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Si(*LVV*) Auger and resonance Auger spectra of SiF_4 molecules with the use of synchrotron radiation

S. Aksela,* K. H. Tan, H. Aksela,* and G. M. Bancroft
Department of Chemistry and Centre for Chemical Physics, University of Western Ontario, London, Ontario, Canada N6A 5B7
and Canadian Synchrotron Radiation Facility, Synchrotron Radiation Center, University of Wisconsin—Madison,
Stoughton, Wisconsin 53889

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Si(*LVV*) Auger spectra have been obtained for gas-phase SiF_4 molecules with use of tunable photon energies from monochromatized synchrotron radiation. In addition to the normal Auger spectrum excited by photon energies well above the Si $2p$ photoionization threshold, resonance Auger spectra were measured by exciting the Si $2p$ electrons to different empty molecular and Rydberg orbitals. Our normal Auger spectrum shows more detailed structure than those previously reported using electron-beam excitation, and verifies the double structure corresponding to the case where the two final-state holes are either in the same or different Si—F bonding orbitals. In the resonance Auger spectra, the peaks shift by 7.5 ± 0.5, 4.6 ± 1.0, or 2.7 ± 1.0 eV to higher energies when the Si $2p$ electron is excited to the $\sigma^*[3s(a_1)]$, $4s$, and $3d$ orbitals, respectively.

INTRODUCTION

Synchrotron-radiation excitation of the normal Auger spectra has some important advantages over the electron-beam excitation usually used. First, the photon energy can be selected to ionize only the appropriate energy levels, whereas the high-energy electron beam causes ionizations also in deeper core levels. These ionizations furthermore cause cascade Auger processes resulting in extra satellite structure in the spectra.[1,2] Second, the background, mainly due to inelastically scattered primary beam electrons, is usually much higher and more rapidly increasing with decreasing electron kinetic energies for electron-beam excitation.

However, the most important advantage of synchrotron radiation in the study of Auger spectra is that core electrons can be selectively excited into the bound resonance states below the ionization threshold. The created core hole is filled by the Auger-type process in which the excited electron either takes part or remains as the spectator. These resonance Auger spectra can provide important complementary information on the electronic structure in atoms and molecules.

Because of the very low cross sections, only a few resonance Auger spectra have been reported for atoms[3-7] and molecules.[8] Ungier and Thomas have also studied very recently resonance Auger spectra of some simple molecules using electron-electron coincidence spectroscopy.[9]

SiF_4 is an interesting molecule for an Auger and resonance Auger study for several reasons. First, the $2p$ ionization threshold at ~ 112 eV is in a convenient energy range for the Tantalus storage ring and our grazing incidence monochromator. Second, the Si(*LVV*) Auger spectrum of SiF_4 has been reported recently by Rye and Houston,[10] but their electron-excited spectrum suffers from relatively poor resolution and a strong and rapidly changing background making the intensity ratios of the peaks inaccurate. Rye and Houston found an interesting double structure in the spectrum which was explained to correspond to the cases where the two final-state holes appear either in the same or separate Si-F bonding orbitals. Third, the absorption spectrum in the vicinity of the Si $2p$ ionization edge has been studied by several workers.[11-14] These spectra are characterized by several peaks above and below the $2p$ ionization limit, and these peak positions are required for the present study. Several assignments of the absorption features have been made. The most recent assignment[13] of the doublet with energies at 106.1 and 106.7 eV is to the $2p\rightarrow\sigma^*[3s(a_1)]$ core to antibonding valence transition. These peaks are followed by a broad maximum (~ 110 eV) consisting of several peaks below the ionization threshold. The first of these is assigned[13] to the $2p\rightarrow\sigma^*[3p(t_2)]$ transition, while the remaining peaks are assigned to $2p\rightarrow4s$, $2p\rightarrow3d(t_2)$, $2p\rightarrow3d(e)$, $2p\rightarrow5s$, and $2p\rightarrow4d$ Rydberg transitions. Above the $2p$ ionization limit (~ 112 eV) the absorption spectrum shows two strong maxima around 117 and 133 eV. These are known to arise from the enhanced scattering of the outgoing photoelectrons from quasibound states (shape resonances).

