

# Electron capture in collisions of $\text{N}^{2+}$ and $\text{O}^{2+}$ ions with $\text{H}(1s)$ at low impact energies

P. Barragán, L. F. Errea, L. Méndez,\* I. Rabadán, and A. Riera

*Laboratorio Asociado al CIEMAT de Física Atómica y Molecular en Plasmas de Fusión.*

*Departamento de Química, Universidad Autónoma de Madrid, Madrid-28049, Spain*

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## Abstract

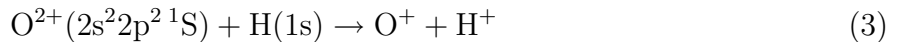
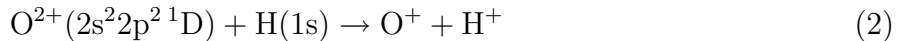
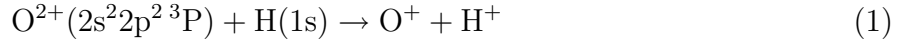
We present *ab initio* quantal calculations of electron capture cross sections for collisions of ground and metastable states of  $^{14}\text{N}^{2+}$  and  $^{16}\text{O}^{2+}$  ions with  $\text{H}(1s)$ , at collision energies  $10^{-2} < E < 10^2$  eV/amu. The calculation for  $\text{N}^{2+} + \text{H}$  updates the previous one of Barragán et al. (Phys. Rev. A 70, 022707 (2004)) at  $0.1 < E < 0.3$  eV/amu. Total cross sections for both systems show large values of about  $5 \times 10^{-15}$  cm<sup>2</sup> at  $E < 0.1$  eV/amu, where they exhibit resonant structures.

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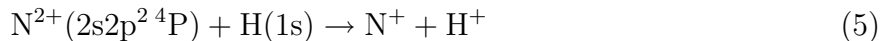
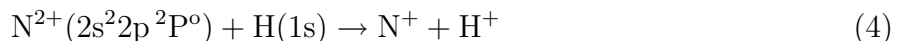
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\*To whom correspondence should be addressed: [l.mendez@uam.es](mailto:l.mendez@uam.es)

Electron capture (EC) in collisions of  $O^{2+}$  and  $N^{2+}$  ions with H are important processes in astrophysical (e.g. in planetary nebulae [1]) and controlled fusion plasmas, where state-selected cross sections for collisions of ions in both ground and metastable states are required to model edge plasma regions [2]. Since low impact energies are needed in these applications, we have extended to lower energies the semiclassical calculation of [3] for the EC reactions:



and we have recalculated and extended to lower energies the cross sections for reactions:



previously evaluated in ref. [4]. In the present work we focus on total cross sections. We study the shape of these cross sections at low energies, the presence of resonant structures and we discuss the limitations of previous calculations. The applications in fusion and astrophysics are presented in refs.[5] and [5] respectively.

In general, the calculation of EC cross sections at low energies requires the use of very precise molecular wavefunctions, and a quantal treatment for the dynamics, including reaction coordinates (see [7] and references therein), to ensure that the expansion fulfills the collision boundary conditions. Although several definitions of these coordinates have been proposed (see [8]), they have been only applied to single electron systems (see [9]), and the common reaction coordinate method is the only treatment employed for many-electron systems. In the present work we have defined the reaction coordinate in terms of the switching function of [10].

We have evaluated the molecular wavefunctions by applying a multireference configuration interaction (CI) treatment by means of the program MELD [11]. In this method, one constructs a basis of configurations by allowing single and double excitations from a set of reference configurations; these are antisymmetrized products of SCF molecular orbitals, which are linear combinations of Gaussian type orbitals. The gaussian basis and the set of reference configurations have been chosen (details can be found in [3, 4]) in order to

