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Low-energy electron attachment to SF_6 molecules: Vibrational structure in the cross-section for SF_5^- formation up to 1 eV

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Abstract

In high resolution electron attachment to SF_6, cusp structure due to interchannel coupling has been observed in the cross-section for SF_5^- formation at the thresholds for vibrational excitation of the $SF_6(v_1)$ mode up to $v_1 = 10$. It is superimposed on the broad band peaking around 0.55 eV which has been previously attributed to attachment into a repulsive potential surface and subsequent direct dissociation. The newly observed vibrational structure as well as electron energy loss spectra (which exhibit strong excitation of the v_1 mode up to high quantum numbers) indicate, however, that both the channel yielding the long-lived SF_6^- anions and the dissociative $SF_5^- + F$ channel have a common primary attachment process, mediated by the same SF_6^- scattering state and strongly coupled to the process of vibrational excitation.

1. Introduction

Negative ion formation in electron attachment to SF_6 molecules at low energies ($E < 2$ eV)

$$e^-(E) + SF_6 \rightarrow [SF_6^-] \rightarrow \text{negative ions} \qquad (1)$$

is an important process in gaseous dielectrics and has been studied for a long time [1]. Near zero electron energy E, metastable SF_6^- ions are formed by s-wave attachment with a $E^{-1/2}$ behaviour of the cross-section [1–7] which reaches a value of 2×10^{-17} m^2 at $E = 1$ meV [1,3,7]. Towards higher energies (300 meV), the yield for SF_6^- formation strongly decreases by more than four orders of magnitude, and dissociative electron attachment (DEA) yielding SF_5^- ions takes over with a peak maximum located between 0.3 and 0.6 eV [1,8–12]. The shape of the DEA cross-section was found to be strongly dependent on the gas temperature T_G (i.e. on the internal rovibrational energy of the SF_6 target molecules), exhibiting a shift and a rise of the peak cross-section towards zero energy with increasing T_G [1,9–12]. To account for these observations, the higher energy peak in SF_5^- production has been attributed to attachment into a repulsive potential surface and subsequent direct dissociation [9,12,13,16]. Spence and Schulz [17], however, had found that the energy-integrated cross-section for *total* anion formation at energies below 2 eV is independent of gas temperature from 300 to 1200 K. They postulated that there is a common electronic SF_6^{-*} complex initially formed and that the branching ratios for dissociation of this complex depend upon the thermal energy initially in the SF_6 molecule. Later, an analysis of temperature dependent swarm data [18] adopted this hypothesis. We note that at electron energies above about 2 eV, other fragment anions including F^-, F_2^-, and SF_4^- are formed through several repulsive resonances [1,13].

Characteristic vibrational cusp structure due to channel interaction has been theoretically predicted [14] and experimentally confirmed [3] to occur in the SF_6^- attachment spectrum at the onset for vibrational excitation of the symmetric stretch mode $v_1 = 1$ (see also [7,15]). In this Letter, we report the new observation that this structure is clearly present in the cross-section for SF_5^- formation up to *high* vibrational thresholds (up to at least $v_1 = 10$), i.e. into the

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region of the higher energy peak with the maximum located at 0.55 eV for SF_6 molecules at room temperature ($T_G = 300$ K). This vibrational structure indicates, in contrast to other interpretations, that the attachment process, yielding long-lived SF_6^- anions and dissociated $SF_5^- + F$ products, is dominated by the same SF_6^- scattering state throughout the low energy range from zero to about 1 eV. Electron energy loss spectra are reported – extending previous work of Rohr [19] and Randell et al. [20] – which exhibit strong near-threshold vibrational excitation of SF_6 in the symmetric stretch mode v_1 up to high quantum numbers and thus support the idea of substantial interchannel coupling between the attachment and vibrational excitation channels.

2. Experimental

Relative cross-sections for SF_5^- formation have been measured at Kaiserslautern with the EXtended Laser Photoelectron Attachment (EXLPA) method over the energy range 0–2 eV at an energy resolution of about 20 meV. As described in a recent paper [12], threshold electrons (energy < 0.2 meV) are created by resonant two-step photoionization of potassium atoms in a volume which is separated from the attachment region by 53 mm. The electrons are accelerated to variable energies and magnetically guided to the electrically field-free reaction chamber which contains the SF_6 target either as a diffuse gas (temperature $T_G = 300$ K) or in a collimated seeded supersonic beam (12.5% SF_6 in helium) from a differentially pumped nozzle. For the diffuse target gas, anions due to electron attachment are extracted by a pulsed electric field, during which the photoelectron current is interrupted, and detected by a quadrupole mass spectrometer, followed by an electron multiplier and counting electronics. With the supersonic target beam, directed along the mass spectrometer axis, no anion extraction field is used, taking advantage of the initial kinetic energy of the SF_6 molecules.

