# $L_{2,3}M_1$ satellites in soft x-ray emission

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A systematic study of the  $L_{2,3}M_1$  (3s-2p) transition in elements from Cl to Cr reveals a satellite of unusually large intensity (~30% of parent line). Previous work on some of these elements led to the suggestion that the satellite is due to a two-electron shakeup (configuration-interaction) process. We compare our measurements of position, width, and intensity of the satellite as a function of atomic number with the predictions of this model and extend the theory to explain the apparent disappearance of the satellite outside this range.

#### I. INTRODUCTION

Shakeup effects are a part of all measured soft x-ray emission (SXE) spectra. A shakeup process is one in which a core transition perturbs and excites one or more of the surrounding electrons. Two extreme examples of this are the one-electron excitation (Auger process) and the collective excitation of electrons (plasmon sideband). Until recently, only these two processes were known to give rise to satellites of measureable intensity. Previous  $emission^{1-8}$  and x-ray photoelectron spectroscopy work<sup>9,10</sup> (XPS) gave rise to the suggestion that a twoelectron shakeup process might explain the satellite observed below the 3s-2p line of certain elements. It is unusual to have a two-electron shakeup process which is identifiable and which dominates both the one-electron and collective effects. This led us to undertake a comparison between the predictions of the two-electron shakeup model and our measured spectra. SXE data on the  $L_{2,3}M_1$  transition show an unusually large feature on the low-energy side of the parent line which we believe to be a satellite due to this two-electron shakeup process.

Cooper and LaVilla<sup>1</sup> observed a low-energy satellite in the  $L_{2,3}M_1$  x-ray emission spectra of Ar and KCl and were the first to propose a two-electron shakeup or "semi-Auger" process as the explanation for the feature. The present theory, using the configuration interaction method, was initially worked out by Dyall and Larkin<sup>7,11,12</sup> (DL) for similar satellites observed in photoelectron spectra and atomic Ar x-ray emission spectra.<sup>13</sup> The work of DL indicates that these satellites and their intensities are dependent of the physical characteristics of the ionizing probe. In this paper we modify the DL theory for atomic x-ray emission and apply it to the emission from solids. The results are compared to the properties of the satellites observed in SXE as a function of atomic number Z.

#### **II. EXPERIMENTAL**

In SXE spectroscopy one bombards the sample with electrons creating core holes. X rays are produced when higher-energy electrons make transitions into the core holes. If the core level is narrow, the x rays emitted give an approximate picture of the energies of the upper core levels or of the density of states of the valence band. In our experimental apparatus, a Pierce-type electron gun operating typically at 3 kV and a beam current of 1 mA is used to excite the sample. The x rays are dispersed via grazing incidence diffraction from a holographically etched toroidal grating and are detected by a phosphorcoated photodiode array. The energy resolution over the range of interest varies from 0.06 to 0.18 eV. Further details are described elsewhere.<sup>14-16</sup>

Samples used were typically of 99.9 + % purity and were etched in acid to decrease surface impurities. The sample chamber is held at  $10^{-9}$  Torr so that contamination during measurement is not a problem. Metal halides were used when necessary. Alkali-metals and the alkaline-earth halides decompose rapidly to pure metal under an electron beam.<sup>3</sup> The general trend of the susceptibility to radiation damage that we observe indicates that the higher the ionicity of the bond, the more likely decomposition will occur. On the Mulliken electronegativity scale,<sup>17</sup> the cutoff seems to be an ionicity of 0.30, that is, any compound with an ionicity above this will decompose readily. The ionicities of transition metal oxides generally exceed this critical value.

The data shown in Figs. 1-4 for Ar, K, Sc, and V, respectively, each contain three prominent peaks. The horizontal coordinate is photon energy in eV and the vertical is intensity in arbitrary units. All data have been corrected for instrumental and optical effects. The first peak (lowest in energy) is the satellite while the second and third are the spin-orbit split transition  $(L_3M_1 \text{ and } L_2M_1,$ respectively). The full set of data taken in this study ranged from P to Fe, but only elements Cl to Cr inclusive show the satellite with any discernible strength.

What is most surprising is the strength of the satellite. Most shakeup effects in SXE are on the order of 1% of the parent line. The intensity reported here is the integrated intensity of the satellite relative to the total observed transition probability. Fits to the data were carried out using a Lorentzian of the form

$$P_L(E, A, \mu, \Gamma) = \frac{A}{2\pi} \frac{\Gamma}{(E - \mu)^2 + (\Gamma/2)^2} , \qquad (1)$$

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FIG. 1. Ar satellite and 3s-2p transition. A is the satellite, B the  $L_3M_1$  transition, and C the  $L_2M_1$  transition. The feature on the high-energy shoulder of the parent line is a Mo transition from the substrate used.



