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Resonances in the differential excitation functions of five electronic states of Ne in the autoionization region*

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We have observed many resonances by electron spectrometry in the differential excitation functions of the states $3s$, $3s'$, $3p'$, $4s'$, and $3d$ of neon in the autoionization region. These measurements are compared to the data obtained by other spectroscopic methods and their interpretation is discussed. For the main resonances, the energies and natural widths are determined. Moreover, their effective strengths and profile indices are presented for the various decay channels in comparison with the data obtained in the ionization channel.

I. INTRODUCTION

Negative-ion resonances in the region of autoionization states of rare gases have been detected by various methods of electron impact. (1) By transmission spectroscopy, the first observation was performed by Kuyatt *et al.*[1] in helium, and in a systematic manner in all rare gases by Sanche and Schulz.[2] (2) By electroionization spectroscopy, the first manifestation of this phenomenon was reported by Grissom *et al.*[3] in helium, and afterwards, with more precision, by Marmet and co-workers[4-6] in most of the rare gases. (3) With methods of optical detection, Heideman *et al.*[7] observed these resonances in a few optical excitation functions of helium, and Marchand and Veillette in the photon detection of helium[8] and argon.[9] (4) Burrow and Schulz[10] detected this phenomenon in helium by threshold spectroscopy, and also Grissom *et al.* in helium[3] and neon.[11] (5) Recently such resonances were also observed by metastable detection.[12]

Consequently it is expected to observe this phenomenon also in the measurements of differential excitation functions by electron spectrometry, i.e., in the current of the electrons inelastically scattered at a given angle and with a fixed energy loss (corresponding to the excitation of an electronic state of the scatterer). Actually at the threshold of these excitation functions, many negative-ion resonances (compound states), whose parents are the states of the neutral atom located below the ionization threshold, were observed: In helium, the first observation was reported by Schulz and Philbrick.[13] Afterwards this study was completed by Chamberlain,[14,15] and by Ehrhardt and co-workers.[16-18] Swanson *et al.* carried out that type of measurements in krypton[19] and xenon.[20] The equivalent was also achieved by metastable detection[21] and by photon detection.[22-24]

However, in the autoionization region of these excitation functions, only Simpson *et al.*[25] detected these negative-ion resonances by electron spectrometry. They measured the differential excitation functions of the states $2\,^3S$, $2\,^1S$, and $2\,^1P$ of helium in the region 56–59 eV and observed the resonances caused by the states of He^- at 57.1 eV and 58.2 eV. No other similar measurement was carried out in the autoionization region of other gases; only Chamberlain[15] reported to have tried without success to detect resonances in argon, in the region 25–29 eV.

On the other hand, the electron energy loss measurements performed by Simpson *et al.*[26] in this energy range must be mentioned. They reported the first observation of optically forbidden transitions in the autoionization region of all the rare gases (including neon). These transitions appear in their spectra as resonant energy losses superimposed on the continuous background due to ionization losses, exactly as the absorption resonances observed by Codling *et al.*[27] in their photoabsorption measurements. These autoionizing-state resonances of the neutral atom are the parent states of the short-lived negative-ion states observed in the present work. For the sake of simplification, the term "resonance" will be used throughout this work with the meaning of "negative-ion resonance," as did Schulz[28] in his review on the resonances in electron impact.

In this paper, we report the detection of numerous resonances appearing in the differential excitation cross section of five states of neon in the autoionization region. The measurements are achieved by electron spectrometry in the forward direction. The energy positions of all the observed features are compared with available data obtained by means of other spectroscopies[2,6,11] which are able to detect such resonances. The effective strengths, the profile indices, the energies and the widths of the resonances are presented for the various decay channels and compared to the data obtained by electroionization.[6] Designations are proposed for the various states of Ne^-. The possibility of detecting perturbations caused by the presence of states of the neutral atom is discussed.

The processes by which the compound states can be observed may be seen in the following way. After a temporary negative ion has been formed in an excited state, this compound state can decay into channels energetically available.[28] Thus the resonance is likely to cause interferences in the measurements of processes whose thresholds are lower or near the resonance energy. The only resonances observed so far in differential excitation functions in the autoionization region[25] are type I (Feshbach) resonances.[29] However, according to Taylor *et al.*,[29,30] type II (shape) resonances are likely to appear in excitation functions, and actually strong manifestations of them were observed in differential excitation functions below the ionization threshold in helium by Ehrhardt *et al.*[16,17] This was afterwards confirmed by many theorists.[31-33] Thus type II resonances can be expected to appear even in the autoionization region.

