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SATELLITE LINES IN THE 5s-5p PHOTOELECTRON SPECTRUM OF XENON (*)

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Résumé. — On utilise le rayonnement synchrotron d'ACO dans le domaine de 75-100 eV pour obtenir des spectres de photoélectron de la couche externe du xénon. On observe des raies satellites, attribuées à des états excités de l'ion, à côté des raies intenses correspondant à la création des états $5s^2\ 5p^5\ ^2P$ et $5s\ 5p^6\ ^2S$ de Xe^+. On compare les intensités des raies satellites aux intensités des pics principaux lorsque l'énergie du rayonnement incident varie. Il y a plusieurs processus d'excitation des satellites, i.e. shake-up, super Coster-Kronig, corrélations dans l'état initial et corrélations dans l'état final impliquant l'électron éjecté dans le continuum. On présente dans ce papier des calculs théoriques basés sur un processus déterminé, super Coster-Kronig, qui donnent pour l'énergie de liaison de la couche 5s une valeur en bon accord avec l'expérience, et un ordre de grandeur correct pour les intensités relatives des raies correspondant aux états finaux de l'ion $5s^2\ 5p^4\ 5d\ ^2S$ et $5s^2\ 5p^4\ 6d\ ^2S$.

Abstract. — ACO synchrotron radiation has been used to obtain photoelectron spectra of the outer shell of xenon in the 75-100 electron volts energy range. Satellite peaks corresponding to the formation of excited ionic states have been observed, in addition to the main peaks corresponding to the production of the $5s^2\ 5p^5\ ^2P$ and $5s\ 5p^6\ ^2S$ states of Xe^+. A comparison is presented of the intensities of the satellites relative to the main lines for different energies of the incident radiation. There are several ways to excite satellite lines, *via* core rearrangement, virtual super Coster-Kronig processes, ground state correlation and final state cattering processes involving the primary photoelectron. In this paper we present theoretical calculations involving one particular process, namely virtual Coster-Kronig process, which gives good agreement with experimental 5s binding energy and gives the correct order of magnitude for the relative intensities of the satellites corresponding to the $5s^2\ 5p^4\ 5d\ ^2S$ and $5s^2\ 5p^4\ 6d\ ^2S$ final states.

1. **Introduction.** — When a system of closed-shell atoms is photoionized with monochromatic radiation, the photoelectron spectrum reveals intense peaks, the so-called main peaks, at kinetic energies

$$E_{kin} = h\nu - E_B$$

where E_B is the binding energy of the ejected electron. Commonly the spectrum reveals also a set of satellite peaks at lower kinetic energies associated with each main peak. These satellite lines correspond to transitions in which the ion is left in an excited state. The existence of these lines is strictly forbidden in the one-electron model of the atom and, consequently, they are an explicit manifestation of electron correlation effects.

There are several mechanisms leading to satellite lines and, accordingly, these lines have been designated as shake-up [1-5], conjugate shake-up [5-10] and correlation or configuration interaction satellites [6, 9-13]. In the shake model the ejection of an electron into the continuum causes the remaining electrons to see a new potential. As a result of the sudden change in the potential, one electron in the

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orbital $n_2\ l_2\ j_2$ may be excited to an orbital $n'_2\ l_2\ j_2$ or may be ejected in the continuum, leading to the double ionization of the atom [14, 15] : in this case the process is commonly referred to as *shake-off*. This theoretical treatment has been mostly used to calculate the electron shake-off probabilities for photoionization in all shells of all rare gases [16]. The results seem to be in good agreement with the experiments when ionization takes place in a core shell and the shake-off process in the outer shell [16-18]. But this statement has to be applied with some caution. For simultaneous excitation of a 2p electron in neon following photoionization in the 1s shell [2-4], the shake theory seemed to reproduce correctly the experimental values [3]. However this agreement has recently been considered to be fortuitous [19] and configuration interaction in the initial and final states had to be considered for complete agreement with experiment. The shake model fails completely to explain the experimental results when both electrons belong to the same outer shell [14-18, 20, 21]. Then electron-electron correlations play a major role. Also the existence of conjugate shake-up lines cannot be explained in the theoretical approach of the shake theory.

