# Analysis of secondary-electron emission in beam-foil experiments with molecular ions

N. R. Arista, M. M. Jakas, G. H. Lantschner, and J. C. Eckardt
*Centro Atómico Bariloche, Comisión Nacional de Energía Atómica, 8400 Bariloche, Argentina*
(Received 17 June 1986)

We analyze the possibility of large oscillatory wake effects in secondary-electron emission induced by molecular ions, which were proposed in previous interpretations of results derived from beam-foil experiments. We show that theoretical expectations are in disagreement with the description of such oscillatory effects in terms of wake phenomena. Our analysis of various experimental evidences suggests a different origin for the effect. We propose a simple explanation for the molecular effect in secondary-electron emission, which is based on previous knowledge of electron emission from solid targets, and of molecular effects in the energy loss of ion clusters in solid foils.

## I. INTRODUCTION

Molecular effects in secondary-electron emission (SEE) from solids have been studied by several workers[1–4] by measurements of the electron yields due to atomic- and molecular-ion impact.

This molecular effect can be characterized by a difference in the electron yield due to molecular ions ($\gamma_m$) and the sum of the yields of the constituent ions ($\gamma_a=\gamma_1+\gamma_2$ for diatomic ions), incident with equal velocities. The parameter commonly used to characterize the effect is the ratio $R=\gamma_m/\gamma_a$. Deviations in the values of $R$ below unity have been reported.[3]

More recently, a molecular effect has been observed also in transmission experiments.[5–7] In this case the authors present an explanation where only the electrons ejected downstream in the beam direction are considered, and the effect is attributed to oscillatory wake effects on the target electron density following the projectile. We consider this explanation unsatisfactory in light of other experimental evidence and theoretical arguments.

The existence of wake effects in the dynamics of ion-cluster transmission in solids, and in the energy loss of the emerging ions, has been studied by several authors[8–13] both theoretically and experimentally. These studies provide a good framework to analyze the possibility of a real oscillatory behavior in the SEE yield. We find that in the conditions of the transmission experiments mentioned above, no oscillatory behavior in the electron emission can be expected, and we discuss a more plausible explanation for the results.

## II. WAKE EFFECTS

There are at least two important theoretical arguments that preclude a consistent description of an oscillatory behavior in SEE in terms of plasma wakes, as reported in Refs. 5–7.

The first one refers to the phenomenon of plasmon damping. The plasma resonance for carbon can be represented by a frequency $\omega_p=23$ eV/$\hbar$ and a damping constant $\gamma=11$ eV/$\hbar$ (Ref. 14). The calculation of the wake potential was given earlier by Neufeld and Ritchie,[8] and it was more recently reviewed by Echenique, Ritchie, and Brandt;[9] they obtain the following expression:

$$\Phi_w(\rho,z)=\frac{2Ze\omega_p}{v}\sin\left[\frac{\omega_p z}{v}\right]K_0\left[\frac{\omega_p\rho}{v},\frac{v}{v_F}\right]\exp\left[\frac{\gamma z}{2v}\right] \tag{1}$$

for points $z$ behind the ion position ($z<0$). The function $K_0(x,y)$ is defined in Ref. 9.

In Fig. 1 we show the values of the wake potential $\Phi_w$ and electric field $\mathbf{E}_w=-\nabla\Phi_w$ for points along the ion trajectory ($\rho=0$) and for $z<0$. It can be observed that the width of the plasma resonance produces a strong damping of the wake potential. This is in disagreement with the results shown in Ref. 5, where the damping is quite small.

Moreover, it can be shown that if an average over a uniform angular distribution is performed, to represent the case of molecular ions incident with random orientations, the interference effects are further washed out for internuclear distances $r_{12}>v/\omega_p$ (Ref. 12).

The second argument refers to the threshold for

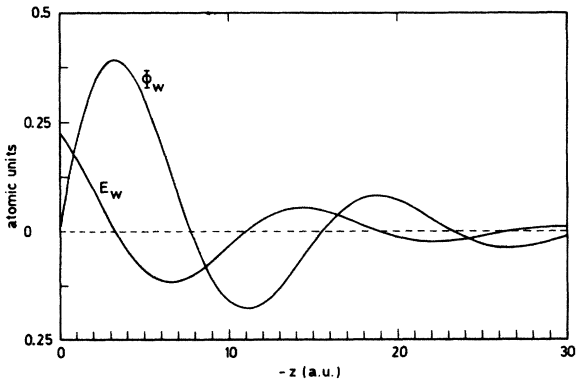


FIG. 1. Oscillatory dependence of the wake potential $\Phi_w$ and electric field $\mathbf{E}_w=-\nabla\Phi_w$ for points along the ion trajectory ($\rho=0$, $z<0$) in atomic units. Calculations correspond to 100-keV/amu ions in carbon foils.

