Electron-impact study in valence and autoionization resonance regions of argon

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The recently built electron-energy-loss spectrometer (EELS) (typical FWHM 60 meV) was employed to measure the EELS spectra of argon in the discrete and autoionization resonance regions at a 2.5-keV impact energy and a mean scattering angle of 0°. Relative differential optical oscillator strength spectra were established by multiplying the EELS spectra by the known Bethe-Born conversion factor of the spectrometer, and were normalized at a single point in the smooth continuum using the absolute values reported by Chan *et al.* [Phys. Rev. A **46**, 149 (1992)]. The absolute oscillator strengths corresponding to these energy regions are reported and compared with previously published experimental and theoretical values. The values of parameters q, ρ^2, E_r, and Γ for autoionization resonances involving states $3s3p^6np\ {}^1P_1^0$ ($n=4$, 5, and 6) have been determined.

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I. INTRODUCTION

The information of absolute optical oscillator strengths of argon in discrete and continuum electronic excitation regions is significant in important areas of application such as plasma physics, biophysics, and astronomy. Moreover, as a heavier atom, argon shows significant nonhydrogenic behavior, so the study of the electronic excitation of argon provides valuable quantitative information for the testing and development of theoretical methods. Previous works about argon have been quoted in detail by Chan *et al.* [1].

For the excitation of argon in a discrete region, theoretical calculations [2–6,9–11] and experimental data [12,13,15–21,24,25,27] have primarily given oscillator strength values for the $3s^23p^5(^2P_{3/2})4s$ and $3s^23p^5(^2p_{1/2})4s'$ states, with considerable variations between them. Experimentally, because of the extremely narrow natural linewidths in the discrete region, Beer-Lambert photoabsorption measurements are no longer available since significant errors may have arisen due to the "line-saturation" (i.e., bandwidth) effect. The line-saturation effect, which affects the measurement of the absolute photoabsorption cross section, has been discussed in detail in Refs. [28–31]. Several alternative experimental methods that have avoided the line-saturation problem have been employed to determine discrete optical oscillator strengths of argon, such as self-absorption [16,23,27], lifetime [12,14,17,24], pressure-broadening profile [13,19,20], and electron-impact-based methods [1,15,18,21,22,25,26]. It should be mentioned that these techniques are somewhat complex and are often severely restricted in their range of application except for the electron-impact-based methods.

In the autoionization resonance region of argon, Fano [32,33] introduced the parametrization of autoionization resonances. Burke and Taylor [34] have calculated the parameters of some autoionizing states involving the excitation of the inner-valence $3s$ electrons of argon using the R-matrix method. Several experimental groups [1,35–43] have studied some details of the autoionization region.

In this paper, our interest is concentrated on these energy regions for the discrete transitions and autoionization resonances involving excitation of the inner-valence $3s$ electrons of argon, i.e., corresponding to 11.0–16.1- and 25.0–30.1-eV energy regions, respectively. Using the highly accurate electron-energy-loss spectroscopy (EELS) method, which is described in detail in Ref. [31], the absolute optical oscillator strength density spectra for the electronic transitions corresponding to these regions are reported. The absolute optical oscillator strength values for discrete transitions and the quantities q, ρ^2, and linewidth Γ for the autoionization resonances are determined. These parameters were defined by Fano [32,33] to characterize each profile of the absorption lines in the atomic and molecular autoionization continuum. The results are compared with other experimental and theoretical values.

II. EXPERIMENTAL METHOD

The recently built high-resolution electron-energy-loss spectrometer, the design and operational details of which have been described in Ref. [44], was employed to obtain the electron-energy-loss spectra. Briefly, the apparatus consists of an electron gun, a hemispherical electrostatic monochromater, a rotable energy analyzer of the same type, and a reaction chamber. All of these components are enclosed in four vacuum chambers, respectively. Figure 1 shows a schematic diagram of the experimental apparatus. The impact energy can be varied from 1 to 5 keV. The measurements were operated at a 2.5-keV impact energy [typical full width at half maximum (FWHM) of 60 meV] and a mean scattering angle of 0° (the acceptance angle is 2.0×10^{-4} sr). The spectra measured in the experiment were the sum of many repetitive scans, and they have been recorded with an energy-loss interval of 10 meV. The background pressure in the vacuum chambers was 3.0×10^{-5} Pa and when the gas sam-