Another way for describing the breakdown of the independent single particle model is the formalism of configuration interaction (CI) [10, 13, 22, 23]. One distinguishes between initial state configuration interaction (ISCI) and final state configuration interaction (FSCI). FSCI might be divided into final ionic state configuration interaction (FISCI) and continuum state configuration interaction or interchannel coupling (CSCI), that is final state scattering processes. In FISCI, only the configuration interaction between the final ionic states of the $(N-1)$ electron system is considered, whereas in CSCI the interaction between different continuum channels of the full N-electron system (i.e. ion plus photoelectron in this case) is taken into account. A completely equivalent formulation can be given within the framework of many body Green's function theory [24-28].

Generally all of the possible CI will be present, but, in certain cases, only one (or some) will be dominant. For example, for the group of IIA-IIB elements, FISCI is expected to be relatively unimportant, because the final ionic states have only one electron outside a closed shell [10]. FISCI should be an important way to describe satellites in the photoionization of rare gases in the 5s and 5p shell of Xe in this work, in the 3s shell of Ar [6, 23] and also in inner shells like the 4s and 4p shells of Xe [4, 25, 27]. Other considerations propose that interchannel coupling, CSCI, is expected to be important for photoionization near the threshold and for ions with closely spaced states involving the same shell [20, 21, 26, 29]. This last consideration leads us to focus on the problem of the photon energy dependence of the relative intensities of the satellites; more exactly the important parameter is the ratio ε between the energy of the photoelectron and the energy required to make an additionnal excitation. It can be expected that CSCI will go to zero for increasing ε. Therefore, for large values of ε, the correlation satellites are due only to ISCI and FISCI. Only three systematic investigations of satellite intensity as a function of ε have been performed, in the case of the 2p shell of Ne [5] and of the 1s shell of He [7, 30] : in these experimental results a plateau for these intensities is reached at high photon energy, whereas, at low photon energy, both elements show a completely different behavior.

This work will focus on correlation effects in the outer shell of Xe. Earlier works have measured the intensities of these satellites using AlKα radiation ($h\nu$ = 1 487 eV) [4, 6]; here we will present results obtained from photoelectron spectra of Xe ionized in the outer shell using synchrotron radiation of 75 to 93 eV energy. In addition a theoretical analysis involving FISCI will be proposed.

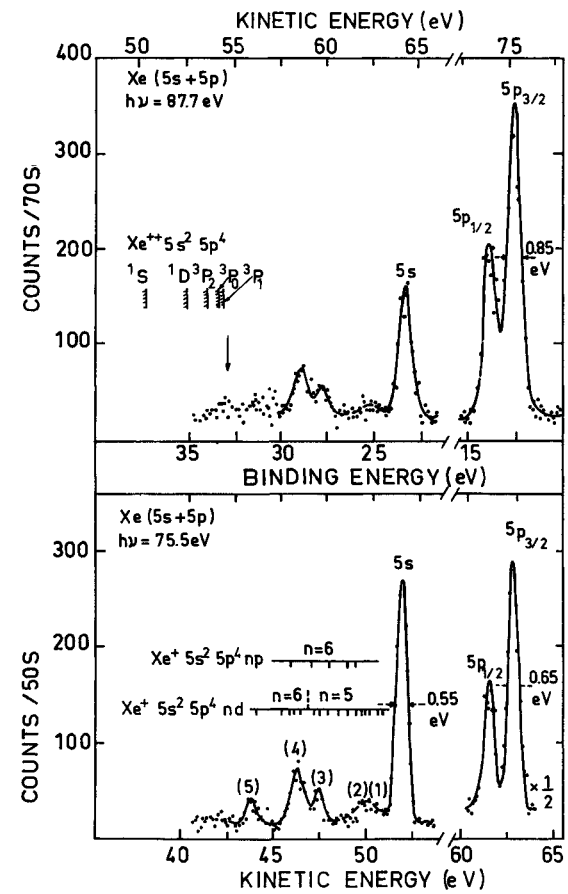

FIG. 1. — Typical photoelectron spectra following Xe 5p-5s photoionization. Peaks labelled $5p_{3/2}$, $5p_{1/2}$ and 5s are due to the ejection of a single electron from the 5p and 5s subshells. The number in brackets are the designations previously used for satellites in an experiment performed at high incident photon energy [4]. Top of the figure : $h\nu$ = 87.7 eV, with a monochromator band pass of 0.64 eV; the five dashed vertical lines indicate the double ionization limits (^{1}S, ^{1}D and ^{3}P terms) taken from optical data (ref. [36]); the full arrow shows the energy position of the ^{3}P limit calculated in this work. Bottom : $h\nu$ = 75.5 eV, with a band pass of 0.30 eV; the lines give the energy positions of the Xe^+ excited states taken from optical data (ref. [36]).