 

plasmon excitation. A charged particle moving with velocity $v$ can produce excitations with phase velocities $\omega/k \leq v$. Since plasmon excitation is well defined only for wave vectors $k$ smaller than a given value $k_c$ (i.e., wavelengths longer than $2\pi/k_c$), it may be shown that a threshold velocity $v_{\rm th} \cong \omega_p/k_c$ is needed to induce collective excitations.[15] The value of $v_{\rm th}$ can be calculated using Lindhard's dielectric function $\epsilon(u,z)$, where $u=\omega/kv_F$ and $z=k/2k_F$. The plasma resonance is given by the condition $\epsilon_1(u,z)=0$, and the threshold for plasmon excitation is determined by $\epsilon_1(u_c,u_c-1)=0$. This yields the relation[16]

$$\frac{1}{(u_c-1)^2}\left[u_c \ln\left[\frac{u_c}{u_c-1}\right]-1\right]=2\pi v_F \,, \tag{2}$$

and the threshold velocity is determined by $v_{\rm th}=u_c v_F$.

For carbon targets we take $v_F \cong 1.2$ a.u., and obtain $v_{\rm th}=1.6$ a.u. This corresponds to a minimum energy of 60 keV/amu. The main effects shown in Refs. 5–7 occur at energies below this value. Hence, they cannot be attributed to wake phenomena.

## III. MULTIPLE SCATTERING EFFECTS

Since multiple scattering modifies the distribution of the internuclear distances between the molecular fragments during their transit through the solid, a discussion of these effects would be important for any analysis of processes depending on the internuclear distance at the exit.

To give only a short description of the effects of multiple scattering on the dynamics of an ion cluster, we consider the motion of ions of C and O resulting from the incidence of $CO^+$ ions on solid foils, and we make the following simplifying assumptions:

(a) The average distance $r_{12}$, between the C and O ions traversing a foil of thickness $t$, is calculated using a screened interaction of the form

$$V(r)=Z(\mathrm{O})Z(\mathrm{C})r^{-1}\exp(-r/a) \,, \tag{3}$$

where $Z(\mathrm{C})$ and $Z(\mathrm{O})$ are effective ion charges, and $a=\alpha v/\omega_p$, with $\alpha \cong 1$.

(b) The uncertainty in the distance between the nuclei proceeds from the multiple scattering of the fragments according to the expression

$$\Delta r_{12}=[r_{1/2}^2(\mathrm{C})+r_{1/2}^2(\mathrm{O})]^{1/2} \,, \tag{4}$$

where $r_{1/2}(Z)$ is the half-width at half maximum lateral deviation of nucleus $Z$ due to multiple scattering[17] (in particular, we notice that this would apply if multiple scattering acts independently on each nucleus and incoherently of the Coulomb force).

Figure 2 shows the distances $r_{12}$ between the C-O fragments as functions of the dwell time, for velocities $v=0.5$, 1.5, and 2.5 a.u. Calculations of $r_{12}$ and $\Delta r_{12}$ are plotted separately. The multiple scattering uncertainties $\Delta r_{12}$ were calculated according to Ref. 17, using Lenz-Jensen interatomic potentials.

We can see that multiple scattering effects are weakly dependent on the ion velocity, while the Coulomb explosion shows a much greater sensitivity below $v=1.5$ a.u. In any case, for dwell times larger than 15 fs, a considerable uncertainty in the determination of the C-O separation due to multiple scattering can be expected.

