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Young-Type Interference in Projectile-Electron Loss in Energetic Ion-Molecule Collisions

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Under certain conditions an electron bound in a fast projectile ion, colliding with a molecule, interacts mainly with the nuclei and inner shell electrons of atoms forming the molecule. Because of their compact localization in space and distinct separation from each other, these molecular centers play in such collisions a role similar to that of optical slits in light scattering leading to pronounced interference in the spectra of the electron emitted from the projectile.

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The wave-particle duality, which states that all atomic objects exhibit particle as well as wave properties, is one of the basic concepts of quantum mechanics. Proposed initially by de Broglie [1] in 1923, this concept was confirmed a few years later in the electron diffraction experiments [2,3]. Since then, a large number of investigations have been performed in order to observe the wave nature of not only electrons but also heavier particles such as, for example, neutrons, atoms, dimers, and even fullerenes C_{60} [4]. Most of these measurements were aimed at a demonstration of Young's double-slit phenomena, in which the coherent addition of the amplitudes of two (or many) paths, leading to interference, is related to the wavelike particle behavior.

In the atomic world the natural analog of Young's slits is represented by diatomic molecules. Starting with the works [5,6], particularly significant interest has been focused on studying interference phenomena involving homonuclear molecules [7–22].

These studies dealt with two principally different interference scenarios. In one of them, the attention was focused on interference in the spectra of electrons emitted from the molecule in the course of photoionization [6–13] and consequent Auger decay [14], as well as in ionization by electrons [15] and heavy ions [16–18]. Note that in such a scenario, unlike Young's experiment, the wave is not diffracted by the ''slits'' but rather emerges from them. In the second scenario, which was realized in [19–22] for electron capture and proton scattering and is a more direct analog of Young's optical experiment, interference is caused by coherent scattering of the incident projectile on the atomic centers of the molecule.

In this Letter we propose yet another way to collision-induced interference. It falls into the second scenario but, similarly to [16–18], deals with interference in electron emission spectra. It is realized in collisions of molecules with partially stripped multiply charged projectile ions, in which the electron(s) of the projectile is emitted.

Compared to the electron emission, studied in [16–18], the present case possesses important differences. In particular, in the situation, considered in [16–18], the electron wave is launched from the slits, which are not really separated and well localized. Indeed, electrons of molecules like H_2 are delocalized over the entire molecular volume and are mainly located not on the atomic nuclei but rather between them. The majority of electrons, emitted by the impact of a fast charged projectile, are the so-called soft electrons, which have low energies and are ejected from the entire volume occupied by the electrons in their initial state. As a result, the whole volume of the molecule participates in forming the emission pattern. In contrast, as will be shown below, the emission from the projectile, because of large momentum transfers involved, occurs due to a coherent scattering of the electron of the projectile on the *nuclei* of the molecule (partially screened by the inner shell electrons) and, therefore, the slits are very well separated and localized in space that can lead to pronounced interference effects in the emission pattern.

Below, based on the relativistic time-dependent perturbation approach, we shall derive the cross section for electron loss in collisions with homonuclear dimers. The possibility of interference effects will be demonstrated by calculating the cross section for fast hydrogenlike magnesium $Mg^{11+}(1s)$ and $S^{15+}(1s)$ ions colliding with N_2 dimers.

Atomic units are used throughout except where otherwise stated.

Since the collision between an ion carrying an electron and a molecule in general represents a very complex many-body problem, our consideration will be based on a simplified model which, however, takes into account all essential physics of the collision process in question. Within this model, in order to describe electron transitions in the projectile we shall use the first order perturbation theory in the interaction between this electron and the

molecule. Such an approximation is a good one, provided $Z_p \gtrsim Z_A$, where Z_p and Z_A are, respectively, the nuclear charges of the ion and the atoms in the molecule, and one merely wishes to describe projectile-electron transitions, without paying attention to what happens with the molecule in such collisions.

Further, we shall only consider molecules whose atoms have relatively large atomic numbers, $Z_A \gg 1$. Under the simultaneously fulfilled conditions $Z_A \gg 1$ and $Z_p \gtrsim Z_A$, the main contribution to the projectile-electron transitions in collisions with the molecule is given by the screening target mode, in which the projectile electron interacts with the molecule ''frozen'' during the short collision time in its initial state [23].

