Investigation of the interference effect in the case of low energy electron emission from O_2 in collisions with fast bare C-ions

2012 J. Phys. B: At. Mol. Opt. Phys. 45 215207

(http://iopscience.iop.org/0953-4075/45/21/215207)

IOP Publishing | Journal of Physics B: Atomic, Molecular and Optical Physics

J. Phys. B: At. Mol. Opt. Phys. **45** (2012) 215207 (6pp) | doi:10.1088/0953-4075/45/21/215207

Investigation of the interference effect in the case of low energy electron emission from O_2 in collisions with fast bare C-ions

Saikat Nandi[1], **A N Agnihotri**[1], **C A Tachino**[2], **R D Rivarola**[2], **F Martín**[3,4] **and Lokesh C Tribedi**[1]

[1] Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai 400 005, India
[2] Institute de Fisica Rosario (CONICET-UNR), Universidad Nacional de Rosario, 2000 Rosario, Argentina
[3] Departamento de Química, Modulo 13, Universidad Autónoma de Madrid, E-28049 Madrid, Spain
[4] Instituto Madrileño de Estudios Avanzados en Nanociencia, Cantoblanco, E-28049 Madrid, Spain

E-mail: lokesh@tifr.res.in

Received 5 May 2012, in final form 8 August 2012
Published 1 October 2012

Abstract

We have measured the double differential cross sections (DDCSs) for low energy electron emission from O_2 under the impact of 51 MeV bare carbon ions. This study is aimed at investigating the Young-type interference in electron emission from a multi-electronic diatomic molecule. The DDCS spectra, differential in emission energy and angle, are compared with state-of-the-art continuum distorted wave-eikonal initial state (CDW-EIS) calculations. The DDCS ratios (i.e. O_2/2O) do not produce any oscillatory behaviour due to the interference effect unlike that observed in the case of H_2. The forward–backward angular asymmetry parameter, deduced from the measured DDCS values, is found to be a monotonically increasing function of electron velocity and does not show any oscillation. These observations are in qualitative agreement with the prediction of the molecular CDW-EIS model which uses a linear combination of atomic orbitals. The apparent absence of the oscillation in the spectra is qualitatively explained in terms of cancellation of contributions arising from different molecular orbitals.

(Some figures may appear in colour only in the online journal)

1. Introduction

Light waves, when made to pass through two slits of dimension comparable to wavelength of the incident light, interfere with each other to produce alternate bright and dark fringes known as the interference pattern. The two slits act as two coherent sources of light. Similarly, electrons from coherent sources can also interfere with each other to produce interference patterns due to their wave nature [1]. In this regard, the diatomic molecule acts as a natural double slit. Cohen and Fano [2] predicted the existence of the interference effect for photoionization in diatomic molecules. Jain and Khare [3] and Jain *et al* [4] showed theoretically the existence of such oscillations in the cross-sectional spectrum of electron elastic scattering from a hydrogen molecule. Since then, there has been significant progress in the study of interference oscillation in the case of the H_2 molecule, both experimentally [5–11] and theoretically [12–16]. An angular dependence has also been observed in the frequency of the interference oscillation [17, 18]. Moreover, the existence of a double-frequency component in the oscillatory structure has also been debated [19–21]. There are several different ways to deduce the interference oscillation from the measured double differential cross sections (DDCSs). One of the ways is to look for the oscillatory structure in the molecular DDCS spectrum in comparison to the atomic one [5, 6, 22]. In such a case, the atomic DDCS data are obtained, experimentally [6, 23] or theoretically. Besides, the asymmetry parameter quantifying the forward–backward angular asymmetry can also be used to detect the primary or first-order oscillation. The asymmetry parameter, which

Table 1. Measured values of the DDCS in units of Mb eV^{-1} sr^{-1} at different angles. Maximum possible error in the data is about 22%.