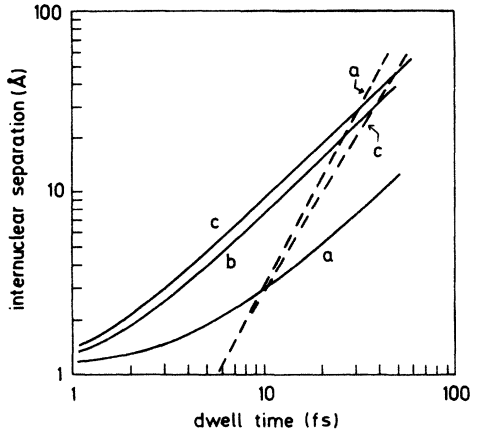


FIG. 2. Internuclear distance $r_{12}$ (solid lines) and statistical uncertainties $\Delta r_{12}$ (dashed lines) between the C-O fragments as functions of the dwell time, for velocities (a) $v=0.5$, (b) $v=1.5$, and (c) $v=2.5$ a.u.

From these calculations we can assess the importance of multiple scattering effects for the analysis of previous experiments dealing with molecular ions in solid foils. In particular, for the experiments of Refs. 5–7, our calculation shows that for those measurements carried out with foil thickness greater than 8 $\mu$g/cm$^2$ the uncertainty $\Delta r_{12}$ in the internuclear distance becomes comparable or even greater than $r_{12}$. This would produce a significant smearing of the wake potential acting at the exit.

## IV. AN ALTERNATIVE EXPLANATION

In Fig. 3 we show the ratios of electron yields, $R=\gamma(\mathrm{CO}^+)/[\gamma(\mathrm{C}^+)+\gamma(\mathrm{O}^+)]$ corresponding to molecular- and atomic-ion bombardment of carbon foils, using the results reported in Fig. 7 of Ref. 6. A common

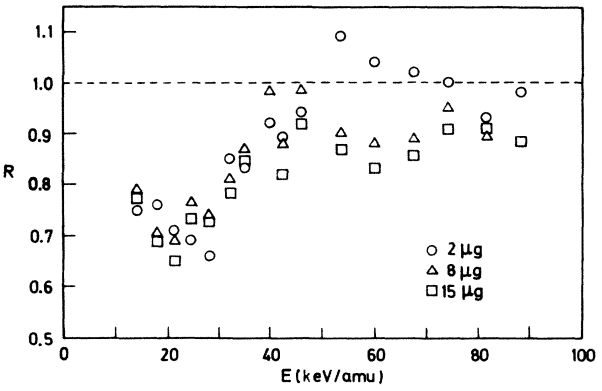


FIG. 3. Ratios of secondary-electron yields, $R=\gamma(\mathrm{CO}^+)/[\gamma(\mathrm{C}^+)+\gamma(\mathrm{O}^+)]$ corresponding to molecular and atomic bombardment of carbon foils, according to the results of Ref. 6.

feature in the data is a gradual increase in the value of $R$ for energies between approximately 20 ad 50 keV/amu, *almost independently of the foil thickness.* The data for higher energies do not show a clear behavior, they rather approach a value close to unity (with a maximum spread of $\pm 15\%$).

We stress that this gradual increase of $R$ in this energy range is the main effect observed in these experiments. The oscillatory behavior reported in Refs. 5–7 could only be obtained through the assignment of *ad hoc* phase shifts to each set of data.

Our alternative explanation of this effect is based on earlier observations of molecular effects in the energy loss of ion clusters transmitted through thin solid foils at various energies,[11–13] and on previous studies of molecular effects in SEE in various velocity ranges.[1–4]

As is known from earlier work,[11,12] the vicinage effect in the energy loss of swift ion clusters becomes positive ($R > 1$) for rather large velocities ($v > 1$ a.u.) and for internuclear distances $r_{12} \lesssim v/\omega_p$, due to interferences of long-wavelength plasma waves. That is, the velocity range where manifestations of wake effects can be important. At lower velocities (namely $v < v_{th} = 1.6$ a.u. for C) the dominant mechanism for energy loss is the excitation of single electrons. For low velocities and for the range of internuclear distances probed in the experiments, the interference in single excitations gives a negative effect on the energy loss[12,13] (i.e., $R < 1$). The proportionality between energy loss and SEE (Refs. 18 and 6) could then be used to predict a similar effect in SEE. This would be in qualitative agreement with the behavior shown in Fig. 3. Actually, the reduction of the energy loss alone ($\sim 15\%$) would only partially explain the larger effects in SEE shown in Fig. 3.