2. **Experiment.** — Photoelectron spectra following photoionization of Xe were analyzed with a cylindrical mirror analyzer. In the energy range from 75 to 93 eV, synchrotron radiation emitted by the ACO storage ring was used [31]. The continuum radiation was monochromatized by means of a one meter grazing incidence monochromator [32]. The band pass of the monochromator was varied between 0.3 and 0.6 eV, depending upon the photon energy and the counting rate. The photon flux, determined by measuring the current of photoelectrons emitted from a gold foil, was between 10^9 and 10^{10} photons/s-Å for wavelengths extending from 100 to 250 Å, depending upon the type of mirror used (plane or cylindrical) to deflect the radiation coming from the ring. The cylindrical mirror analyzer was used with its axis parallel to the incident radiation, which is the only experimental set-up giving independence from the state of polarization of the incident radiation [33]. The analyzer accepted electrons about the magic angle of 54°44', which makes the results independent of any angular distribution effect [44]. The resolution, which can be varied continuously, was set at about 1 %. A detailed description of this spectrometer will be given elsewhere.

For all data presented here, the target gas pressure was maintained at about 1.5×10^{-4} torr in the source volume. A careful study of the influence of the pressure on electron scattering losses has been carried out for the accurate determination of the Xe 5p branching ratio [34]. The total scattering cross section of Xe [35] is decreasing rapidly with increasing electron energy above 7 eV and its differences are small for different electron kinetic energies varying in the 44-53 eV range. Therefore the pressure effects are negligible in this case within the error limits.

3. **Results and discussion.** — Figure 1 shows photoelectron spectra of Xe obtained with 87.7 eV and 75.5 eV incident radiation. Both spectra in this figure are essentially identical. Satellite peaks are labelled in reference to the nomenclature adopted by Gelius [4]. The energies for excited states taken from optical data [36] are reported in the figure 1 as well as in the figure 2 which displays the 75.5 eV photoelectron spectrum taken with better counting statistics. Due to problems in the line identification, in these figures all possible states arising from ^3P, ^1D, ^1S parent states of Xe^{2+} are shown. They give a set of close-spaced optical levels for each of the peaks (1) to (4). This large number of lines could be reduced by selection rules of angular momentum and parity conservation only when specific excitation mechanisms can be assumed. With respect to the $5s^2\ 5p^5\ ^2P$ photopeak, the $5s^2\ 5p^4\ 6p\ ^2P$ final states would correspond to the so-called shake-up lines, while the $5s^2\ 5p^4$ 5d SLJ, $5s^2\ 5p^4$ 6d SLJ and $5s^2\ 5p^4$ 6s SLJ states may correspond to the so-called

