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State-selective electron capture by O^{3+} ions from atomic and molecular targets

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State-selective electron-capture processes in low-energy collisions of O^{3+} recoil ions with He, Ar, D_2, O_2, H_2O, and CO_2 at impact energies between 0.3 and 1.2 keV and scattering angles between 0° and 6° have been studied using a differential energy-gain spectrometer, capable of measuring simultaneously the energy-gain and the scattering angle of projectile products in ion-atom and -molecule collisions. The energy-gain spectra show that the dominant reaction channels are due to capture into excited states of the projectile product O^{2+}. In this work, no clear evidence of the processes involving long-lived metastable state O^{3+} ($2s2p^2\ ^4P$) was observed. The energy-gain spectra are interpreted qualitatively in terms of the reaction windows, which are calculated using the single-crossing Landau-Zener model and the extended version of the classical over-the-barrier model. The energy dependence of cross sections for electron capture are also measured and found to be nearly constant with changing collision energy. The data are also compared with available measurements and calculations and theoretical results based on the multichannel Landau-Zener model.

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I. INTRODUCTION

The study of single-electron-capture processes in collisions of multiply charged ions with neutral atoms and molecules has recently received considerable theoretical and experimental attention. Multiply charged oxygen ions, in addition to carbon and nitrogen ions, are important species present in the terrestrial atmosphere [1] and in the interstellar medium [2], as well as in the envelopes of many stars. The interaction of oxygen ions with water molecules and carbon dioxide is of interest in studies of astrophysics, comets, and planetary atmospheres [3,4]. Furthermore, electron capture by multiply charged ions present in the solar wind from atmospheric and cometary gases such as H_2O and CO_2 has been suggested among important mechanisms for the charge balance of astrophysical plasmas and the origin of the x-ray emission from comets and planetary atmospheres [5,6]. Electron capture is also an important process in determining the emissivity and ionized structure of a wide variety of atmospheric atoms and molecules. State-selected studies of the electron-capture processes can therefore contribute significantly to an understanding of the observed spectra of these atomic and molecular ions [7,8].

Only a few measurements of state-selective single-electron capture by triply charged oxygen ions from atomic and molecular targets have been made by using a translational energy-gain spectroscopy technique. Wilson *et al.* [9] have measured the energy change spectra for electron capture by O^{3+} ions in atomic hydrogen at impact energies of 4.2, 6.0, 9.0, and 12 keV. Bangsgaard *et al.* [10] have performed energy-gain measurements and have been determined total and partial absolute cross sections for single-electron capture by O^{3+} ions from He for projectile energies between 250 and 1500 eV. Thompson *et al.* [11] studied single-electron capture in collisions of 12-keV O^{3+} ions from He. Other experimental and theoretical investigations into electron capture by triply charged oxygen ions are mostly from atomic and molecular hydrogen. In the case of molecular targets, despite its importance in improving the atomic and molecular database necessary for the accurate modeling of astrophysical plasmas [3] and also in stimulating theoretical treatments in this energy range, there have been no previous experimental measurements of cross sections, differential in translational energy gain and projectile scattering angle, for state-selective electron capture at low energies.

In the present work, using the translational energy-gain spectroscopy technique, we have measured the energy-gain spectra and absolute total cross sections for single-electron capture in collisions of O^{3+} recoil ions with atomic (He and Ar) and molecular (D_2, O_2, H_2O, and CO_2) targets at impact energies between 300 and 1200 eV and scattering angles between 0° and 5°. The data have been obtained on a differential energy spectrometer, which has been described by Yaltakaya *et al.* [12]. Briefly, oxygen ions were produced from water molecules in a recoil ion source by using 25-MeV F^{4+} ions from the Western Michigan University tandem Van de Graaff accelerator as a pump beam. An einzel lens was used to focus the ion beam extracted from the ion source into a 180° double-focusing magnet. After mass selection the ion beam was again focused by two pairs of deflectors and directed into a gas cell containing low-pressure target gas to ensure single-collision conditions. Ions scattered through a nominal angle θ into a solid angle ($\Delta\Omega$) of about 3×10^{-3} sr were energy analyzed by means of a 90° double-focusing electrostatic analyzer (ESA) and then detected by a one-dimensional position-sensitive channel-plate detector, which is located at the focal plane of the ESA. The scattering angle θ is selected by means of an aperture (1 mm diameter) in front of the ESA. For the measurement of absolute total cross sections for single-electron capture, the target gas pressure in the collision cell was measured by a capacitance manometer (MKS Baratron) and was typically ≤ 2 mTorr to ensure single-collision conditions. In addition, an angular acceptance of about ±12° was used after removing the angular

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selector in front of the ESA. The total experimental uncertainties for absolute values of the total cross sections were obtained by the quadratic sum of the statistical deviations, determination of target thickness, and counting efficiency.