Energy (eV)	30°	45°	60°	75°	90°	105°	120°	135°	150°
11	1.3722	0.9370	0.9902	1.2821	1.1413	0.5855	0.4453	0.7745	0.4327
15	0.9577	0.6054	0.6755	0.9855	0.7970	0.4249	0.3546	0.3685	0.2521
21	0.6047	0.4020	0.4479	0.6305	0.5633	0.2797	0.1983	0.2717	0.1592
25	0.4826	0.3388	0.3454	0.4894	0.3611	0.2015	0.1668	0.1799	0.1009
35	0.3133	0.2402	0.2327	0.3131	0.2357	0.0938	0.0961	0.1035	0.0507
40	0.2978	0.1898	0.1860	0.2694	0.2356	0.1276	0.0786	0.0863	0.0523
60	0.1688	0.1138	0.1320	0.1997	0.1534	0.0699	0.0408	0.0381	0.0242
80	0.1204	0.0806	0.1041	0.1545	0.1182	0.0457	0.0250	0.0254	0.0145
100	0.0702	0.0503	0.0705	0.1276	0.0851	0.0346	0.0173	0.0160	0.0087
140	0.0462	0.0351	0.0301	0.0835	0.0538	0.0182	0.0080	0.0073	0.0051
200	0.0212	0.0198	0.0278	0.0543	0.0288	0.0078	0.0033	0.0040	0.0018
260	0.0124	0.0108	0.0167	0.0320	0.0182	0.0031	0.0022	0.0013	0.0014
300	0.0106	0.0095	0.0113	0.0262	0.0128	0.0031	0.0011	0.0013	0.0013
340	0.0084	0.0078	0.0111	0.0235	0.0091	0.0021	0.0010	0.0007	0.0004
400	0.0050	0.0054	0.0085	0.0163	0.0055	0.0020	0.0008	0.0011	0.0006
440	0.0112	0.0083	0.0096	0.0209	0.0088	0.0043	0.0032	0.0049	0.0036
480	0.0129	0.0118	0.0150	0.0230	0.0106	0.0070	0.0055	0.0083	0.0067
500	0.0103	0.0086	0.0097	0.0159	0.0093	0.0045	0.0031	0.0052	0.0037
540	0.0039	0.0031	0.0055	0.0109	0.0025	0.0011	0.0008	0.0006	0.0006
600	0.0019	0.0018	0.0040	0.0084	0.0012	0.0004	0.0002	0.0006	0.0001

is independent of any atomic cross-sectional data, shows the prominent oscillatory structure due to Young-type double slit interference in the case of the hydrogen molecule [24]. Although such studies exist for H_2, multi-electronic systems are less discussed in recent developments, except for a few experimental works [25, 26] using N_2 and O_2 as targets. Also, Tachino *et al* [27] theoretically investigated the possibility of observing the interference effect in the case of the proton impact ionization of N_2. In this work, we have studied the ionization of the oxygen molecule under the impact of fast bare carbon ions. The DDCS values for the O_2 molecule have been divided by those for atomic oxygen, obtained theoretically, in order to derive any primary oscillation. The asymmetry parameter has also been derived and shown to have no notable oscillation. The experimentally obtained DDCS values have further been compared with the continuum distorted wave-eikonal initial state (CDW-EIS) model calculations.

2. Experimental assembly

The experiments were performed with 4.25 MeV u^{-1} C^{6+} ions available from a 14 MV Pelletron accelerator facility at TIFR, Mumbai, India. The experimental setup, used in the present investigation, has already been described in [28]. In brief, it consists of a high vacuum scattering chamber equipped with a motorized turntable and a hemispherical electrostatic analyser [29] for the measurement of the angular and energy distributions of ejected electrons. The scattering chamber was flooded with molecular oxygen as the target gas at a pressure of 0.15 mTorr. The number of electrons at different energies, ejected in a particular direction, were energy analysed in the hemispherical analyser and detected using a channel electron multiplier. The energy dependence of the DDCS was investigated for nine different emission angles, namely 30°, 45°, 60°, 75°, 90°, 105°, 120°, 135° and 150°. The error due to the counting statistics depends on the emission angle as well as the emission energy. For example, the error was higher in the case of large backward angles and also, for large emission energies. In such cases, the maximum statistical error could be about 10–15%. The uncertainty in the gas pressure was about 6–7%. Overall, the maximum absolute error in the data, presented here, is about 20–22%.