Besides these facts, many experimenters reported the observations of threshold features ("Wigner cusps") at the opening of new channels (i.e., at the positions of states of the neutral atom) in total[2] and differential elastic cross sections.[34,35] Moreover in a recent theoretical work, Oberoi and Nesbet[32] calculated that such features can appear in the excitation cross section of helium (below the ionization threshold) in addition to type I and II resonances. Thus it can be anticipated that these features could eventually appear in the autoionization region of excitation functions. But, as it will be shown hereafter, if the energy positions of the observed features are compared with the known positions of the states of the neutral atom, it seems more reasonable to interpret most of them as negative-ion resonances.

II. EXPERIMENT

The electron spectrometer used in these measurements was previously used for a study of electron energy loss in neon below the ionization threshold.[36] Recently it has been extensively described,[37] and therefore only its main features are presented here. This spectrometer, schematically shown in Fig. 1, involves an electron gun, electrostatic lenses, a 127°-cylindrical monochromator and analyzer, a collision chamber with rotatable exit slit, and an electron multiplier detector. The energy resolution of the over-all spectrometer was measured to be as high as 20 meV in short-duration transmission measurements.[37] For the long-duration measurements presented here, the over-all energy resolution has been evidenced to be near 30 meV. The angular resolution of the spectrometer is about ±3°. The scattering length is 3.3 cm. During the present work, the pressure in the scattering chamber was about 2×10^{-3} Torr, corresponding to a gas density of about 7×10^{13} atoms/cm^3. To measure the excitation function to a given state, the energy analyzer is tuned at a fixed energy loss and the incident electron energy is swept. Measurements are achieved with pulse-counting techniques and repetitive energy sweeps.

III. RESULTS

The inelastic channels which were chosen correspond to the excitation of the $3s$, $3s'$, $3p'$, $4s'$, and $3d$ states. According to our study of electron impact excitation of neon below the ionization threshold,[36] these channels are the most prominent transitions for the range of excitation energy involved here (42–50 eV), thus providing the best signal-to-noise ratio in the measurements. For each channel, the accumulation of data lasted from one week to several weeks for the less strong peaks, before it reached a convenient signal-to-noise ratio. On the other hand, these transitions involve the three main angular momentum changes which are possible for the excitation of a p electron from the ground state. These transitions also provide many types of continua for the study of the configuration interactions[38] with the discrete states detected in the autoionization region.

The differential excitation functions of these five states, measured between 42 and 46 eV, are presented in Fig. 2. In addition, the measurement of the excitation function of the $3s'$ state is presented in Fig. 3 from 44 to 50 eV. The angle of observa-

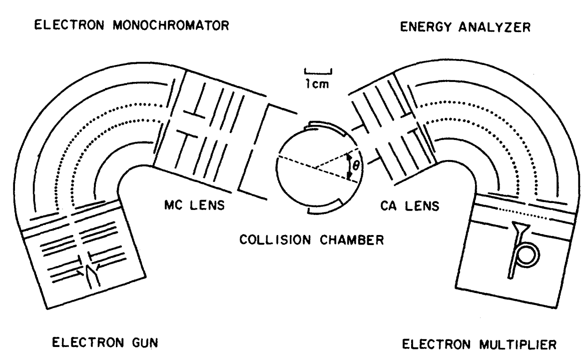

FIG. 1. Schematic diagram of the electron spectrometer.

tion is 0° for all the measurements. These curves have been "straightened through smoothings," a technique used in order to flatten the background of the experimental curves.[39] A "straightened" curve results after subtraction, from the original data, of a reference curve following its general shape. The reference curve is obtained by smoothing the original curve N times ($N=100$ here). The general properties of this technique were fully described and analyzed by Carbonneau *et al.*[39]

The features which appear in the curve of the $3s'$ state (which has the best signal-to-noise ratio) are identified by numbers, and their energy positions are given in Table I. These data are compared with those of Sanche and Schulz,[2] Grissom *et al.*,[11] and Bolduc *et al.*[6] The energy calibration has been achieved by means of transmission measurements of the resonance[1] in neon at 16.04 eV. The accuracy of the energy values is about ±0.06 eV.