II. RESULTS AND DISCUSSION

In the process of single-electron capture by multiply charged ions from atomic or molecular targets, the projectile gains an amount of energy (Q), which depends on the participating electronic states, masses of the projectile and target, and the scattering angle of the projectile ions. In a classical two-body collision, the translational energy of an ion undergoing inelastic scattering differs from the impact energy of the projectile ion E_0 by

$$Q = E - E_0 = \Delta E - \Delta K,$$

where ΔE is the energy defect of the reaction and ΔK is the translational energy given to the target and is given by [13]

$$\Delta K = \frac{M}{M+m}(1-\cos\theta)\left[\frac{2mE_0}{M+m} - \Delta E\right] + \frac{M(\Delta E)^2}{4mE_0}\cos\theta,$$

where m and M are the target and projectile masses, respectively, and θ is the final laboratory scattering angle of the projectile. In the present measurements, the translational energy spectra are expressed in terms of the Q-values and no correction (i.e., ΔK) was added to the measured energy gain.

The energy levels for O^{3+} and O^{2+} ions and atomic gases used in calculating the energy defect ΔE were taken from Bashkin and Stoner [14] and for molecular targets from photoelectron spectroscopy data [15,16]. For O_2 we used the data of Richard-Viard *et al.* [17], for D_2 the calculations of Dunn [18] and Cohen *et al.* [19], for H_2O the data of Reutt *et al.* [20] and Richardson *et al.* [21], and for CO_2 the data of Masuoka *et al.* [22] and Cornaggia and Hering [23]. The product states are identified by the energy-gain Q, measured from the translational energy-gain spectra obtained with a differential energy-gain spectrometer. The energies (ΔE) were calculated assuming that the projectile ion is in its ground state and the molecular targets and their product ions are at the lowest vibrational levels ($\nu=0$). For molecular targets, the energy defects (ΔE) have been calculated on the basis of the Franck-Condon principle. Therefore, the ionization energies of the target are those for vertical processes.

A. Translational energy-gain spectroscopy

O^{3+}-He *collisions.* Figure 1 shows the translational energy-gain spectra for the formation of O^{2+} ions from the reaction of the 300-eV O^{3+} ions with He at different projectile laboratory scattering angles. The observed collision spectrum at 0° scattering angle is dominated by a peak due to transfer excitation from the ground-state O^{3+} $[2s^2(^1S)2p\ ^3P]$ ions into the $2s2p^3\ ^3P$ state of the O^{2+} product ions with contributions from capture into the $2s2p^3\ ^3D$ state. In a transfer-excitation process, which results from a two-electron process, a target electron transferred to the projectile ion with simultaneous excitation of the projectile core. Such processes are also called core-varying single-electron capture (CVSEC) [24]. This channel is observed to be the dominant reaction over the entire collision energy region studied, in agreement with the measurements of Thompson *et al.* [11] and Bangsgaard *et al.* [10]. The broader peak, centered around $Q=6$ eV, is due to core-varying single-electron capture into the $2s2p^3\ ^1D$, ^3S, and ^1P states.

