

## Configuration interaction in argon $KL$ resonances

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Curious "doublet" resonances in the  $KL$  double photoexcitation spectrum of argon are explained as a strong admixture of  $3d^2$  excited states to the principal  $[1s2p]4p^2$  excitation. A simulated "ab initio"  $KL$  photoabsorption spectrum including shake-up and shake-off contributions agrees well with the experimental data.

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Multielectron transitions have been studied extensively in the photoabsorption spectra of argon. Within the prominent  $KM$  absorption structure, starting 20 eV above the argon  $K$  edge, numerous resonances and absorption edges have been identified as transitions to double excitation and excitation-ionization  $[1s3p]$  and  $[1s3s]$  states, on the basis of calculated energies. The analysis of the satellites in the fluorescence spectra at different excitation energies within the structure provided additional confirmation [1]. Subsequently, the absorption structure has been successfully reproduced in *ab initio* calculations of the photoabsorption cross section [2,3].

Decisive evidence of the  $KL$  photoabsorption structure, two orders of magnitude smaller [4], has been presented in a low-noise photoabsorption spectrum by Kuetgens and Hormes [5]. Two absorption edges 10 eV apart, each preceded by a resonance peak, have been shown to appear at the energies of the  $[1s2p]$  double vacancy states, grouped into a singlet and a triplet. An indication of an additional splitting in the more prominent singlet resonance has been confirmed in a high-resolution experiment by Deutsch, Maskil, and Drube [6], where both singlet and triplet resonances have been found split into two equal peaks, 1.7 eV apart. The large-scale features of the spectrum, as already shown in [5], are determined by the dominant singlet-triplet splitting of the  $[1s2p]$  state, shown clearly in the level schemes of doubly ionized as well as singly ionized and excited states. The prominent resonances are reasonably attributed to the  $[1s2p]4p^2$  states and the subsequent Rydberg series. However, their apparent doublet structure remains a puzzle, since half integer spins can only be associated with singly ionized states. The attempt of the authors in [5,6] to attribute the 1.7-eV splitting and other small spectral features indiscriminately to  $[1s2p]$  states of the Ar, Ar<sup>+</sup>, or Ar<sup>2+</sup>, wherever agreement with calculated energies is found, is not justified.

It seemed that a simple and coherent explanation should be found for the puzzling spectral feature which has no counterpart in any of the known absorption spectra of the noble gases [1,2,4,7]. For that purpose we first

remeasured the argon  $KL$  photoabsorption spectrum at the HASYLAB synchrotron radiation facility in Hamburg, and found a full agreement with the results of Deutsch, Maskil, and Drube [6]. Candidate excited states of neutral Ar $[1s2p]$  were recalculated within the MCDF model, using the GRASP computer code [8], with projection into the  $LS$  coupling scheme. In the dipole approximation only  $^1P$  states are accessible from the Ar ground state ( $^1S$ ). Even with that limitation the calculated level scheme is too rich to allow the identification of resonances solely by energies. The calculation of transition probabilities is required for the purpose and, ultimately, the simulated absorption spectrum. Following the procedure of Schaphorst *et al.* [7], a fully relaxed calculation of dipole matrix elements between many-electron determinant wave functions was performed. The coupling coefficients from the GRASP output were used to construct the contributions of the  $^1P$  transition amplitudes in the final atomic states. Figure 1 shows the results of this calculation. The  $[1s2p]4p^2$  multiplet [Fig. 1(a)] is too asymmetric in the singlet part to account for the observed feature. The next member of the Rydberg series, the  $[1s2p]4p5p$  multiplet, shows a similar structure [Fig. 1(b)]. The amplitudes, however, are too low to modify the dominant  $4p^2$  component into the observed shape.

The required modification of the  $4p^2$  level scheme is provided by the configuration mixing. Among configurations  $[1s2p]4s^2$ ,  $[1s2p]3d^2$ ,  $[1s2p]4p5p$ , and  $[1s2p]4s3d$ , only  $[1s2p]3d^2$  was found to mix strongly with  $[1s2p]4p^2$ . Indeed, a pair of levels with equal strengths of both components is found in the singlet group [Fig. 1(c)], giving it a distinctly doublet appearance. The mixing also affects the triplet levels and causes a slight intensity redistribution, suppressing the strongest line. Consequently, two close compound peaks in the triplet group and two resonances in the singlet group prevail over smaller lines.