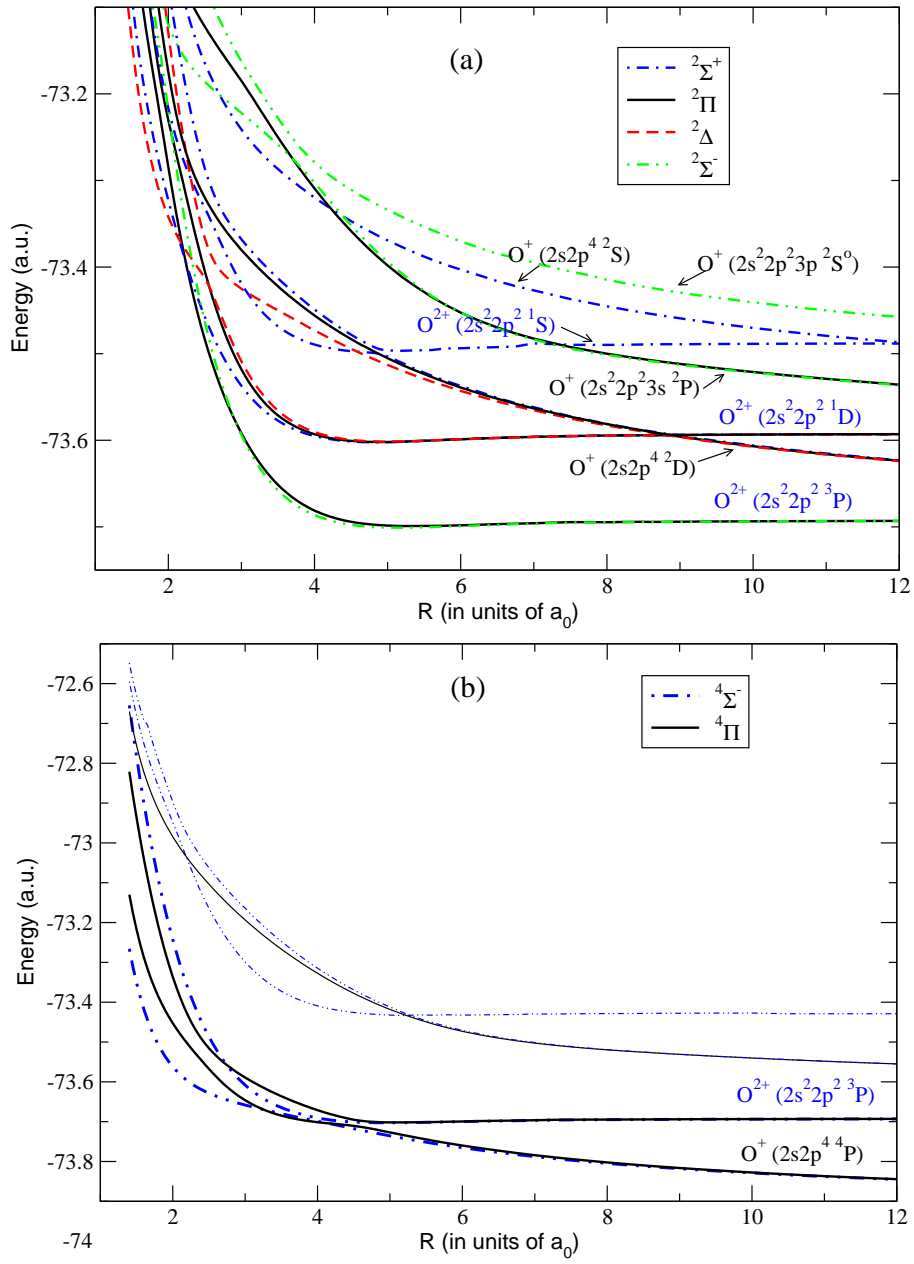


FIG. 1: (Color online) Potential energy curves of the molecular states of the  $\text{OH}^{2+}$  quasimolecule (a) Doublet states; (b) quadruplet states. Indicated in the figure are the states of the  $\text{O}^{2+}$  and  $\text{O}^+$  ions in which dissociate the molecular states in the limit  $R \rightarrow \infty$ .

ensure that the asymptotic energy differences differ in less than 0.2eV from the spectroscopic values (Physical Reference Data, National Institute of Standards and Technology; <http://physics.nist.gov>) for all the states involved. The dynamical couplings have been evaluated as explained in [12] and [13].

