

Article

Valence-Shell Angle-Resolved Electron Energy Loss Spectra of SF₆

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Espectros de perda de energia de elétrons foram obtidos para a molécula de hexafluoreto de enxofre na região da camada de valência (0 a 100 eV) com uma variação angular de 1.0° - 15.0°. Os espectros foram medidos para uma energia de impacto de 1 keV. O estudo com variação angular sugere a presença de três transições proibidas na região de energia abaixo de 40 eV. Acima de 40 eV, estruturas interessantes aparecem para maiores ângulos de espalhamento podendo estar associadas a processos multieletrônicos proibidos. Uma nova estrutura foi observada em 76 eV. A seção de choque elástica absoluta foi obtida através de normalização dos dados relativos com valores experimentais encontrados na literatura. As seções de choque diferenciais inelásticas absolutas foram obtidas para as transições em 11,5 eV e 13,2 eV. A força do oscilador generalizada (FOG) foi determinada para a transição em 11,5 eV no intervalo de K^2 entre 0,05 e 1,3 u.a.. O valor experimental da força do oscilador óptica (0,312) para o pico em 11,5 eV, obtido através de extrapolação da curva da FOG, mostra boa concordância com dados da literatura.

Electron-energy-loss spectra of the SF₆ molecule have been measured in the valence-shell region (0 - 100 eV) covering an angular range of 1.0° - 15.0°. The spectra have been measured at an impact energy of 1.0 keV. The angular dependence of the spectra showed the presence of three forbidden transitions in the region below 40 eV. Above 40 eV interesting features appear at higher scattering angles, which can be associated to nondipole many-body processes. A new feature was observed at 76 eV. The absolute elastic differential cross section has been obtained through normalization of the relative data to the experimental values found in the literature. The absolute inelastic differential cross sections for the 11.5 eV and 13.2 eV transitions have been determined. The generalized oscillator strength (GOS) for the 11.5 eV peak has been also determined in the 0.05 to 1.3 a.u. K^2 range. The experimentally determined value for the optical oscillator strength (0.312) for the 11.5 eV peak obtained through extrapolation of the GOS curve showed good agreement with the data found in the literature.

Keywords: sulphur hexafluoride, electron energy loss spectroscopy, forbidden transitions, generalized oscillator strength

Introduction

Electron-energy-loss spectroscopy (EELS) has already proved to be a powerful technique in the study of valence

and inner-shell electronic excitations. A great number of atoms and molecules have been studied using the electron impact technique at low and high-resolution conditions¹⁻³.

Near zero scattering angle and at high incident electron energies the First Born Approximation (FBA)⁴ is valid, and electrons behave like pseudophotons⁵. As a consequence, the electron energy loss spectra are similar to photoabsorption spectra. In fact, under these conditions the energy-loss spectrum shows the same excitation features, but with the relative intensities equal to the E_{on}^{-3} times the observed intensities in the optical spectrum, where E_{on} is the electron energy-loss (which corresponds to the photon excitation energy). This consequently means that although the observed features in both spectra will be the same, their relative intensities will be different. A small angle, high incident energy EELS spectrum will consequently furnish essential information on the photoabsorption processes of a given atomic or molecular target⁴. Moreover, it is also possible, using a single experimental set-up, to measure absorption data covering a broad energy range extending from the ultraviolet to the X-ray range. This is a very useful feature even when compared to synchrotron radiation measurements, which are usually plagued with harmonic contamination. Electron impact spectra obtained at non-zero scattering angles may additionally furnish invaluable spectroscopic information about dipole-forbidden electronic transitions in the valence as well as in the inner-shell excitation regions⁶. This is a major advantage of this technique over other conventional spectroscopic techniques. Despite the great spectroscopic potentiality of this technique, only a few groups have dedicated their efforts to the study of the angular dependence of valence and core excitations induced by electron impact.

