Electron correlation in the $3p^4 4s$ state of Ar studied via the $2p^5 4s$ resonant Auger spectra

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The $2p_{1/2,3/2}$ electrons of Ar have been excited selectively to the $4s$ orbital and the deexcitation has been recorded by utilizing the Auger resonant Raman effect. Intensity distribution resulting from the mixing between $3p^4 4s$ and $3p^4 3d$ final ionic states of the resonant Auger transitions has been clearly revealed. Experimental results are compared with the results of multiconfigurational Dirac-Fock calculations. [S1050-2947(96)01709-X]

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I. INTRODUCTION

When a core electron is excited to an unoccupied bound state, the vacancy can be filled via a resonant Auger process where the excited electron takes part in the recombination process (participator Auger process) or stays as a spectator (spectator Auger process). Spectator Auger process is usually the dominating decay channel and in the first approximation the spectra are expected to be similar to the normal Auger electron spectra except an overall energy shift due to the shielding by the spectator electron. Quite recently this so-called strict spectator model has been shown to fail completely in explaining the high-resolution resonant Auger electron spectra of Kr and Xe [1–3]. This is because the coupling between the spectator electron and the core holes created by the Auger decay splits the parent levels to energetically close lying daughter levels. Changes in the intermediate coupling conditions cause a remarkable redistribution in the population of the parent final states as compared to the normal Auger electron spectra. The strict spectator model has been, however, assumed to explain rather well the main features of the resonant Auger spectra taken at the first resonance of Ar, the $2p^5 4s$ resonance [4]. Because the final double-core-hole states of Ar are close to the LS coupling the spectator electron does not change the mixing of core-hole states having different total orbital angular momenta since it introduces primarily an electrostatic perturbation [5].

Intensity distribution of the $2p^5 4s$ resonant Auger decay, however, differs remarkably from the predictions based on the strict spectator model, as will be discussed in more detail below. The occurence of spectral features can be traced back to be caused by the strong mixing of final ionic configurations (FISCI). The first hints of this were reported by Meyer *et al.* [6] who observed some broadening of the spectral lines. The same authors also predicted an intensity redistribution in the spectrum caused by the FISCI. Their moderate resolution did not allow for further studies of the strength of the effect. Very recently de Gouw *et al.* measured the same spectra with a slightly better resolution [7]. Their results agreed with those of Meyer *et al.* [6] but were not accurate enough to allow for detailed comparison between experiment and theory.

High-resolution measurements of the Ar $2p^5 4s$ resonant Auger spectra are rather difficult due to high energies of photons and electrons in association with relatively small cross sections for selective excitations of the $2p$ electrons. Recent experimental progress at beamline 51 at the Max-laboratory has provided powerful tools to study the strength of FISCI in the spectra. The spectra now recorded reveal nicely resolved fine structures which form a far more solid basis for comparison between experiment and theory than before [4,6,7]. In Ref. [6] the FISCI between the $3p^4 4s$ and $3p^4 3d$ configurations could be seen indirectly as a shifting and broadening of the $3p^4(^1\mathrm{D})4s$ peak as compared to the $3p^4(^3\mathrm{P})4s$ and $3p^4(^1\mathrm{S})4s$ peaks. Our results show different fine structures in the case of each peak. This means that comparision between the linewidths of the main peaks where the fine structure is smeared out due to a moderate resolution, is not so straightforward. De Gouw *et al.* [7] could actually see two partly separated peaks corresponding to the transitions to the final states with pronounced $3p^4 4s$ or $3p^4 3d$ character in their spectra. They were not, however, able to reveal the fine structure and determine the partial intensities of the lines.

II. EXPERIMENT

A. Measurements

The measurements were carried out on the so-called Finnish beamline (BL 51) [8] at the MAX-laboratory in Lund, Sweden. Details of the measurements of the Ar resonant Auger spectra taken at several resonances will be discussed elsewhere [9]. Therefore only a short summary of the results will be given here.