	O <sup>2+</sup> (2s <sup>2</sup> 2p <sup>2</sup> 3p)+H(1s)	N <sup>2+</sup> (2s <sup>2</sup> 2p 2p <sup>o</sup> )+H(1s)	
	Σ <sup>-</sup> - Σ <sup>-</sup>	Π - Π	Π - Π
$a$ (Hartree/a <sub>0</sub> )	0.0273	0.0466	0.0255
$H_{12}$ (Hartree)	0.00394	0.00757	0.00182
$R_0$ (a <sub>0</sub> )	3.971	4.570	6.439

TABLE I: Values of the parameters  $a$ ,  $H_{12}$  and  $R_0$  obtained by fitting the potential energy curves to a linear model near the avoided crossings.

In O<sup>2+</sup>+H collisions we have employed an expansion in terms of 29 molecular states whose energies are shown in fig. 1. The entrance channel of reaction (1), O<sup>2+</sup> (2s<sup>2</sup>2p<sup>2</sup> 3P) + H(1s), correlates to molecular states <sup>2,4</sup>Σ<sup>-</sup> and <sup>2,4</sup>Π, and, as has been explained in previous works [3, 14, 15], reaction (1) takes place mainly through transitions to the states <sup>4</sup>Σ<sup>-</sup>, <sup>4</sup>Π, dissociating into O<sup>+</sup> (2s2p<sup>4</sup> 4P) + H<sup>+</sup>, in the avoided crossings between the corresponding potential energy curves at  $R \simeq 4$  and 4.5 a<sub>0</sub>, respectively (see fig. 1b and table I). Since, in the doublet subsystem, the energy of the entrance channel does not pseudocross those of the capture channels, this subsystem is less important for reaction (1); however, reactions (2) and (3) take place through transitions between doublet molecular states.

To ensure the validity of our molecular treatment at low impact energies, we have recalculated the potential energy curves of the quadruplet states by employing the [4s3p2d1f] Gaussian basis set centered at the O nucleus of ref. [16], and increasing the precision by reducing the perturbative selection threshold down to  $5 \times 10^{-7}$  Hartree ( $\approx 1.4 \times 10^{-5}$  eV). This leads to an asymptotic energy difference between the channels O<sup>2+</sup> (2s<sup>2</sup>2p<sup>2</sup> 3P) and O<sup>+</sup> (2s2p<sup>4</sup> 4P) that differs in less than 0.18 eV from the experimental value. We have selected iteratively the reference space at each internuclear distance, which yields a total CI space of about 70,000 configurations at  $R < 2.0$  a<sub>0</sub>.

The cross section for reaction (1) is given by:

$$\sigma = 2/9[\sigma(^4\Sigma^-) + \sigma(^4\Pi_+) + \sigma(^4\Pi_-)] + 1/9[\sigma(^2\Sigma^+) + \sigma(^2\Pi_+) + \sigma(^2\Pi_-)] \quad (6)$$

where  $\sigma(i)$  are the capture cross sections for the collision with the molecular entrance channel of this symmetry and multiplicity, and the subindexes  $\pm$  indicate the symmetry of the  $\Pi$  wavefunctions under reflection in the collision plane. The EC total cross section is shown in figure 2, where we have included the semiclassical results of ref. [3] to illustrate the smooth