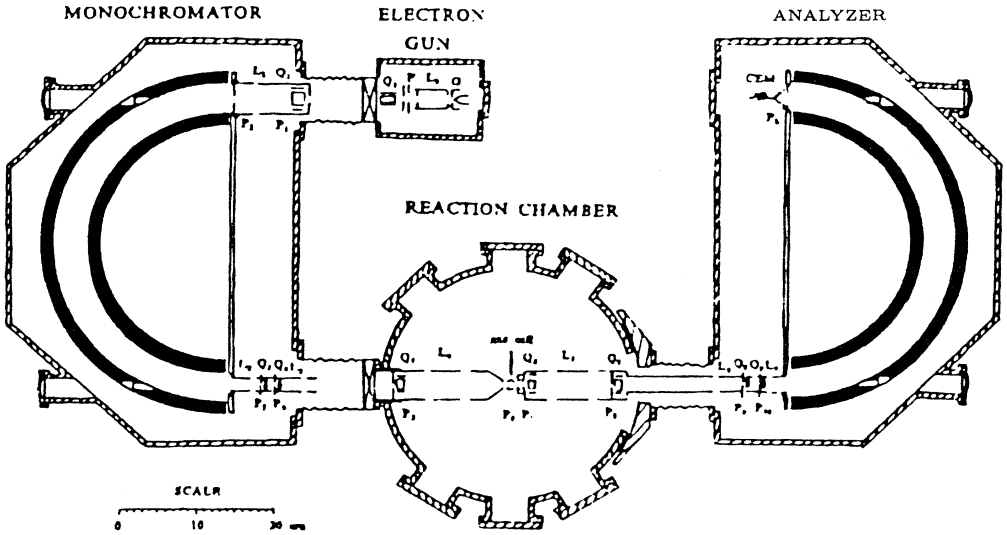

FIG. 1. Schematic diagram of the electron-energy-loss spectrometer.

ple was entered, the pressure in the reaction chamber rose to 8.0×10^{-3} Pa. Relative differential optical oscillator strength spectra were established by multiplying the electron-energy-loss spectra by the Bethe-Born conversion factor of the spectrometer. The method used was referred to in Ref. [31] and determined in Ref. [45]. The absolute optical oscillator strength values were obtained by normalizing at a single point in the smooth continuum using the absolute values recently reported by Chan *et al.* [1].

For all of the above measurements, the contributions from background gases remaining at the base pressure were removed by subtracting the signal when the pressure of argon was one-fifth of the gaseous sample pressure. The energy scale of the spectra were calibrated by using the discrete excitation peak corresponding to $3s^23p^5(^2P_{1/2})4s'$ state of argon (11.828 eV [46]).

III. RESULTS AND DISCUSSIONS

A. Absolute optical oscillator strengths for the discrete excitation region

Figure 2 shows the resulting spectra of argon in the discrete region (11.0–16.1 eV) normalized at 16.0 eV. The assignments of the various members of the nl and nl' series indicated in Fig. 2 were taken from Ref. [46]. The $nd[\frac{1}{2}]$ and $nd[\frac{3}{2}]$ states which converge to the same $^2P_{3/2}$ limit are labeled nd and $n\underline{d}$, respectively. The completely resolved peaks $4s$ and $4s'$ were integrated from the respective peak areas to determine their absolute oscillator strengths. For the partially resolved or unresolved peaks at higher energies, a least-squares curve fitting program was used to determine the absolute optical oscillator strengths of the respective peaks. Because these transitions have extremely narrow natural linewidths, neglecting the influence of the natural line profile of the discrete electronic transitions of argon is reasonable. The experimental line profile is well described by the instrumental function which can be described by the weighted sum of a 96% Gaussian profile and a 4% Lorentzian profile. Figure 2(b) shows the deconvoluted peaks and the absolute differential optical oscillator strength spectrum in the 13.5–16.1-eV energy region.