FIG. 2. K satellite and 3s-2p transition. The deviation of the fit from the data at 262 eV is due to slight oxygen contamination (second-order  $K\alpha$  line).



FIG. 3. Sc satellite and 3s-2p transition. A is the satellite, B the  $L_3M_1$  line, and C the  $L_2M_1$  line.



FIG. 4. V satellite and 3s-2p transition. A is the satellite, B the  $L_3M_1$  line, and C the  $L_2M_1$  line.

TABLE I. Parameters from Lorentzian fit to satellite and parent line.  $\Gamma$  is the full width at half maximum,  $\Delta^{expt}$  is the satellite position relative to the energy centroid of the parent line, *I* is the ratio of the area of the satellite to the total transition probability, and  $\mu$  is the absolute position of the satellite.

the area of the satemet to the total transition probability, and $\mu$ is the absolute position of the satemet.								
Ζ	$\Gamma$ (eV)	$\Delta^{expt}$ (eV)	Ι	$\mu$ (eV)				
Cl	~3	~-6		~176				
Ar	5.1±0.5	$-10.7\pm0.3$	$0.28 \pm 0.04$	209.6±0.3				
K	6.4±0.3	$-14.1\pm0.2$	0.27±0.02	246.7±0.1				
Ca	11.1±0.3	$-16.5\pm0.6$	$0.28{\pm}0.05$	278.5±0.1				
Sc	14±2	$-16.2\pm0.3$	$0.29{\pm}0.05$	332.6±0.4				
Ti	18.7±0.7	$-17.3\pm0.9$	0.24±0.04	379±1				
V	23±2	-18±2	$0.22{\pm}0.01$	428±2				
Cr	28±5	-19.4±0.1	0.18±0.01	481.4±0.1				

for each of the peaks where A is the integrated intensity (area),  $\Gamma$  is the full width at half maximum, and  $\mu$  is the peak position. For all figures the points represent data and the solid line the fit. A summary of the results is presented in Table I.

The results given in Table I include the parameters from the fit as well as some calculated values. The relative position  $\Delta^{expt}$ , for example, refers to the satellite-peak position relative to the energy centroid of the parent line. The intensity I refers to the area of the satellite divided by the total area (satellite and parent line). As Z is increased, a clear pattern emerges: the half width increases, the intensity decreases, and the satellite moves further away from the parent line. Therefore, whatever model is used must explain these trends as well as the apparent disappearance of the satellite before Cl and after Cr.

### **III. THEORY**

The Ar data upon which the DL emission theory is based involves a transition between singly ionized states (Ar II-Ar II). In SXE the transitions are between neutral states (e.g., Ar I-Ar I) since the core holes created by the electron beam are quickly screened by electrons from surrounding atoms. The ions are screened before emission occurs since the screening time is on the order of the inverse plasma frequency ( $\sim 10^{-16}$  s) whereas the core hole lifetimes are considerably longer ( $\sim 10^{-14}$  s). It is for this reason that the DL theory will have to be modified before it can be applied to SXE data.<sup>18</sup>

If we follow the DL theory, then the configuration interaction method gives the initial atomic state function (ASF)  $\Phi_n^i(Z)$  and final ASF is  $\Phi_m^f(Z)$  with energies  $E_n^i$ and  $E_m^f$ . The ASF can be expressed as a linear combination of configuration state functions (CSF)  $\chi(Z)$  so that

$$\Phi_n^i(Z) = \sum_k b_{nk} \chi_{nk}^i(Z) , \qquad (2)$$

$$\Phi_m^f(Z) = \sum_j c_{mj} \chi_{mj}^f(Z) , \qquad (3)$$

where the CSF are Slater determinants of one-electron wave functions and  $b_{nk}$  and  $c_{mj}$  are the coefficients of the CSF in the initial and final ASF, respectively. The transition energies are given by

$$E_{mn} = E_m^f - E_n^i \tag{4}$$

and the transition rates by

$$\boldsymbol{R}_{mn} \propto \boldsymbol{E}_{mn}^{3} \left| \left\langle \Phi_{m}^{f}(\boldsymbol{Z}) \middle| \mathbf{r} \middle| \Phi_{n}^{i}(\boldsymbol{Z}) \right\rangle \right|^{2}, \qquad (5)$$

where  $\mathbf{r}$  is the dipole operator. The relative intensity of two lines which have the same initial state is given by

$$\frac{I_{mn}}{I_{pn}} = \frac{R_{mn}}{R_{pn}} , \qquad (6)$$

where density-of-states effects are assumed to be negligible.