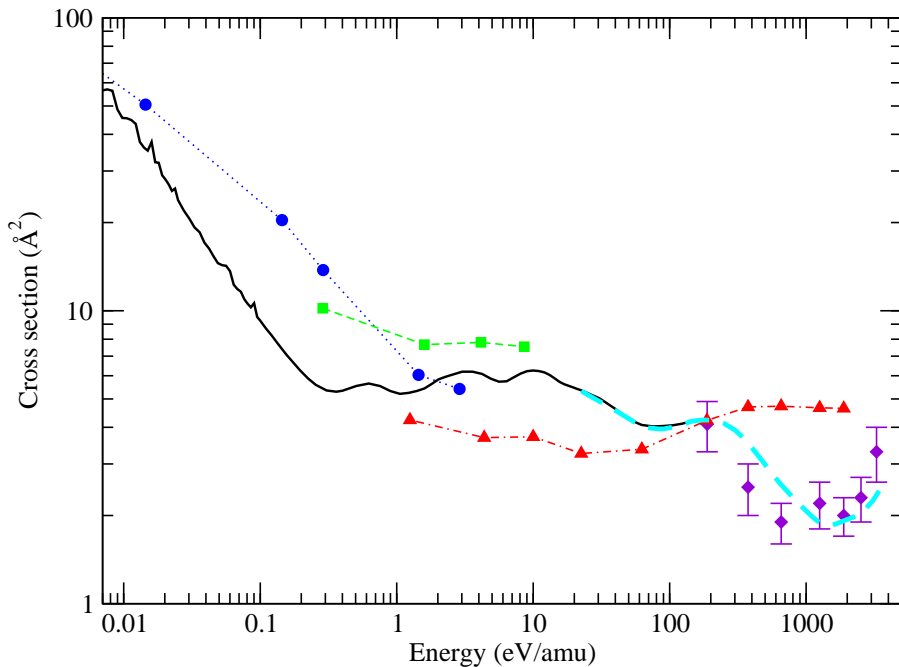


FIG. 2: (Color online) Total EC cross sections in  $O^{2+} (2s^2 2p^2 \ ^3P) + H(1s)$  collisions. Full line, present results; dashed line, semiclassical results of ref. [3];  $\blacksquare$ , quantal calculation of ref. [17];  $\bullet$ , quantal calculation of ref. [15];  $\blacktriangle$ , semiclassical results of ref. [18];  $\blacklozenge$ , experimental results of ref. [19]

joining of both calculations. To discard the possibility of spurious maxima at large  $R$ , we have also checked that the cross section of fig. 2 does not vary when the numerical entrance channel potentials are substituted at  $R > 12a_0$  by polarization-type expressions  $c_1 - c_2 R^{-4}$ . For  $E < 0.2\text{eV/amu}$ , the energy dependence of the EC cross section is similar to that found in the model calculation of [20] for EC in  $N^{3+} + H$  collisions, where the increase of the capture cross section eV can be approximately described by the Langevin model (see [21, 22]) and some spikes are noticeable, most of them due to shape resonances in the adiabatic entrance channel potential, as also found in the calculations of refs. [20, 23, 24], although the resonances are not clearly shown in our cross section because of the superposition of contributions from different initial molecular states. To further illustrate these resonances, we show in figure 3 the contribution  $\sigma(^4\Pi_+)$  to the capture cross section. In this figure we have added the values of the vibro-rotational quantum numbers  $(v, j)$  of the shape resonances; these have been obtained analyzing the contribution of each  $j$ -partial wave to

the total cross section and with the help of the program Level 7.7 [25]. As the energy decreases, the positions of the resonances in the cross section show significant shifts with respect to the energies of the quasibound states calculated with Level 7.7, probably due [26] to the interaction of the entrance channel with the capture channel dissociating into  $O^+(2s2p^4\ ^4P)$  (see fig. 1). In this respect, an indication of the importance of the non-adiabatic interaction is given by the peak at  $E \simeq 0.013\text{eV/amu}$ . To assign this peak, we have obtained a set of diabatic states by means of an unitary transformation [27] that cancels out the radial coupling between the molecular states of the basis set, which is the usual procedure in quantal calculations. Solving the radial Schrödinger equation with the diabatic potential of the entrance channel allows us to assign the peak to the (5,19) quasibound state of this potential.

The calculated EC cross section is smaller than that given by the simple Langevin model (it yields a constant value  $\sigma\sqrt{E} \approx 18.8\text{\AA}^2\text{ eV}^{1/2}$  in fig. 3), because the transition probability is smaller than 1, as assumed in that model for collisions with  $b < b_{\text{max}} = \sqrt{(2\alpha q^2/\mathcal{E})}$ , where  $\alpha$  is the H(1s) polarizability and  $\mathcal{E}$  is the centre of mass energy. To illustrate this, we have calculated the cross section  $\sigma(^4\Pi_+)$  by applying a simple model where the transition probability is estimated by using the Landau-Zener model, with the parameters of table I for the interaction  $H_{12}$ , the difference of energy slopes,  $a = d(H_{22} - H_{11})/dR$ , and the crossing point  $R_0$ . This yields (see e.g. [28]):

$$\sigma = 2\pi \int_0^{b_{\text{max}}} b P(b) db \approx 2\pi \int_0^{b_{\text{max}}} b 2p(1-p) db \quad (7)$$