Among the features which are observed in these

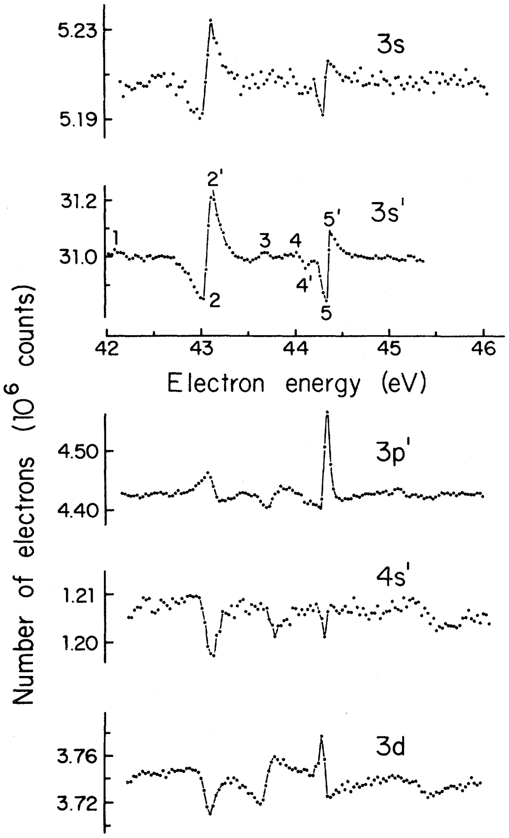

FIG. 2. Measurements of the differential excitation functions of the states $3s$, $3s'$, $3p'$, $4s'$, and $3d$ of neon by electron impact in the autoionization region (42–46 eV; $\theta=0°$). The curves have been straightened by subtraction of the original curve smoothed 100 times. The energy positions of the features are given in Table I.

measurements, a few have the typical resonance shape. Their cross-section variations can be described by the Fano-Cooper formula[40]:

$$\sigma(\epsilon)=\sigma_a[(q+\epsilon)^2/(1+\epsilon^2)]+\sigma_b\,,$$

where $\epsilon=(E-E_r)/\frac{1}{2}\Gamma$. E is the electron energy, E_r the resonance energy, and Γ the natural width of the state. σ_a is the resonant portion of the cross section, σ_b the nonresonant portion, and q is the line-profile index. For the strongest resonances observed in these measurements it is possible to give the characteristics E_r, Γ, q, and also the strength of the resonance.[25] Indeed the cross-section variations $\Delta\sigma$ during the resonance can be determined relative to the off-resonance value σ of the cross section. This effective strength is given by

$$\frac{\Delta\sigma}{\sigma}=\frac{\sigma_a}{\sigma_a+\sigma_b}\,(q^2+1)=\rho^2(q^2+1),$$

where ρ^2 is the resonant fraction of the cross section, according to the definition of Simpson *et al.*[25]

These characteristics have been determined for the resonances No. 2, 3, 5, 7, 10 (for the various decay channels in the case of q and $\Delta\sigma/\sigma$). They are compared with the data obtained in the ionization channel[6] in Tables II and III. Our values have been determined by a graphical method, as suggested by Comer and Read.[41] Since most of the resonances are naturally broad, the usual experimental flattening and broadening effects[25,37,42] were not taken into account, except for the case of the resonance 5 which is particularly sharp. To evaluate the natural width of this resonance, we took into account our energy resolution and the

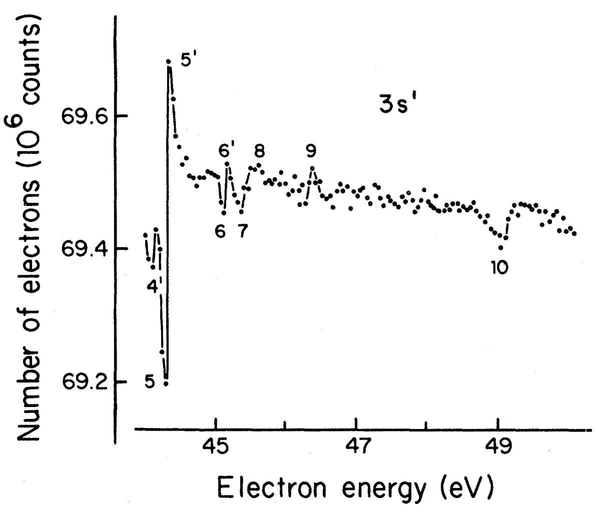

FIG. 3. Continuation of the measurement presented in Fig. 2 for the state $3s'$, from 44 to 50 eV.