The resulting values obtained from the analysis of these spectra are shown in Table I. The experimental errors in this work mainly result from these factors: the errors have arisen from the Bethe-Born factor of the instrument, the statistical uncertainties, the double scattering, the uncertainties of gas pressure, and the uncertainties in making the data absolute. Moreover, the errors from the deconvoluting procedure should be considered for the partially resolved or unresolved peaks. The estimated errors in experimental measurements are listed in parentheses. Because previous works of argon in the discrete region have been tabulated by Chan *et al.* [1], only some major groups' results were listed in Table I. Clearly, the present measurements are in good agreement with the values previously reported by Chan *et al.* [1] for all transitions in Table I. The other electron-impact values obtained by Geiger [22] and Natali, Kuyatt, and Mielczarek [26] which were quoted in Ref. [23] are mostly consistent with the present work while for some states the discrepancies are up to 60%. For the optical measurements the data, using the self-absorption method performed by Westerveld [23], are lower than this work by 4–20%. The data of Lawrence [14] which employ the lifetime method show lower values for the $4s$ and $4s'$ states, while they are in good agreement with our work for the $5s$, $3\underline{d}$, $5s'$, and $3d'$ states. Theoretically, the values obtained by Lee and Lu [7] and Lee [8] show variations compared with our results; however, the values for the resonance $4s$ and $4s'$ lines reported by Aymar, Feneuille, and Klapisch [6] agree well with this work.

B. Absolute optical oscillator strength densities for the inner-valence excitation region

The energy region of 25.0–30.1 eV belongs to the autoionization resonance region involving the excitation of the inner-valence $3s$ electrons of argon. The resulting optical oscillator strength density spectrum, which was normalized in the smooth continuum at 25.0 eV, is shown in Fig. 3(a). One-electron excitation resonances of the $3s3p^6np\ ^1P_1^o$ series (where $n > 3$) are reported in this work and the assignments are taken from Ref. [38]. Although double-excitation resonances which are extremely weak have been observed in this energy region [38], they are not indicated in the present work. Similar to the situation for the electron-energy-loss spectra in the valence-shell excitation region, the present work shows agreement with the work of Chan *et al.* [1] in terms of shape and absolute values. Three very small peaks at 25.25, 27.55, and 28.30 eV can be observed in the present work. The electron-energy-loss spectrum at a 2.5-keV impact energy (80-meV FWHM) and a mean scattering angle of 2.0° in the same energy region was plotted in Fig. 3(b). By comparing Fig. 3(b) with 3(a), the respective ratio of areas for these three peaks to some optically allowed transition is high as the momentum transfer becomes large. For example, the ratio of the area peak at 27.55 eV to $3s3p^64p$ transition changes from 0.12 to 0.73 as the momentum transfer