In the present paper we address our attention to the SF₆ molecule. Although this molecule has been the focus of many studies based on the use of photons⁷⁻¹⁰ and electrons¹¹⁻¹³, the first angle-resolved EELS measurements for this compound were published by Ying *et al.*¹³ at an impact energy of 2.5 keV. In this work absolute absorption transition probabilities of valence-shell (7-70 eV) electronic transitions have been determined covering an angular range up to 8.5°. With the aim of exploring the valence and inner-valence excitation regions of the SF₆ molecule and with the aim of elucidating the still controversial discussion about the assignment of the experimental features, its spectra have been studied using the angle-resolved electron impact technique at 1.0 keV incident energy. The energy-loss range extends from zero to 100 eV and the angular range from 1.0° to 15.0°. Using the absolute elastic differential cross section, our data were normalized and the GOS-curves for the 11.5 eV and 13.2 eV transitions were determined.

Experimental

The experimental apparatus used for the present measurements has been described elsewhere in detail^{14,15}. Basically it consists of a rotatable electron gun, a neutral gas

beam, a Möllenstedt electron velocity analyser¹⁶ fixed on the vacuum chamber wall and a conventional detection system. A crossed-beam geometry is used for the electron-gas collision. The electron beam, produced by a triode-type electron gun, is used without prior energy selection. A typical beam current is 10 μ A and the beam diameter, measured at the scattering region, is approximately 0.5 mm. The gas beam is produced by the expansion of a given sample inside the scattering chamber through a hypodermic needle with a 0.2 mm internal diameter.

The scattered electrons are velocity analyzed by the Möllenstedt analyzer and detected by an electron multiplier (Spiraltron, Galileo Electro Optics). The energy resolution of this system was set to 0.6 eV, as determined by the full width at half maximum (FWHM) of the elastic peak. The analyser viewing cone is determined by two parallel, circular apertures of 200 and 50 μ m diameter, located, respectively, at 120 and 150 mm from the scattering center. The stray magnetic fields in the plane of the measurements have been reduced to less than 10 mG in all directions by three pairs of orthogonal Helmholtz coils.

The residual pressure is of the order of 10⁻⁶ torr. When the gas sample is introduced into the scattering chamber the pressure is raised one order of magnitude. For each angle, the spectra are continuously recorded until a good signal to noise ratio is achieved. The background is corrected by subtracting another spectrum, obtained at the same experimental conditions, but with the gas sample introduced at a side flange, far from the scattering center. The energy-loss scale is calibrated by using the well-known transition 1s-2p of He at 21.22 eV. The scattering zero-degree angle was checked by measuring the elastic peak area over a 15° range on both the right-hand and left-hand sides with respect to the electron analyzer main geometrical axis. Although a precision mechanism allows the determination of the scattered angle with an accuracy of 0.02°, our angular resolution, defined by the set of apertures in front of the Möllenstedt analyzer, is limited to 1.0° in the present experiment. The experimental uncertainties are estimated in the following manner. The maximum statistical uncertainty was 1.5%. Fluctuations in the primary beam current and on the sample pressure were of the order of 1.0% and 0.5% respectively. The determination of the area of the peak is also subject to a 2% error. The elastic cross section which was used in the normalization procedure²⁴ has an associated uncertainty of 8%. The overall uncertainty δ , defined as $\delta = (E \delta_i/2)^{1/2}$, is equal to 9%, including the error in the extrapolation of the GOS-curve, which was of 5%.

In the present work, the incident electron energy was 1000 eV and the electron energy-loss spectra have been obtained in the angular range of 1.0° - 15.0° covering an energy range of 100 eV.

Results and Discussion

The SF₆ molecule, as it is well known, belongs to the O_h symmetry point group. The accepted valence electronic configuration for this molecule is¹³:

$$(4a_{1g})^2 (3t_{1u})^6 (2e_g)^4 (5a_{1g})^2 (4t_{1u})^6 (1t_{2g})^6 (3e_g)^4 (1t_{2u})^6 (5t_{1u})^6 (1t_{1g})^6$$

The experimentally determined ionization potentials are respectively^{17,18}:

$$44.2; 41.2; 39.3; 27.0; 22.9; 19.8; 18.6; 17.0; 15.7$$

Figure 1 shows the valence-shell electron energy loss spectrum of the SF₆ molecule at the scattering angle of 1.5°, covering the 2-50 eV energy range. Several spectral features can be observed. The assignment of these features has been discussed in detail earlier^{7-13,19-21}. They may be attributed to transitions from the ten valence orbitals into unoccupied molecular orbitals and/or Rydberg orbitals, although some discussion still exists in the literature on the nature of these final states. Some points related to this situation will be addressed below as we discuss our angle-resolved electron energy loss data.