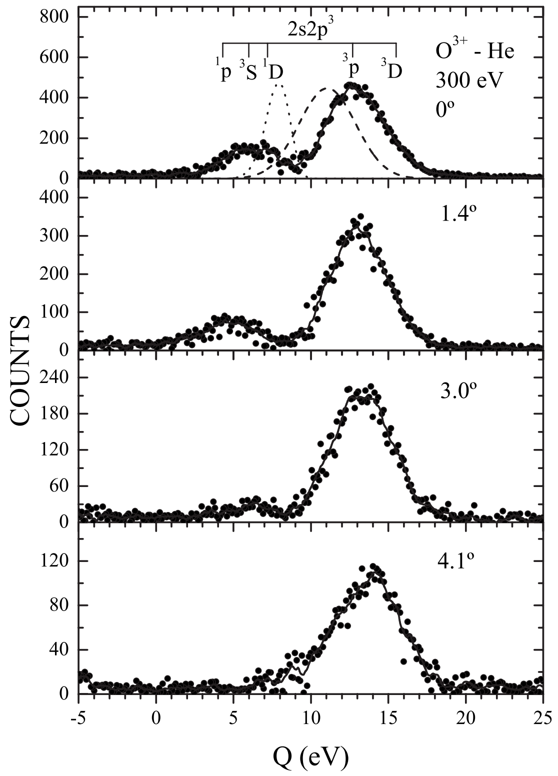

FIG. 1. Translational energy-gain spectra for single-electron capture by 300-eV O^{3+} ions from He at different projectile laboratory scattering angles. Also shown are reaction windows calculated on the basis of a single-crossing LZ model (dotted curve) and the ECOB model (dashed curve).

As the scattering angle is increased, electron capture into the $2s2p^3\ ^3P$ state remains dominant, but the relative importance of single-electron capture into the $2s2p^3\ ^1D$, ^3S, and ^1P states is strongly decreased. This indicates that the angular distributions for capture into the $2s2p^3\ ^3P$ state is strongly peaked in the forward direction in the O^{3+}+He collisions. Figure 1 also shows the calculated reaction windows for 300-eV O^{3+}-He collisions, using both a single-crossing Landau-Zener (LZ) model [25] and the extended version of the classical over-the-barrier (ECOB) model [26]. Calculated peak values have been normalized to our observed peak values in the translational energy-gain spectrum. The reaction window based on the ECOB model is assumed to be Gaussian around the classical value of the minimum-Q value $[Q=(q-1)/R_c]$ (where $R_c=[2(q)^{1/2}+1]/I$, with q is the charge state of the projectile and I is the ionization potential of the target in atomic unit) which is related to the time dependence of the potential barrier height, with a width proportional to $v^{1/2}$, where v is the impact velocity of the projectile. The reaction based on a single-crossing LZ model favors Q values smaller than those observed and is positioned near the

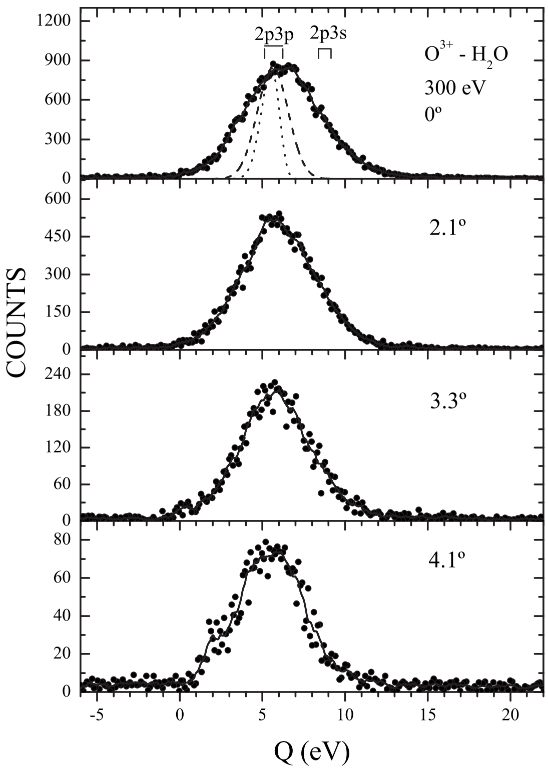

FIG. 2. Translational energy-gain spectra for single-electron capture by 300-eV O^{3+} ions from H_2O at different projectile laboratory scattering angles. Also shown are reaction windows calculated on the basis of a single-crossing LZ model (dotted curve) and the ECOB model (dashed curve).