High-energy parts of the electron spectra taken at $h\nu = 244.4$ eV and 246.5 eV corresponding to the $2p_{3/2} \rightarrow 4s$ and $2p_{1/2} \rightarrow 4s$ excitations, respectively, are shown in Fig. 1. The narrow bandwidth of the synchrotron radiation (about 70 meV), which is clearly smaller than the inherent linewidth of the $2p^5 4s$ states (about 115 meV), enabled us to benefit from the resonant Raman effect [10]. As compared with the spectra reported by Meyer *et al.* [6] and by de Gouw *et al.* [7] the structure in our spectra is much better resolved.

B. Experimental results

The energies and intensities of the resonant Auger lines were determined with the use of a least-squares fitting rou-

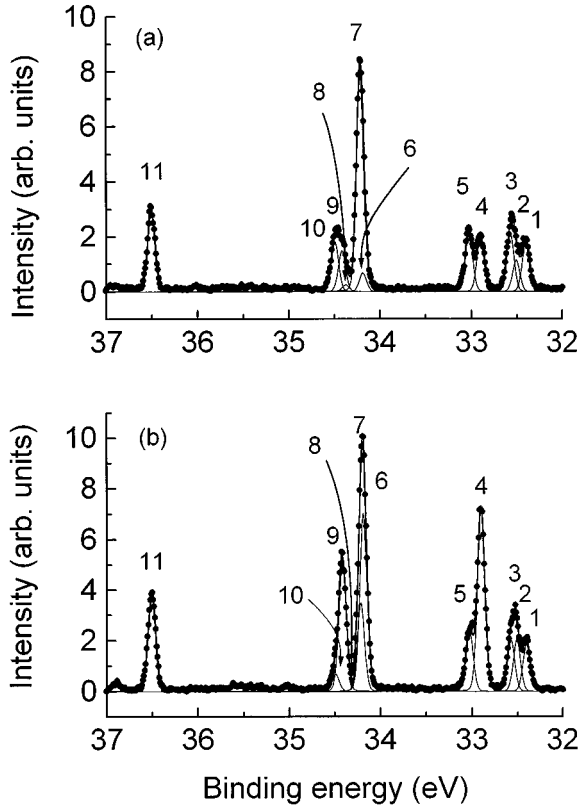

FIG. 1. Kinetic-energy region of lines 1–11 of the electron spectrum of Ar excited (a) by 246.5 eV photons and (b) by 244.4 eV photons. Line numbers refer to Tables I and II.

tine. The energy separation of the individual lines was taken from the energy splitting of the final-state levels determined by optical measurements [11,12]. The splitting was kept fixed during the fitting procedure. The linewidths and shapes were also kept fixed after they had been determined reliably. The first estimate for the linewidth was obtained after fitting the first five lines (lines 1–5 in Fig. 1). After that the eleven lines (lines 1–11 in Fig. 1) were fixed to have a common initial linewidth achieved above. Fitting procedure was continued but now the program was allowed to change the common initial linewidth. A total linewidth of 96 meV for individual line components was eventually achieved. The line shapes were well produced by Voigt functions. The product of the distribution of incoming photons and the Lorentzian lifetime broadening results in a shape of resonant Raman lines with the Lorentzian tails cut off [13]. Such line shapes are in close resemblance to the Voigt functions [14].

Energies and relative intensities of the individual lines are given in Tables I and II, columns 4 and 5. In column 6 the intensities of well resolved prominant peaks, due to partly overlapping line components, are also shown. The accuracy of the relative line intensities is not as good as that of the peak intensities due to the heavy overlap of close lying lines.

The intensity of line 8 (corresponding to the transitions to the final state assigned as $3p^4(^3P)3d\ ^2F_{5/2}$ in optical data [11,12]) is very weak but the presence of the line is clearly seen when the $2p_{1/2}4s$ resonant Auger spectrum is examined in more detail. We have included line 8 also in the case of the $2p_{3/2}^3 4s$ resonant Auger spectrum (Table II) in order to keep the line numbers consistent for the two spectra.