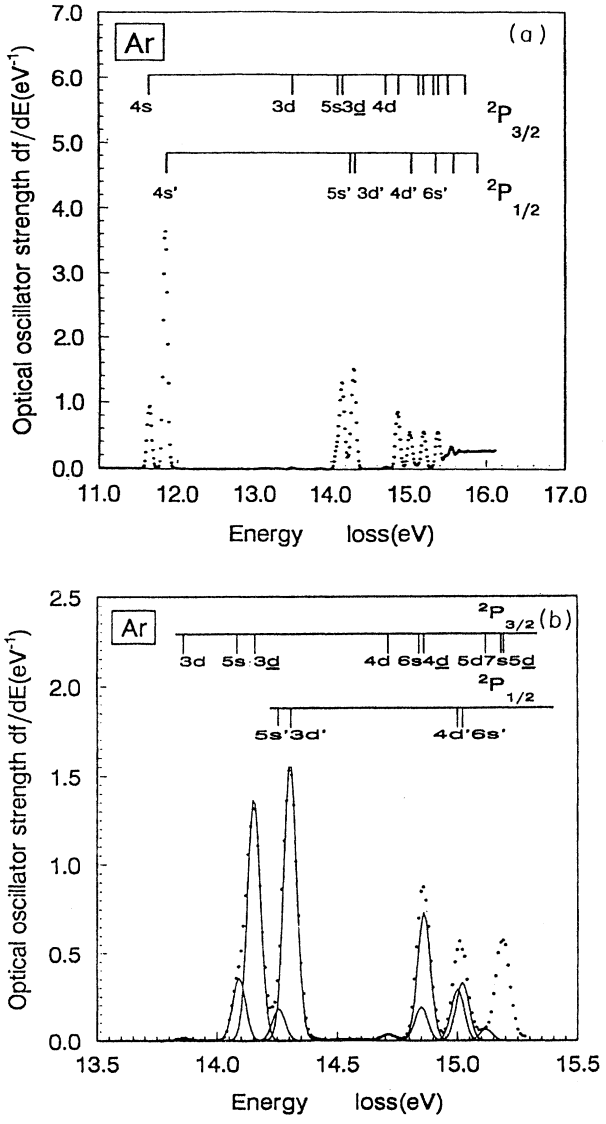

FIG. 2. Absolute optical oscillator strengths of argon in the discrete region. The assignments are taken from Ref. [46]. (a) 11.0–16.1 eV. (b) Expanded view of the 13.5–16.1-eV energy region. The deconvoluted peaks are plotted as solid lines.

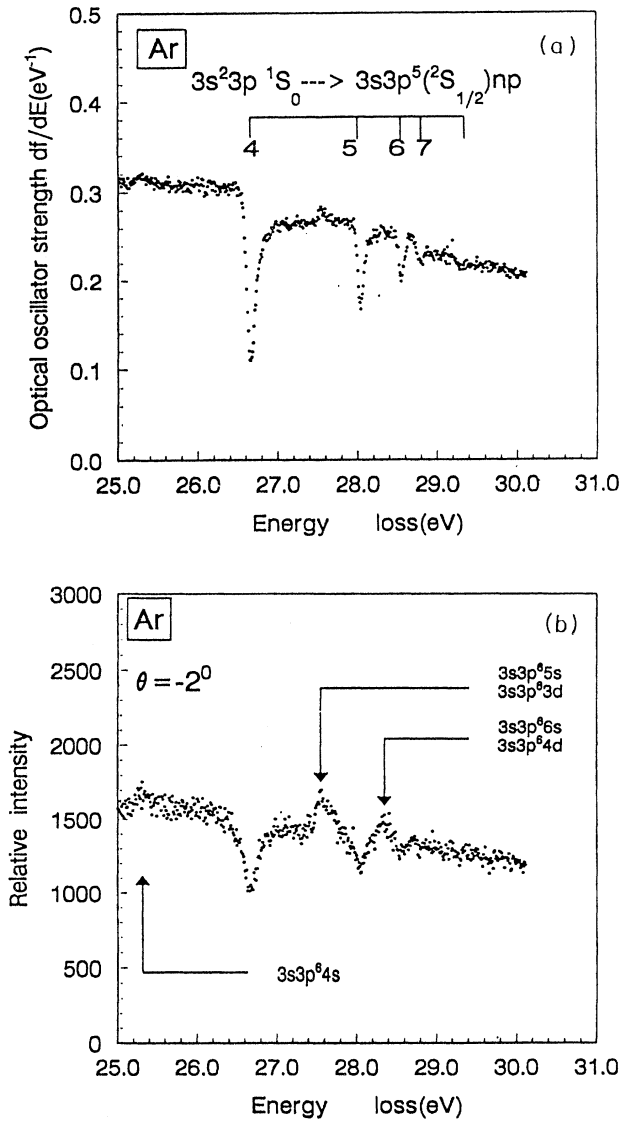

FIG. 3. (a) Absolute optical oscillator strengths of argon in the autoionizing resonance region of 25.0–30.1 eV. (b) The electron-energy-loss spectrum of argon in the 25.0–30.1-eV region obtained at 2.5-keV impact energy (80-meV FWHM) and a mean scattering angle of 2.0°. The states of interest are $3s3p^64s$ at 25.25 eV, $3s3p^65s$, $3s3p^63d$ at 27.55 eV, and $3s3p^66s$, $3s3p^64d$ at 28.30 eV.

TABLE I. Experimental and theoretical absolute optical oscillator strength values of argon in a discrete region from $3s^2 3p^8 \rightarrow 3s^2 3p^5 m$ where m is any of the configurations listed in the column headings.

