PHYSICAL REVIEW A VOLUME 45, NUMBER 7 1 APRIL 1992

Suppressed electron capture in slow $O^+(^4S°, ^2D°, ^2P°)$-He collisions

E. Wolfrum, J. Schweinzer,* and H. Winter
Institut für Allgemeine Physik, Technische Universität Wien, Wiedner Hauptstrasse 8-10, A-1040 Wien, Austria
(Received 21 November 1991)

Total cross sections for single-electron capture from He by keV O^+ $^2D°$ and $^2P°$ metastable ions turned out to be much smaller than for O^+ $^4S°$ ground-state ions, despite a larger energy defect for the latter reaction. We suggest that this unexpected behavior is caused by efficient suppression of electron capture into either metastable ion due to a competing, collisionally induced inelastic transition into the companion metastable state.

PACS number(s): 34.70.+e, 52.20.Hv, 82.30.Fi

I. INTRODUCTION

Single-electron capture (SEC) from neutral atoms by relatively slow singly charged ions (impact velocity smaller than 1 a.u.) generally prefers reaction channels with the smallest available energy defects ΔE [1-3]. This holds if no specific final states are excluded, which could, e.g., be caused by Wigner's spin conservation rule [4-6]. If primary ions in different states (i.e., in their ground state and one or several metastable states) participate in such collisions, reaction energy defects for SEC into given final projectiles states will differ and henceforth so will the various primary-ion state-selective SEC cross sections.

For O^+-He collisions, the following three SEC reactions can be regarded as most probable:

$$O^+(^4S°)+\mathrm{He}\rightarrow O°(^3P)+\mathrm{He}^+-10.96\ \mathrm{eV}\ , \quad (1a)$$

$$O^+(^2D°)+\mathrm{He}\rightarrow O°(^3P)+\mathrm{He}^+-7.64\ \mathrm{eV}\ , \quad (1b)$$

$$O^+(^2P°)+\mathrm{He}\rightarrow O°(^3P)+\mathrm{He}^+-5.94\ \mathrm{eV}\ . \quad (1c)$$

We have studied all three SEC reaction channels by measuring the respective primary state-selective total cross sections absolutely, and by using translational energy spectroscopy for additional related investigations. We found that despite the comparably smaller energy defects for (1b) and (1c) the related cross sections are much smaller than for (1a), although no known selection rule can be held responsible for this unexpected result.

In the present Rapid Communication we analyze this remarkable finding which may be characterized as suppressed electron capture.

II. EXPERIMENTAL METHODS

A. Ion production

Primary O^+ ions have been produced in a Nier-type ion source by electron impact on H_2O, for which the resulting ion kinetic-energy spread was considerably smaller than for other source feeding gases as, e.g., O_2 or CO_2, which improved the achievable energy resolution of translational energy spectra (TES).

Beside the ground-state $O^+(2s^22p^3)^4S°$ there exist two long-lived excited metastable states, i.e., $O^+(2s^22p^3)^2D°$ and $O^+(2s^2p^3)^2P°$ (τ =3.6 h and 4.57 s [7], with excitation energies 3.32 and 5.02 eV above the $^4S°$ ground state [8], respectively).

By systematically varying the ion-source electron-impact energy E_e, the two metastable ion-beam fractions could be changed accordingly (see below). After their acceleration to a desired collision energy E and magnetical mass analysis the primary ions were guided into the collision chamber, while possible charge-exchanged neutral atoms were removed from the beam by means of two sets of deflection plates.

B. Measurement of metastable ion-beam fractions

For determination of the primary-ion-beam composition at various values of E_e, the techniques of collisionally induced state excitation or deexcitation (cf., e.g., [9,10]) and ion-beam attenuation (cf., e.g. [11,12]) have been combined. The ratio of the two O^+ metastable state fractions has been derived from TES for their inelastic scattering on He. Generally, excitation or deexcitation between two states, i and k, gives rise to TES signals $I(i\rightarrow k)$ and $I(i\leftarrow k)$ displayed symmetrically on the exothermic and endothermic sides of the respective elastic peak, with their intensity ratio $F(i)/F(k)$ given by (cf. above references)

$$\frac{F(i)}{F(k)}=\frac{(2J_i+1)}{(2J_k+1)}\frac{I(i\rightarrow K)}{I(i\leftarrow k)}\ . \quad (2)$$

J_i and J_k are the total angular momenta of the respective states.