A nearly homogeneous magnetic field of about 0.002 T (produced by two current coils in near-Helmholtz configuration outside the vacuum chamber) is applied along the potassium beam direction. This field guides the electrons (typical current about 40 pA) towards the reaction volume and further to the off-axis electron collector. For a proper operation of the experiment, great care has to be taken to avoid any collisions of the electron beam with surfaces on its way from the photoionization chamber to the collector since these would yield spurious low-energy electrons and thus lead to unwanted attachment processes, especially critical in energy ranges, where the attachment cross-section is small. This requires a careful alignment of the position of the focussed ionization laser (diameter 0.12 mm). Under optimal conditions, an electron energy width slightly below 20 meV has been achieved so far in the EXLPA experiments, and the drop of the SF_6^- cross-section towards higher electron energies could be followed over five orders of magnitude [12].

Electron energy loss spectra for vibrational excitation of SF_6 molecules were measured at Fribourg with an optimized high resolution spectrometer with hemispherical analyzers [21]. The resolution was about 10 meV in the energy loss mode, that is about 7 meV in the incident beam, at beam currents around 40 pA. The gas was introduced through a single nozzle with 0.25 mm diameter, made of molybdenum and kept at about 30 °C during the measurements. The energy of the incident beam was calibrated on the 19.365 eV $He^-(^2S)$ resonance [22] and is accurate to within ±10 meV. The absence of artifact structures in the cross-sections was verified by recording elastic and vibrational excitation (VE) cross-sections in CO_2.

3. Results and discussion

In this Letter, we concentrate on the attachment yield for SF_5^- formation. The complete results for both SF_6^- and SF_5^- formation, including *absolute* cross-sections for the diffuse gas target at $T_G = 300$ K, will be published elsewhere [23]. For comparison with the SF_5^- data, we mention that at $T_G = 300$ K the yield for SF_6^- ions strongly dominates at very low energies, but decreases monotonically by several orders of magnitude up to $E = 300$ meV; the SF_5^- yield starts to become higher than the SF_6^- yield at energies above about 280 meV.

In Fig. 1, we present the SF_5^- anion yield measured for the diffuse target gas ($T_G = 300$ K; SF_6 density around 5×10^{10} cm^{-3}) at a resolution of about 20 meV over the energy range 0.02–1.3 eV (channel width 1 meV). The data accumulation time per channel was 48 s; the shown spectrum (3 meV/channel) was obtained after summing over three adjacent channels of 1 meV width. At energies above 0.3 eV the overall shape of the spectrum is quite similar to that reported by Brion [8] in which the higher energy peak is found to be located at 0.53(4) eV while in other previous work the latter peak was found at energies between 0.3 and 0.4 eV [1,9–11]. We explain these differences (at least in part) by different internal (vibrational) energies of the SF_6 molecules. In electron beam experiments with a hot cathode as electron source [9–11] the target chamber (especially if rather close to the hot filament) will be at a temperature higher than 300 K; moreover, it is possible that the target has a hotter component due to molecules which originate from hotter surfaces close to the cathode and make their way to the reaction volume. Even a small fraction of hotter SF_6 molecules can make a significant contribution to the SF_5^- signal at low electron energies because of the strong rise of the low energy cross-section with gas temperature.

The main result in Fig. 1 is the interesting, previously not observed, vibrational structure in the SF_5^- attachment spectrum which is clearly visible up to about 1 eV. Closer inspection of the vibrational structure reveals – as indicated by the vertical dashed lines in Fig. 1 – a close correspondence of the experimentally observed downward cusps with the energy positions of the vibrational v_1 thresholds up to