	$(^2P_{3/2})4s$	$(^2P_{1/2})4s'$	$(^2P_{3/2})3d$	$(^2P_{3/2})5s$	$(^2P_{3/2})3\underline{d}$	$(^2P_{1/2})5s$	$(^2P_{1/2})3d'$
			Experiment				
(1) Electron-impact method							
Present work	0.0676	0.259	0.0010	0.0241	0.0929	0.0122	0.106
	(0.0040)	(0.015)	(0.0003)	(0.0029)	(0.0078)	(0.0032)	(0.010)
Chan *et al.* [1]	0.0662	0.265	0.0013	0.0264	0.0914	0.0126	0.106
	(0.0033)	(0.013)	(0.0001)	(0.0026)	(0.0091)	(0.0013)	(0.011)
Geiger [22]	0.066	0.255	0.0025	0.032	0.108	0.0108	0.097
Natali, Kuyatt, and Mielczarek [26]	0.070	0.278	0.0010	0.028	0.092	0.0124	0.110
(2) Optical methods							
Westerveld *et al.* [23]	0.063	0.240	0.00089	0.025	0.079	0.0106	0.086
(Absolute self-absorption)	(0.005)	(0.02)	(0.00007)	(0.002)	(0.006)	(0.0008)	(0.007)
Lawrence [14]	0.059	0.228		0.028	0.093	0.013	0.107
(Lifetime: delayed coincidence)	(0.003)	(0.021)		(0.002)	(0.006)	(0.003)	(0.015)
			Theoretical				
Aymar, Feneuille, and Klapisch [6]							
(1) dipole length	0.071	0.286					
(2) dipole velocity	0.065	0.252					
Lee and Lu [7]	0.080	0.210	0.0016	0.045	0.045	0.039	0.128
Lee [8]	0.059	0.030	0.0011	0.034	0.053	0.025	0.11

	$(^2P_{3/2})4d$	$(^2P_{3/2})6s$	$(^2P_{3/2})4d$	$(^2P_{1/2})4d'$	$(^2P_{1/2})6s'$	$(^2P_{3/2})5d$	$(^2P_{3/2})7s$	$(^2P_{3/2})5\underline{d}$
				Experiment				
(1) Electron-impact method								
Present work	0.0025	0.0129	0.0493	0.0197	0.0223	0.0043	0.0411	
	(0.0002)	(0.0023)	(0.0043)	(0.0020)	(0.0018)	(0.0006)	(0.0074)	
Chan *et al.* [1]	0.0019	0.0144	0.0484	0.0209	0.0221	0.0041	0.0426	
	(0.0002)	(0.0014)	(0.0048)	(0.0021)	(0.0022)	(0.0004)	(0.0043)	
Natali [26]	0.004	0.0094	0.048	0.015	0.0224	0.0032	0.0139	0.0234
Theoretical								
Lee and Lu [7]	0.0026	0.023	0.039	0.032	0.013	0.0043	0.013	0.030
Lee [8]	0.0031	0.014	0.036					

changes from 0.0056 to 0.23 a.u. It is clear that these three small peaks come from optically forbidden transitions. These nondipole transitions have arisen from the finite but very small momentum transfer. According to the data of the energy levels of argon calculated by Brion and Olsen [40] and Bergmark *et al.* [41], the peak at 25.25 eV is assigned to $3s3p^6 4s$ and the peak at 27.55 eV is then assigned to the configurations $3s3p^6 5s$ and $3s3p^6 3d$, while the peak at 28.33 eV is attributed to the transitions $3s \rightarrow 6s$ and $3s \rightarrow 4d$, but the intensity that has arisen from the $3s \rightarrow nd$ transitions is small. The energies of these transitions are listed in Table II. The structure in this energy region has also been studied by Rude and Lang [42], Simpson, Chamberlain, and Mielczarek [36], Gerbert, Morgenstern, and Niehaus [43], and Brion and Olson [40]. In the work of Simpson, Chamberlain, and Mielczarek [36], the energy positions of these three peaks were 25.8, 27.55, and 28.1 eV, and they did not identify the assignment of the peak at 28.1 eV. However, in the work of Chan *et al.* [1] the impact energy is 3.0 keV, the

TABLE II. Experimental and theoretical energy levels of optically forbidden transitions for argon (energies in eV).

	$3s \rightarrow 4s$	$3s \rightarrow 5s$	$3s \rightarrow 3d$	$3s \rightarrow 6s$	$3s \rightarrow 4d$
Experiment					
This work	25.25	27.55		28.33	
Simpson, Chamberlain, and Mielczarek [36]	25.8	27.55		28.1	
Theoretical					
Bergmark *et al.* [41]	25.22				
Brion and Olsen [40]		27.51	27.57	28.30	28.30

acceptance angle is 3.0×10^{-5} sr, and the momentum transfer is so small that these peaks cannot be detected in their work.