In principle, this method should lead to all ion-beam fractions, if the above ratios can be determined for transitions between pairs of all states present in the ion beam. For O^+-He collisions, the following three excitation or deexcitation reactions are of importance:

$$O^+(^4S°)+\mathrm{He}\leftrightarrow O^+(^2D°)+\mathrm{He}\pm 3.32\ \mathrm{eV}\ , \quad (3a)$$

$$O^+(^4S°)+\mathrm{He}\leftrightarrow O^+(^2P°)+\mathrm{He}\pm 5.02\ \mathrm{eV}\ , \quad (3b)$$

$$O^+(^2D°)+\mathrm{He}\leftrightarrow O^+(^2P°)+\mathrm{He}\pm 1.7\ \mathrm{eV}\ . \quad (3c)$$

Excitation or deexcitation between the O^+ $^4S°$ ground state and either of the metastable doublet states in collisions with He [i.e., reactions (3a) and (3b)] is forbidden by Wigner's spin conservation rule and thus could not be

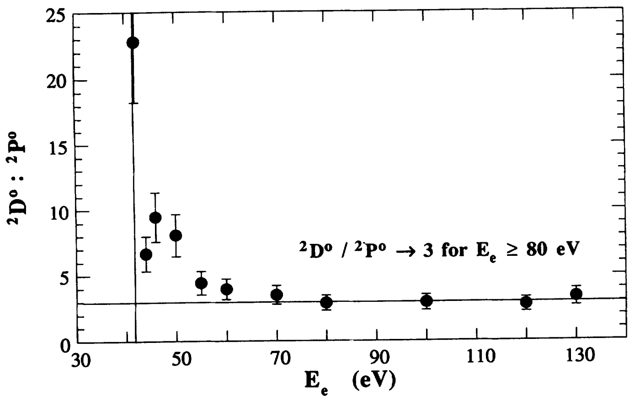

FIG. 1. $^2D^o : {}^2P^o$ metastable state ratio evaluated from TES vs electron impact energy E_e.

observed in the TES. Figure 1 presents the $^2D^o$-to-$^2P^o$ metastable ion states ratio versus electron impact energy E_e as obtained from our TES measurements. For $E_e < 42$ eV no $^2P^o$ fraction could be found anymore, and beyond 80 eV the $^2D^o$:$^2P^o$ ratio reached a constant value of about 3. With O_2 as target particles, for which Wigner's rule does not strongly apply, all three ion-beam fractions in O^+ beams produced by electron impact on O_2 have been measured [10].

Unfortunately, the limited energy resolution of our translational energy spectrometer (detailed descriptions of the instrument have been given in [13,14]) did not permit separation of all essential peaks in TES for such collisions, also because of their considerable broadening due to participation of a number of vibrational target states.

We therefore measured the total metastable ion-beam fraction by means of the ion-beam attenuation technique, using H_2 as well as N_2 as attenuation gases, since for both molecules the SEC cross section for the O^+ ground-state ions is much smaller than for the metastable ions [11,15]. Figure 2 shows a typical attenuation curve for 2-keV O^+ ions in N_2, with E_e set to 30, 35, and 130 eV, respectively. Such measurements have been carried out for different ion-impact energies and yielded total metastable ion-beam

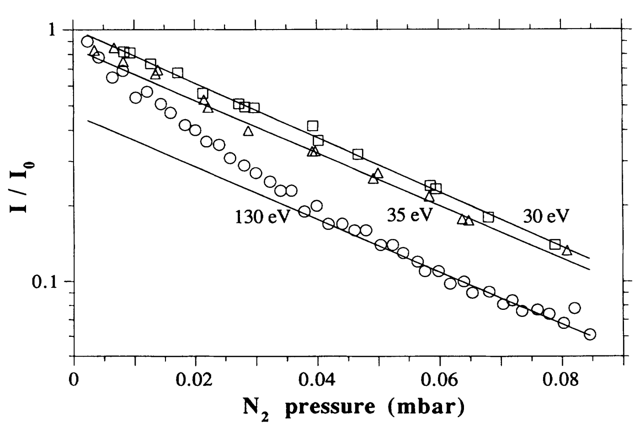

FIG. 2. Attenuation characteristics for O^+ ions in N_2, with ion beams produced by 30-, 35-, and 130-eV electron impact on H_2O, respectively.