with

$$p = \exp[-2\pi H_{12}^2/av_R] \quad (8)$$

and the radial velocity in the crossing point:

$$v_R = v \left( 1 - \frac{V_1(R_0)}{\mathcal{E}} - \frac{b^2}{R_0^2} \right)^{\frac{1}{2}} \quad (9)$$

has been calculated by employing the numerical value of the adiabatic entrance channel potential  $V_1(R_0)$ . The reasonable agreement of this model with the numerical values (fig. 3) indicates that the cross section is determined by  $b_{\text{max}}$  and the transition probability in the crossing region. Besides, as in ref. [22], the energy dependence of the Landau-Zener probability (8) qualitatively explains the relatively small deviation of the EC cross section from the  $E^{-1/2}$  behaviour.

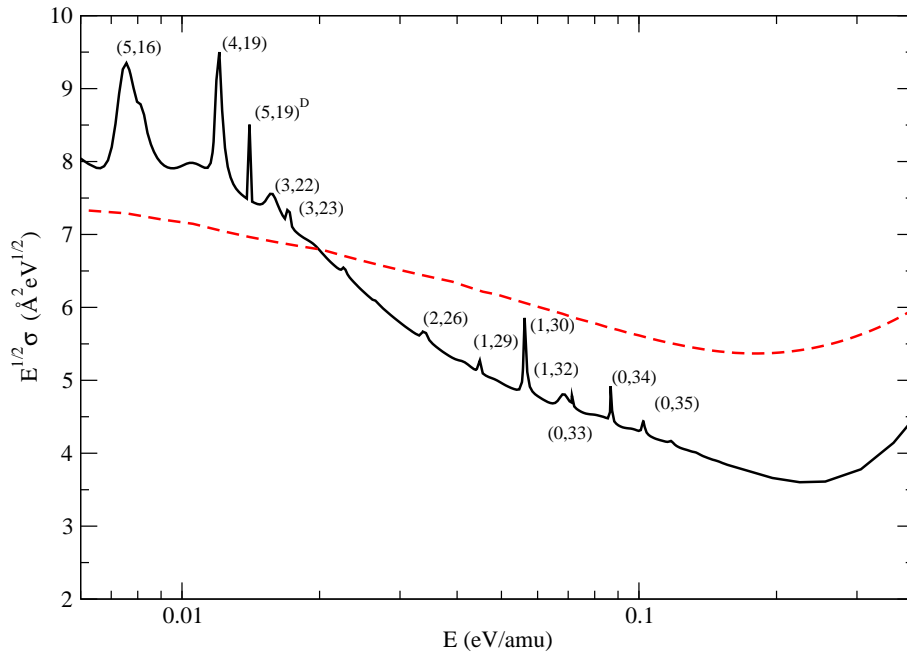


FIG. 3: (Color online) Contribution  $\sigma(^4\Pi_+)$  of the  $^4\Pi_+$  subsystem to the cross section for reaction (1). The labels  $(v, j)$  in the peaks are the vibrorotational quantum numbers of the resonant states of the adiabatic potential of the molecular entrance channel, with the exception of the resonance labelled with the superscript D, which corresponds to a quasibound state of the diabatic entrance channel. The dashed line is the result of applying a two-state semiclassical model (see text) .

At collision energies  $0.5 < E < 10\text{eV/amu}$ , the cross section for reaction (1) shows an oscillatory structure as a function of the energy, caused by an interference effect between transitions in the two avoided crossings at  $R \simeq 2.5$  and  $4.6 a_0$  in the  $^4\Pi$  subsystem. The differences between our cross sections and the previous ones of refs. [18] and [15] are due to the more precise molecular wavefunctions employed in the present work, as pointed out in [3].