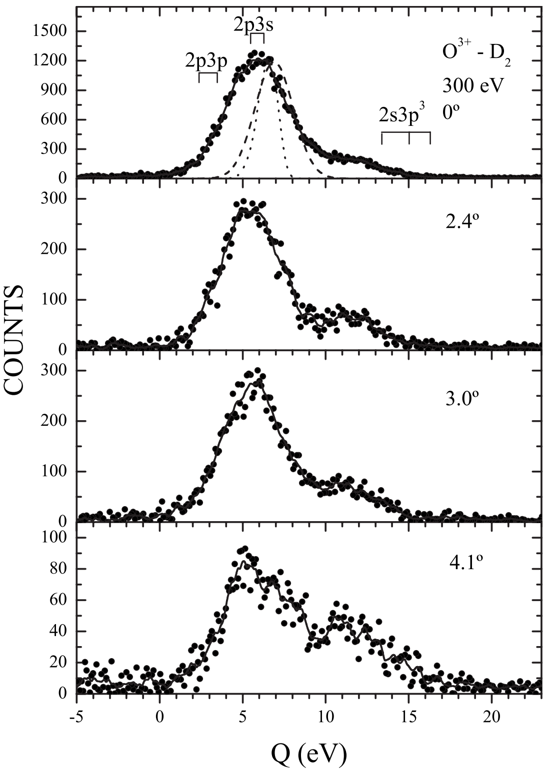

FIG. 3. Translational energy-gain spectra for single-electron capture by 300-eV O^{3+} ions from D_2 at different projectile laboratory scattering angles. Also shown are reaction windows calculated on the basis of a single-crossing LZ model (dotted curve) and the ECOB model (dashed curve).

$2s2p^3\ ^1D$ state, while the reaction window based on the ECOB model maximizes at about 11 eV and accommodates most of the dominant channel.

O^{3+}-H_2O *collisions.* Figure 2 shows the translational energy-gain spectra of the product O^{2+} ions in the O^{3+}-H_2O collisions at impact energy of 300 eV and different projectile laboratory scattering angles. The zero-angle spectrum shows only one broad peak; this peak correlates with nondissociative pure single-electron capture into the $2p3p$ state of O^{2+} ions with production of H_2O^+ in the ground state ($X\ ^2B_1$). There is some contribution from an unresolved reaction at about 9 eV, involving capture into the $2p3s$ state of O^{2+}. As the scattering angle increases, single-electron capture into the $2p3p$ state remains dominant. This indicates that the angular distributions for capture into the $2p3p$ state is peaked in the forward direction in O^{3+}-H_2O collisions. The reaction windows coincide very well with the position of dominant channel and underestimate the contribution of the $2p3s$ channel.

O^{3+}-D_2 *collisions.* Figure 3 shows translational energy-gain spectra for single-electron capture by 300-eV O^{3+} ions from D_2 at different scattering angles. The observed collision spectrum at 0° scattering angle is dominated by a peak corresponding to nondissociative single-electron capture from the ground-state O^{3+} $[2s^2(^1S)2p\ ^3P]$ ions into the $2p3s$ state of the O^{2+} with production of D_2^+ in the ground state ($X\ ^2\Sigma_g^+$), with contributions from capture into the $2p3p$ state. The smaller peak at about 13 eV correlates with transfer excitation into the $2s2p^3$ states, but contributions due to dissociation of D_2^{+*} into $D^+ + D$ (1 s) cannot be ruled out. Beijers *et al.* [27] have used photon emission spectroscopy to determine absolute state-selective cross sections for single-electron capture by O^{3+} ions colliding on atomic and molecular hydrogen at impact energies between 45 and 752 eV/amu. Cross sections for capture into $2p3p$ states were found to be comparable to that for $2p3s$ states, which are in disagreement with the present data where the relative populations of $2p3p$ and $2p3s$ states are, respectively, 8.1% and 75% at 300 eV impact energy.

As the scattering angle is increased, single-electron capture into the $2p3s$ state of O^{2+} remains dominant, whereas the relative importance of the reaction channel correlated with capture into the $2s2p^3$ state increases. This indicates that the angular distribution for capture into $2p3s$ is more strongly peaked in the forward direction than for a process associated with capture into the $2s2p^3$ state. The reaction windows coincide very well with the position of the dominant reaction channel.