Moreover, provided the condition $Z_p \gtrsim v$ is fulfilled (v is the collision velocity), the momentum q transferred in the collision becomes so large (on the molecular scale) that the outer electrons of the molecule are not able to screen the nuclei of the molecule. Therefore, the main contribution to the electron loss arises from the interaction with the nuclei of the molecule partially screened by the inner shell electrons. Additionally, large momentum transfers occur in collisions where the typical distances R between the nuclei and the electron of the projectile are small ($R \sim 1/q \ll 1$), which makes the Coulomb field of the partially screened nuclei effectively short ranged. Thus, the projectile electron undergoes transitions due to the interaction with well localized centers of force which, in addition, are well separated in space. In addition, since the inner electrons are basically atomic electrons, one can treat the molecule as a sum of free atoms and use the atomic parameters for the description of the field produced by the molecule in the collision.

Taking all this into account, the scalar potential describing the field of the molecule in its rest frame K' can be written as

$$\Phi'_M(\mathbf{r}') = \sum_{j=1}^{2} \frac{Z_j \phi_j(|\mathbf{r}' - \mathbf{R}'_j|)}{|\mathbf{r}' - \mathbf{R}'_j|}, \tag{1}$$

where $\mathbf{r}'$ is the observation point of the field and $\mathbf{R}'_j$ is the coordinate of the nucleus of the jth atom ($j = 1, 2$), Z_j the charge of the nucleus, and

$$\phi_j(x) = \sum_l A_j^l \exp(-\kappa_j^l x), \tag{2}$$

with the screening parameters A_j^l ($\sum_l A_j^l = 1$) and κ_j^l tabulated in [24,25].

It is convenient to treat the projectile-electron transitions using the reference frame K in which the nucleus of the projectile is at rest. We take the position of the nucleus as the origin of K and assume that in this frame the center of mass of the molecule moves along a straight-line classical trajectory $\mathbf{R}(t) = \mathbf{b} + \mathbf{v}t$, where $\mathbf{b} = (b_x, b_y, 0)$ is the impact parameter, $\mathbf{v} = (0, 0, v)$ is the collision velocity, and t is the time. Using Eqs. (1) and (2) and the Lorentz transformation for the potentials, we obtain that the electromagnetic field of the molecule in the frame K is described by the potentials

$$\Phi_M(\mathbf{r}, t) = \gamma \Phi'_M(\mathbf{s}_j) \qquad \mathbf{A}_M(\mathbf{r}, t) = \left(0, 0, \frac{v}{c}\Phi_M\right), \tag{3}$$

where $\mathbf{r} = (\mathbf{r}_\perp, z)$ with $\mathbf{r}_\perp \cdot \mathbf{v} = 0$ the coordinate of the point of observation of the field in the frame K, c is the speed of light, and $\gamma = 1/\sqrt{1 - v^2/c^2}$ is the collisional Lorentz factor. Further,

$$\mathbf{s}_j = (\gamma(z - vt_j), \mathbf{r}_\perp - \mathbf{b}_j), \tag{4}$$

where $\mathbf{b}_j = \mathbf{b} + \delta\mathbf{b}_j$ is the impact parameter for the nucleus of the jth atom of the molecule, t_j is the time of its closest approach to the origin, and $\mathbf{s}_j$ is the vector connecting the position of the jth atomic nucleus of the molecule and the electron of the ion (as is viewed in the rest frame of the molecule).

Using the first order perturbation theory in the interaction of the electron of the ion with the molecular field, described by the potentials (3), one can show that the cross section $\sigma_{\rm fi}$ for the projectile-electron transitions occurring in collisions with the molecule is given by

$$\sigma_{\rm fi} = 4\sigma_{\rm fi}^{(A)} \cos^2\left(\frac{\mathbf{q}' \cdot \mathbf{l}_0}{2}\right). \tag{5}$$

Here, $\sigma_{\rm fi}^{(A)}$ is the cross section for the projectile-electron transitions occurring in collisions with the corresponding single atom, $\mathbf{q}' = (\mathbf{q}_\perp, \frac{\omega_{\rm fi}}{\gamma v})$ is the momentum transferred to the projectile ion (as viewed in the rest frame of the molecule) with $\omega_{\rm fi}$ being the transition frequency for the electron of the ion, and $\mathbf{l}_0 = (l_0, \vartheta_M, \varphi_M)$ is the vector connecting the positions of the atomic nuclei of the molecule in its rest frame. In what follows we shall count the polar orientation angle ϑ_M of the molecule from the direction of the projectile velocity $\mathbf{v}$. In addition, we set $\varphi_M = 0°$.