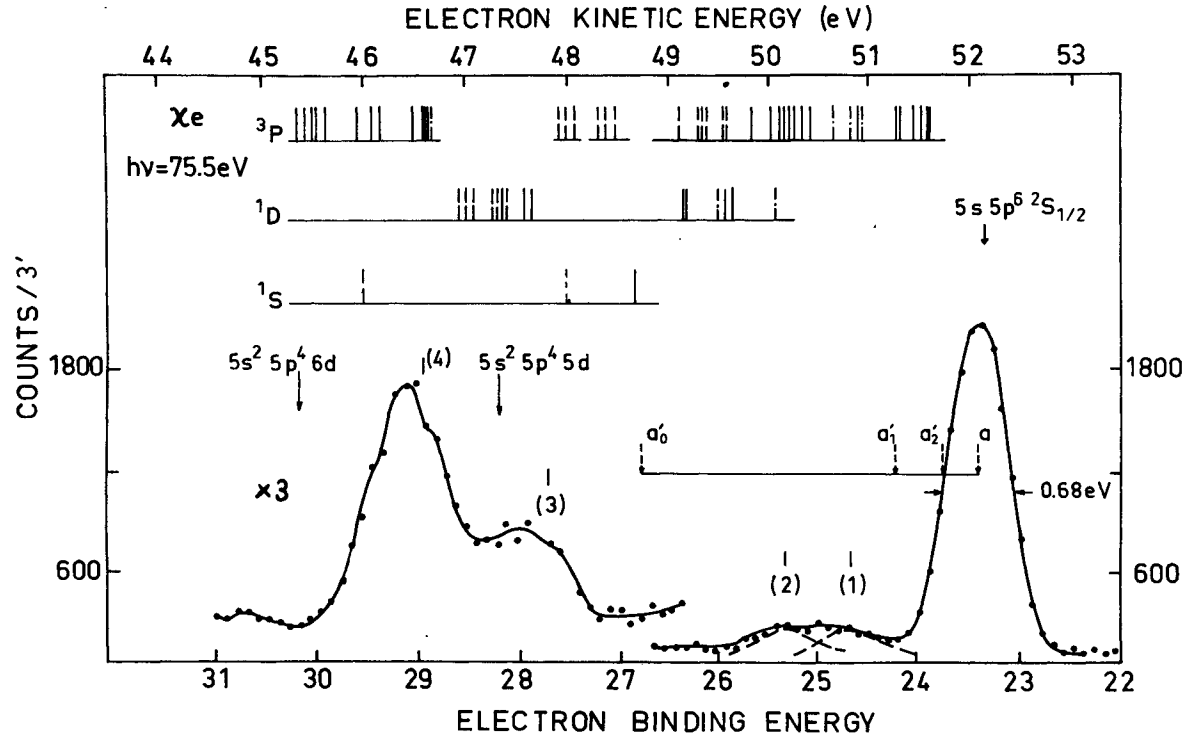

FIG. 2. — Xe photoelectron spectrum with $h\nu$ = 75.5 eV; the left part is magnified by a factor of 3. On the right part, the four dashed arrows indicate the various calculated positions for the binding energies of the 5s level. (See table III.) The two arrows on the left are the energy positions of the $5s^2\ 5p^4$ 5d and $5s^2\ 5p^4$ 6d excited states calculated in this work. In the upper side of the figure, the tick marks give the positions of these excited states of Xe^+ taken from optical data [36]. The terms ^1S, ^1D, ^3P refer to the core configuration $5s^2\ 5p^4$. On the right of the $5s^2\ 5p^4$ 5d arrow, these final states correspond to $5s^2\ 5p^4$ 5d final states. On the left side, the lines correspond to the $5s^2\ 5p^4$ 6d final states. The dashed lines correspond to the $5s^2\ 5p^4$ 6p final states, the dash dotted marks to the $5s^2\ 5p^4$ 6s final states. The two shoulders visible on both sides of satellite (4) have been observed in all spectra obtained with good counting statistics.

TABLE I

Experimental line energies in the Xe *photoelectron spectrum. Satellite lines are labelled* (1), (2), (3), (4) *and* (5), *as previously done in ref.* [4]. *The* $5p_{3/2}$ *optical value was used for calibration. Columns* 1 *and* 2 *give the values obtained at high photon energy* (1 487 eV) *by Gelius* (Ref. [4]) *and Spears et al.* (Ref. [6]); *columns* 3 *and* 4 *correspond to the experimental values measured in the present work*; *column* 5 *gives the possible final states taken from the optical spectroscopy tables* (Ref. [36]). *All energies are in electron volts.*