The angular dependence of the valence-shell spectra (2-50 eV) is shown in Fig. 2. The angular range is from 1.5° to 5.5°. Several spectral changes can be observed as the scattering angle, θ , is increased, *i.e.*, as the momentum transfer is increased. The intensity of features located at

11.5 eV, 16.0 eV, 17.0 eV, 19.5 eV and 23.0 eV clearly decreases with increasing scattering angle. On the other hand, features at 13.2 eV and 28.0 eV show an opposite behaviour *i.e.* they become more intense as the momentum transfer increases. This opposite behaviour demonstrates the different nature of the processes involved in these transitions. The decrease in the relative intensities of these features with momentum transfer suggests a dipole-allowed nature for these transitions, while the increase in the relative intensities with momentum transfer could suggest a nondipole nature associated to the transitions.

Recently Ying *et al.*¹³ have measured angle-resolved valence-shell (7-70 eV) spectra of the SF₆ molecule at an impact energy of 2.5 keV. Comparison to our data shows however some discrepancies concerning the nature of the transitions. Firstly the feature at 23.0 eV of Fig. 2, which shows, as mentioned before, an allowed behaviour, is considered by Ying *et al.*¹³ to have nondipole character. It has been however assigned by Sze and Brion¹² as having a contribution from the $[1t_{2u}, 5t_{1u}] \rightarrow 2t_{2g}$ and $5a_{1g} \rightarrow 6t_{1u}$ allowed transitions. A nondipole behavior is to be expected for the feature at 28.0 eV, which shows a prominent increase in intensity as the scattering angle approaches 5.5° (see Fig. 2). The 28.0 eV peak has been assigned to the $4t_{1u} \rightarrow 2t_{2g}$ transition^{12,13}.

In disagreement with the work of Ying *et al.*¹³, we observe a strong increase in intensity as the momentum transfer increases for the the 13.2 eV transition, indicating

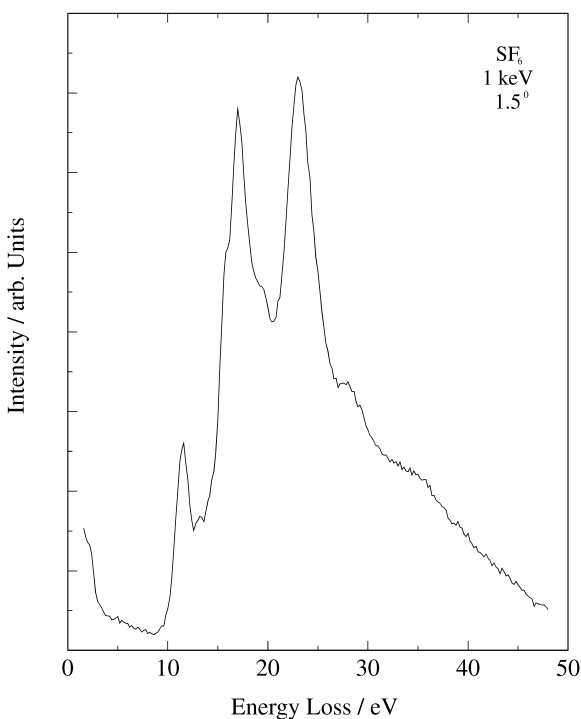


Figure 1. Electron energy-loss spectrum of the SF₆ molecule measured at 1.5°.