TABLE I. O^+ ion-beam compositions for electron impact on H_2O at $E_e = 30$, 40, and 130 eV, respectively.

E_e (eV)	$F(^4S^o)$ (%)	$F(^2D^o)$ (%)	$F(^2P^o)$ (%)
30	100		
40	64 ± 3.6	33 ± 5.6	
130	47 ± 7.1	40 ± 8	13 ± 3

fractions of zero for $E_e \leq 30$ eV, about 33% at 40 eV and above 50% at 130 eV. With the ratio of metastable fractions already known, the ion-beam composition could then be determined for different values of E_e as listed in Table I.

In clear disagreement with our results, Hughes and Tiernan [16], applying a different technique, found for O^+ ion beams produced by 60-eV electron impact on H_2O a total metastable fraction of about 90%. While the reason for this difference remains unclear, it does not concern the subject of present interest (see below).

C. Measurement of primary-ion state-selective total SEC cross sections

Such measurements have been carried out with primary O^+ ion beams of different, well-defined compositions. Assuring single collision conditions in the He-filled collision cell, immediately behind the latter the charge-exchanged neutral atoms were separated from the remaining ions and counted by equipment described in [17]. From the related signals, absolute SEC cross sections have been derived [12,18] by calibrating the He target thickness with reference to SEC data for He^+-He collisions at 2.75 keV impact energy [19]. For a mixed primary-ion beam the resulting apparent SEC cross section σ_{mix} is related to the primary-ion state-selective SEC cross sections of interest by

$$\sigma_{mix} = \sum_{i=1}^{3} F(i)\sigma_i \,. \tag{4}$$

By measuring such apparent cross sections with ion beams of grossly different compositions (cf. Table I), all three state-selective SEC cross sections should thus have become known.

III. RESULTS AND DISCUSSION

To our big surprise, no measurable SEC cross sections for either of the two metastable O^+ states could be found. After measuring SEC cross sections for a pure ground-state ion beam ($E_e = 30$ eV), apparent SEC cross sections σ_{mix} at 40- and 130-eV electron impact energy decreased in accordance with the respective smaller ground-state ion fractions.

Total errors for σ_g (ground-state primary ions) are due to the applied calibration cross sections ($\pm 2.5\%$) and the statistical errors of our measurements (about $\pm 5\%$).

Figure 3 shows that our ground-state-related SEC cross sections are in fair agreement with Ref. [20], where the

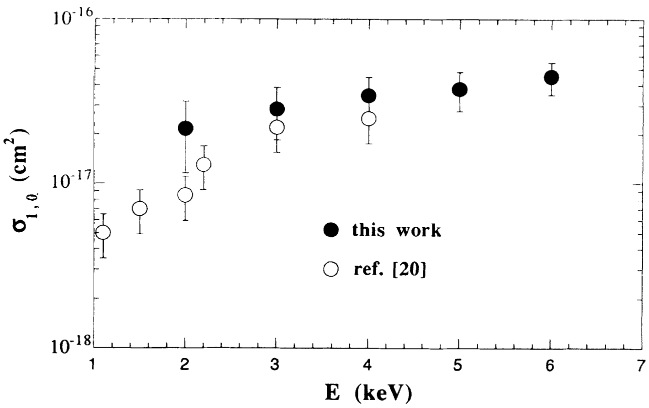

FIG. 3. Absolute SEC cross sections for $O^+(^4S^o)$-He collisions compared with data from Ref. [20].

growth method has been applied for SEC determination. In the latter reference, for a mixed-ion beam with undefined metastable fraction higher cross sections have been found than for a pure ground-state ion beam produced from CO_2 at 21-eV electron impact energy.