3. DDCS distribution at the fixed emission angle

Typical numerical values for the measured DDCS have been shown in table 1. The absolute electron DDCS as shown in figure 1 decreases over three or four orders of magnitude as the energy of the emitted electrons increases between 10 and 600 eV. The emitted electrons in the low energy region are generally produced in soft collisions between the projectiles and the target atoms. These electrons were produced in collisions involving a high impact parameter and hence, are emitted with high cross sections. The experimental data were compared with the theoretical calculations using the CDW-EIS model for multi-electronic, diatomic targets [27].

A simple molecular representation for the triplet initial molecular state was used for the theoretical calculations. Contributions from different alpha and beta orbitals were considered, according to the corresponding electron spin character. For each molecular orbital (MO) of the target, the corresponding initial bound wavefunction was written as a linear combination of Slater-type orbitals (STOs) centred on each nucleus of the diatomic molecule. The optimization of the STO characteristic exponents was done by employing Gaussian 98 [30]. In particular, these values were calculated by using a minimal STO-6G basis set within the Hartree–Fock (HF) approximation, where each STO is represented as a linear combination of six Gaussian-type orbitals (GTOs). The equilibrium internuclear distance and the orbital energies for each MO of the target were calculated by employing a larger unrestricted HF 6-311G* basis set. DDCSs were