In this paper, we report accurate Si(*LVV*) Auger spectra, and for the first time in a molecule report, resonance Auger spectra with the spectator electron in more than one virtual level. Calculations are useful for confirming our energy shifts and assigning the orbital in which the spectator electron resides.

EXPERIMENTAL

The measurements were carried out using the Canadian Synchrotron Radiation Facility at the Tantalus storage ring in Stoughton, Wisconsin, as described in a recent paper.[15] A 600 line/mm holographic grating from JY, Inc., was used in the Grasshopper Mark IV monochromator. The Leybold-Heraeus electron spectrometer was used with 100-eV pass energy, giving an electron spectrometer con-

TABLE I. Binding energies in SiF_4.

Orbital[a]	This work	Others	Character[b]
$1t_1$	16.48	16.46[b]	1.00 F $2p$
$5t_2$	17.54	17.55[b]	0.97 F $2p$, 0.03 Si $3p$
$1e$	18.08	18.09[b]	1.00 F $2p$
$4t_2$	19.52	19.51[b]	0.86 F $2p$, 0.02 F $2s$, 0.12 Si $3p$
$5a_1$	21.59	21.4[c]	0.75 F $2p$, 0.05 F $2s$, 0.20 Si $3s$
$3t_2$		39.27[c]	0.91 F $2s$, 0.09 Si $3p$
$4a_1$	39.46	40.6[c]	0.86 F $2s$, 0.14 Si $3s$
Si $2p$	111.60	111.79[d] 111.70[e]	

[a] Notation includes core levels.
[b] Reference 17.
[c] Reference 21.
[d] Reference 19.
[e] Reference 20.

tribution to the observed linewidths of ~0.6 eV.

The gaseous SiF_4 was forced through a 30-μm single-hole capillary into the interaction region and the electrons were detected at the magic angle of 54.7° so that electron intensities are independent of β and the polarization of the incident radiation. The gas pressure in the interaction region was approximately 10^{-3} torr, and in the sample chamber about 1×10^{-4} torr. Hence the inelastic scattering of the emitted electron can be neglected.

The energy calibration of the photoelectron spectra was based on the known energies[16] of valence-band photopeaks of N_2. The spectra of SiF_4 and N_2 were recorded simultaneously. The spectrometer offset for Auger energies was obtained from the known Auger energies of Xe *NOO* and Kr $M_{4,5}$ *NN* spectra.

RESULTS AND DISCUSSION

The binding energies for the valence-band and core levels up to Si $2p$ were measured and the values are given in Table I. Our results are in a good agreement with those of Jonas *et al.*[17] and Yates *et al.*[18] for the outer valence-band levels and with the Si $2p$ binding energies of Perry and Jolly[19] and Kelfve *et al.*[20] The binding-energy value for F $2s$ agrees with the results of Perry and Jolly.[21]

Normal Auger spectra

The Si(*LVV*) Auger spectrum was measured at photon energies of 117, 123, 130, and 133 eV, which are all well above the $2p$ ionization threshold (111.6 eV). The photon energies 117 and 130 eV correspond to the strong shape resonance maxima, whereas 123 eV hits the minimum between them. With the experimental accuracy, no significant changes in peak positions and relative intensities were observed in the Auger spectra excited by these different photon energies.

Figure 1 shows the Auger spectrum taken at 130 eV photon energy. The spectrum is characterized by broad lines [4—5 eV full width at half maximum (FWHM)] covering the large kinetic energy range from 20 to 75 eV. The spectrum was least-squares fitted to Gaussian line components, and the results are given in Table II. The broadening of the Auger lines is mainly due to the initial- and final-state multiplet splitting and vibrational broadening, and only very slightly affected by lifetime effects and the spectrometer broadening. The $2p$ spin-orbit splitting of 0.6 eV is much smaller than the experimental linewidth. Therefore the spectrum shows an unresolved $L_{2,3}VV$ spectrum. The linewidth is comparable to the total width of the outer valence band and roughly twice the energy separation between the $4t_2$ and $5a_1$ orbitals (Table I), which have prominent Si character. This is in agreement with the simple model that the line shape of *LVV* transitions is a self-convolution of valence-band orbitals having the same character as the core-hole state. The interatomic transitions between orbitals of Si and F characters have very low probability.[22]

The photolines from F $2s$ and Si $2p$ levels cause some overlap with the Auger spectrum. At 130 eV photon energy, F $2s$ is just on the high-energy side of the Auger spectrum and the tail of Si $2p$ photoline causes increasing background to the spectra on the low-energy side.