Final states in Tables I and II can also be occupied via direct photoionization. The $3s$ photoionization cross section is distributed between the main and correlation satellite channels due to the FISCI and channel interaction between the $3s3p^6\epsilon l$ and $3s^2 3p^4 nl\epsilon l'$ channels. The photoelectron spectrum, recorded below the resonances, was used to estimate the contribution of the direct photoionization in the population of the final states under study in present work. It turned out, however, that less than two percent of the total intensity at resonances is gained via direct photoionization.

TABLE I. Relative transition rates of the Ar $2p_{3/2}^3 4s$ resonant Auger transitions.

Final ionic state	J	Line no.	Energy (eV) [11,12]	Intensity (%) Expt.		Expt. [7]	Expt. [6]	Calc III		Calc IV		Calc V [6]
$3p^4(^3P)4s$	5/2	1	32.40	5.3(1)	16.4(3)	12.4	13	5.9	18.3	5.9	16.0	20
	3/2	2	32.51	5.8(3)				6.1		6.1		
	1/2	3/3′	32.57	5.3(2)				0.3/6.0		3.8/0.2		
$3p^4(^3P)4s$	3/2	4	32.90	20.6(5)	27.2(4)	24.7	29	24.3	32.1	24.3	34.1	31
	1/2	5	33.03	6.6(2)				7.8		9.8		
$3p^4(^1D/^3P)4s/3d$	3/2	6/6′	34.19	21.7(8)	29.3(8)	28.1	46	14.9/9.1	29.0	14.9/9.1	29.0	25
	5/2	7	34.21	7.6(1.1)				5.0		5.0		
	5/2	8	34.38	0.2(2)	16.2(1)	23.6		0.0	10.3	0.0	10.3	15
	3/2	9	34.42	14.4(4)				9.1		9.1		
	5/2	10	34.49	1.6(4)				1.2		1.2		
$3p^4(^1S)4s$	1/2	11/11′	36.50	10.9(2)	10.9(2)	11.2	12	7.9/2.4	10.3	9.9/0.7	10.6	9

TABLE II. Relative transition rates of the Ar $2p_{1/2}4s$ resonant Auger transitions.

Final ionic state	J	Line no.	Energy (eV) [11,12]	Intensity (%) Expt.		Expt. [7]	Expt. [6]	Calc III		Calc IV		Calc V [6]
$3p^4(^3P)4s$	5/2	1	32.40	7.0(1)	20.0(8)	15.6	19	9.3	24.9	9.3	24.5	24
	3/2	2	32.51	4.9(2)				5.5		5.5		
	1/2	3/3′	32.57	8.1(4)				0.4/9.7		9.1/0.6		
$3p^4(^3P)4s$	3/2	4	32.90	8.4(3)	17.6(2)	15.6	19	10.2	20.8	10.2	20.4	22
	1/2	5	33.03	9.2(2)				10.6		10.2		
$3p^4(^1D/^3P)4s/3d$	3/2	6/6′	34.19	9.0(5)	35.4(5)	36.3	53	3.5/3.6	33.7	3.5/3.6	33.8	25
	5/2	7	34.21	26.4(8)				26.6		26.6		
	5/2	8	34.38	1.1(4)	14.5(5)	20.8		0.2	8.9	0.2	8.9	18
	3/2	9	34.42	6.1(9)				2.5		2.5		
	5/2	10	34.49	7.3(1)				6.2		6.2		
$3p^4(^1S)4s$	1/2	11/11′	36.50	12.5(1)	12.5(1)	11.7	9	8.9/2.8	11.7	11.8/0.7	12.5	11

This indicates that the interference of direct photoionization and resonant Auger channels is of minor importance.