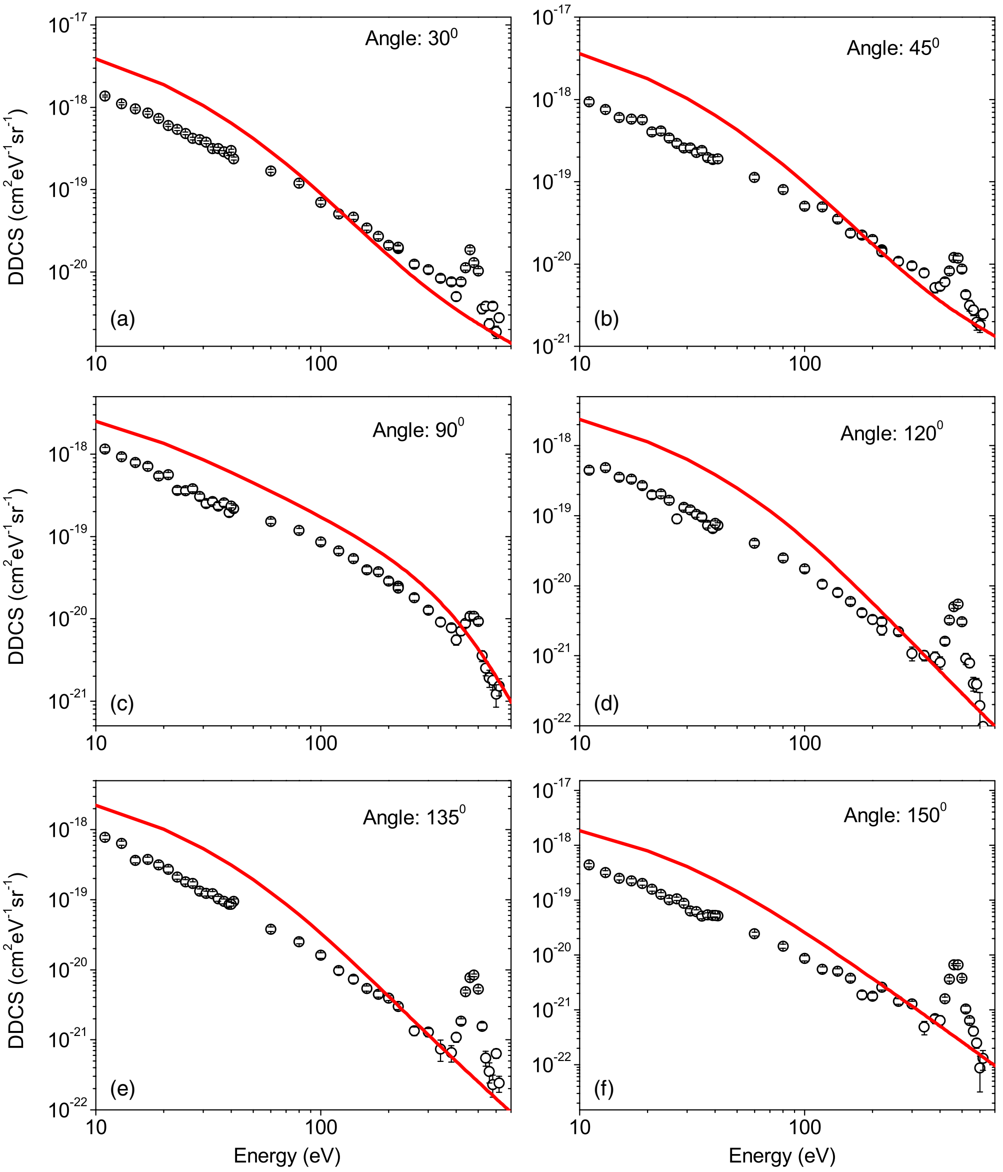

Figure 1. The absolute electron DDCS for emission angles 30°, 45°, 90°, 120°, 135° and 150°. The solid line in each plot corresponds to the prediction of the CDW-EIS model.

calculated in the CDW-EIS model by integrating the post-form of the scattering matrix element, over the transverse momentum transfer and averaging over the molecular solid angle. DDCS for the complete molecule was obtained as a linear combination of partial DDCSs corresponding to each MO.

The overall shapes of the DDCS spectra, obtained for different emission angles, have been qualitatively well reproduced by the CDW-EIS model. However, the theoretical values seem to overestimate the data everywhere, except in some regions. Large deviation of the experimental data from theoretical values can be observed in the low energy region. The theoretical values exceed the data at the lowest energy by a factor of about 2–4, for various angles. For energies more than 100 eV, a quantitative agreement can be found for the extreme forward and extreme backward angles (see figures 1(a), (b), (e) and (f)). The pronounced peak around 480 eV corresponds to the K-LL Auger line for oxygen. The theoretical model does not take into account the Auger emission process which is the cause for apparent large quantitative deviations in the higher energy (>400 eV) region.

4. Deducing the interference oscillation

4.1. DDCS ratios between molecular and atomic targets

The effect of interference is generally too small to be observed accurately from the DDCS spectrum itself. So, it becomes necessary to divide the DDCS values obtained from the molecular oxygen with those obtained for atomic oxygen under an identical condition. Here, we have divided our experimental DDCS values as obtained for the collision system 4.25 MeV u^{-1} C^{6+}+O$_2$ by two times the DDCS values for atomic oxygen target (2O). It should be noted that, in the absence of the experimental DDCS for atomic O, it was calculated theoretically using the CDW-EIS model. Figure 2 shows such