Comparison with the previous electron-beam-excited spectrum of Rye and Houston[10] demonstrates the advan-

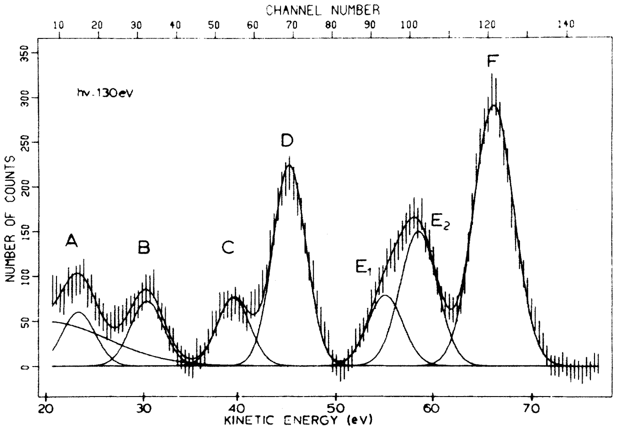

FIG. 1. Si *LVV* Auger spectrum excited by 130-eV photons. A linear background, ranging from 170 counts on the left-hand side to 110 counts on the right, has been subtracted from the data. The spectrum has been least-squares fitted to Gaussian line components shown by solid curves. The broad shape on the low kinetic energy end represents the tail of Si $2p$ photolines.

TABLE II. Si LVV Auger energies and relative intensities.

Line	Energy (eV)		Relative intensity (%)	
A	23.7(5)		5.7	
B	30.7(3)		7.0	
C	39.6(3)		7.3	
D	45.4(2)		21.7	
E_1	55.2(3)	57.5	8.1	24.4
E_2	58.7(3)		16.3	
F	66.3(2)		33.9	

tage of photon excitation with respect to the background. In their spectrum, the Auger peaks ride on a very strong and rapidly increasing background. In our spectra, the background is much lower and varies slowly with the kinetic energy. However, the main structures of the spectra are rather similar but the intensity ratios are different and peaks are better resolved in the present spectrum. The spectrum consists of three doublets. In accordance with the assignments of Rye and Houston,[10] the highest kinetic-energy peak (F) at 66.3 eV corresponds to Si $L_{2,3}VV$(FF′) type transitions with the two final-state holes in the outer valence-band $5a_1$ and $4t_2$ orbitals which have significant Si character, but the two holes reside around different fluorine atoms. The peak E is broader and clearly asymmetric and is fitted by two components E_1 and E_2. It is identified as the Si $L_{2,3}VV$(FF) transitions, where the two final-state holes are located in the same Si—F bonding orbitals. The stronger electrostatic interaction between the holes in the same orbitals give rise to an average energy shift of 8.8 eV between the peaks (F) and the centroid of the structure (E). The peaks D and C at 45.4 and 39.6 eV arise from $L_{2,3}VV'$(FF′) transitions, where one of the final-state holes is in the outer valence orbitals ($5a_1$,$4t_2$) and the second in the deeper-lying F($2s$) $3t_2$ and $4a_1$ orbitals. The holes are in different Si—F bonds for D and in the same for C. The peaks are separated by 5.8 eV which is clearly less than the energy separation in the former doublet. The intensity ratio of the lower to higher energy peak of 0.34 is also substantially lower than 0.72 which was obtained for the E to F ratio. The third B-A doublet is analogously identified as the transitions where both final-state holes are in the F($2s$) $3t_2$ and $4a_1$ orbitals. Peaks are now separated by 7 eV, but the intensity ratio is inaccurate due to the difficulties in accounting properly for the tail of the Si $2p$ photoline overlapping with peak A.