TABLE I. Energy positions (eV) of the features observed in the channel $3s'$ of neon.

Feature No.	Roy *et al.* (Present work) Diff. excitation	Sanche and Schulz (Ref. 2) Transmission	Grissom *et al.* (Ref. 11) Trapped electrons	Bolduc *et al.* (Ref. 6) Ionization	Designation
1	42.11	41.98	42.10	...	$2p^4(^3P)[3s^23p(^2P)]$
2, 2′	43.02–43.13	43.00–43.11	...	43.12	$2s2p^63s^2(^2S)$
3	43.67	43.61–43.73	...	43.72 [a]	$2s2p^63s3p(^2P)$
4, 4′	44.01–44.11	...	44.00	...	$2p^4(^3P)[3s3p^2(^2S)]$
5, 5′	44.33–44.39	44.33–44.38	44.00–44.51	...	$2p^4(^3P)[3s3p^2(^2D)]$
6, 6′	45.15–45.22	45.18	...	...	$2s2p^63p^2(^2S)$
7	45.39	45.43	45.53	45.41	$2p^4(^1D)[3s^23p(^2P)]$
8	45.63	...	...	...	$2s2p^63p^2(^2D)$
9	46.39	...	...	...	$2s2p^64s^2(^2S)$
10	49.05	49.03	...	49.02	$2p^4(^1S)3s^23p(^2P)$

[a] This feature was afterwards attributed to autoionization (Ref. 49).

Doppler broadening (for the appropriate combination of mass, temperature, and incident energy[42]). In addition to the broadening effect, these experimental factors also attenuated the amplitude of this particular resonance, because of its sharpness (flattening effect), thus changing its effective strength. If the over-all effects of these factors are simulated by the convolution with a Gaussian distribution,[42,43] the strength of a resonance with this line shape is attenuated by about 25% while it is broadened from 35 to 50 meV. The values given in Table II for this resonance (No. 5) have accordingly been corrected.

A measurement was carried out in the elastic channel (at $\theta=15°$) between 42 and 46 eV, until a signal-to-noise ratio comparable to the $3s'$ measurement (Fig. 2) was obtained. This measurement was not carried out in the forward direction, since at 0° the tuning of the spectrometer for the measurement of the elastically scattered electrons actually permits the detection of the primary electrons[1] (transmission measurements). In such conditions, it becomes impossible to separate the elastic channel from the inelastic ones since the cross section involved is the total scattering cross section.[2] No feature could be observed in this channel. Thus we concluded that the manifestations of these resonances in the elastic channel are smaller than 0.1%. Nevertheless we think that in absolute value the cross-section changes could be of the same order as in the ionization channel[6]; but this is far beyond the detection limit of the present measurement. This observation is in agreement with the work of Simpson *et al.*,[25] who found no effect of the resonances in helium in the elastic channel at angles $\geqslant 3°$. The conclusions based on these measurements at 15° are indicated in Table II under the heading "Elastic."

TABLE II. Effective strengths and profile indices of the resonances in the various channels.

Resonance No.	Characteristics	$3s$	$3s'$	$3p'$	$4s'$	$3d$	Elastic [a]	Ionization [b]
2	q	1.15 ± 0.10	1.25 ± 0.05	−1.8 ± 0.4	−0.5 ± 0.1	−0.4 ± 0.2	...	0.7 ± 0.3
	$\Delta\sigma/\sigma$ (%)	0.86	1.2	1.1	1.0	1.1	$\ll 0.1$	0.01
3	q	...	...	0.7 ± 0.2	−0.3 ± 0.2	0.8 ± 0.1	...	−0.8 ± 0.2 [c]
	$\Delta\sigma/\sigma$ (%)	<0.1	<0.1	0.84	0.55	1.2	$\ll 0.1$	0.03 [c]
5	q	0.6 ± 0.1	0.80 ± 0.05	2.8 ± 0.3	0.0 ± 0.2	1.4 ± 0.1	...	...
	$\Delta\sigma/\sigma$ (%) [d]	0.7	1.0	5.0	0.8	2.0	$\ll 0.1$	...
7	q	...	...	...	...	...	...	−0.8 ± 0.2
	$\Delta\sigma/\sigma$ (%)	...	0.10	...	...	...	...	0.04
10	q	...	0.2 ± 0.1	...	...	...	...	−1.5 ± 0.2
	$\Delta\sigma/\sigma$ (%)	...	0.10	...	...	...	...	0.11

[a] These measurements have been carried out at $\theta=15°$.
[b] Reference 6.
[c] This feature was afterwards assigned to autoionization (Ref. 49).
[d] These values have been corrected for experimental flattening effects.