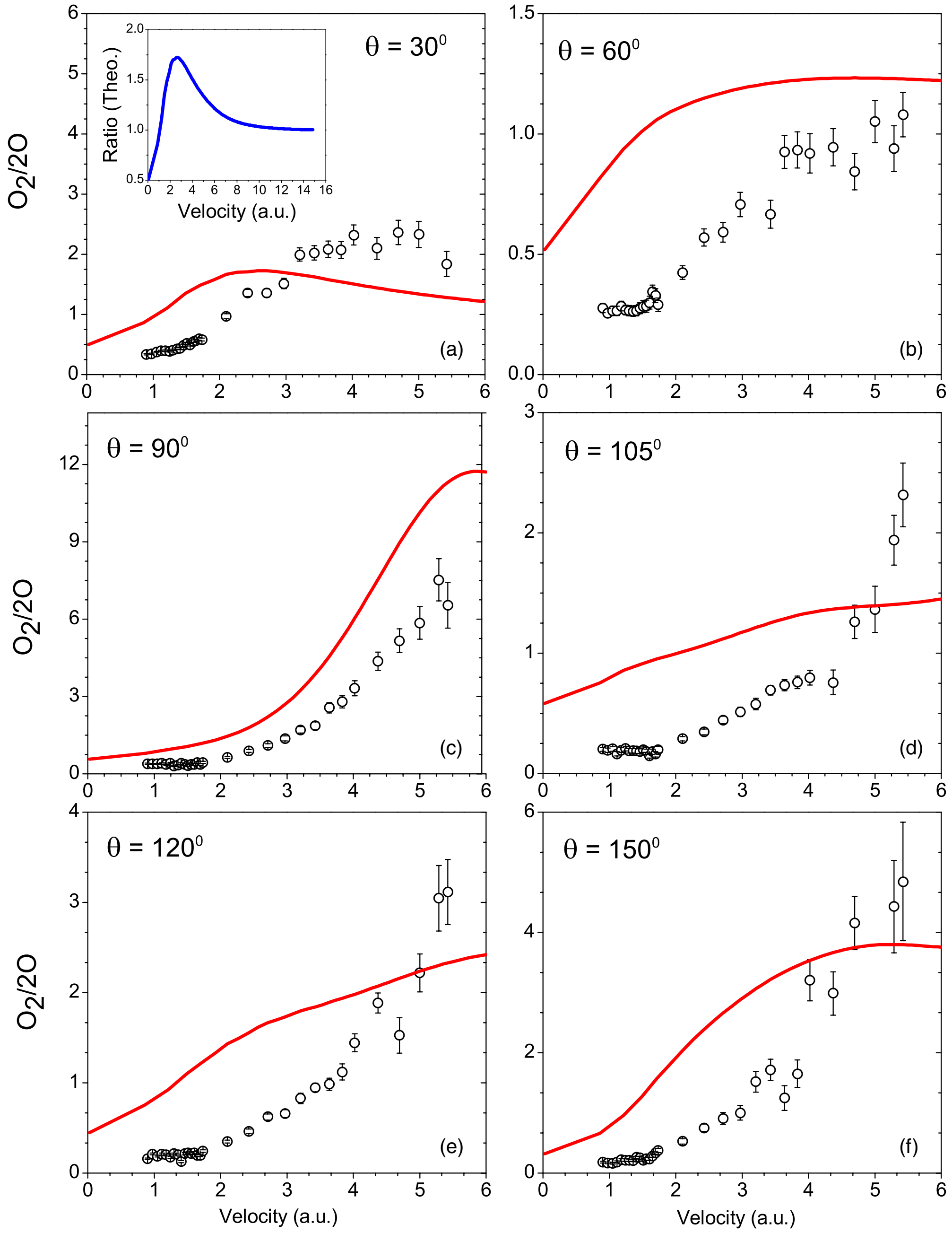

Figure 2. The ratio of molecular DDCS (for O_2) from the experiment to atomic DDCS (for O) from the CDW-EIS model at different angles, such as 30°, 60°, 90°, 105°, 120° and 150°. The solid line in each plot corresponds to the theoretical calculations using the CDW-EIS model. The inset in (a) corresponds to the theoretical values of the DDCS ratios over a velocity range of 0–15 au.

plots of DDCS ratios (i.e. $O_2/2O$) for different emission angles: 30°, 60°, 90°, 105°, 120° and 150°. It can be seen that there is no appreciable oscillation as a function of the electron velocity. In most of the cases the ratio shows a monotonically increasing behaviour. In the case of 30°, the experimental data show some kind of variation which cannot be termed as oscillation. The CDW-EIS model also predicts a peak-like structure at around 2.5 au, whereas in the experimental data the peak can be seen at around 4.5 au. The theoretically calculated values saturate beyond 5 au (see the inset of figure 2(a)).