We remark that SEC cross sections for H^+-He collisions [21] are of similar size as the data shown in Fig. 3, which came as no surprise since for both collision systems the SEC reaction energy defects are quite similar. Comparison with $Ar^+ + He$ collisions, which for SEC involve an energy defect of -8.8 eV and SEC cross sections of about 2×10^{-17} cm^2 in the appropriate impact energy region [22] let us expect at least similar cross sections for reactions (1b) and (1c) because of the still smaller energy defects involved.

SEC by the two metastable O^+ doublet state ions [i.e., reactions (1b) and (1c)] can be considered independently from reaction (1a), because He collisionally induced transitions between either metastable state and the O^+ ground state are forbidden by Wigner's spin conservation rule (see Sec. IIB).

We believe that the unexpectedly small SEC cross sections for both metastable primary ions are caused by competition from reaction channels (3c), which involve considerably smaller reaction energy defects than the SEC reactions [1(b) and (1c)].

At small internuclear distance the collisional system can be regarded as a quasimolecule $(O\text{-}He)^+$. The knowledge of (not yet available) exact molecular potential energy curves would be helpful to assess the importance of particular inelastic transitions. The Wigner-Witmer rules [23] must be applied for building up the quasimolecular states of interest from both the atomic states of the united atom (UA) (i.e., Ne^+) and the separated atoms (SA), respectively.

However, to give at least a qualitative explanation of the experimentally obtained results, rude estimates for the involved molecular binding energies at small internuclear distance have been indicated in Figs. 4(a) and 4(b) for the quartet and doublet systems, respectively. Correlation between UA and SA states is achieved by simply connecting states with the same symmetry, starting with the lowest

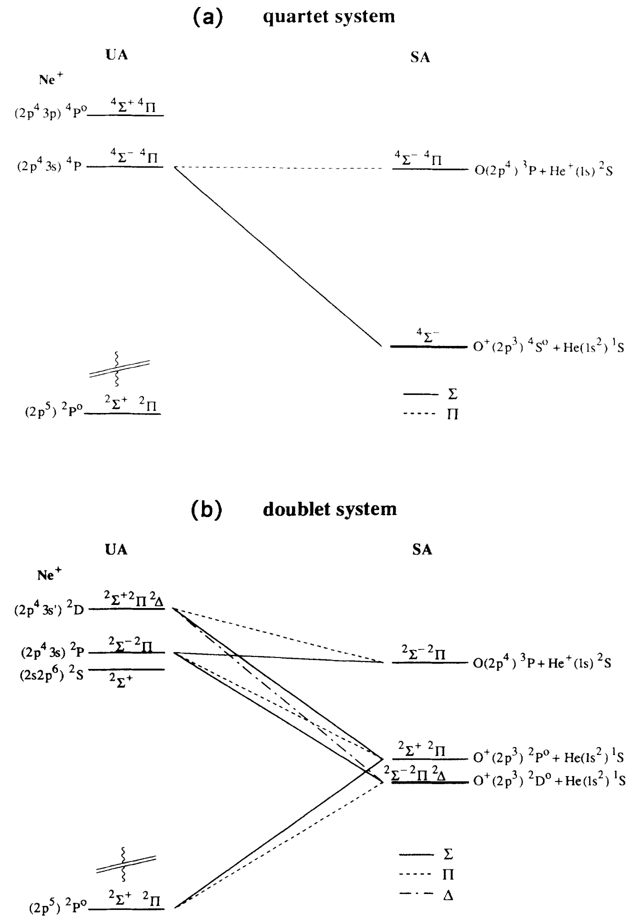

FIG. 4. Correlation diagrams for (a) the quartet collision system and (b) the doublet collision system. Only the lowest states of relevance have been shown.

SA states, respectively.