Peak	*hv* eV	1 487 (Ref. [4])	1 487 (Ref. [6])	87.7 this work	75.5 this work	Possible final states according to Ref. [36]
—		—	—	—	—	—
$5p_{3/2}$		12.130	12.130	12.130	12.130	$5s^2\ 5p^5\ {}^2P_{3/2}$
$5p_{1/2}$		13.43 (1)		13.44 (5)	13.41 (5)	$5s^2\ 5p^5\ {}^2P_{1/2}$
5s		23.40 (1)	23.43	23.39 (5)	23.34 (5)	$5s\ 5p^6\ {}^2S_{1/2}$
(1)		24.61 (5)			24.6 (1) [a]	$[5s^2\ 5p^4({}^3P)\ 6s]\ {}^2P$; $5s^2\ 5p^4({}^3P)\ 5d\ {}^4F$
(2)		25.24 (5)			25.3 (1)	$[5s^2\ 5p^4({}^3P)\ 5d]\ {}^2P,\ {}^4P,\ {}^2D$ $[5s^2\ 5p^4({}^1D)\ 6s]\ {}^2D$
(3)		27.95 (5)	27.6 (1)	28.0 (1)	27.6 (1) [b]	$[5s^2\ 5p^4({}^3P)\ 6p]\ {}^2D,\ {}^2P$; $[5s^2\ 5p^4({}^1S)\ 5d]\ {}^3P$
					28.3	$[5s^2\ 5p^4({}^1D)\ 5d]\ {}^2P,\ {}^2S$; $5s^2\ 5p^4({}^1S)\ 6s\ {}^2S$
(4)		29.02 (5)	29.01 (5)	29.0 (1)	28.8 [b]	$[5s^2\ 5p^4({}^3P)\ 6d]\ {}^4D$; $[5s^2\ 5p^4({}^3P)\ 7s]\ {}^4P$
					29.3 (1)	$[5s^2\ 5p^4({}^3P)\ 6d]\ {}^4F$; $[5s^2\ 5p^4({}^1S)\ 6p]\ {}^2P$
(5)		31.44 (5)			31.5 (1)	$[5s^2\ 5p^4({}^1D)\ 6d]\ {}^2F,\ {}^2D$

[a] The estimated error on satellite energies includes the error on the energy of the 5s peak used for satellite calibration.

[b] A tentative deconvolution split in two components each of the satellites (3) and (4).

conjugate shake-up lines (CSCI satellites), belonging to the same photopeak; but these later final states might also be attributed to FISCI satellites due to virtual super Coster-Kronig processes between $5s\ 5p^6\ {}^2S$ and $5p^4\ md, ms\ {}^2S$. It is not possible to achieve a line identification and interpretation of their origin by comparing the predicted energy line positions [36] with the experimental data. Only satellite (5) may be assigned without ambiguity to $5s^2\ 5p^4\ 6d\ {}^2L$ final states. The results for the mean excitation energies relative to the ground state of Xe for peaks (1) to (5) are summarized in table I; the relative experimental intensities of the various satellite peaks are given in table II.

Using diagrammatic techniques for the self energy and spectral function of a core hole [25, 37] (i.e. working in the high energy limit), we have calculated the energies of the main 5s, 5p photolines and of some satellites [37]. These values are reported in table III. Taking into account relativistic effects and monopole relaxation through the HF ΔSCF method (but not correlation energy), the agreement is reasonable for $5p_{3/2}$ and $5p_{1/2}$ binding energies and the discrepancy can probably be accounted for by correlation energy (about 1 eV). But for the 5s level, the difference is much too large to be explained by the ground state correlation energy, as has already been indicated by Gelius [4]. It has been previously shown that super Coster-Kronig processes, which can be characterized as final ionic state configuration interaction (FISCI), may reduce the intensities of core peaks and give prominent satellite structure in the case of 4p and 4s photoelectron spectra of Xe and Ba [25, 27]. A similar mechanism, namely $5s\ 5p^6 \rightleftarrows 5p^4\ md^2$ super Coster-Kronig dipole fluctuations, is introduced here to

TABLE II

Relative intensities of the main and satellite peaks in the Xe 5s-5p *photoelectron spectrum. Columns* 1 *and* 2 *give the values measured in this work at* 75.5 eV *and* 87.7 eV *photon energy. In column* 3 *are shown the results of Gelius* (Ref. [4]) *obtained at high photon energy* (1 487 eV). *The values are normalized to the* 5p *and to the* 5s + 5p *photoelectron peaks. The relative intensities are proportional to the respective peak area, corrected for the change, with electron kinetic energy, of the spectrometer transmission.*

	Experimental values			Theoretical values
	hv = 75.5 eV This work	*hv* = 87.7 eV This work	*hv* = 1 487 eV Ref. [4]	This work
	—	—	—	—
$5p_{3/2}$	100	100	100	
$5p_{1/2}$	48	51	50	
5s	60	41	33	
5s	100	100	100	100
(1) + (2)	12 (2)		13 (4)	
(3)	11 (2)		25 (4)	
(4)	34 (2)		52 (4)	
(3) + (4)	45 (4)	68 (7)	77 (8)	21
(5)	15 (2)		20 (3)	8
(1) to (5)/5s	72 (8)		110 (15)	
$\frac{\text{(1) to (5)}}{\text{5s + 5p}}$	21 (3)		20 (3)	10 [a]

[a] See Ref. [43].