The non-appearance of the oscillation can be explained by the CDW-EIS model and in terms of the MOs present in the O_2 molecule. The argument has already been presented in the recent theoretical work by Tachino *et al* [27] for the multi-orbital molecule N_2. In general, contributions from different alpha and beta orbitals, depending upon their electron spin character, are considered separately to calculate the DDCS values for each of the MOs. The contribution from each MO was divided by the corresponding contributions from the asymptotic atomic state of the orbital. It has been observed that, the DDCS ratio, obtained in this way, corresponding to different orbitals, shows a smooth oscillation as a function of the electron emission energy. For the alpha MO (as well as for the beta MO), the oscillations in all the internal orbitals (1s orbitals) are almost in phase with themselves. The same is true for the external orbitals (2s and 2p orbitals) also. However, the oscillations in the inner orbitals are phase shifted with respect to the outer orbitals. As all the contributions are added, it produces the graphs as shown in figure 2.

It was shown in an earlier work by Stolterfoht *et al* [5] that the interference oscillation for electron emission from a H_2 molecule is proportional to $\left(1+\frac{\sin(kd)}{kd}\right)$, where k is the electron velocity and d is the internuclear separation. Evidently, it can be seen that one complete sinusoidal oscillation corresponds to a variation from 0 to 2π for the argument kd. For O_2, we have $d = 2.28$ au and hence, the full oscillation is expected for k varying from 0 to 2.8 au (i.e. over 0–76 eV). We have plotted the experimental data over a velocity range of 0–5.5 au. However, no such behaviour could be seen in any emission angle. Winkworth *et al* [26] have reported a higher frequency component in the DDCS ratio, which was termed as second-order oscillation, whereas no primary oscillation was observed by these authors. In this work, neither primary nor secondary oscillation can be visible.

4.2. Angular asymmetry parameter

In order to quantify the forward–backward angular asymmetry, arising from the two-centre effect (TCE), one can introduce the parameter $\alpha(k)$ as [31]

$$\alpha(k,\theta) = \frac{\sigma^{(2)}(k,\theta) - \sigma^{(2)}(k,\pi-\theta)}{\sigma^{(2)}(k,\theta) + \sigma^{(2)}(k,\pi-\theta)}. \tag{1}$$

Now, the DDCSs, $(\sigma^{(2)}(k,\theta))$ for an electron with energy $\epsilon_k = \frac{k^2}{2}$ (au), to be ejected into the solid angle $d\Omega_k$, can be expanded in terms of the Legendre polynomials as follows:

$$\sigma^{(2)}(k,\theta) \equiv \frac{d^2\sigma}{d\epsilon_e\, d\Omega_e} = \sum_L \beta_L(k) P_L(\cos\theta), \tag{2}$$

where θ is the ejection angle with respect to the direction of the projectile beam. It has been shown by Fainstein *et al* [31] that only the values of L upto $L = 2$ contribute significantly to α-parameter. Hence, keeping the first three of terms in the above expression we obtain,

$$\alpha(k,\theta) = \frac{\sum_j \beta_{2j+1}(k)}{\sum_j \beta_{2j+1}(k)} \simeq \frac{\beta_1(k)}{\beta_0(k) + \beta_2(k)}. \tag{3}$$

Evidently, it gives the relative importance of the asymmetric contributions. Since the angular distribution varies slowly near 0 and π, we have used the DDCS measured at 30° and 150° to obtain the approximate value of $\alpha(k,\theta)$. Figure 3 shows the velocity dependence of the angular asymmetry parameter at $\theta = 30°$. In this case, the α-parameter varies from 0.5 to 0.9 over an energy range of 5–400 eV for ejected electrons. The theoretical values are scaled up by a factor of 1.42 (see the dashed line in figure 3) to display the qualitative agreement with experimental data. It should be noted that the amount of asymmetry as predicted by the CDW-EIS model is less (which varies from 0.2 to 0.8) compared to that obtained from the experiment. The asymmetry parameter is found to increase monotonically with electron velocity. This feature, in general, can be explained in terms of a TCE as has been described earlier [31, 32]. However, this is in contrast to the earlier result for H_2, wherein a prominent oscillatory behaviour was noted in the velocity dependence of the α-parameter [10, 24]. For H_2, this has been well understood in terms of the interference phenomenon. It was then shown [10] that using a Cohen–Fano type of oscillatory function [2], i.e. $\sin(kd)/(kd)$ in the expression for DDCS, one could reproduce the observed oscillation. Although oxygen is a diatomic molecule, the reason for the absence of oscillatory behaviour in the α-parameter can be attributed to the contributions of several MOs, as already explained in the last section. The experimental observation is in qualitative agreement with the theory, in general. However, one may note that a very weak oscillation is still preserved in the theoretical calculations.