C. Comparison with previous results

Our values (column 6 in Tables I and II) can be compared with relative intensities of the main peaks reported by de Gouw *et al.* [7] given in column 7 (we normalized their values to 100% to agree with our normalization) and those of Meyer *et al.* [6] given in column 8. In addition to the fact that we are able to determine the intensities of individual transitions (column 5) our relative intensities of the main peaks also differ from the values reported earlier [6,7]. De Gouw *et al.* [7] noted that their relative intensities of the main peaks at higher binding energies are in general higher than the values reported by Meyer *et al.* [6]. De Gouw *et al.* assigned this to be due to the lack of transmission correction in the spectra of Meyer *et al.* However, our relative intensities for the main peaks are closer to those of Meyer *et al.* even though we measured our spectra much in the same way as de Gouw *et al.* We argue that in a short energy range the transmission does not change as strongly as proposed by de Gouw *et al.* The differences between the relative intensities of the main peaks in the spectra reported here, by Meyer *et al.* [6] and by de Gouw *et al.* [7] are probably caused by the fact that the main peaks are not well resolved in the spectra of Refs. [6,7]. This makes it very difficult to determine the relative intensities of peaks properly. In addition, statistics of the spectra reported by de Gouw *et al.* is not as good as ours, and the line shape in their spectra seems to be somewhat asymmetric due to asymmetric transmission profile of the electron analyzer [15]. We have used Voigt line shapes for each individual line in the fitting procedure, which reproduced well the measured symmetric line shapes. De Gouw *et al.* used pure Lorentzian line shapes for peaks even though the shapes obviously were far from Lorentzian. At the $2p_{1/2}4s$ resonance our spectrum is also free from the contribution due to the $2p_{3/2}\rightarrow 3d$ excitation.

III. DISCUSSION

A. Calculations

Relative transition rates were calculated using a single-channel multiconfiguration Dirac-Fock method [16]. In order to follow how the intensity distribution changes in passing from the normal Auger electron spectra to the resonant Auger electron spectra and how strong is the effect of FISCI, four different calculations were carried out. First, the normal Auger, second the spectator Auger, third and fourth the spectator Auger transitions including the FISCI (with two different basis sets) were reproduced. For the final ionic state we included all the jj-coupled configurations which result from the nonrelativistic configurations $3p^4 4s$ (calculation II), and configurations $3p^4 4s$, $3p^4 5s$ and $3p^4 3d$ (calculation III). The results of calculations I–III are depicted in Figs. 2 and 3. Results of calculation III are also given in Tables I and II, columns 9 and 10. Furthermore, we have used the total angular momentum values J to assign the final states in Tables I and II since the final states are strongly mixed of several LS-coupled states which all have a common value of J. Therefore, the LS-coupling scheme is not adequate in describing the states.

Final-state configurations $3p^4 ns$, $n=6,7,\ldots$ and $3p^4 md$, $m=4,5,\ldots$ were not included in our calculation III. This calculation is however, assumed to reproduce the intensity distribution of the transitions to the final states with $J=3/2$ and 5/2 reasonably well. There is, however, one extra peak in the calculated spectra labeled as 6′ which indicates the need of larger basis set. We have given the intensity of line 6′ together with the intensity of line 6 in Tables I and II since we assume that the use of a larger basis set would transfer most of the intensity of line 6′ to the line 6 nearby.

For the $J=1/2$ states the intensity distribution was recalculated using the mixing coefficients from a calculation with a larger basis set which included configurations $3s3p^6$, $3p^4 ns$, $3p^4 md$ where $n=$4-7 and $m=$3-7, $3s3p^4 3d4s^1$,