Note that expressions similar to Eq. (5) have been obtained in the past (but without the relativistic correction) for other atomic processes involving ''two slits'' (see, e.g., [26]).

In Fig. 1 we present the electron loss cross section, $d^3\sigma/dp_{\rm lg}dp_{\rm tr}d\varphi_p$, differential in the longitudinal ($p_{\rm lg} = \mathbf{p} \cdot \mathbf{v}/v$) and transverse ($p_{\rm tr} = \sqrt{p^2 - p_{\rm lg}^2}$) momenta, and the azimuthal emission angle φ_p of the electrons emitted from 7.8 MeV/u $Mg^{11+}(1s)$ projectiles in collisions with N_2 molecules. The cross section is obtained by integrating over the vector of the transverse momentum transfer $\mathbf{q}_\perp$. In the figure this cross section is given in the target frame as a function of $p_{\rm lg}$ and $p_{\rm tr}$ for the emission into the plane spanned by the molecular axis and projectile velocity (i.e., for $\varphi_p = 0°$). The molecular polar orientation angle is $\vartheta_M = 90°, 20°$ (the upper row, from left to right),

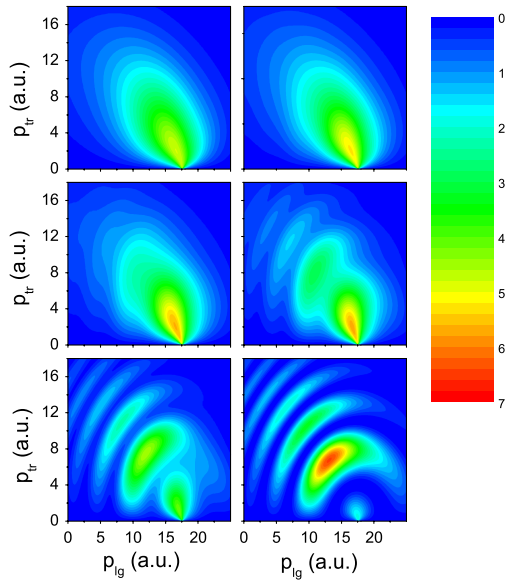

FIG. 1 (color online). The spectra of electrons (in a.u.) emitted into the plane spanned by the molecular axis and projectile velocity from 7.8 MeV/u $Mg^{11+}(1s)$ ions colliding with N_2 molecules. The spectra (from left to right and from top to bottom) correspond to $\vartheta_M = 90°$, 20°, 15°, 10°, 5°, and 0°. The angle ϑ_M is counted from the direction of the projectile velocity **v**.

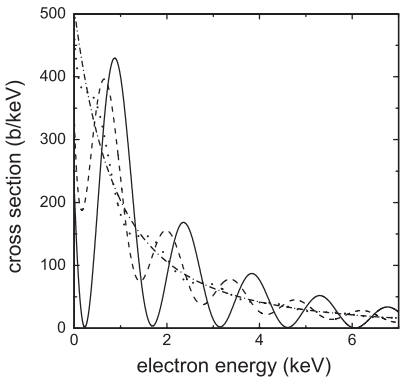

FIG. 2. Energy spectrum of electrons emitted under the zero azimuthal angle from 7.8 MeV/u $Mg^{11+}(1s)$ ions colliding with N_2. The spectrum is given in the projectile frame. Solid, dashed, and dotted curves correspond to collisions with the molecules oriented in the target frame under the polar angle $\vartheta_M = 0°$, 5°, and 10°, respectively. For a comparison, the dash-dotted curve shows the spectrum in collisions with N atoms multiplied by 2.

15°, 10° (the second row, from left to right), and 5°, 0° (the lower row, from left to right). At small ϑ_M the spectra exhibit very clear structures, which arise due to interference caused by the coherent interactions between the electron of the projectile and the two atomic centers of the molecule [27].

At an impact energy of 7.8 MeV/u ($v = 17.6$ a.u.) the typical momentum transfer to the electron of the ion, which is necessary for its removal out of the ion, is ~6–8 a.u. This magnitude is substantially larger than the typical momenta of the outer electrons of nitrogen. This means that within the screening target mode the projectile-electron transitions are governed mainly by the interaction between this electron and the target nuclei (partly screened by the K-shell electrons). Moreover, since the momentum transfers are large, the relative contribution of the so called antiscreening mode to the projectile-electron loss process, which scales roughly as $\sim Z_A$, is by about $Z_A = 7$ times smaller than that $\sim Z_A^2$ arising due to the screening mode (see, e.g., [23]). Thus, the outer target electrons have a minor effect on the projectile-electron transitions, and, therefore, the latter ones can indeed be regarded as occurring due to the interaction with two "slits," which are well localized and well separated from each other within the space occupied by the molecule.