TABLE III. Energies and widths of the resonances.

Resonance No.	E_r (eV) Diff. excitation	E_r (eV) Ionization [a]	Γ (eV) Diff. excitation	Γ (eV) Ionization [a]
2	43.09± 0.05	43.12± 0.05	0.13 ± 0.03	0.15± 0.05
3	43.72± 0.06	43.72± 0.05 [b]	0.15 ± 0.05	0.19± 0.03 [b]
5	44.36± 0.05	···	0.035± 0.015 [c]	···
7	45.39± 0.08	45.41± 0.05	0.20 ± 0.05	0.20± 0.03
10	49.05± 0.08	49.02± 0.05	0.35 ± 0.10	0.50± 0.03

[a] Reference 6.
[b] This feature was afterwards assigned to autoionization (Ref. 49).
[c] The evaluation of the natural width of this resonance takes into account the broadenings caused by the Doppler effect and by the width of the electron energy distribution.

IV. INTERPRETATION

In order to achieve an interpretation of these results, we have compiled the electronic states of neon in the region of autoionization. The energy positions of these states are given in Fig. 4 and have been taken from the experimental data of Simpson *et al.*,[26] Bergmark *et al.*,[44] Codling *et al.*,[27] Comer and Read,[45] Edwards and Rudd,[46] and Bolduc *et al.*[6,38] A few theoretical values are available from the works of Weiss,[47] and also Parcell *et al.*[48] The energy diagram presented in Fig. 4 illustrates the relationships between the states of the neutral atom (column Ne) and the ten states of the negative ion possibly observed here (column Ne^-), corresponding to the ten features appearing in Figs. 2 and 3. The Roman numerals at the far right indicate the resonance type. The oblique lines between the two columns relate the states of Ne^- to their parent states (or the states of reference for the determination of the resonance type). A negative slope indicates a type-I resonance and a positive slope a type-II resonance. As established by Taylor *et al.*,[29] when the parent atom has many terms with the same electronic configuration, the reference for the determination of the resonance type is the energy position of the lowest term. The designations of the states are also given in Table I.

The first feature appearing in our measurements has been observed by other experimenters.[2,11] Sanche and Schulz[2] have attributed it to a negative-ion state associated with the parent $2s^2 2p^4(^3P)3s^2$, whose position was calculated by Weiss[47] at 42.18 eV and observed by Bolduc and Marmet[38] at 42.22 eV. The resonance No. 2 has been attributed by Sanche and Schulz,[2] and by Bolduc *et al.*[6] to the state $2s2p^6 3s^2(^2S)$ of Ne^-, the parent state being located at about 0.3 eV above, according to electron energy loss measurements,[26,44,45] ion impact excitation measurements,[46] and calculations.[47]

It is interesting to notice that the strength of the resonance (see Table II) is 100 times higher in differential excitation than in ionization.

The case of feature No. 3 is somewhat special. It appears very weakly in most of the channels, except in the channel 3*d* where it is very strong. We think that this feature is a shape resonance (type II) with the configuration $2s2p^6 3s3p(^2P)$. Its energy position relative to the parent state and to the other features (see Fig. 4) strongly favors