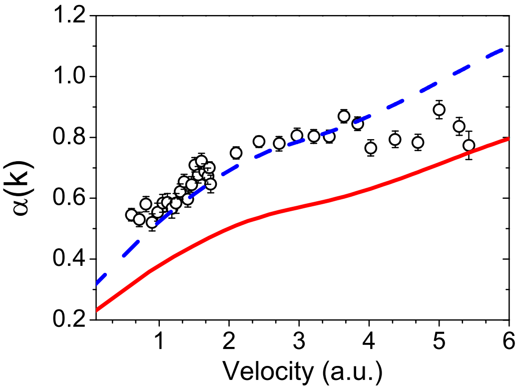

Figure 3. Velocity dependence of the angular asymmetry parameter. The solid line corresponds to the CDW-EIS calculation. The dashed line corresponds to theoretical values scaled up by a factor of 1.42.

5. Conclusions

We have studied low energy electron emission from the oxygen molecule in collisions with fast bare C-ions of energy 51 MeV. The main motivation of this work was to look for Young-type interference oscillation in the electron emission spectrum from the multi-electronic diatomic molecule. It has been shown in this work, both experimentally and theoretically, that there is no sign of any primary oscillation in the DDCS ratios of $O_2/2O$. This observation, which differs from the earlier report, has been explained by the CDW-EIS model in terms of a multi-orbital picture. Moreover, the forward–backward angular asymmetry parameter also does not show any prominent oscillation. The monotonically increasing behaviour, observed in the $\alpha(k)$-parameter, is well known in terms of the two-centre effect (TCE). The absolute values of the electron DDCS, for different electron energies and angles, are compared with the CDW-EIS model which shows certain deviation from the former, especially at low energies.

Acknowledgments

We would like to thank the Pelletron accelerator team for smooth operation of the machine. One of the authors (FM) acknowledges the financial support from the MICINN projects FIS2010-15127 and CSD 2007-00010. CAT, RDR and FM acknowledge the Programa de Cooperación Interuniversitaria e Investigación Científica entre España e Iberoamérica AECID project A2/039631/11. In addition, CAT and RDR acknowledge the Consejo Nacional de Investigaciones

Científicas y Técnicas de la República Argentina (PIP number 11220090101026).