The Auger energy is often written for CVV transitions in the form

$$E_{\rm Aug}=E_B(C)-E_B(V)-E_B(V)-U_{\rm eff}\ ,$$

where E_B's are the core and valence orbital binding energies and $U_{\rm eff}$ describes the effective two-hole interaction energy. In this formalism, the observed energy splittings between the components of the doublets describe directly the changes in $U_{\rm eff}$ when the holes are in different or the same Si—F bonds. Using the binding energies from Table I we calculate the following $U_{\rm eff}$ values for the indicated peaks: F, 4.2 eV; E, 13.1 eV; D, 6.4 eV; C, 12.1 eV; B, 2.0 eV; and A, 9.1 eV.

The $U_{\rm eff}$ values fall clearly into two groups, 4.2, 6.4, and 2.0 eV and 13.1, 12.1, and 9.1 eV, with the average values 4.2 and 11.4 eV and corresponding to the final configurations FF′ and FF. These values compare rather well with the values 7 and 15 eV of Rye *et al.* Their absolute values are somewhat higher due to their slightly different binding- and kinetic-energy values but the differences between the two sets agree within the errors of the two measurements. It should be noted that the $U_{\rm eff}$ values contain, besides the Coulombic interaction energy between the two holes, the extra-atomic relaxation energy contribution. It is probably not constant, but should increase when the holes are in more tightly bound orbitals. The calculated $U_{\rm eff}$ values show rather significant variations. For a more accurate study, the energies and intensities should also be calculated for all different molecular Auger line components, which form the broad lines of the spectra. This is, however, outside the scope of this study.

Resonance Auger spectra

The resonance Auger spectrum consists of two parts. The high-energy part corresponds to autoionization tran-

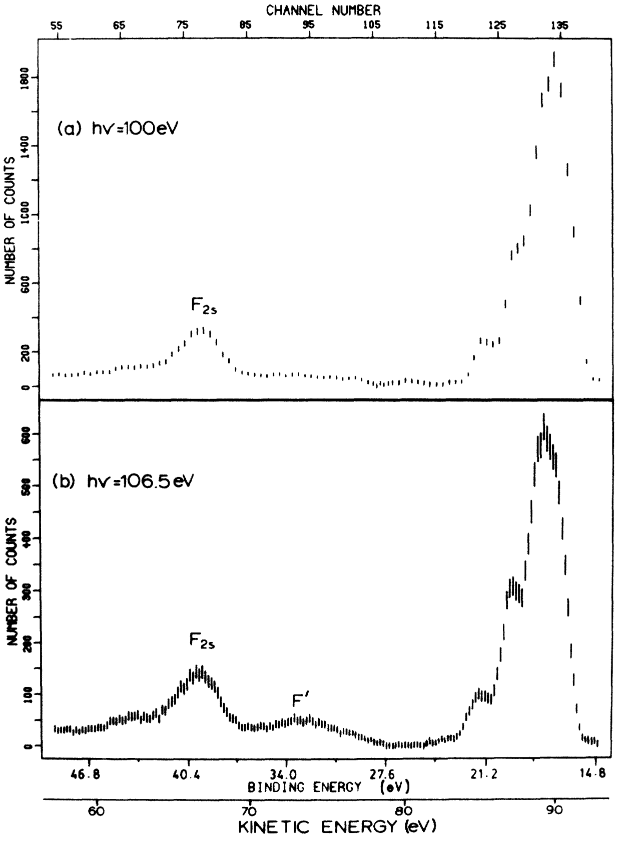

FIG. 2. F $2s$ and valence-band regions taken at 100 eV photon energy which is well below the Si $2p$ ionization threshold (top), and at 106.5 eV which corresponds to the strongest $2p\rightarrow\sigma^*[3s(a_1)]$ resonance absorption maximum (bottom). The valence-band peaks are on the right of both figures. Peak indicated by F' is the most intense resonance Auger peak corresponding to the peak F in the normal Auger spectrum shown in Fig. 1. The kinetic-energy scale is given for (b).

sitions involving the excited electron. The final states for this process are single-hole states where one electron is removed from an orbital and other electrons remain unexcited. Hence these states can also be reached through the ordinary photoemission process, and the corresponding spectrum is the valence-band photoemission spectrum ($E_B = 16$—24 eV) using this resonance excitation energy. The other part is created by processes in which the excited electron remains as the spectator during the decay when one electron fills the core hole and the other is emitted. These final states can be reached also by the photoemission process and a simultaneous excitation of another electron to some higher state (shake up).