The calculations for  $\text{N}^{2+} + \text{H}$  collisions (figures 4 and 5) have employed the 56-term basis set previously used in the quantal calculation of ref. [4], with the only difference of a finer grid of internuclear distances, as explained below. The mechanisms of reactions (4) and (5) have been discussed in detail by Barragán et al. [4]. Total cross section for reaction (4) is obtained from expression:

$$\sigma = 1/12[\sigma(^1\Sigma^+) + \sigma(^1\Pi_+) + \sigma(^1\Pi_-)] + 3/12[\sigma(^3\Sigma^+) + \sigma(^3\Pi_+) + \sigma(^3\Pi_-)] \quad (10)$$

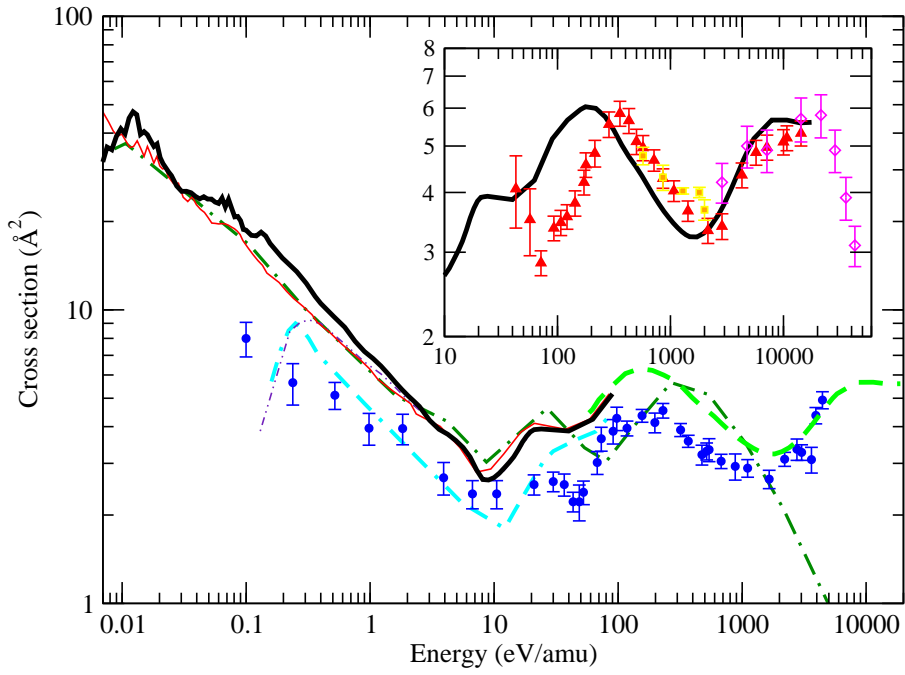


FIG. 4: (Color online) Total EC cross sections in  $N^{2+} (2s^22p \ ^2P^o) + H(1s)$  collisions. Solid lines, present calculation: Thick line, results for collisions with H, thin line, results for collisions with D. Experimental results:  $\square$ , [19];  $\diamond$ , [29];  $\blacktriangle$ , [30];  $\bullet$ , [31]. Previous theoretical results:  $\cdot - \cdot -$ , [32];  $\cdot - \cdot -$ , [33];  $- \cdot - \cdot -$ , quantal calculation of ref. [4];  $- \cdot - \cdot -$ , semiclassical calculation of ref. [4].

At low energies, this reaction takes place through transitions in the avoided crossing between the triplet states dissociating into  $N^{2+} (2s^22p \ ^2P^o) + H(1s)$  and  $N^+ (2s2p^3 \ ^3D^o) + H^+$ . Triplet and quintet molecular states are required to evaluate the cross section for reaction (5):

$$\sigma = 3/24[\sigma(^3\Sigma^-) + \sigma(^3\Pi_+) + \sigma(^3\Pi_-)] + 5/24[\sigma(^5\Sigma^-) + \sigma(^5\Pi_+) + \sigma(^5\Pi_-)] \quad (11)$$

and the main mechanism involves transitions from the molecular entrance channels to states correlating to  $N^+ (2s^22p3s \ ^3P^o) + H^+$  and  $N^+ (2s2p^3 \ ^3S^o) + H^+$ . As in  $O^{2+} + H$  collisions, the cross sections for both reactions reach similar values at low impact energies and show resonant structures (see figs. 4 and 5).