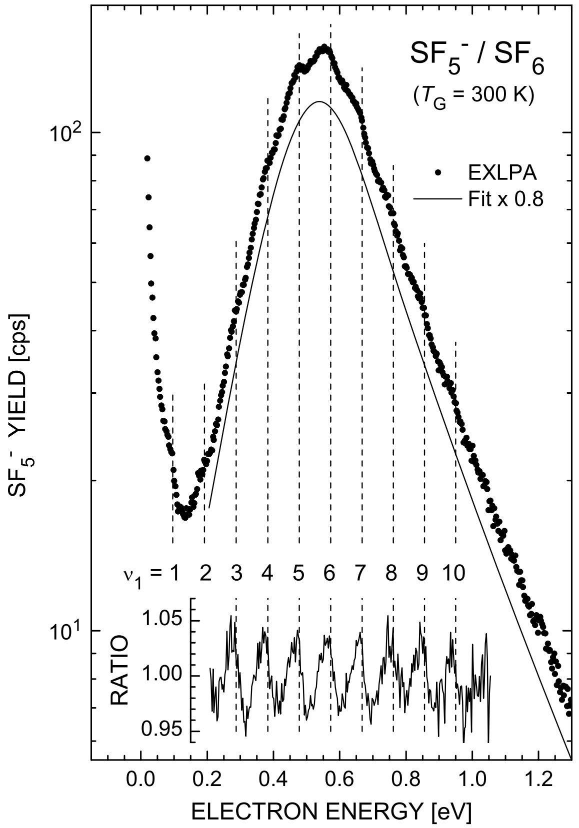

Fig. 1. Energy dependent yield for SF_5^- formation over the range 0.02–1.3 eV (3 meV per channel), as measured with the EXLPA method with a diffuse SF_6 target at $T_G = 300$ K (resolution about 20 meV). Note the vibrational structure at the onsets for excitation of the symmetric stretch vibrational mode v_1, indicated by vertical dashed lines. The structureless curve below the data represents an average fit function to the data (multiplied by 0.8 for clearer visibility). In the lower part, we show the ratio between the measured SF_5^- yield and the average fit function.

at least $v_1 = 10$. These threshold energies were measured by electron energy loss spectroscopy in Fribourg (see below). For clearer illustration we show in the lower part of Fig. 1 the ratio between the measured SF_5^- yield and the average fit function to the measured data over the range 0.2–1.2 eV: at the thresholds for v_1 vibrational excitation the ratio exhibits sharp drops which we attribute to interchannel coupling with the VE channels.

The symmetric stretch mode v_1 (a_{1g} symmetry) of the octahedral $SF_6(O_h)$ molecule has been theoretically considered [14,15] and experimentally [3,12,15] observed to be very important in the primary s-wave attachment process as signalled, e.g., by the prominence of the $v_1 = 1$ downward cusp in the SF_6^- attachment spectrum. By comparison, the feature in the SF_6^- attachment spectrum associated with the infrared active v_3 antisymmetric stretch mode is rather weak [3,12,15]. The presence of the v_1 related vibrational structure in the SF_5^- attachment spectrum up to high vibrational onsets reflects substantial interaction of the v_1 vibrational excitation channels with the *dissociative* attachment process over the full energy range of Fig. 1.

In Fig. 2, we show electron energy loss spectra (EELS) for SF_6, measured over the incident energy range 0.05–1.3 eV at the scattering angle 135° with a constant residual electron energy of $E_r = 0.1$ eV, thus emphasizing the threshold region. The energy loss spectrum is dominated by excitation of the symmetric stretch vibration, as demonstrated for quantum numbers up to $v_1 = 12$. (This is also true for a spectrum measured at $\theta = 30°$ except for the fact that now excitation of one quantum of the strongly IR-active v_3 mode is somewhat more intense than that for $v_1 = 1$.) From several EELS such as those in Fig. 2 ($E_r = 0.1$ and 0.5 eV) as well as from EELS measured at constant incident energy we found that the energy positions $E_0(v_1)$ of the peaks belonging to excitation of the v_1 vibrational mode (relative to the $v_1 = 0$ level) are accurately described by the expression:

$$E_0(v_1) = hc\omega_0 v_1 - hc\omega_0 x_0 v_1^2 \tag{2}$$

with $hc\omega_0 = 96.1(3)$ meV (i.e. the spectroscopic value [24]) and $hc\omega_0 x_0 = 0.11(3)$ meV. The energy positions for the v_1 vibrational onsets in Figs. 1 and 3 were drawn according to expression (2).