It turns out that the peak intensities from the valence band ionization, which includes both direct and resonant photoionization, is about 10 times as large as the intensity for spectator Auger decay. This is illustrated in Fig. 2, where we show the main part of the resonance Auger spectrum taken at photon energies of 100 and 106.5 eV. The spectrum taken at 100 eV photon energy [Fig. 2(a)] is the ordinary photoelectron spectrum because the photon energy is well below the resonance excitation energies. The 106.5-eV photons create the $2p \rightarrow \sigma^*[3s(a_1)]$ resonance excitation, and the corresponding Auger spectrum is shown in Fig. 2(b). The F 2*s* photoline overlaps with the resonance Auger spectrum, but the peak *F*′ is clearly resolved. Figure 3 shows the low-kinetic-energy part of the spectra in more detail. Before beginning a detailed discussion of the resonance Auger spectrum we would like to make some observations on the 100-eV spectrum. Around the F 2*s* line we can see several weaker "shake-up" lines (S_1—S_4) which are caused by excitations associated with the photoionization. The 45-eV binding-energy line (S_1) on the left-hand side of F 2*s*, especially, is relatively strong. The final states of these satellite lines can be reached also by the resonance Auger spectrum. It can be seen that the peak *F*′ gains most intensity at the 106.5-eV resonance excitation while the intensity of the 45-eV peak increases noticeably. On the low-kinetic-energy and high-binding-energy side of the F 2*s* peak, new peaks can be distinguished.

The opening of the new channels to the same final states at the resonance excitation energy should change the relative intensities of the line components of the valence band, too. The valence-band region of the spectra excited by 100- and 106.5-eV photons is shown in Fig. 4. The spectrum at the resonance photon energy (bottom) should also involve autoionization transitions where the excited electron is involved in the electron emission process. However, within 5% the relative intensities of the valence-band photopeaks are the same, indicating that autionization is not important. This is in clear contrast to the case of BF_3 studied very recently by Kanamori *et al.*[8] These SiF_4 valence-band spectra strongly suggest that valence-band intensity changes on SiO_2 at the Si 2*p* absorption threshold are due to Auger peaks[23] rather than

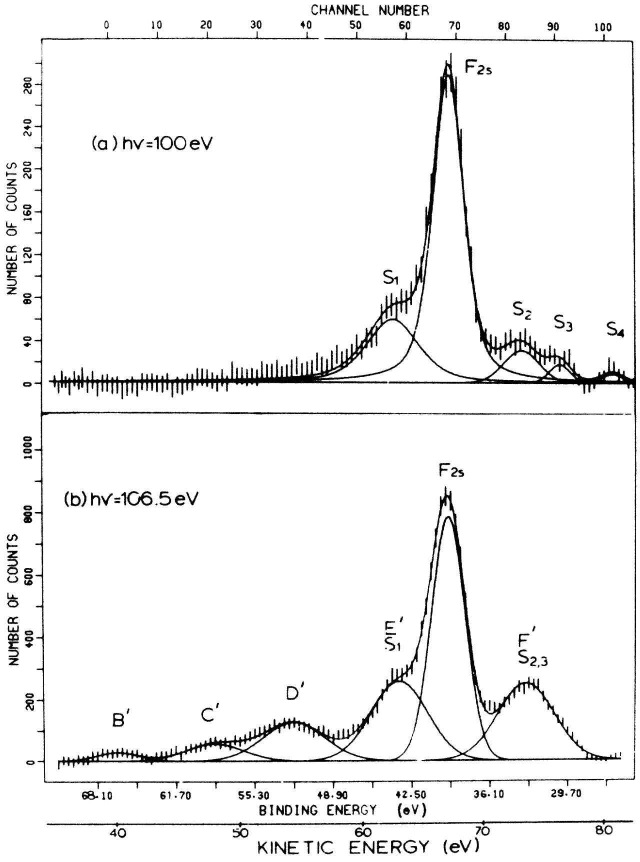

FIG. 3. The resonance Auger region taken at 100-eV and 106.5-eV photon energies. The 100-eV spectrum (top) shows the F 2*s* photoline and satellite lines (S_1—S_4). At the resonance excitation energy 106.5 eV, the resonance Auger peaks (B'—F') also appear. The kinetic-energy scale is given for (b).