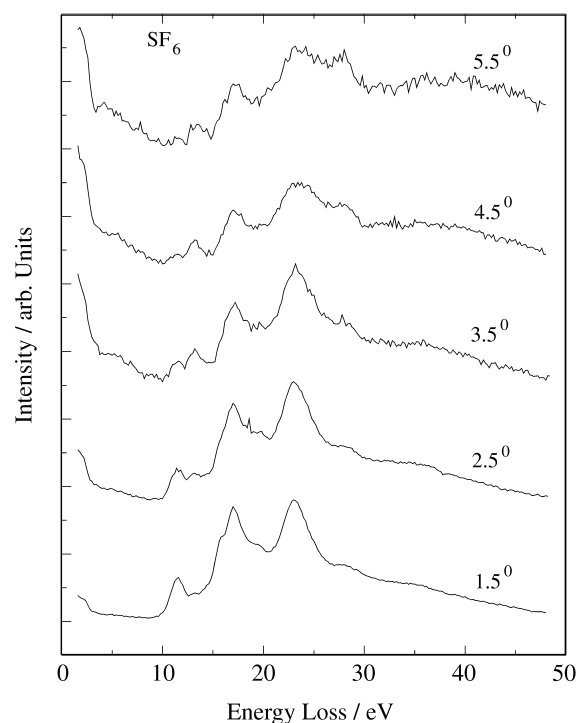


Figure 2. Angle-resolved electron energy-loss spectra of the SF₆ molecule measured at 1.0 keV and covering the angular range of 1.5° to 5.5°.

a nondipole character. Trajmar and Chutjian²¹ have also observed a forbidden transition at the same excitation energy, working at large scattering angles and very low impact energies. The experimental value of 12.8 eV measured by these authors has been assigned as the $1t_{1g} \rightarrow 7a_{1g}$ forbidden transition²⁰. Sze and Brion¹² have also observed a very weak structure at about 12.5 eV in their spectrum measured at 3.0 keV and 0° . They have suggested a forbidden character for this feature. The theoretical work of Hay²² also predicts a nondipole transition to fall into this energy range.

Although many different studies have been performed on the valence-shell region of the SF₆ molecule, most are however concentrated in the 10-40 eV energy range⁸⁻¹¹, only a few extending to about 60 eV¹² or 70 eV as in the recent work of Ying *et al.*¹³. None of them, with exception of the work of Ying *et al.*¹³, has measured the angular dependence of the valence spectra. The region below 10 eV, has received little attention and has not been studied using angle-resolved EELS, shows the intensification of the band located at about 6 eV in Fig. 2 as the momentum transfer increases. This behaviour could suggest the existence of a dipole-forbidden transition. A feature located at 6.494 eV was observed as a very weak structure in the work of Sze and Brion¹², although no possible assignment was suggested by these authors.

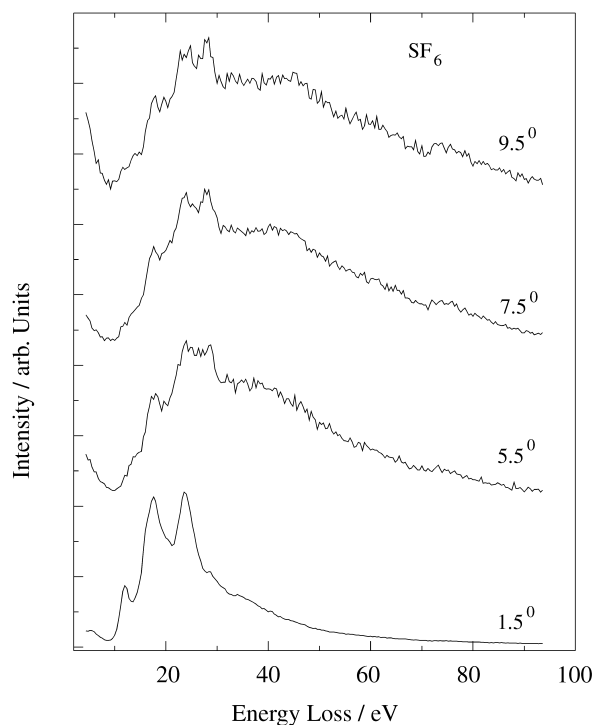


Figure 3. Angle-resolved electron energy-loss spectra of the SF₆ molecule measured at different scattering angles covering an excitation energy range of 0 to 100 eV.