TABLE III

Theoretical 5s *and* 5p *binding energies. Column* 1 *gives the Koopmans' relativistic value, basically relativistic* HF (see Ref. [37]), *calculated in this work, columns* 2 *and* 3 *the monopole relaxation value obtained here (Hartree-Fock) and in a previous calculation by Gelius (Hartree-Fock-Slater,* Ref. [4]); *in column* 4 *are reported the values calculated for the* 5s *binding energy taking into account* : a) *super Coster-Kronig fluctuations involving the* $5s^2\ 5p^4\ nd$ *and* $5s^2\ 5p^4\ \varepsilon d$ *final states*; b) *involving in addition the* $5s^2\ 5p^4\ 4d^9\ mf$ *final states*; *column* 5 *shows the optical values* (Ref. [36]). *All values are in electron volts. Labels* a, a'_0, a'_1 *and* a'_2 *refer to figure* 2.

Shell	Relativistic HF (frozen core)	Relativistic ΔSCF (monopole relaxation) This work (HF)	Gelius (HFS)	Relativistic ΔSCF + dipole relaxation *a)*	*b)*	Optical data
	—	—	—	—		—
$5p_{3/2}$	12.04	11.4	10.99			12.130
$5p_{1/2}$	13.45	12.8	12.33			13.436
5s	27.57	26.8 (a'_0)	26.2	24.3 (a'_1)	23.7 (a'_2)	23.397 (a)

calculate the 5s satellite intensities relative to the main peak in the 5s spectrum of Xe. Columns 4 and 5 give the values calculated for this 5s binding energy when dipolar fluctuations of the 5s core hole are taken into account. Here the mechanism is virtual because the Xe^+ $5s\ 5p^6$ final state is lower in total energy than the Xe^+ $5s^2\ 5p^4$ SLJ final states.

In the calculation we considered a diagrammatic expansion of the self-energy (direct plus exchange) of a 5s hole, describing how a 5s hole jumps to an intermediate 5p state while exciting another 5p electron to *m*d, εd intermediate states. The self-energy has been renormalized to including infinite sequences of diagrams representing the RPAE (Random Phase Approximation with Exchange), the screened electrostatic interaction between the two 5p holes and the electrostatic interaction of the super Coster-Kronig d-electron with the two 5p holes (V^{N-2} potential). The calculation is closely related to a FISCI analysis involving the final states $5s\ 5p^6$, $5s^2\ 5p^4\ md$, $5s^2\ 5p^4\ \varepsilon d$ and also including correlation effects within the RPAE. The calculated average $5p^2$ threshold energy (~ relativistic HF ΔSCF) gives reasonable agreement (the difference being correlation energy) with the values obtained from optical data [36] (see Fig. 1). Using this value for the $5s^2\ 5p^4$ threshold leads to an additional 5s dipole relaxation shift of 2.5 eV and to 35 % reduction of intensity for the 5s level relative to the monopole relaxation case (the monopole relaxation energy shift is only 0.8 eV). Agreement with experiment is then greatly improved in what concerns the position of the peak.

We also considered some effects of correlation between the 4d and 5s, 5p shells, which can be very important due to the high polarizability of the 4d shell. Such effects had to be taken into account to explain the variation of the single 5s-5p photoionization cross sections in the energy range between 60 and 100 eV in Xe [29] (and also in Ba [25, 26]). Good agreement has been obtained for Xe between experimental results [38] and RPAE calculation including such correlation effects. Considering the effect of the 4d shell via virtual Coster-Kronig processes involving

$$5s\ 5p^6 \rightleftarrows 5s^2\ 5p^5 4d^9\ mf, \varepsilon f,$$

transitions gives an additional shift of 0.6 eV, and thus a total dipole relaxation shift of 3.1 eV. The binding energy of the main 5s photoelectron peak then becomes 23.7 eV. This is quite close to the experimental value and shows that monopole relaxation does not determine the Xe valence spectrum. Note however, that the ordinary ground state correlation energy of about 1 eV has not been included.