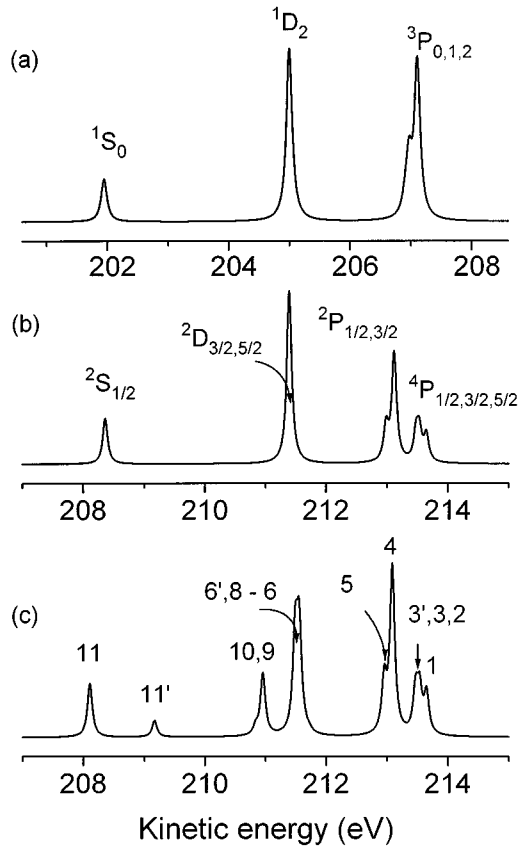

FIG. 2. Calculated spectra for the transitions (a) $2p^3_{3/2} \rightarrow 3p^4$ (referred to as calculation I in the text), (b) $2p^3_{3/2}4s \rightarrow 3p^4 4s$ (calculation II), and (c) $2p^3_{3/2}4s \rightarrow 3p^4 4s,3d$ (calculation III).

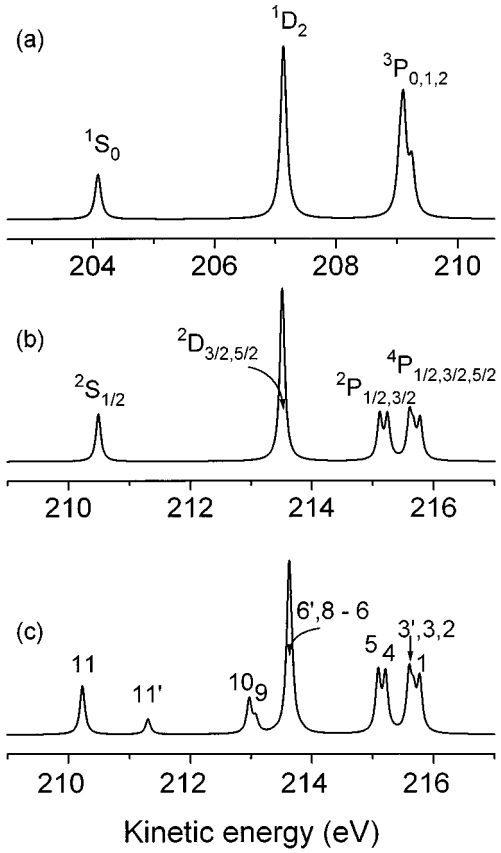

FIG. 3. Calculated spectra for the transitions (a) $2p_{1/2} \rightarrow 3p^4$ (referred to as calculation I in the text), (b) $2p_{1/2}4s \rightarrow 3p^4 4s$ (calculation II), and (c) $2p_{1/2}4s \rightarrow 3p^4 4s,3d$ (calculation III).

$3p^2 3d^2 4s$, $3s3p^4 4s^2$, $3s^0 3p^6 4s$, $3s3p^4 3d^2$, and $3s3p^4 4p^2$ (referred to as calculation IV). When the new mixing coefficients were used, most of the intensity distributed between the $J=1/2$ lines 3, 3′, 11, and 11′ [see Figs. 2(c) and 3(c)] was transferred to the lines 3 and 11. This is seen when comparing the values of columns 9 and 10 with those of columns 11 and 12 in Tables I and II. Because the $3s3p^6$, $J=1/2$ state is also involved in the mixing, the FISCI plays the most prominant role when $J=1/2$. Its omission in the calculations results in incorrect eivenvalues and eigenvectors for the $3p^4 4s$ configuration as well. In calculation IV the mixing of the $3p^4 4s$ and $3p^4 3d$ states with $J=1/2$ is much weaker than suggested by calculation III.