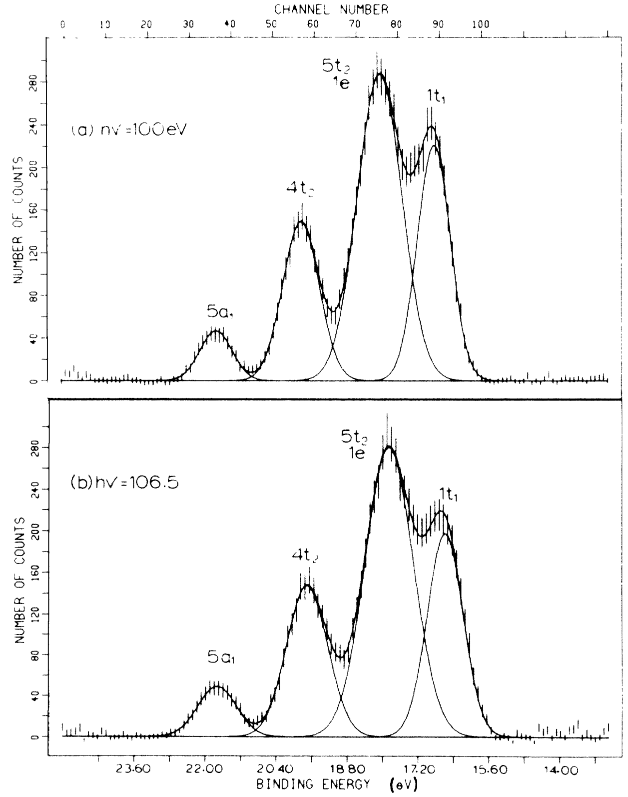

FIG. 4. The valence-band region taken at 100- and 106.5-eV photon energies.

resonance processes as claimed earlier.[24]

We will restrict our discussion in the following to the low-kinetic-energy part of the resonance Auger spectrum (Fig. 3). In the spectrum excited by 106.5-eV photons, the peak F' on the right-hand side of the F 2*s* photopeak is best resolved. It can be identified to correspond to the peak F in the normal Auger spectrum. Its position has shifted 7.2 eV to high kinetic energies. Also, the peaks E', D', and C' correspond to peaks E, D, and C in the normal Auger spectrum although they are weaker and not well resolved, and thus the accuracies of their shifts are lower than for peak F'. Peak E', for example, overlaps with the strong S_1 structure shown in Fig. 3(a). An average energy shift of peaks F'–C' is 7.7 eV. We, however, estimate that a realistic shift value is 7.5±0.5 eV, giving more weight to the most reliable value obtained for peak F'. This shift is clearly higher than corresponding shifts for Xe and Kr rare gases, but about the same as for B *KVV* transitions in BF_3.[8]

Figures 5(a) and 5(b) show the resonance Auger spectra taken at 109 and 111 eV mean photon energies, respectively. At 109 eV photon energy and about 2 eV bandwidth, $2p \rightarrow [3p(5_2)]$ and $2p \rightarrow 4s$ excitations are possible. The best-resolved peak D'' now is present at the kinetic energy of 50.6 eV. We take it to correspond to the peak D in the normal Auger spectrum relative to which it has now shifted by 5.2 eV. The next best-resolved peak E'' is shifted 3.4 eV relative to peak E. Due to the partial overlap of peak E'' with the structure S_1, this value may not be as reliable as the one obtained for D''. As a weighted average we thus estimate an energy shift of 4.6±1.0 eV. By $h\nu = 111$ eV, the best-resolved peak D''' is at 48.5 eV and its shift relative to D is 3.1 eV. Correspondingly E''' shows a shift of 2.3 eV, and an average shift of peaks E'–C' is 2.7±1.0 eV. These two photon energies best excite the $2p \rightarrow 4s$ and $2p \rightarrow 3d$ Rydberg-type transitions, respectively.