The initial configuration set includes a mixture of configurations with the overall symmetry based on screened 2s and 2p holes. The DL theory indicates that

the mixing is small so that a single configuration (2p hole) can be used. Similarly, the final state will include configurations with symmetries based on screened 3s and 3p holes, i.e.,

$$\Phi_{m}^{f}[3s^{2s+1}\mathbf{L}] = c_{ml}|[3s]\rangle + \sum_{j} c_{mj}|[3p^{2}]nd\rangle + \sum_{k} c_{mk}|[3p^{2}]ns\rangle + \sum_{l} c_{ml}|[3s3p]np\rangle + \sum_{p} c_{mp}|[3s^{2}]ns\rangle , \qquad (7)$$

which DL showed can be approximated by the first three terms. We will assume, as did DL, that the satellite structure can be broadly discussed in terms of the singleconfiguration processes which contribute most to the intensity of the satellite.

The configurations used were determined by assuming the screening by the conduction electrons can be simulated by adding an electron to the lowest unoccupied orbital of the ion. Unfortunately, we did not have ready access to a multiconfigurational Hartree-Fock code, so that all calculations were done using a single-configuration Hartree-Fock program based on the code of Froese-Fischer.<sup>19</sup> An admixture of configurations will, of course, have a different energy than a single configuration but if the admixture is dominated by one configuration then the energy difference will be small (  $\sim$  few eV). It is not clear how the intensities from a single-configuration calculation can be compared to those from a multiconfigurational calculation, so they were not calculated. The results for the first few configurations of Cl to Cr are given in Table II. The appropriate LS term is given for both the initial and final states. In those cases where more than one LS term was allowed by the dipole selection rule, Hund's rules were followed and the lowest energy state was chosen. Unfortunately, the code could not calculate all configurations allowed so these have not been included. These configurations, as well as those neglected by the application of Hund's rules will, of course, contribute to the satellite structure.

### **IV. DISCUSSION**

The comparison between the theoretical and experimental relative position as a function of Z is given in Fig. 5. The theoretical results lie close to the data and show the same general trend with Z. This agreement supports the proposed explanation of the satellite. There are two shortcomings to this model, however. First, only single configurations were used to calculate the initial and final states, and second, the wave functions produced by the Fischer code are self-consistent atomic wave functions not solid-state wave functions. Since the energy levels will differ between the atomic and solid-state case, we have chosen to model the relative position rather than the absolute position thereby taking advantage of the self-consistency of the wave functions and their corresponding energies. Despite these limitations, the model is still quite promising.

Figures 6(a) and 6(b) show the experimental widths and intensities, respectively, as a function of Z. The trend is