References

[1] Feynman R P, Leighton R B and Sands M 1975 *Lectures on Physics* vol 3 (New Delhi: Narosa) p 1425
[2] Cohen H D and Fano U 1966 *Phys. Rev.* **150** 30
[3] Jain D K and Khare S P 1977 *Phys. Lett.* A **63** 237
[4] Jain A, Tripathi A N and Srivastava M K 1979 *Phys. Rev.* A **20** 2352
[5] Stolterfoht N *et al* 2001 *Phys. Rev. Lett.* **87** 023201
[6] Misra D, Kadhane U, Singh Y P, Tribedi L C, Fainstein P D and Richard P 2004 *Phys. Rev. Lett.* **92** 153201
[7] Tanis J A, Hossain S, Sulik B and Stolterfoht N 2005 *Phys. Rev. Lett.* **95** 079301
[8] Misra D, Kadhane U, Singh Y P, Tribedi L C, Fainstein P D and Richard P 2005 *Phys. Rev. Lett.* **95** 079302
[9] Frémont F, Hajaji A, Naja A, Leclercq C, Soret J, Tanis J A, Sulik B and Chesnel J-Y 2005 *Phys. Rev.* A **72** 050704
[10] Misra D, Kelkar A H, Kadhane U, Kumar A and Tribedi L C 2006 *Phys. Rev.* A **74** 060701
[11] Schmidt L P H, Schössler S, Afaneh F, Schöffler M, Stiebing K E, Schmidt-Böcking H and Dörner R 2008 *Phys. Rev. Lett.* **101** 173202
[12] Galassi M E, Rivarola R D, Fainstein P D and Stolterfoht N 2002 *Phys. Rev.* A **66** 052705
[13] Nagy L, Kocbach L, Póra K and Hansen J P 2002 *J. Phys. B: At. Mol. Opt. Phys.* **35** L453
[14] Galassi M E, Rivarola R D and Fainstein P D 2004 *Phys. Rev.* A **70** 032721
[15] Fojón O A, Fernández J, Palacios A, Rivarola R D and Martín F 2004 *J. Phys. B: At. Mol. Opt. Phys.* **37** 3035
[16] Fernández J, Fojón O, Palacios A and Martín F 2007 *Phys. Rev. Lett.* **98** 043005
[17] Tanis J A, Chesnel J-Y, Sulik B, Skogvall B, Sobocinski P, Cassimi A, Grandin J-P, Adoui L, Hennecart D and Stolterfoht N 2006 *Phys. Rev.* A **74** 022707
[18] Chatterjee S, Misra D, Kelkar A H, Fainstein P D and Tribedi L C 2010 *J. Phys. B: At. Mol. Opt. Phys.* **43** 125201
[19] Stolterfoht N, Sulik B, Skogvall B, Chesnel J Y, Frémont F, Hennecart D, Cassimi A, Adoui L, Hossain S and Tanis J A 2004 *Phys. Rev.* A **69** 012701
[20] Sisourat N, Caillat J, Dubois A and Fainstein P D 2007 *Phys. Rev.* A **76** 012718
[21] Misra D, Kelkar A H, Chatterjee S and Tribedi L C 2009 *Phys. Rev.* A **80** 062701
[22] Chatterjee S, Misra D, Kelkar A H, Tribedi L C, Stia C R, Fojón O A and Rivarola R D 2008 *Phys. Rev.* A **78** 052701
[23] Tribedi L C, Richard P, DeHaven W, Gulyás L, Gealy M W and Rudd M E 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** L369
[24] Misra D, Kelkar A H, Kadhane U, Kumar A, Singh Y P, Tribedi L C and Fainstein P D 2007 *Phys. Rev.* A **75** 052712
[25] Baran J L, Das S, Járái-Szábo F, Póra K, Nagy L and Tanis J A 2008 *Phys. Rev.* A **78** 012710
[26] Winkworth M, Fainstein P D, Galassi M E, Baran J L, Dassanayake B S, Das S, Kayani A and Tanis J A 2009 *Nucl. Instrum. Methods Phys. Res.* B **267** 373
[27] Tachino C A, Martín F and Rivarola R D 2011 *J. Phys. B: At. Mol. Opt. Phys.* **45** 025201
[28] Misra D, Thulasiram K V, Fernandes W, Kelkar A H, Kadhane U, Kumar A, Singh Y P, Gulyas L and Tribedi L C 2009 *Nucl. Instrum. Methods Phys. Res.* B **267** 157
[29] Gealy M W, Kerby G W III, Hsu Y-Y and Rudd M E 1995 *Phys. Rev.* A **51** 2247
[30] Frisch M J *et al* 1999 *Gaussian 98, Revision A11* (Pittsburgh, PA: Gaussian Inc.)
[31] Fainstein P D, Gulyás L, Martín F and Salin A 1996 *Phys. Rev.* A **53** 3243
[32] Tribedi L C, Richard P, Gulyás L and Rudd M E 2001 *Phys. Rev.* A **63** 062723