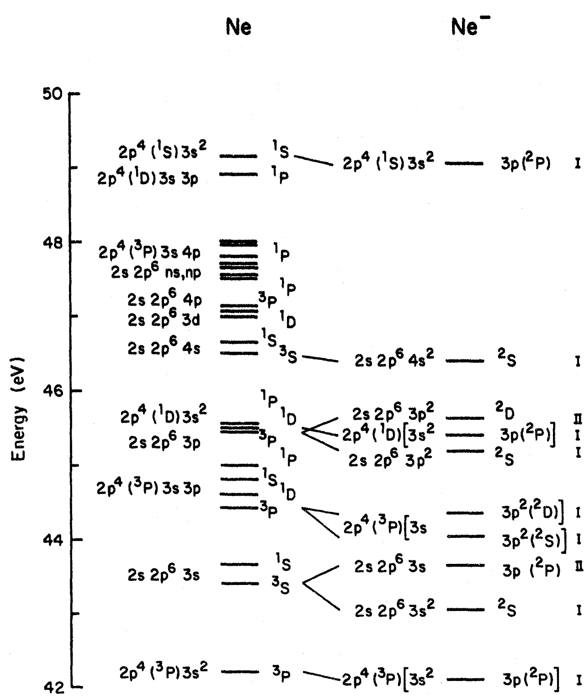

FIG. 4. Energy diagram of the states of Ne, and the ten states of Ne^- possibly observed here. The Roman numerals at the far right indicate the resonance type, and the oblique lines between the two columns relate the states of Ne^- to their parent states (or the states of reference for the determination of the resonance type).

this interpretation since the two subsequent resonances are clearly at a better position to be associated with the doubly excited states $2p^4(^3P)3s3p$. Nevertheless, since the profile of this feature is not clearly determined, it could be a threshold feature caused by the state $2s2p^63s(^1S)$ whose energy position is very close to it.[26,44-47] The feature which appears in the ionization channel[6] at this energy has afterwards been assigned to autoionization.[49]

The two following features (4 and 5) can be associated with the group of states $2p^4(^3P)3s3p$ with a convenient electron affinity. The first one was reported by Grissom *et al.*[11] without designation, while the second one was observed by Sanche and Schulz,[2] and by Grissom *et al.*[11] The latter have proposed the configuration $2p^4(^3P)3s3p^2$ for this Ne^- state. At about these energy positions, Bolduc *et al.*[6] have also observed features; they assigned them to autoionizing states. While in the transmission measurement[2] and in ours these features appear very sharp, in electroionization and trapped-electron measurements they are clearly broader. This observation is in agreement with the interpretation of Bolduc *et al.*[6] On the other hand, feature No. 5 specially deserves attention; as noted by Sanche and Schulz,[2] it is very narrow and strong. The energy width of the state has been evaluated at about 0.035 eV, maybe less, and its strength reaches 5% in the $3p'$ channel. Since this resonance is so strong, this can be an indication that the first member of the group of states $2p^4(^3P)3s3p$ could be located at about 44.4–44.6 eV.

The small feature observed by Sanche and Schulz[2] at 45.18 eV probably corresponds to feature No. 6 of Fig. 3. Its designation could be $2s2p^63p^2(^2S)$, associated with the state $2s2p^63p(^3P)$ located 0.3 eV above.[45,46] The configuration $2s2p^63p3d$ is also possible. The position of resonance No. 7 is in agreement with the observations of the other experimenters[2,6,11]; Bolduc *et al.*[6] have assigned it to the state $2s^22p^4(^1D)3s^23p$, the parent being located at 45.51 eV.

Resonance No. 8 cannot be associated with a parent state located at higher energy because this would provide an electron affinity excessively large. Thus we believe that this state can be a shape resonance, associated with the state $2s2p^63p$. The designation would be $2s2p^63p^2$ with the ^2D term because of the restriction of the angular momentum for that type of resonance.[28,29] It is for this reason that the ^2S term has been assigned to resonance No. 6.

Since the state $2s2p^63s^2(^2S)$ appears so strongly in this measurement (No. 2), one can expect the next member of the series to be observed too. Actually feature No. 9 appears at about 0.15 eV below the position[46] of the state $2s2p^64s(^3S)$. The variations of the electron current caused by this feature are very small, but it has been checked that its strength is clearly larger than the statistical noise. The last feature has already been observed experimentally[2,6] and assigned by Bolduc *et al.*[6] to the state $2s^22p^4(^1S)3s^23p(^2P)$; they observed the parent state at 49.14 eV. It is interesting to notice that this resonance appears with about the same strength in the ionization channel (Table II).

Within the energy band located between features 9 and 10 (see Fig. 4), there are many states of the neutral atom observed experimentally,[6,26,27,44-46] while no feature clearly appears in our results. On the other hand, in the data obtained by transmission,[2] threshold,[11] and ionization[6] spectroscopies, numerous features are detected in that region. We think that these features are caused by autoionization (i.e., by the presence of the states of the neutral atom) as interpreted by Bolduc *et al.*[6] Since they do not appear in our results, this confirms the fact that the features which have been evidenced in our differential excitation measurements are mainly (may be all of them) caused by negative-ion resonances.

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