Another interesting region in the study of the valence excitations of the SF₆ molecule is the region above 40 eV shown in Fig. 3. This is a very complex energy region because not only one-electron processes are allowed but also processes involving more than one electron. All double ionisation potentials lie in this region. A more detailed discussion of the excitation processes appearing in this high energy domain will be presented in a forthcoming publication²³.

Figure 3 shows the valence-shell spectrum of the SF₆ molecule at higher scattering angles and covering the energy range of 2-100 eV. Above 40 eV three new features can be observed. The fact that they are not observed at small angles could indicate a forbidden nature for these features. The band at about 44 eV was measured by Sze and Brion¹² and Ying *et al.*¹³, although its assignment is controversial. While a $3t_{1u} \rightarrow 2t_{2g}$ transition was assigned by Sze and Brion¹², a forbidden character ($5a_{1g} \rightarrow 4e_g$) was suggested by Ying *et al.*¹³. Our data corroborate the nondipole character of this transition. The feature at about 60 eV was first observed by Ying *et al.*¹³ and attributed to a nondipole many-body transition. Although very weak, it can be observed by higher θ in Fig. 3. The feature at about 76 eV in Fig. 3 was observed for the first time. It can be also related to nondipole processes although its assignment is very difficult without help from first principle calculations.

The relative experimental inelastic differential cross sections for the 11.5 eV and 13.2 eV peaks have been made absolute using the elastic differential cross section (EDCS), which was normalized to the absolute elastic differential cross section found in the literature²⁴. In this procedure the interpolation was done using the absolute curve at the scattering angle of 10.5° . At this angle our cross section value was made equal to the absolute value obtaining thus the normalization factor. The experimental elastic differential cross section was measured over a 15° angular range and at an impact energy of 1 keV. Figure 4 shows the absolute elastic differential cross section for the SF₆ molecule plotted together with the absolute curve. These results are also presented in Table 1 as a function of the scattering angle θ . From these values, it was possible to determine the absolute inelastic differential cross sections for the 11.5 eV and 13.2 eV transitions, which are shown as a function of the scattering angle θ in Fig. 5 and Table 2.

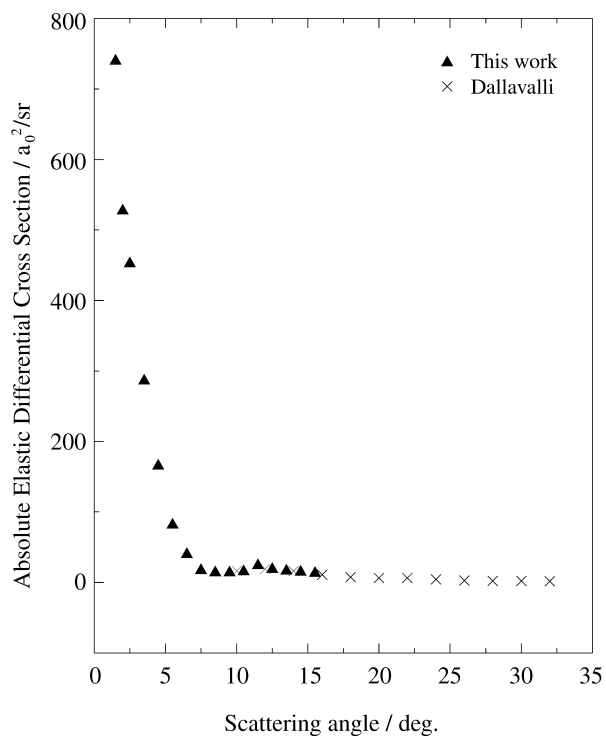
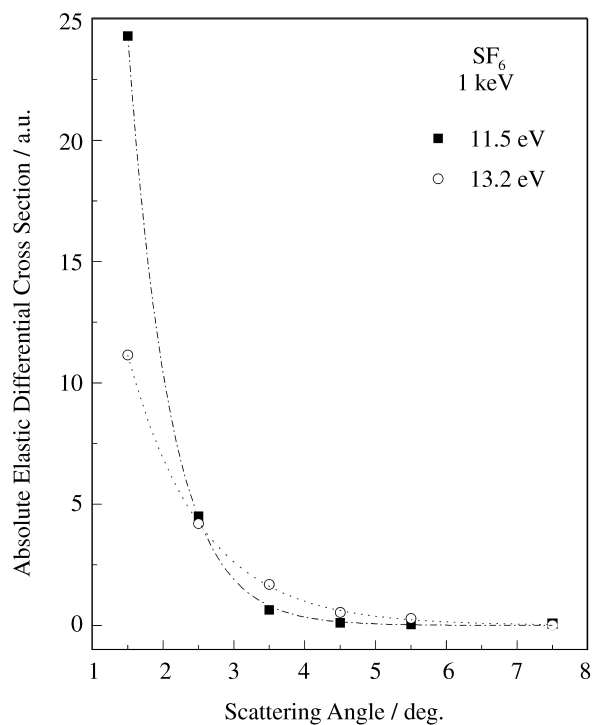
It is now possible to obtain the absolute generalized oscillator strength for both transitions in SF₆. Within the First Born Approximation (FBA), the generalized oscillator strength, $f_{on}(K)$, is directly associated to the differential inelastic cross section $(d\sigma/d\Omega)_{on}$ for the excitation of an atomic or molecular target from the ground state $\underline{0}$ to an excited state \underline{n} through the expression⁴:

Table 1. Elastic Differential Cross Section for the SF₆ molecule.

θ	EDCS (a_0^2/sr)	
	This work	Dallavalli ²⁴
1.5	739.54	
2.0	527.13	
2.5	451.94	
3.5	286.05	
4.5	165.12	
5.5	81.40	
6.5	39.53	
7.5	17.05	
8.5	13.95	
9.5	13.95	
10.0		16.37
10.5	15.50	
11.5		
12.0		19.37
12.5	18.61	
13.5	16.28	
14.0		16.15
14.5	14.73	
15.5	13.18	
16.0		10.90
18.0		7.33
20.0		6.51
22.0		6.18
24.0		4.54
26.0		2.72
20.0		1.92
30.0		1.85
32.0		1.71

Table 2. Absolute Inelastic Differential Cross Section for the 11.5 eV and 13.2 eV transitions in SF₆.

Scattering angle (deg.)	Inelastic differential cross section (a.u.)	
	11.5 eV	13.2 eV
1.5	24.3	11.15
2.5	4.5	4.2
3.5	1.6	1.7
4.5	0.11	0.54
5.5	0.042	0.29
7.5	0.074	0.077

**Figure 4.** Absolute elastic differential cross section of SF₆ as a function of the scattering angle θ .**Figure 5.** Absolute inelastic differential cross section of SF₆ for the 11.5 eV and 13.2 eV transitions as a function of the scattering angle θ .

$$f_{\text{on}}(\mathbf{K}) = \frac{E_{\text{on}}}{2} \left(\frac{|\mathbf{k}_i|}{|\mathbf{k}_f|} \right) |\mathbf{K}|^2 \left(\frac{d\sigma}{d\Omega} \right)_{\text{on}} \quad (1)$$

where E_{on} is the excitation energy, \mathbf{K} is the momentum transfer, $\mathbf{K} = \mathbf{k}_i - \mathbf{k}_f$, and \mathbf{k}_i and \mathbf{k}_f are respectively the initial and the final momentum vectors for the scattering electron. The GOS is independent of the incident energy at energies for which the FBA is valid. Lassetre and Skerbele²⁵ have additionally demonstrated that the GOS tends to the optical oscillator strength (OOS) in the $K^2 = 0$ (optical) limit.

As a first step in the determination of the GOS, the area of the 11.5 eV and 13.2 eV peaks were obtained through a standard deconvolution procedure¹⁵. The ratio between the areas of the inelastic and (absolute) elastic peak was determined in the 1.5° to 8.0° angular range. From these values it was possible to determine the GOS using Eq. 1. This method has been successfully employed in the determination of the GOS values for several atoms and molecules¹⁵. A recent theoretical analysis, based on the use of an universal extrapolation formula for the GOS²⁶, has shown, for example, that our experimental values extrapolate nicely to the optical oscillator strength for the C 1s transition in the CO₂ molecule²⁷. The absolute GOS results are presented in Fig. 6 and Table 3 as a function of K^2 . The GOS-curve as a function of K^2 was extrapolated to $K^2 = 0$ using the universal formula of Msezane and Sakmar²⁸. A value of 0.312 was obtained for the optical oscillator strength from the data of Fig. 6 for the 11.5 eV transition. This result shows good agreement with the photoabsorption data of Blechschmidt *et al.*⁷ (0.33) and Lee *et al.*⁸ (0.20) and also with the EELS data of Hitchcock and van der Wiel¹¹ (0.21) and Ying *et al.*¹³ (0.23).