For a full comparison of the calculated  $KL$  cross section with the experimental spectrum, the contributions of shake-up and shake-off transitions were estimated. The

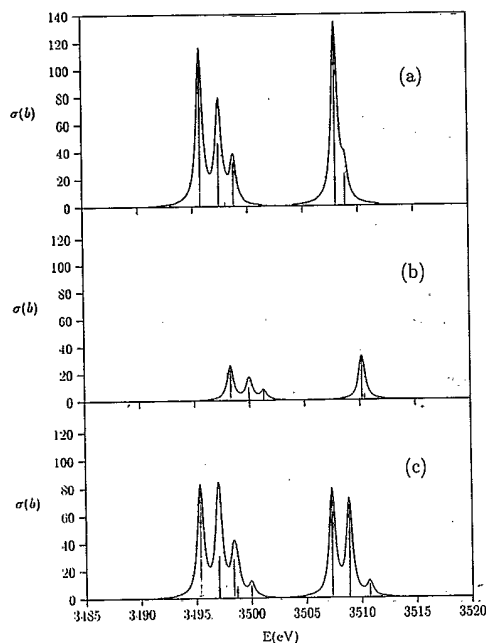


FIG. 1. Comparison of intensities for the resonant transitions of configurations: (a)  $[1s2p]4p^2$ , (b)  $[1s2p]4p5p$ , and (c)  $[1s2p]4p^2 + [1s2p]3d^2$  mixed configuration.

partial cross section per unit energy interval to  $4p\epsilon p$  continuum states was estimated from transition probabilities to the high-Rydberg states  $[1s2p]4pnp$ . The estimate is based on the assumption that the oscillator density of discrete states below an edge matches smoothly the oscillator density of the continuum at the edge [9]. The triplet-singlet pair of absorption edges is constructed in this way. The shake-off contribution, on the other hand, is approximated by an exponential saturation with the high-energy limit of 1.86% of the  $1s$  one-electron ioniza-

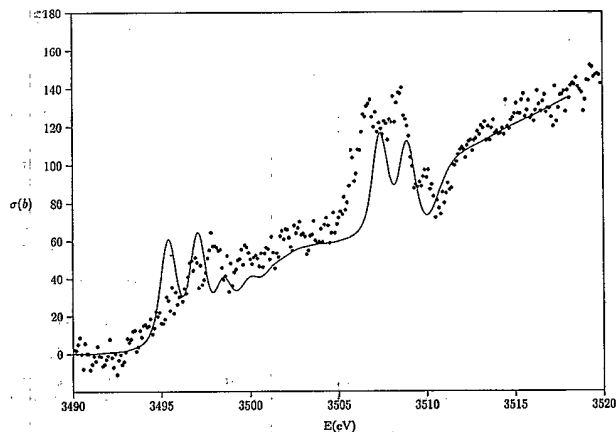


FIG. 2. Comparison between measured and calculated cross section of  $KL$  double-electron transitions in argon.

tion cross section, as predicted in the sudden limit [10], and the characteristic rise interval of 300 eV.

Figure 2 shows the calculated photoabsorption cross section compared to our measured spectrum. The calculated cross section is convoluted with a Lorentzian with 0.69 eV natural width and a Gaussian of 0.7 eV width to account for the experimental resolution. Our *ab initio* calculation of the argon  $KL$  absorption cross section explains for the first time the small resonance splitting and stands in a good overall agreement with the high-resolution measurements. The relatively simple singlet group of resonances are reconstructed particularly well. The complex shape of the triplet absorption structure, resulting from a large number of levels, and the discrepancy of about 1 eV in the energy position of its resonances call for a more elaborate configuration mixing. It is interesting that the  $4p^2 + 3d^2$  mixing does not cause an analogous splitting in the  $KM$  absorption spectrum [2]. There the much smaller singlet-triplet splitting of  $[1s3p]$  states keeps both groups of levels intertwined and obscures the small configuration interaction.

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