Theoretical values reported by Meyer *et al.* [6] are also given in Tables I and II. Their calculation (referred to as calculation V, column 13) included the final-state configurations $3p^4 4s$ and $3p^4 3d$.

B. Comparison between experiment and theory

1. Overall intensity distribution

Intensity distribution of the resonant Auger spectra changes clearly when an extra electron stays as a spectator during the decay as can be seen when comparing the profiles in Figs. 2(a) and 3(a) to those in Figs. 2(b) and 3(b). Calculations tend to overestimate the decay rate to the ^3P and underestimate the rate to the ^1D parent multiplet in both the normal and resonant Auger decay. Similar results were reported earlier for both the normal [16] and resonant Auger spectra [6]. The discrepancy, however, seems to decrease in passing from the normal to the resonant Auger spectra. This may indicate that correlation effects, which have been completely omitted in the case of the normal Auger spectrum, are of importance.

The difference between the profiles in Figs. 2(b) and 3(c) shows that the FISCI results in a redistribution of the intensity of the transitions to the $3p^4(^1D)4s$ final state. Our results confirm the earlier assumptions [6] that the extra splitting of the peaks due to the transitions to the $3p^4\ ^1D$ parent state is due to the mixing of $3p^4 4s$, $J=3/2,5/2$ and $3p^4 3d$, $J=3/2,5/2$ final ionic states (lines 6–10). The calculated intensity distribution is close to the measured one, when the mixing coefficients from calculation IV are used for $J=1/2$.

High resolution allows us to see how big is the difference between the spectra resulting from the decay of the two resonant states, $2p_{1/2}4s$ and $2p^3_{3/2}4s$. Not only does the branching ratio between the ^4P (lines 1–3) and ^2P (lines 4 and 5) peaks change, but also the branching ratio between the peak due to lines 6 and 7 and the peak due to lines 8–10 is different. The difference in the intensity distribution between the two spectra seems to be better reproduced by our calculations than by the calculations reported in Ref. [6].

TABLE III. Relative transition rates as a sum over J of the individual rates.

Final state	Initial state			
	$2p_{3/2}^3 4s$		$2p_{1/2}4s$	
	Calc.	Expt.	Calc.	Expt.
$J=1/2$	24.4	22.8	32.4	29.8
$J=3/2$	63.5	62.5	25.3	28.4
$J=5/2$	12.1	14.7	42.3	41.8

The measured intensity of the peak composed of lines 1–3 agrees better with our calculations than with those of Meyer *et al.* [6] in the case of the $2p_{3/2}^3 4s$ resonant Auger spectrum. For the $2p_{1/2}4s$ resonant Auger spectrum our value is almost identical with that of Meyer *et al.*, both of them being too big when compared to our experimental value. The relative intensity of the peak composed of lines 4 and 5 is somewhat better reproduced by our calculations than by the calculations of Ref. [6] when the $2p_{1/2}4s$ resonant Auger spectrum is concerned. In the case of $2p_{3/2}^3 4s$ resonant Auger spectrum the opposite is true. Our value for the sum of the intensities of lines 6 and 7 is clearly closer to the experimental value. Meyer *et al.* [6] seem to be able to reproduce the sum intensity of lines 8–10 better than we, especially in the case of the $2p_{3/2}^3 4s$ resonant Auger spectrum. Intensity of line 11 in both spectra is somewhat better reproduced by the calculations of this work than of Ref. [6].