In the 0.0 to 1.4 a.u. K^2 range, comparison with the experimental results of Ying *et al.*¹³ shows a reasonable agreement only at $K^2 = 0.0139$ a.u. ($\theta = 0.5^\circ$) and $K^2 = 1.13$ a.u. ($\theta = 4.5^\circ$). At the moment no additional explanation can be furnished as to the observed disagreement at $K^2 = 0.349$ a.u. ($\theta = 2.5^\circ$)²⁹.

In the case of the 13.2 eV curve (see Fig. 6), a different behavior is observed. Due to the absence of enough experimental points covering the K^2 range smaller than 0.2 a.u., it can not be definitively concluded that the GOS curve tends to zero as the scattering angle decreases. However, based on the experimental data of Figs. 2 and 3 and also on the discussion previously presented, it seems reasonable to reinforce the dipole-forbidden character of such a transition.

Conclusions

Angle-resolved EELS data have been measured for the valence-shell region (0-100 eV) of the SF₆ molecule at an impact energy of 1.0 keV. At large scattering angles non-dipole transitions have been identified, in agreement with previously published results. Particularly above 40 eV,

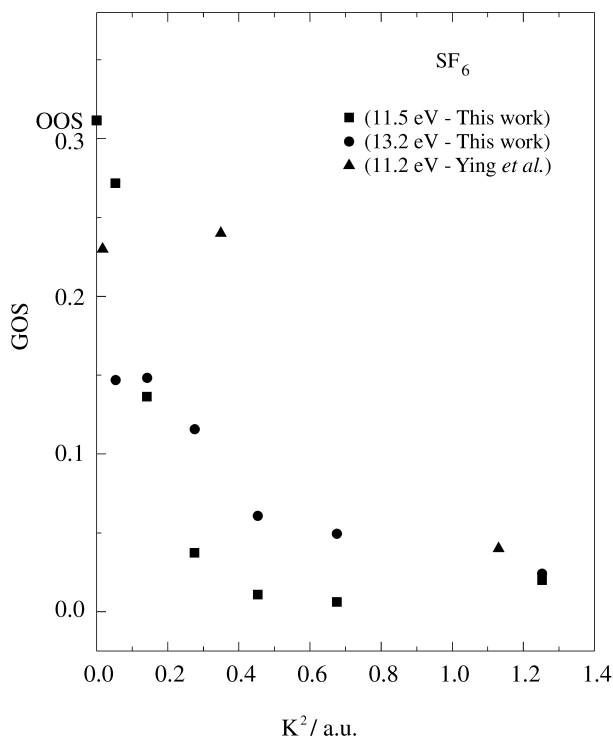


Figure 6. Generalized oscillator strength (GOS) as a function of K^2 for the 11.5 eV and 13.2 eV transitions in SF₆. The experimental data of Ying *et al.*¹³ are also presented (see text).

Table 3. Generalized Oscillator Strength for the 11.5 eV transition in SF₆.

K^2 (a.u.)	GOS
0.0	0.312
0.053	0.272
0.142	0.136
0.275	0.037
0.453	0.011
0.675	0.006
1.253	0.020

three bands associated with forbidden processes involving many-body excitations were observed. A band at 76 eV has been observed for the first time. The absolute generalized oscillator strength for the 11.5 eV excitation has been determined in the K^2 range from 0.05 to 1.3 a.u. and a value of 0.312 for the optical oscillator strength was derived. The absolute elastic differential cross section has also been determined, through normalization of the relative data using the absolute data found in the literature. The absolute inelastic differential cross sections for the 11.5 eV and 13.2 eV transitions have been also determined.

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