C. Analysis of resonance profiles

The Beutle-Fano profile of an isolated autoionizing resonance was represented by Fano [32,33] in the form

$$\sigma=\sigma_a(q+\varepsilon)^2/(1+\varepsilon^2)+\sigma_b \ . \quad (1)$$

Here $\varepsilon=E-E_r/\Gamma/2$ and the quantities E and E_r represent the energy loss and idealized resonance energy, respectively. $\Gamma/2$ a.u. is the half-width of the natural line of the autoionization state and q is called the line profile index which defines the shape of the absorption cross section. Finally, σ_a and σ_b represent two portions of the cross section that correspond to transitions of the continuum that do and do not interact with the discrete autoionization state, respectively. It is also convenient to define a correlation coefficient ρ^2 as it gives the proportion of the continuum which interacts with the autoionizing state,

$$\rho^2=\sigma_a/(\sigma_a+\sigma_b) \ . \quad (2)$$

Using the R-matrix theory, these resonance parameters of $3s3p^64p\ ^1P_1^o$ and $3s3p^65p\ ^1P_1^o$ have been determined by Burke and Taylor [34]. The experimental result was the only one reported by Madden, Ederer, and Codling [38] using a synchrotron radiation source.

Because the experimental profile of an autoionization resonance is the convolution of the natural line-shape function as described in Eq. (1) and the instrumental function of the spectrometer, a nonlinear least-squares parameter-fitting program including the calculation of the standard deviation of each parameter was used to fit the observed resonance transitions. It should be noted that these resonances were analyzed on the assumption that each resonance was considered independent. By comparing previously calculated cross sections [32,47] which allowed the members of the series to interact with each other through the same continuum with Eq. (1), it shows that the other resonance in the series contributed a negligible amount to the cross section in the neighborhood of any given resonance. Thus the one resonance approximation is appropriate for this analysis.

Table III shows the resulting data including previous work. The resonance energies determined in this work are very close to the data reported by Madden, Ederer, and Codling [38], so the energy (29.237 eV) of the series limit of Madden, Ederer, and Codling [38] was used to determine the quantum defect δ of each resonance line. The error caused by this procedure is estimated as 2%. From Table III it can immediately be seen that the quantities Γ, ρ^2, δ, and $n^{*3}\Gamma$ ($n^*=n-\delta$, i.e., the effective quantum number) are consistent with each other except for the single configuration calculation. The experimental data show that the quantity ρ^2, $n^{*3}\Gamma$ is constant for the members of this series within experimental error. But for the quantity q the present work gives a tendency to decrease for higher series, while the resulting work of Madden, Ederer, and Codling [38] shows a slightly tendency to decrease for a higher series and is constant within experimental error. Comparing with the theoretical values which only calculated the resonance parameters of $3s3p^64p$, $3s3p^65p$, the configuration interaction calculations seem to have the same tendency as ours. Fano and Cooper [33] suggested that q be a unique value for all lines of the Rydberg series that converge to the same level of the residual ion state, but they also emphasized that this thinking is tentative. In Fig. 4 a theoretical calculation of how the profile of the $3s3p^65p\ ^1P_1^o$ resonance is affected as q changes from

TABLE III. Experimental and theoretical profile parameters for the $3s3p^6np$ ($n=4,5,6$) series.