O^{3+}-Ar, CO_2, *and* O_2 *collisions*. Figure 4 shows the translational energy-gain spectra for single-electron capture by 300-eV O^{3+} ions from Ar, CO_2, and O_2 at 0° scattering angle. In O^{3+}+Ar collisions, the dominant peak corresponds to capture from the ground-state O^{3+} $[2s^2(^1S)2p\ ^3P]$ ions into the $2p3s$ state of the O^{2+} with production of Ar^+ in the ground state ($3p^5\ ^2P$). The structure on the lower-energy side of the

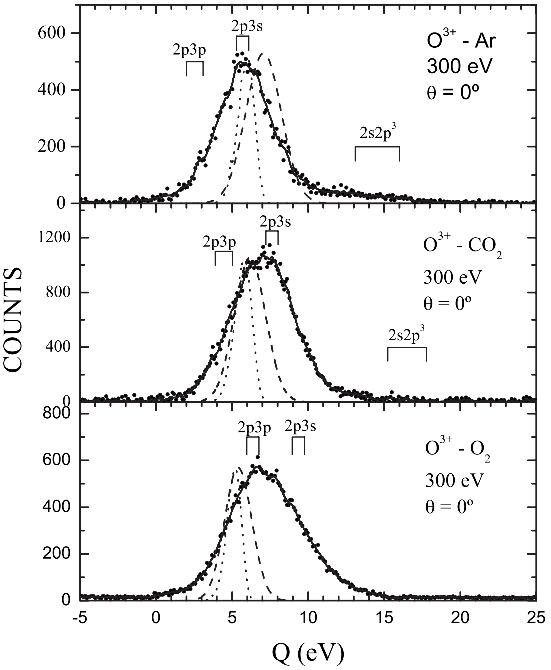

FIG. 4. Translational energy-gain spectra for single-electron capture by 300-eV O^{3+} ions from Ar, CO_2, and O_2 at 0° scattering angles. Also shown are reaction windows calculated on the basis of single-crossing LZ model (dotted curve) and the ECOB model (dashed curve).

dominant peak corresponds to capture into the $2p3p$ state. The smaller peak at about 13 eV correlates with capture into the $2s2p^4$ states of O^{2+}, with contribution from transfer ionization process (single-electron-capture simultaneous ionization of the target product).

For the O^{3+}-CO_2 collisions, the observed collision spectrum is dominated by a peak due to nondissociative single-electron capture into the $2p3s$ state of O^{2+} with production of CO_2^+ in the ground state $(^2\Pi_g)$. There are also some contributions from an unresolved reaction at about 5 eV involving capture into $2p3p$ states. The reaction windows favor Q values smaller than the observed channel.

For the O^{3+}-O_2 collisions, the observed spectrum is dominated by nondissociative single-electron capture from ground-state incident ions O^{3+} $[2s^2(^1S)2p\ ^3P]$ ions into the $2p3p$ state of the O^{2+} with production of O_2^+ in the ground state $(X\ ^2\Pi_g)$. The unresolved structure on the higher-energy side of the dominant peak corresponds to capture into $2p3s$ states.

Figure 4 also shows calculated reaction windows calculated on the basis of a single-crossing LZ model and ECOB model. For O^{3+}-Ar collisions, the reaction window based on a single-crossing LZ model correctly predicts the position of the dominant reaction channel. The reaction window based on the ECOB model underestimates the contribution of the $2p3s$ channel and favors channels with higher Q values compared to the dominant reaction channel. For the O^{3+}-CO_2 and O_2 collisions, although they accommodate most of the dominant peak, the calculated reaction windows favor Q values smaller than those observed and underestimate the contribution of the dominant channels.

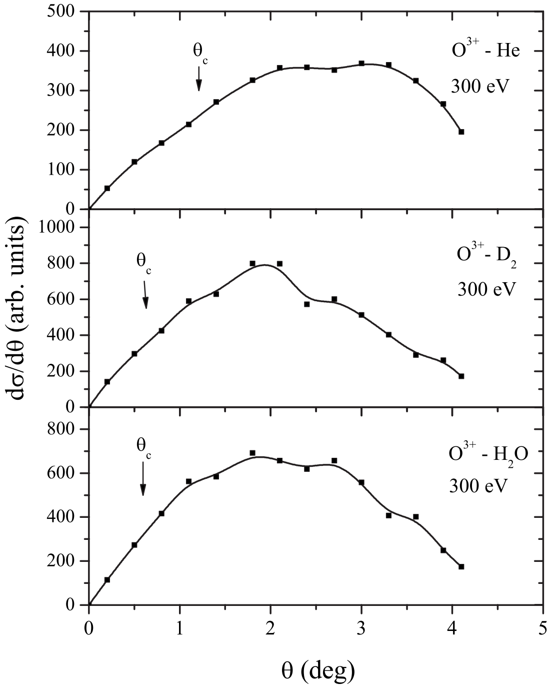

FIG. 5. Experimental differential cross sections $(d\sigma/d\theta)$ for single-electron capture by 300-eV O^{3+} ions from He, D_2, and H_2O. Spline lines are drawn to guide the eye.