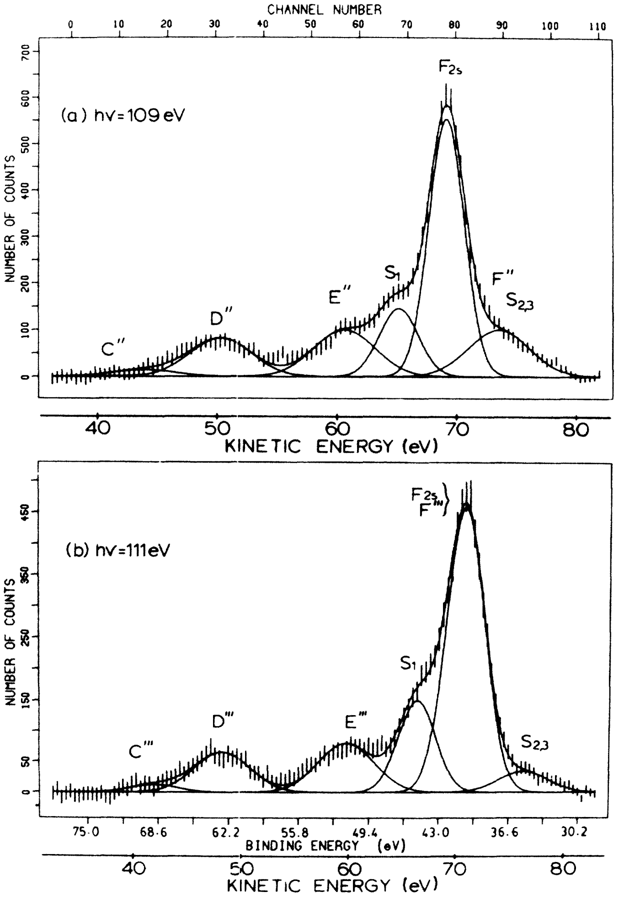

FIG. 5. The resonance Auger spectra excited by (a) 109-eV and (b) 111-eV photon energies.

Comparison with atomic calculations

We have used Grant's Dirac-Fock[25] program to calculate Auger and resonance Auger as well as Si 2*p* and 3*p* binding energies for atomic Si. The average calculated $L_{2,3}M_{2,3}M_{2,3}$ Auger energy for the normal Auger spectrum is 86.3 eV, while the average energies for the resonance Auger spectra with spectators Si 4*s* and Si 3*d* electrons are 91.0 and 90.0 eV, respectively. Thus, the calculated kinetic-energy shift of 4.7 eV is in excellent agreement with the experimental shift of ~4.6 eV for the 109-eV excitation which corresponds to the $2p \rightarrow 4s$ primary process. The calculated shift of ~3.7 eV at 111-eV photon energies (where the spectator electron is 3*d*) is once again in reasonable agreement with the experimental shift. The free-atom calculations thus reproduce the kinetic-energy shifts rather well in the SiF_4 molecule when the resonance excitation leads to Rydberg states. An atomic model was found to give good predictions for the shake-up energies of some molecules.[26,27] The calculations also strengthen the evidence for the latest resonance peak assignments.[13] For example, the calculations support the assignment of the peak at 106.5 eV to $2p \rightarrow \sigma^*[3s(a_1)]$ rather than $2p \rightarrow 4s$ as in previous work.[12]

The calculated atomic $L_{2,3}VV$ Auger energy is about 20 eV larger than the experimental energy value for the highest energy peak (F) in Fig. 1. This unusually large shift is mainly caused by the chemical shift of 2*p* and valence levels in SiF_4 relative to Si atoms. The large valence level shift of 10–15 eV is significantly larger than the shift of 2*p* levels (4–6 eV), resulting in this Auger shift.

From the calculated average binding energies of 7.3 and 22 eV for $3p^{-1}$ and $3p^{-2}$ hole states, the value of 7.4 eV is obtained for $U_{\rm eff}$ in a free Si atom. This value is between the values 4.2 and 13.1 eV for delocalized and localized peaks F and E from SiF_4.

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[*] On leave from Department of Physics, University of Oulu, 90570 Oulu 57, Finland.

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