At small polar orientation angle of the molecule the spectrum displays clear ringlike structures. The center of the rings is located at the point $\mathbf{p}_C = (p_{\rm tr} = 0; p_{\rm lg} = m_e v)$, where m_e is the electron mass, implying that each ring is formed by electrons which in the rest frame of the projectile have close energies. Indeed, the origin of these structures can be traced back by considering the energy spectrum of the emitted electrons in the rest frame of the projectile which is shown in Fig. 2. It is seen that the energy spectrum exhibits oscillations (especially pronounced at very small ϑ_M) due to the alternation of the parts with constructive and destructive interferences. It is not difficult to convince oneself that the ringlike structures in the momentum spectrum originates namely from these oscillations.

Note also that by considering the electron loss in the rest frame of the projectile (and using the time-dependent approach) one can view the corresponding structures in the emission spectra as arising due to interference in time [7] where the time slits act on the scale of $\lesssim 1/\omega_{\rm fi} \sim 10^{-19}$ s. Therefore, in the case under consideration the interference pictures in the target and projectile frames can be thought of as realized in position-momentum and time-energy domain, respectively. Such pictures complement each other.

In the rest frame of the projectile the loss process occurs as a result of the action of two electromagnetic pulses which arrive one after the other. By varying the projectile charge, collision velocity, and molecular target, one can change the strength and effective duration of the first and second pulses as well as the delay between them. This "pulse-pair" process closely resembles "pump-probe" processes involving lasers. In contrast to the latter ones, however, in the collision one can easily reach the time scale well below 10^{-18} s.

Calculations for projectiles having different nuclear charges show that the range of the molecular orientation angle ϑ_M, at which the interference effects are clearly visible in the emission pattern, decreases when Z_p increases. This can be easily understood if we recall that

the size of the electron orbit in the initial state scales as $1/Z_p$. Therefore, a more tightly bound electron can interact simultaneously with both molecular centers only if the transverse size of the molecule $l_{\mathrm{tr}} = l_0 \sin\vartheta_M$ becomes smaller.

As seen in Figs. 1 and 2, the most pronounced interference pattern in the emission spectrum arises at small orientation angles of the molecule. Therefore, in order to verify predicted effects in an experiment, it is very desirable to single out those loss events, which occur at small orientation angles, from the rest. This can be achieved by the determination of the molecular orientation *ex post*, which has been successfully applied in many experimental situations where molecular targets dissociated or Coulomb exploded after photoionization and strong-field ionization or due to electron or ion impact induced ionization.

In the collisions, considered above, by far a dominant contribution to the total electron emission is given by electrons ejected from the target. Therefore, an important question to address is whether in the momentum space there exists a substantial overlap between the electrons emitted from the projectile and those ejected from the target, which would mask the above predicted interference effects. In order to answer it we have estimated the emission from the N_2 molecules. We found that, since $v > Z_p$ and $v \gg Z_A$, the overlap in the case, considered in Figs. 1 and 2, is small and the interference pattern in the electron emission from the projectile is not ''damaged'' by the electrons ejected from the target.

In conclusion, we have considered interference effects in the electron emission accompanying energetic collisions of ionic projectiles with molecular targets. In contrast to all the previous studies of this subject, which were focused on interference in the electron emission from the target, we were searching for signatures of the interference effects in the electron emission from the projectile. We have shown that this emission may possess very clear interference structures which are caused by the coherent interactions between the electron of the projectile and the atomic centers of the molecule. Under certain conditions (which were discussed in detail above) this interaction is basically the one between the electron of the projectile and the nuclei of the atomic centers (partially screened by the inner shell atomic electrons). This means that the interference arises from the scattering of the projectile electron on atomic slits, which are well localized in space and distinctly separated from each other, playing a role rather similar to that of the optical slits in the Young-type experiments with photons. In the rest frame of the projectile the process can be viewed as caused by two electromagnetic pulses arriving one after the other and acting on the subattosecond scale.

Owing to recent advances in the experimental techniques it has become feasible to test the above theoretical predictions. In particular, this will be done in forthcoming experiments at MPI-K (Heidelberg, Germany).

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[27] Note that in the 90° case the emission pattern is practically identical to that in collisions with two independent nitrogen atoms.