In the case of the quartet system, the most probable inelastic reaction channel by far belongs to SEC [cf. Fig. 4(a)] and correlation of the SA initial state with UA ground state is forbidden (cf. above). In contrast to this, for the doublet system [cf. Fig. 4(b)] correlation of the SA $(^2D^o, ^2P^o)$ entrance states with the strongly bound Ne^+ ground state is possible. However, the energy difference between these states and the one leading to O^0-He^+ in the SA limit is large and henceforth SEC along this path seems rather improbable. In addition to the SEC channels which correlate to excited UA states similar as in the case of the quartet system, further reaction channels corresponding in the SA limit to excitation or deexcitation processes are open, with transitions among them being rather likely because of the comparably small energy differences involved. Therefore, transitions from either initial doublet state into SEC final channels are inhibited by transitions related to excitation or deexcitation between both metastable O^+ primary states.

However, the experimentally observed, rather strong (by at least a factor of 10) suppression of the doublet SEC channel in comparison to the quartet system cannot be explained by these simple considerations to full satisfaction. Detailed close coupling calculations are in progress to explain more quantitatively the observed, unexpectedly low cross sections for SEC by the metastable O^+ ions.

In light of our observations, a total metastable O^+ ion-beam fraction of 90% as measured by Hughes and Tiernan [16] seems rather unlikely, but if true would make the present findings even more remarkable.

ACKNOWLEDGMENTS

This work has been supported by Kommission zur Koordination der Kernfusionforschung at the Austrain Academy of Sciences. It was performed within the coordinated research program on Atomic and Molecular Data for Fusion Edge Plasmas of the International Atomic Energy Agency Vienna under Contract No. 5235/CF.

*Present address: Max Planck Institut für Plasmaphysik, D-8046 Garching bei München, Germany.

[1] H. S. W. Massey, Rep. Prog. Phys. **12**, 248 (1949).
[2] J. B. Hasted, Proc. R. Soc. London, Ser. A **205**, 421 (1950).
[3] Y. N. Demkov, Zh. Eksp. Teor. Fiz. **45**, 195 (1963) [Sov. Phys. JETP **18**, 138 (1964)].
[4] E. Wigner, Nachr. Akad. Wiss. Göttingen Math. Phys. Kl. **IIa**, 375 (1927).
[5] J. H. Moore, Phys. Rev. A **8**, 2359 (1973).
[6] J. H. Moore, Phys. Rev. A **10**, 724 (1974).
[7] M. Lavollée and G. Henri, J. Phys. B **22**, 2019 (1989).
[8] S. Bashkin and J. O. Stoner, *Atomic Energy Levels and Grotrian Diagrams* (North-Holland, Amsterdam, 1975).
[9] N. Kobayashi, T. Nakamura, and Y. Kaneko, J. Phys. Soc. Jpn. **52**, 2684 (1983).
[10] M. Hamdan and A. G. Brenton, J. Phys. B **22**, 2289 (1989).
[11] B. R. Turner, J. A. Rutherford, and D. M. J. Compton, J. Chem. Phys. **48**, 1602 (1968).
[12] E. Unterreiter, J. Schweinzer, and H. Winter, J. Phys. B **24**, 1003 (1991).
[13] J. Schweinzer and H. Winter, J. Phys. B **22**, 893 (1989).
[14] F. Aumayr, J. Schweinzer, and H. Winter, J. Phys. B **22**, 1027 (1989).
[15] Y. Xu, E. W. Thomas, and T. F. Moran, J. Phys. B **23**, 1235 (1990).
[16] B. M. Hughes and T. O. Tiernan, J. Chem. Phys. **55**, 3419 (1971).
[17] J. Schweinzer and H. Winter, J. Phys. B **23**, 3881 (1990).
[18] F. Aumayr, G. Lakits, and H. Winter, Phys. Rev. A **33**, 846 (1986).
[19] R. Hegerberg, T. Stefansson, and M. T. Elford, J. Phys. B **11**, 133 (1978).
[20] T. Kusakabe, Y. Mizumoto, K. Katsarayama, and H. Tawara, J. Phys. Soc. Jpn. **59**, 1987 (1990).
[21] P. M. Stier and C. F. Barnett, Phys. Rev. **103**, 896 (1956).
[22] T. J. M. Sluyters, E. de Haas, and J. Kistemaker, Physica **25**, 1376 (1959).
[23] G. Herzberg, *Spectra of Diatomic Molecules* (Van Nostrand Reinhold, New York, 1950).