Z	Initial states	Final states	$\Delta^{theor}$ (eV)	Z	Initial states	Final states	$\Delta^{\text{theor}}$ (eV)
Cl	$2p^{5}(^{2}P)3p^{62}P$	$3s^{1}(^{2}S)3p^{6}{}^{2}S$		Ti	$2p^{5}(^{2}P)3d^{3}(^{4}F)^{5}G$	$3s^{1}(^{2}S)3d^{3}(^{4}F)^{5}F$	
		$3p^{4}(^{1}D)3d^{1}$	-6.19		-	$3p^{4}(^{3}P)3d^{4}(^{5}D)$	-12.38
		$3p^{4}(^{1}D)4d^{1}$	+2.76			$3p^4(^3P)3d^4(^3G)$	-8.64
		$3p^{4}(^{1}D)5d^{1}$	+1.96			$3p^{4}(^{3}P)3d^{4}(^{3}F)$	-8.82
	a 5/2m / 13m	a 1/2m / 12m				$3p^{4}(^{3}P)3d^{4}(^{3}D)$	-7.15
Ar	$2p^{3}(^{2}P)4s^{13}P$	$3s^{-1}(^{2}S)4s^{-1}S$				$3p^4(^1D)3d^4(^5D)$	-11.56
		$3p^{4}(^{1}D)4s^{1}(^{2}D)3d^{1}$	-1.06			$3p^{4}({}^{3}P)3d^{3}({}^{4}F)[{}^{6}D]4d^{1}$	-11.58
		$3p^{+}(^{1}D)4s^{1}(^{2}D)4d^{1}$	-2.47			$3p^{4}(^{3}P)3d^{3}(^{4}F)[^{4}D]4d^{1}$	-25.84
K	$2p^{5}(^{2}P)4s^{2}P$	$3s^{1}(^{2}S)4s^{2}{}^{2}S$				$3p^{4}(^{3}P)3d^{3}(^{4}F)[^{6}F]4d^{1}$	-11.50
	•	$3p^{4}(^{1}D)3d^{1}$	-4.69			$3p^{4}(^{3}P)3d^{3}(^{4}F)[^{4}F]4d^{1}$	-23.01
		$3p^{4}(^{1}D)4d^{1}$	-8.44			$3p^{4}(^{3}P)3d^{3}(^{4}F)[^{6}F]5s^{1}$	-10.78
Ca	$2n^{5}(2D)2d^{1}3E$	$2n^{1/2}S(2d^{13}D)$				$3p^{4}(^{3}P)3d^{3}(^{4}F)[^{6}F]6s^{1}$	-11.92
Ca	2р (Г)3а Г	$3n^{4}(^{3}D)2d^{2}(^{3}E)$	8 07				
		3p(T)3a(T) $3p^{4}(^{3}p)3d^{2}(^{3}p)$	- 7.32				
		$3p^{4}(^{3}P)3d^{2}(^{1}D)$	-1.52				
		$3p^{4}(^{1}D)3d^{2}(^{3}F)$	-7.58	V	$2p^{5}(^{2}P)3d^{4}(^{5}D)^{6}F$	$3s^{1}(^{2}S)3d^{4}(^{5}D)^{6}D$	
		$3p^{4}(^{1}D)3d^{2}(^{3}P)$	-676			$3p^{4}(^{1}D)3d^{5}(^{6}S)$	-14.61
		$3p^{4}(^{3}P)3d^{1}[^{4}P]4d^{1}$	-9.47			$3p^{4}(^{3}P)3d^{4}(^{5}D)[^{7}P]4d^{1}$	-9.76
		$3p^{4}(^{3}P)3d^{1}[^{4}D]5s^{1}$	- 5 32			$3p^{4}(^{3}P)3d^{4}(^{5}D)[^{7}D]4d^{1}$	-14.23
		$3p^{4}(^{3}P)3d^{1}[^{4}D]6s^{1}$	-6.38			$3p^{4}(^{3}P)3d^{4}(^{5}D)[^{7}F]4d^{1}$	-11.35
		$3p^{4}(^{3}P)3d^{1}[^{2}D]6s^{1}$	- 17 11			$3p^{4}(^{3}P)3d^{4}(^{5}D)[^{5}P]4d^{1}$	-29.98
		$3p^{4}(^{1}D)3d^{1}[^{2}D]6s^{1}$	- 14.02			$3p^{4}(^{3}P)3d^{4}(^{5}D)[^{5}D]4d^{1}$	-27.57
		Sp (D) Su [D]	14.02			$3p^{4}({}^{1}D)3d^{4}({}^{5}D)[{}^{5}P]4d^{1}$	-28.13
Sc	$2p^{5}(^{2}P)3d^{2}(^{3}F)^{4}G$	$3s^{1}(^{2}S)3d^{2}(^{3}F)^{4}F$				$3p^{4}({}^{1}D)3d^{4}({}^{5}D)[{}^{5}D]5s^{1}$	-25.72
		$3p^{4}(^{3}P)3d^{3}(^{4}F)$	-10.35			$3p^{4}(^{3}P)3d^{4}(^{3}F)[^{5}D]5s^{1}$	-23.31
		$3p^{4}(^{3}P)3d^{3}(^{2}G)$	-7.14			$3p^{4}(^{3}P)3d^{4}(^{3}D)[^{5}D]5s^{1}$	-21.38
		$3p^{4}(^{3}P)3d^{3}(^{2}F)$	- 8.91			$3p^{4}(^{3}P)3d^{4}(^{3}P)[^{3}D]5s^{1}$	-20.75
		$3p^{4}(^{3}P)3d^{3}(^{2}D)$	- 5.94			$3p^{4}({}^{1}D)3d^{4}({}^{5}D)[{}^{5}D]6s^{1}$	-26.94
		$3p^{4}(^{1}D)3d^{3}(^{4}F)$	-9.66			$3p^{4}(^{3}P)3d^{4}(^{3}F)[^{5}D]6s^{1}$	-23.31
		$3p^{4}(^{1}D)3d^{3}(^{4}P)$	-9.14			$3p^{4}(^{3}P)3d^{4}(^{3}D)[^{5}D]6s^{1}$	-22.61
		$3p^{4}({}^{1}D)3d^{2}({}^{3}P)[{}^{3}P]4d^{1}$	-9.83			$3p^{4}(^{3}P)3d^{4}(^{3}P)[^{5}D]6s^{1}$	-21.98
		$3p^{4}(^{3}P)3d^{2}(^{3}F)[^{5}F]5s^{1}$	-7.69				
		$3p^{4}(^{3}P)3d^{2}(^{3}F)[^{3}F]5s^{1}$	-18.63				
		$3p^{4}({}^{1}D)3d^{2}({}^{3}F)[{}^{3}F]5s^{1}$	-17.01	ļ			
		$3p^{4}({}^{1}D)3d^{2}({}^{3}P)[{}^{3}F]5s^{1}$	-16.56	Cr	$2p^{5}(^{2}P)3d^{5}(^{6}S)^{7}P$	$3s^{1}(^{2}S)3d^{5}(^{6}S)^{7}S$	
		$3p^{4}(^{3}P)3d^{2}(^{1}G)[^{3}F]5s^{1}$	-17.69			$3p^{4}({}^{1}D)3d^{5}({}^{6}S)[{}^{6}D]4d^{1}$	-29.97
		$3p^{4}(^{3}P)3d^{2}(^{1}D)[^{3}F]5s^{1}$	-13.45			$3p^{4}(^{3}P)3d^{5}(^{4}D)[^{6}D]4d^{1}$	-23.58

