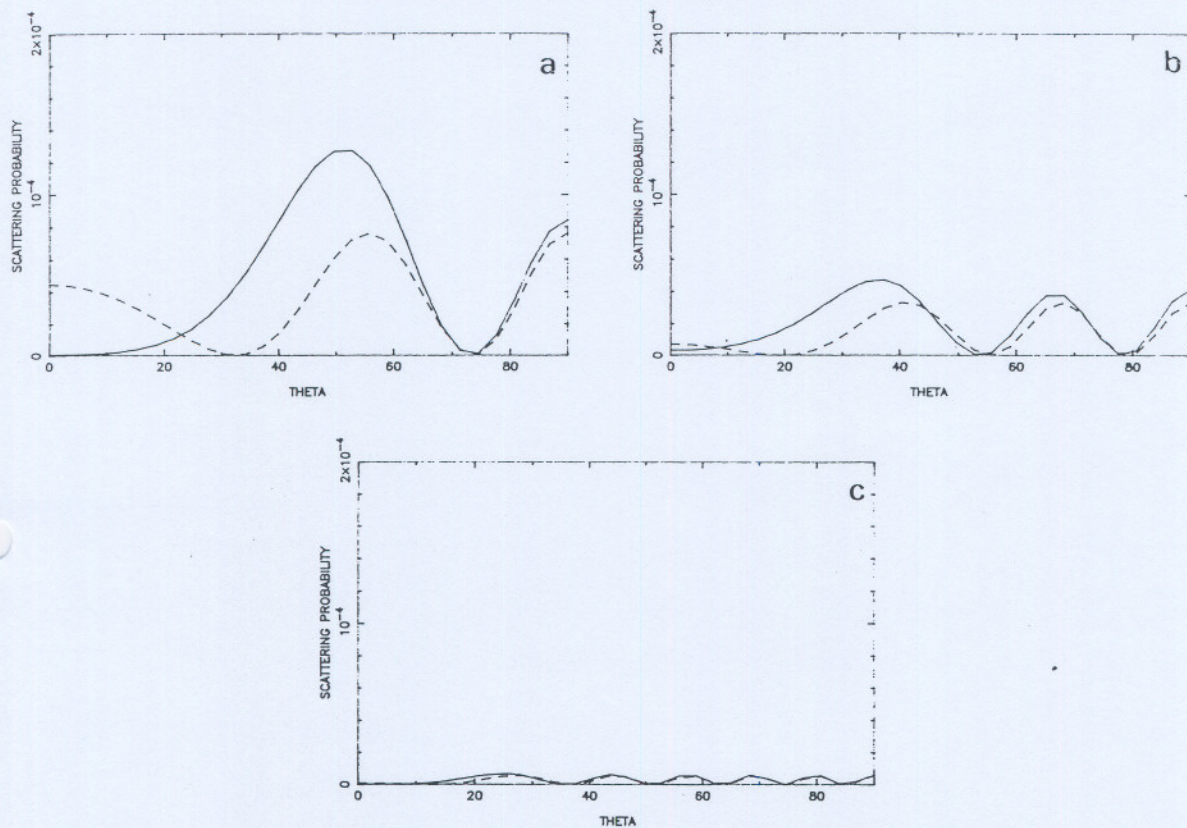


Fig. 2. The solid and dashed curves show second- and first-order perturbation theory calculations, respectively, of the scattering probabilities of target valence electrons versus  $\theta$ , obtained from eq. (16) for  $Z = 1$  and for (a)  $\nu = 2.5$ , (b)  $\nu = 4.0$  a.u. and (c)  $\nu = 8.0$  a.u.

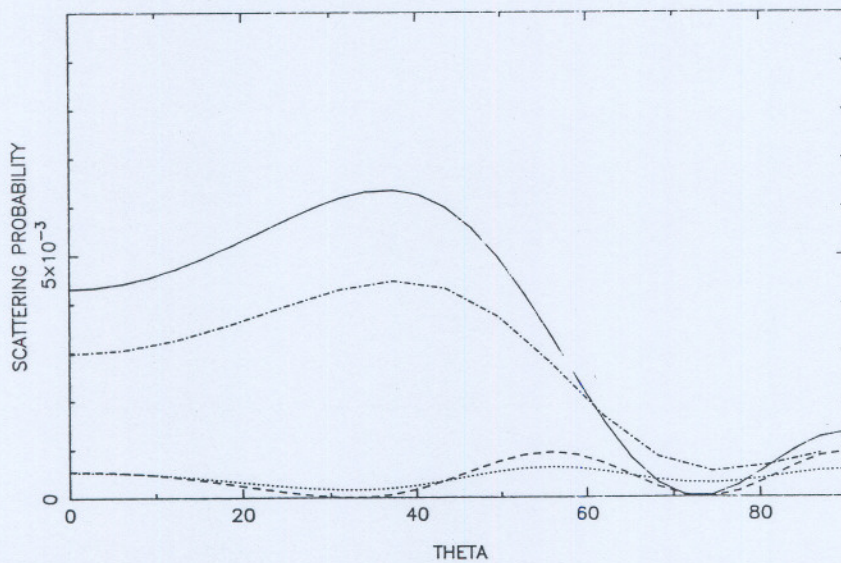


Fig. 3. The solid and dashed curves show second- and first-order perturbation theory calculations, respectively, of the scattering probabilities versus  $\theta$ , obtained from eq. (16) for  $Z_0 = 7$ ,  $\nu = 2.5$  a.u. and the effective charge given by eq. (17). The dashed-dotted and dotted curves show second- and first-order perturbation theory calculations, respectively, obtained from eq. (11) for  $Z_0 = 7$  and  $\nu = 2.5$  a.u.