We must then consider other possible contributions to this effect. To this end we turn our attention to processes occurring when the molecular ions enter the foil. Obviously, the initial charge corresponding to the incidence of $CO^+$ ions is smaller than for the impact of separate $C^+$ and $O^+$ ions; therefore, a reduction of the yield is expected. Previous observations of molecular effects in SEE from a single surface[3,4] are consistent with a picture based on reduced energy loss and initial-charge effects. In fact, from the results of Ref. 3 we find values of $R$ of about 12–40% below unity, which are indeed comparable with the effect shown in Fig. 3. In addition, at even lower velocities, where potential emission dominates, other experiments also yield a reduced emission for molecular ions.[2] Therefore, processes occurring at the *entrance surface* of the foil may be the most important in determining the molecular effect. This should be taken into account in the analysis of beam-foil experiments, since so far these experiments have actually measured the total emission from both surfaces of the foil.

In conclusion, we consider that theoretical and experimental evidences do not support the existence of an oscillatory behavior in the SEE of molecular ions, at least under the conditions of previous beam-foil experiments. The only neat molecular effect seems to be that of a reduction in the $R$ values for low velocities, an effect that can be explained without recourse to wake phenomena. In addition, we expect that much of this effect should occur at the entrance surface. We consider that further experimental evidences could still be obtained from separate measurements of the emission in the forward and backward directions of the foil.

## ACKNOWLEDGMENTS

This work was supported in part by the International Atomic Energy Agency and Secretaría de Ciencia y Tecnología of Argentina.

[1] A. G. Hill, W. W. Buechner, J. S. Clark, and J. B. Fisk, Phys. Rev. **55**, 463 (1939); L. N. Large, Proc. Phys. Soc. London **81**, 1101 (1963).

[2] F. M. Propst and E. Luscher, Phys. Rev. **132**, 1037 (1963); A. A. Dorozhkin, A. N. Mishin, and N. N. Petrov, Bull. Acad. Sci. USSR, Phys. Ser. **38**, 60 (1974).

[3] R. A. Baragiola, E. V. Alonso, O. Auciello, J. Ferrón, G. Lantschner, and A. Oliva Florio, Phys. Lett. **67A**, 211 (1978).

[4] D. Hasselkamp, S. Hippler, and A. Scharmann, Nucl. Instrum. Methods **B2**, 475 (1984).

[5] H. J. Frischkorn, K. O. Groeneveld, P. Koschar, R. Latz, and J. Schader, Phys. Rev. Lett. **49**, 1671 (1982).

[6] H. J. Frischkorn and K. O. Groeneveld, Phys. Scr. **T6**, 89 (1983).

[7] H. J. Frischkorn, P. Koschar, J. Kemmler, R. Latz, J. Schader, and K. O. Groeneveld, Nucl. Instrum. Methods **B2**, 35 (1984).

[8] J. Neufeld and R. H. Ritchie, Phys. Rev. **98**, 1632 (1955).

[9] P. M. Echenique, R. H. Ritchie, and W. Brandt, Phys. Rev. B **20**, 2567 (1979).

[10] Z. Vager, D. S. Gemmell, and B. J. Zabransky, Phys. Rev. A **14**, 638 (1976); D. S. Gemmell, Nucl. Instrum. Methods **194**, 255 (1982).

[11] W. Brandt, A. Ratkowsky, and R. H. Ritchie, Phys. Rev. Lett. **33**, 1329 (1974); W. Brandt and R. H. Ritchie, Nucl. Instrum. Methods **132**, 43 (1976).

[12] N. R. Arista and V. H. Ponce, J. Phys. C **8**, L188 (1975); N. R. Arista, Phys. Rev. B **18**, 1 (1978).

[13] J. C. Eckardt, G. Lantschner, N. R. Arista, and R. A. Baragiola, J. Phys. C **11**, L851 (1978); J. Steinbeck and K. Dettmann, *ibid.* **11**, 2907 (1978).

[14] H.-J. Hagemann, W. Gudat, and C. Kunz (unpublished); J. Opt. Soc. Am. **65**, 742 (1975).

[15] J. Lindhard, K. Dan. Vidensk. Selsk., Mat.-Fys. Medd. **28**, No. 8 (1954); J. Lindhard and A. Winther, *ibid.* **34**, No. 4 (1964).

[16] C. Erginsoy, Brookhaven National Laboratory Report No. BNL-50083 (1968) (unpublished), p. 30.

[17] A. D. Marwick and P. Sigmund, Nucl. Instrum. Methods **126**, 317 (1975).

[18] R. A. Baragiola, E. V. Alonso, and A. Oliva Florio, Phys. Rev. B **19**, 121 (1979).