TABLE II. LS terms and calculated relative positions  $\Delta^{\text{theor}}$  from the 3s-2p parent line for each configuration. Negative values indicate a transition whose energy is less than that of the parent line.

unmistakable; both curves appear to be nearly linear. The minor anomaly in the Ca intensity is probably due to carbon contamination. The carbon  $K\alpha$  line lies at approximately 280 eV and contributes to the intensity observed. As mentioned before, we did not model the width or intensity. We believe the width increases with Z due to more configurations being mixed in. The configurations will, in general, have different energies so that as more configurations are added in, a broader feature is expected. One drawback to this explanation is the expected shape of the satellite. One would expect such a scheme to produce an asymmetric satellite peaked

at the energy whose configuration has the largest dipolematrix element and tailing off as the matrix elements go to zero. Since the Coster-Kronig type of Auger process is known to produce satellites of considerable intensity,<sup>20,21</sup> the configuration in which a 3p electron is moved to the 3d orbital is expected to have the largest matrix element. Thus, the addition of configurations should produce a satellite peaked at the  $|[3p^2]3d^{+1}\rangle$  configuration energy and tail off as other configurations are added in. Instead what is observed is a symmetric peak that can usually be well described by a simple Lorentzian as we have shown. We have no explanation for the simplicity of the shape of



FIG. 5. Comparison of experimental satellite position (solid line) with the results of the model described in text.

this satellite.

The apparent disappearance of the satellite is easily explained. Figure 5 shows that as Z is lowered,  $\Delta^{\text{theor}}$  and  $\Delta^{\text{expt}}$  both go to zero somewhere around Z=17. This means that the shakeup configurations are no longer energetically allowed. The satellite disappears as Z is increased due to increased broadening (higher Z mixes more configurations) and to decreased intensity (smaller dipole matrix elements). The satellite may still be present at higher Z but is indistinguishable from the bremsstrahlung background. To check this, a satellite for Mn was extrapolated from the parameters of Table I. This satellite was then subtracted from and compared to our measured spectrum for Mn. There was no discernible change indicating that a broad satellite may be present but unobservable for this and nearby elements.

In summary, we have compared the modified DL theory with our experimental SXE data on the 3s-2p satellite. With regard to the satellite position and observability as a function of Z, the theory compares favorably with the data. Further testing of the model can be done using a multiconfigurational atomic structure calculation to see if the intensity and width can be better understood. In addition, the above experiment and calculations can be repeated for the analogous transitions  $M_{2,3}N_1$  in elements near Rb and  $N_{2,3}O_1$  in elements near Cs, since similar shakeup configurations are energetically possible



FIG. 6. (a) Experimental full width of satellite as a function of Z. (b) Experimental relative intensity of satellite as a function of Z.

in those cases. Previous emission experiments<sup>22</sup> on atomic Kr indicate the presence of an  $M_{2,3}N_1$  satellite.

## ACKNOWLEDGMENTS

We would like to thank the other members of this laboratory, P. Bruhwiler, R. Carson, D. Husk, P. Livins, A. Mansour, J. Nithianandam, and C. Tarrio, and Dr. A. S. Brill for helpful suggestions and discussions throughout the course of this work. We are also grateful to Dr. H. P. Kelly and M. Kutzner for the use of the Fischer code. This research was supported in part by the National Science Foundation (Grant No. DMR 85-15684).

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