For  $E > 0.2\text{eV/amu}$ , the present results agree with the previous ones of ref. [4] and, in particular, the cross section for reaction (4) agrees with the semiclassical results. To further illustrate the comparison of our cross sections with the available experimental results, they are shown in the inset of figure 4 for  $E > 10 \text{ eV}$ . However, at  $E < 0.2\text{eV/amu}$ , the cross



sections reported in our previous work decrease rapidly and this behaviour disappears in the new calculation that uses a finer grid of internuclear distances. We have found that in the previous work, the diabatic potential of the entrance channel, obtained after interpolation and integration of the radial couplings, showed a small spurious maximum of about 0.1eV at large  $R$ , which gave rise to the rapid decrease of the cross section. A similar difficulty could explain the corresponding maximum in the calculation of ref. [33]. In this work we have checked our calculation, as explained above for  $O^{2+} + H$  collisions to ensure that our new results do not present this limitation in the energy range of fig. 4.

Our cross section for reaction (4) and those from previous calculations [32, 33] show a Langevin-type behavior  $\sigma \sim E^{-1/2}$  at low energies. As explained in [4], the differences between the theoretical results are mainly due to the accuracy of the energy gap between the energies of the entrance molecular channel and that dissociating into  $N^+(2s2p^3 \ ^3D^o)$ . In particular, the change of this gap from 0.079eV [33] to 0.099eV [4] leads to an increase of the capture cross section by a factor of 1.3 at  $E \approx 1\text{eV}/\text{amu}$ . Given the sensitivity of the cross section to the accuracy of the molecular data, we have checked our calculation by enlarging the Gaussian basis set of ref. [4]; we have added one s function with exponent  $\alpha = 0.0176$ , one p function ( $\alpha = 0.0176$ ) and two d functions ( $\alpha = 0.04, 8.25$ ), centered on the N nucleus; we have decontracted the f orbitals on the same nucleus, and we have added a contracted d function on the H nucleus taken from ref. [16]). We have also reduced the selection threshold in the CI step to  $7 \times 10^{-8}$ Hartree, which leads to a CI space that includes about 85,000 configurations in the avoided crossing region  $R \approx 6.5 \text{ a}_o$ . A test on the accuracy of our calculation is provided by the comparison between the electronic gradient matrix elements calculated numerically and by applying the relationship:

$$\langle i|\nabla|j \rangle = (E_j - E_i) \langle i|\mathbf{r}|j \rangle, \quad (12)$$

which is fulfilled by exact eigenfunctions of the electronic Hamiltonian. Our values for the  $Z$  component of the electronic gradient (the only non-vanishing) between the  $^3\Pi$  wavefunctions of the entrance and the main exit channels are  $0.0110 \text{ a}_o^{-1}$  (numerical) and  $0.0109 \text{ a}_o^{-1}$  (from eq. (12)) at  $R = 6.4\text{a}_o$ . Although the enlargement of the basis set yields energy decreases of about  $8 \times 10^{-2}\text{eV}$  in the avoided crossing region, the energy gap changes in less than  $3 \times 10^{-3}\text{eV}$ . In order to estimate the error in the capture cross sections associated to these changes in the energy gap we have employed the Landau-Zener model that leads to variations

of the cross section smaller than 10%.

The merged-beams experiment of ref. [31] reported a cross section smaller than the theoretical ones, and, in contrast to results for other collisions, the absolute value of the slope of the logarithmic  $\sigma - E$  plot (fig. 4) is smaller than the Langevin value 0.5 at  $E < 2\text{eV}/\text{amu}$ . In order to compare with these experimental values, we have evaluated the EC cross section for the reaction with Deuterium, which is slightly lower than that with H, as previously found for other collision systems in refs.[21, 34, 35]. The ratio  $\sigma_H/\sigma_D$  is smaller than 1.2 and decreases when the energy increases for  $E > 0.1\text{eV}/\text{amu}$ , while the simple Langevin model predicts a constant ratio of 1.4. These results qualitatively agree with the kinematic model of [34] at  $E > 0.1\text{eV}/\text{amu}$ , but not at lower energies where the semiclassical method of this model is probably not appropriate. It can be noted from fig. 4 that the decrease due to the isotope effect does not significantly improves the agreement with the experimental results. Another possible explanation of the difference with the experiment could be an inaccurate treatment of transitions to other molecular states. In particular transitions to molecular states dissociating into  $\text{N}^+(2s2p^3\ ^3\text{P}^\circ)$ , whose energies show avoided crossings with those of the molecular entrance channels at  $R \simeq 11\ a_0$ , should become relevant at low velocities, but we have shown in ref. [4] that they are not significant in the energy range of the experiment. Moreover, since these transitions take place at very large internuclear distances compared to that of the most important avoided crossing ( $\approx 6.4a_0$ ), interferences between both are unlikely, so that transitions at distant avoided crossings increase the EC cross section, and cannot explain the discrepancy with the experimental data. On the other hand, the results of fig. 5 suggest that a possible contamination by metastable ions would not be noticeable at  $E < 10\text{eV}/\text{amu}$ .