2. Intensities of individual lines

The relative intensities of lines 1–5 are quite well reproduced by our calculations. Our calculated intensity ratio of lines 6 and 7 agrees quite well with the experimental value for the $2p_{1/2}4s$ resonant Auger spectrum. For the $2p_{3/2}^3 4s$ resonant Auger spectrum our calculations overestimate the intensity of line 6 and underestimate the intensity of line 7. This is not necessarily due to the incompleteness of the multiconfigurational Dirac-Fock (MCDF) calculations, but could also indicate that our fitting procedure was not able to solve the relative intensities in the case of lines 6 and 7. The branching ratio for lines 9 and 10 is well reproduced by theory for the $2p_{3/2}^3 4s$ resonant Auger spectrum whereas the opposite holds true for the $2p_{1/2}4s$ resonant Auger spectrum. Here the accuracy of the experimental values is not very high, however.

The relative intensities of the individual lines have now been reported. This makes it possible to sum the intensities over the total angular momentum J and compare the sums with the theoretical ones. As seen from Table III the theory seems to reproduce the observed sum intensities fairly well. This indicates that the Auger amplitudes are well reproduced by theory, whereas the discrepancies in the case of individual partial rates are related to the problems in calculating the mixing coefficients properly.

3. Energy splitting

The capacity of the MCDF calculations to account for FISCI can be tested not only by comparing the calculated and experimental values for the intensity distribution but also for the energy splitting. We will use the energy splitting taken from calculation III in the following. The calculations overestimate the energy difference between the peaks ^4P (lines 1–3) and ^2P (lines 4 and 5) by about 0.06 eV and between the peak ^2P and the peak composed of lines 6–8 by about 0.31 eV. The calculated energy splitting of the individual line components inside the peaks ^4P and ^2P differs less than 0.01 eV from the measured values. Both the intensity distribution and the energy splitting of lines 1–5, which is less influenced by FISCI, is well reproduced by the present calculations. This indicates that the theory accounts very well for the spectator-core coupling.

The calculated energy splitting between the lines 6 and 7 is by 0.026 eV too big and the one between lines 7 and 8 by 0.076 eV. The calculations overestimate the energy splitting between lines 8 and 9 by 0.267 eV and between lines 9 and 10 by 0.04 eV. The position of line 8 is thus misplaced by theory, but since the line is very weak (see Tables I and II), this has no real effect when the calculated and experimental profiles are compared to each other. Here the FISCI is more important than in the case of lines 1–5. Even though the main features in the spectra are accounted for by calculations with limited basis set, finest details are not reproduced properly.

The calculated energy splitting between the ^2S (line 11) and the peak composed of lines 9 and 10 is already by 0.72 eV too big. The larger basis set used in calculation IV reduces the splitting considerably resulting in a better agreement with the experiment. Thus for $J=1/2$ the limited basis set is too rough.

4. General

Well resolved structure in the $2p^5 4s$ resonant Auger spectra of Ar has allowed us to carry out a detailed comparision between experiment and theory. The experimental and calculated relative intensities of individual lines, being a very sensitive test to the accuracy of method to include FISCI, have been found to be in a fairly good agreement to each other. Earlier works [6,7] did not report relative line intensities but the peaks intensities only, and thus the comparision could not reach the individual J values. Our results clearly indicate, that especially for $J=1/2$ final states an extended basis set is needed to arrive at a satisfactory reproduction of the experiment. Some minor discrepancies still remain between experiment and theory, indicating that the basis set used in the calculations is not large enough. A slightly better accuracy on the experimental side might also help in further development of the method. The eigenvectors used in our calculations are available from the authors.

IV. CONCLUSIONS

Fine structure in the $2p^5 4s$ resonant Auger spectra has been well resolved by utilizing the Auger resonant Raman effect which allowed us to record the spectra with a linewidth narrower than the natural lifetime broadening. A remarkably better accuracy is also obtained for peak intensities in present work as compared to Refs. [6,7], where the data handling was hampered due to lower resolution, lower sta-

tistics, and, curious line shape. Comparision between MCDF calculations and experimental results showed that the existence of extra peaks in the spectra could be explained to be due to strong mixing between $3p^4 4s$ and $3p^4 3d$ final ionic states. Achieved experimental resolution is now good enough to give an opportunity to test and develop theoretical calculation methods as far as configuration interaction is concerned.

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