Good agreement between theory and experiment for the energies does not mean that the theoretical model is fully appropriate. The shift and the strength of the main 5s peak, for example, results from an integration over the whole satellite spectrum and is comparatively insensitive to details of the satellite spectrum. The critical test of the model is connected with the intensities (and to a lesser degree the energy) of the satellites. In the present first principle calculation, we find the intensity ratio $5s\ 5p^6 : 5p^4\ 5d : 5p^4\ 6d$ to be 100/21/8 and the energy positions of the satellites as shown in figure 2. It must be noted that intensities and energies refer to the same kind of average of levels in the final states [25, 37]. Therefore, the theoretical results can be compared with experimental results only for the corresponding groups of levels. As seen in figure 2, the theoretical positions are possibly compatible with the optical values, but in view of the large multiplet splitting, any attempt to a detailed comparison is meaningless at present.

From the theoretical intensity ratio, one might assume that peaks (3) and (4) belong to the $5p^4\ 5d$ final states and peak (5) to $5p^4\ 6d$, which, however, disagrees with the optical level assignment. On the other hand, if one uses the Moore tables assignment, peak (3) belongs to the $5p^4\ 5d$ and (4) + (5) to $5p^4\ 6d$ final states, which is in conflict with the theoretical ratio. In addition, production of these satellite lines via the virtual super Coster-Kronig process would lead to final ionic states of the same symmetry : ^2S; according to the Moore tables, only peak (3) could be explained in this way; under these circumstances peak (4) would remain unexplained [39]. Inclusion of configurations like $5s^2\ 5p^4\ nd^2$ in the representation of the ground state might lead to final ionic states of the type $5s^2\ 5p^4\ nd$ SLJ. However preliminary results [40] of such a multiconfigurational calculation seem to indicate that it could not account for such a large discrepancy. This situation could not be cleared up by

inclusion of multiplet splitting from the start, nor by including the $5p^4$ *ms* levels, which have been omitted in the present calculations. These remarks might suggest that some of the $5s^2\ 5p^4\ 5d$ SLJ configuration are missing or are wrongly assigned in the Moore tables [41].

The theoretical intensities are summarized in table II for a comparison with experiment. Theory seems to give a too small intensity for the satellites. On the other hand, these calculations are quite sensitive to change in energy difference between $5s\ 5p^6$ and $5s^2\ 5p^4\ 5d$ final states : a small change in this energy difference can lead to theoretical intensities higher by a factor of two for these satellites. Also there are many other processes that have to be investigated and may contribute to the intensities of the satellites. Thus the theoretical intensities presented here should be considered as preliminary results, giving only an estimate of the intensity of the process.

As noted before, there are several ways to excite any given satellite lines, e.g. via core rearrangement, virtual Auger and (super) Coster-Kronig types of processes (FISCI), ground state correlation processes (ISCI) and final state scattering processes involving the *primary* photoelectron (CSCI). In this paper we have concentrated on one particular process (super Coster-Kronig fluctuations of the core hole, FISCI) of particular importance for the shift of the 5s level. However, the other processes mentioned above can be important for the intensity of these satellites (ISCI at all photoelectron energies, CSCI at low photoelectron energy) and must be investigated. Also an interesting feature is the non separability of the 5s-5p spectra. The $5s^2\ 5p^4\ 5d$ satellite, for example, can be connected with a 5s hole via a virtual super Coster-Kronig type of transition (FISCI) and, at low energy, with a 5p hole via inelastic scattering of the photoelectron (conjugate shake-up, for instance). In addition, ground state correlation processes (ISCI) leading directly to two 5p holes cannot be directly referred to either the 5s or the 5p levels and the 5s-5p spectrum should therefore be treated in a unified manner.

In this view, comparing the experimental intensities measured at various photon energies, we can see that the sum of the intensities of the satellites (1) to (5), which represents the satellite contribution to the total (5s + 5p) photoionization cross section, is practically the same at high (1 487 eV) and low (75 eV) photon energy, i.e. respectively 20 % and 21 %. We can however note a tendency to obtain lower values relative to the 5s peak when the photoelectron energy decreases. This is true for the overall intensity as well as for the individual intensity of each satellite, except for peaks (1) and (2). Further experiments at lower photon energies are needed for a better understanding of the satellite spectrum [42].

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