	$3s3p^64p\ ^1P_1^o$				$3s3p^65p\ ^1P_1^o$			$3s3p^66p\ ^1P_1^o$	
	This work	Madden [38]	Burke [34] SC	Burke [34] CI	This work	Madden [38]	Burke [34] CI	This work	Madden [38]
E_r (eV)	26.605	26.614			27.994	27.996		28.509	28.509
δ	1.727	1.723	1.639	1.678	1.692	1.690	1.651	1.682	1.682
	(0.034)				(0.034)			(0.033)	
Length			1.58	−0.33			−0.26		
q	−0.31	−0.22[a]			−0.21	−0.21		−0.18	−0.17
	(0.03)	(0.05)			(0.03)	(0.02)		(0.03)	(0.03)
Velocity			0.90	−0.29			−0.22		
Γ (meV)	76.1	80[a]	15	68	28.0	28.2	24.3	13.0	12.6
	(5.0)	(5)			(2.1)	(1.3)		(3.3)	(1.2)
Length			0.905	0.855			0.85		
ρ^2	0.87	0.86[a]			0.86	0.83		0.87	0.85
	(0.05)	(0.04)			(0.05)	(0.02)		(0.14)	(0.03)
Velocity			0.899	0.861			0.86		
$n^{*3}\Gamma$	0.89	0.94[a]	0.20	0.85	1.01	1.02	0.913	1.19	1.01
(eV)	(0.08)	(0.05)			(0.09)	(0.05)		(0.44)	(0.10)

[a]The quoted errors of the parameters for this resonance are estimated probable errors. The other errors of Madden are standard deviation determined by a computer.

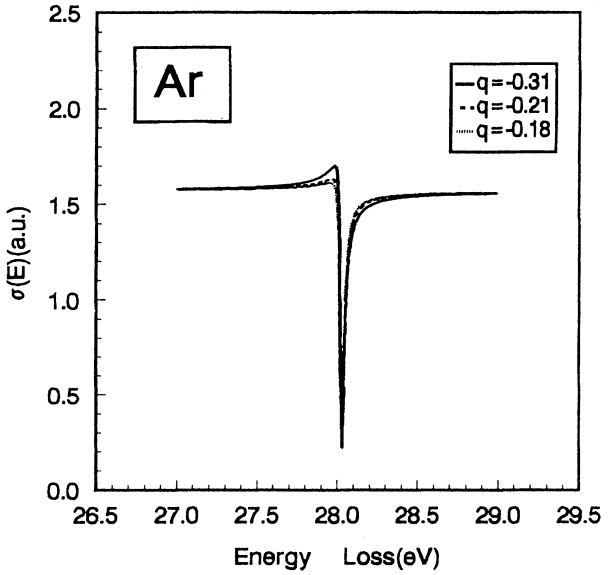

FIG. 4. Theoretical resonance profiles which best fit the $3s3p^6 5p$ peak. The figures show how the profile is affected as q changes from -0.31 to -0.18.

-0.31 to -0.18 is shown. It is clear that the shape of the resonance profile is not very sensitive when q varies in this range of values.

The major factors which affect errors in determining these quantities are listed as the following. Since the instrumental function cannot be accurately described, it may result in errors for the quantities Γ and ρ^2. However, it is sufficient to determine the profile index q which defines the shape of the absorption cross section, but the nondipole transition's existence may affect the determination of the quantity q. For the resonance of $3s3p^6 4p$ and $3s3p^6 5p$, the errors caused by instrumental resolution are small since the spectrometer resolution is sufficiently narrow. While for $3s3p^6 6p$, the uncertainty is much larger because of its extremely narrow natural linewidth. For the quantity E_r, the above factors have a negligible influence on its determination, and its uncertainty mostly arises from the experimental energy-loss interval which is 10 meV. The errors in parentheses for the present work result from above factors except for the influence of these nondipole transitions.

IV. CONCLUSION

The absolute optical oscillator strength data of argon in both discrete and autoionizing resonance regions are given using the recently built electron-energy-loss spectrometer at an impact energy of 2.5 keV (the typical FWHM is 60 meV) and a mean scattering angle of 0°. The electron-energy-loss spectra were transformed into the absolute oscillator strength spectra by multiplying the Bethe-Born conversion factor of the spectrometer and normalizing it at a single point in the smooth continuum region. In the discrete region, the present result were compared with the theoretical calculations and experimental data. Absolute optical oscillator strength densities for the autoionization resonance region involving the inner-valence $3s$ electron excitation have also been determined. The present work in these regions are, in general, in good agreement with the electron-impact-based data of Chan *et al.* [1]. The quantities q, ρ^2, Γ, and E_r of the autoionization resonance profile parameters obtained by deconvoluting the experimental peaks were compared with theoretical values and experimental data. It shows that the quantity q has a tendency to decrease for higher series members that are unsimilar to the previous experimental results [38]. The other quantities are well consistent with each other.

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