B. Differential cross sections

The experimental total differential cross sections $(d\sigma/d\theta)$ for single-electron capture by 300-eV O^{3+} ions from He, D_2, and H_2O are shown in Fig. 5. The differential cross sections were determined using the translational energy-gain technique, by calculating the area under the peaks in the energy-gain spectra at different projectile laboratory scattering angles using a curve-fitting program. The general features of the distributions are qualitatively explained in terms of semiclassical model based on Coulomb potential curves, which have been described in detail by Andersson *et al.* [28]. The traditional two-state model has been used to estimate the critical angle θ_c, which corresponds to capture at an impact parameter equal to the crossing radius, by assuming that capture occurs at a localized curve crossing between the potential energy curves for entrance and exit channels. For small angles, $\theta_c = Q/2E$, where Q is the exoergicity of the collision and E is the collision energy. This angle separates the events scattered at smaller angles due to capture on the way out of the collision and events scattered at larger angles due to capture on the way into the collision. The critical angle for each dominant channel is indicated by an arrow in Fig. 5.

For 300-eV O^{3+}-He collisions, the distribution contains a broad peak lying slightly outside the critical angle $\theta_c = 1.21°$ for capture into the excited state $2s2p^3\ ^3P$ of the O^{2+} ion. This distribution represents contributions from electron capture that takes place on the way into the collisions. For

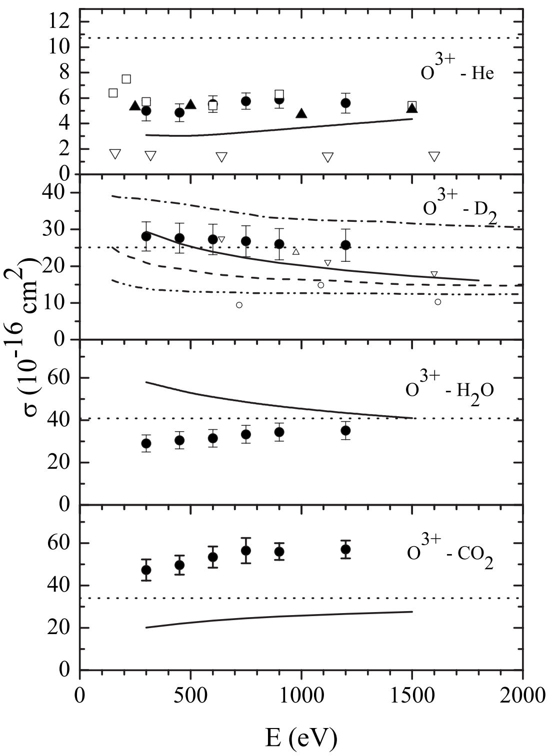

FIG. 6. Total cross sections for single-electron capture by O^{3+} ions from He, D_2, H_2O, and CO_2, present work (●); Ishii *et al.* [29] (□); Bangsgaard *et al.* [10] (▲); Janev *et al.* [30] (▽); Phaneuf *et al.* [33] (△); Beijers *et al.* [27] (◯). Dotted curves: COB model. Dash-dotted curves: EA—Wang *et al.* [34]. Dash-double-dotted curves: IOSA—Wang *et al.* [34]. Dashed curves: VSA—Wang *et al.* [34]. Solid curves: MCLZ calculations.

300-eV O^{3+}-D_2 collisions, the spectrum shows a smooth rise to a broad maximum around 2° outside $\theta_c = 0.61°$, assuming that electron capture occurs into $2p3s$ of the O^{2+} ion. The distribution is due to capture on the way into the collision. For 300-eV O^{3+}-H_2O collisions, the distribution contains a peak slightly greater than $\theta_c = 0.59°$, which corresponds to the $2p3p$ capture channel, indicating that capture takes place on the way into the collision.