To summarize, we report EC cross sections for collisions of  $\text{O}^{2+}$  and  $\text{N}^{2+}$  with  $\text{H}(1s)$ , by applying a molecular expansion in terms of multireference-CI wavefunctions, which allows us to cover a large energy range. In particular, low energy cross sections exhibit, for both systems, Langevin-type increases, previously found in one-electron systems. We have corrected our previous results for  $\text{N}^{2+} + \text{H}(1s)$  collisions, at energies between 0.1 and 0.3eV. Although our EC cross section for these collisions show a better qualitative agreement with merged-beams experiments than that of ref. [4], some discrepancies remain in the low-energy region, where the experimental values depart from the  $1/\sqrt{E}$  dependence found in all calculations at low energies. We have also shown the presence of resonant structures in the total EC

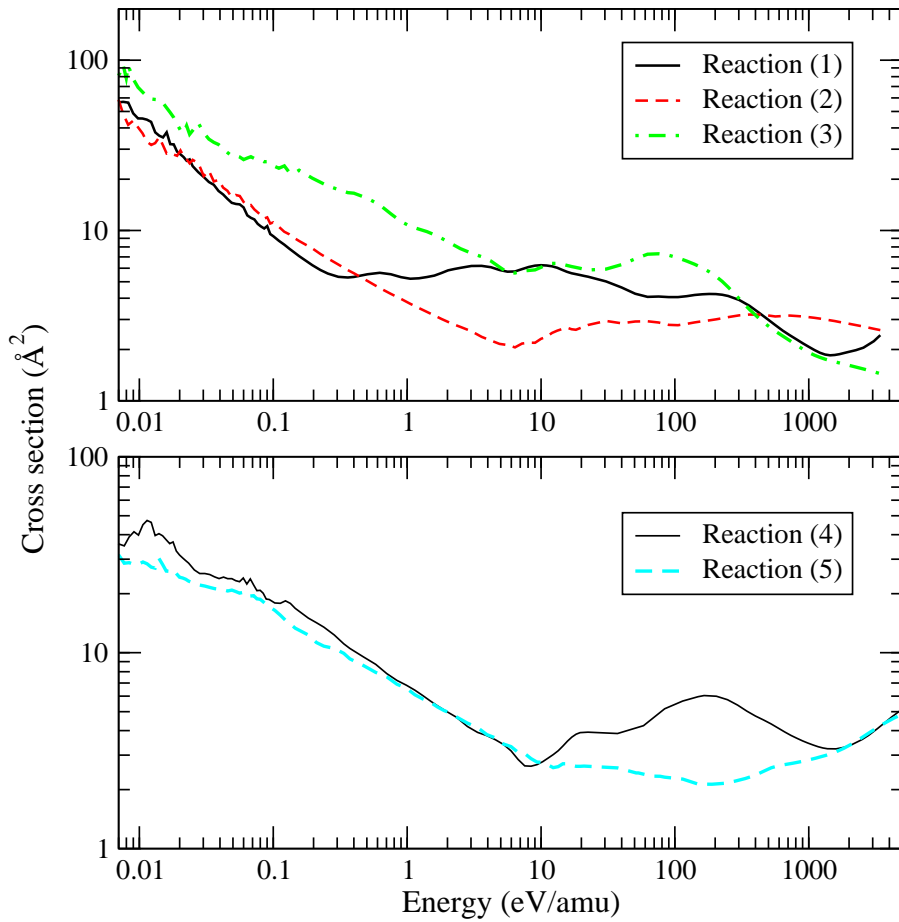


FIG. 5: (Color online) Total cross sections for EC processes for ground state ions (reactions (1) and (4)), and metastable ions (reactions (2)(3) and (5)).

cross section for both collision systems.

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