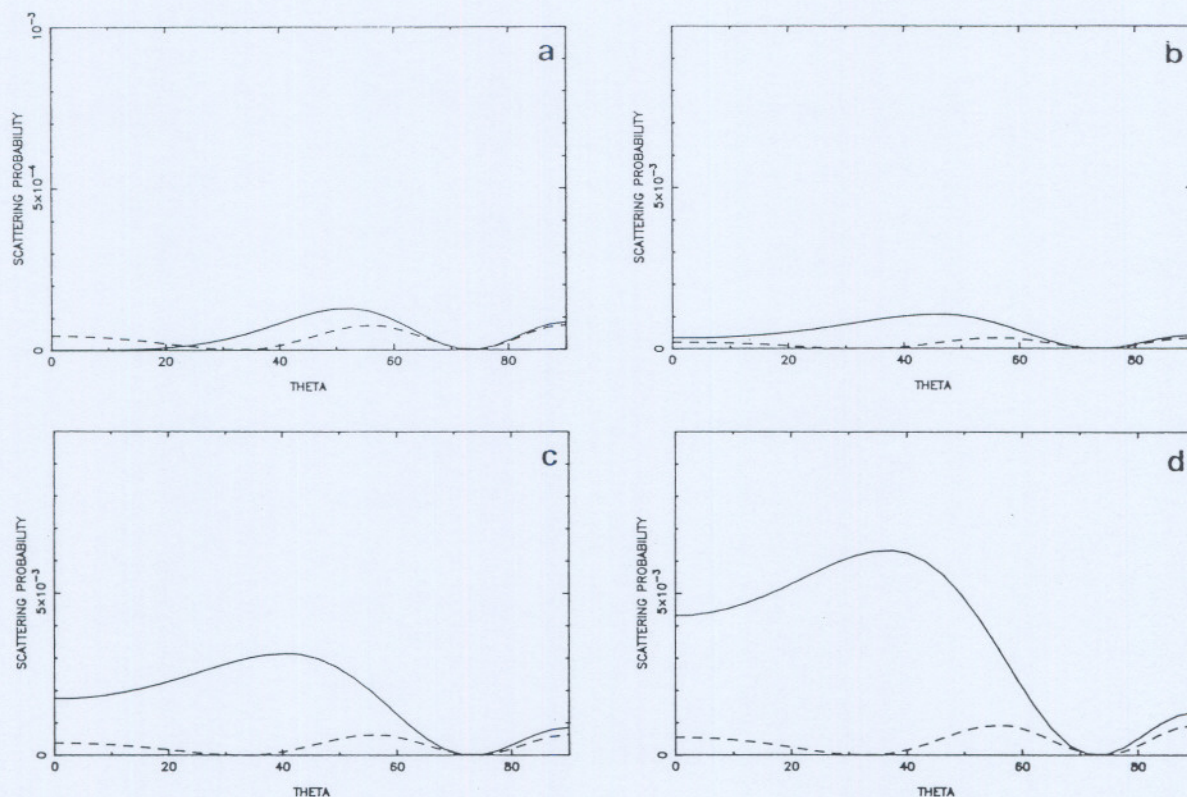


Fig. 4. Perturbation theory calculations of the scattering probabilities of target valence electrons versus  $\theta$ , obtained from eq. (16) for  $v = 2.5$  a.u. and effective charges given by eq. (17), with (a)  $Z_0 = 1$ , (b)  $Z_0 = 3$ , (c)  $Z_0 = 5$  and (d)  $Z_0 = 7$ . Second- and first-order calculations are represented by solid and dashed curves, respectively.

where  $Z_0$  is the bare ion charge. Notice from this figure that the specific contributions from all the initial states to the average scattering probability do not change qualitatively its angular structure. It can be concluded, also, that the second-order term is quite large compared with the first-order one. And, on the other hand, agreement with the experimental data is not very good [10]. It may be due to the fact that for such effective charges and velocities second-order perturbation theory is not valid any more. Indeed, the smaller the effective charge the smaller the second-order terms, as it is obvious from figs. 4a–d which show the way the results change as the effective charge increases, for a fixed velocity.

#### 4. Conclusions

It has been shown that second-order perturbation theory is more accurate for small charges and high velocities, as we expected. On the other hand, as the ion velocity increases, the angular structure of the scattering probability changes appreciably. However, the available experimental data have been obtained for rather large effective charges, so new experiments performed for  $Z = 1$  and at different velocities would be very useful in order to investigate the applicability of linear theories and capture and loss theories, and to detect the effects of electron density fluctuations. We hope our work will provoke such experiments.

In future work, we shall investigate the nonlinearity appearing for high values of  $Z$ . Indeed, the fact that in order to calculate the average scattering probability it is sufficient to compute the transition probability of the electron having zero velocity in the laboratory system enables to perform third-order perturbation theory calculations. Furthermore, the wave functions of the continuum for the two-center

problem, exact solutions of the Schrödinger equation, may be computed, so the exact scattering probabilities of electrons on two Coulomb centers could be obtained, though this would be a cumbersome task.

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