C. Total cross sections

The measured total cross sections for single-electron capture by O^{3+} ions from He, D_2, H_2O, and CO_2 are shown in Fig. 6 together with other available measurements and calculations. For O^{3+}-He collisions, the total cross sections show no significant dependence on the collision energy. Our data are in reasonably good agreement with the experimental results of Bangsgaard *et al.* [10] and Ishii *et al.* [29]. The data presented in this study, however, are considerably higher than the recommended cross sections of Janev *et al.* [30] and found to be 60% higher than the multichannel Landau-Zener calculations [25], based on the Taulberg expression for the coupling matrix H_{12} [31]. The measured cross sections are about a factor of 2 smaller than the classical over-the-barrier (COB) model [32] prediction of 10.737×10^{-16} cm^2 assuming the capture probability $A = 0.5$. An average value for A of 0.253 is found experimentally by dividing the measured total cross section by the classical cross section (πR_c^2). This small value of A is probably due to the population of core excited states. The responsible coupling matrix element (H_{12}), which couples the incoming channel with the outgoing reaction channels leading to these core excited states, is small and thus will yield measured cross sections smaller than πR_c^2.

For $O^{3+} + D_2$ collisions, the total cross sections slowly decrease with increasing collision energies. Our data are in good agreement with the measurements of Phaneuf *et al.* [33] and the recommended cross sections of Janev *et al.* [30] for $O^{3+} + H_2$ collisions, but are a factor of 2 higher than the results of Beijers *et al.* [27]. The present measurements are in good agreement with the COB model prediction of 27.24 $\times 10^{-16}$ cm^2 with $A = 0.5$ and multichannel Landau-Zener (MCLZ) calculations at low energies (≤ 900 eV) and show similar behavior (cross sections slowly decrease with increasing collision energies). Recently Wang *et al.* [34] performed a nearly complete *ab initio* study of single-electron capture in collisions of ground-state O^{3+} $[2s^2(^1S)2p\ ^3P]$ ions with molecular hydrogen using a fully quantum-mechanical, molecular-orbital (QMO) coupled-channel method, adopting the so-called infinite-order sudden approximation (IOSA), the vibrational sudden approximation (VSA), and the electronic approximation (EA). The data presented here are considerably lower than those of EA calculations by a factor of 1.35 and found to be on average 1.42 and 3.33, respectively, higher than VSA and IOSA predictions.

For $O^{3+} + H_2O$ and CO_2 collisions, the total cross sections increase with increasing impact energy, a pattern that is well documented for such collisions at low energies [31,35]. This can be understood by the reaction window, which broadens with increasing energies and therefore increase the probability of capture channels with large Q values. For $O^{3+} + H_2O$ collisions, the energy dependence of cross sections is in direct contradiction with the MCLZ calculation, which decreases with increasing energy, and the COB model overpredicted the total cross section for capture by at least of a factor of 1.2. For $O^{3+} + CO_2$ collisions, The MCLZ results are at least a factor of 2 smaller than the experimental results and show the same energy dependence, whereas the predictions of the COB model lie about 30%–40% below the experimental results.

III. CONCLUSIONS

Doubly differential cross sections, in energy and angle, for single-electron capture by O^{3+} ions from He, Ar, D_2, O_2, H_2O, and CO_2 have been studied by means of translational energy gain spectroscopy at collision energies between 0.3 and 1.2 keV. Translational energy gain spectra for single-electron capture by O^{3+} ions from He indicated that the dominant reaction channel was correlated with transfer excitation into the $2s2p^3\ ^3P$ state, while for the H_2O, D_2, Ar, O_2, and CO_2 targets the dominant channels were due to capture into $2p3p$, $2p3s$, $2p3s$, $2p3p$, and $2p3s$ states, respectively. The energy-gain spectra were interpreted qualitatively in terms of the reaction windows that were calculated using the single-crossing LZ model and the ECOB model. The reaction windows provide the best description of the observed spectra for all collision systems. However, for O^{3+}-He collision system, the reaction window based on a single-crossing LZ model did not describe the position of dominant processes, likely due to the involvement of core-varying transitions. In these collision systems, there is no clear evidence of molecular excitation and dissociation except possibly for the D_2 target. The energy dependence of the absolute total cross sections of the present work was compared with the available data and theoretical calculations. No reported data on single-electron capture by O^{3+} ions from D_2, H_2O, and CO_2 are, however, available for comparison.

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