The spectrum in Fig. 2 shows that the excitation of the Raman-active v_1 mode has a large cross-section at threshold up to very high quanta and is thus likely to have a substantial effect on other cross-sections through interchannel coupling at vibrational thresholds, also up to high quanta. The spectrum also shows that v_1 is the only mode with this property – v_3 and a few other modes are weaker and, more importantly, are excited only in single quanta. These observations lend support to the idea that the vibrational structure observed in the SF_5^- attachment spectrum is caused by substantial coupling of the channels leading to the long-lived SF_6^- ion and, at higher energy, to the SF_5^- fragment, with the vibrational excitation channels.

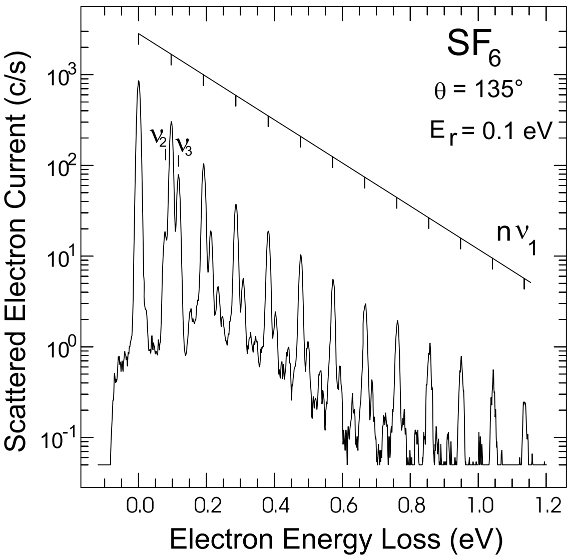

Fig. 2. Electron energy loss spectra for SF_6 molecules ($T_G = 330$ K), measured with constant residual energy $E_r = 0.1$ eV over the incident energy range 0.05–1.3 eV at the scattering angle $\theta = 135°$.

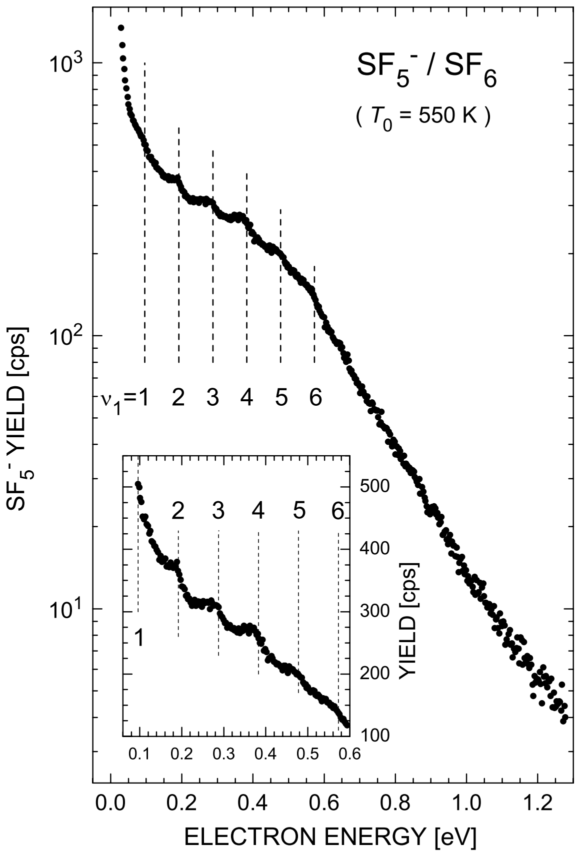

Fig. 3. Energy dependent yield for SF_5^- formation over the range 0.03–1.2 eV (3 meV per channel), as measured with the EXLPA method for a supersonic beam target (12.5% SF_6 in He; $p_0 = 1$ bar) at a nozzle temperature of $T_0 = 550$ K. Note the vibrational cusp structure at the onsets for the symmetric stretch excitation v_1 of neutral SF_6.

Previous interpretations of the SF_5^- spectra attributed the peak at higher energies to transitions from the neutral ground state to a repulsive anion surface through which (direct) dissociation occurs. The broadening of the higher energy peak towards lower energies with rising gas temperature was attributed to the fact that – within this scenario – attachment transitions out of vibrationally excited molecules need less energy (when proceeding at the turning point located at higher internuclear distances). This kind of explanation has been invoked in much of the previous work in which red-shifts of DEA bands were observed with rising gas temperature [25–28].

The new finding of the $SF_6(v_1)$ vibrational structure in the SF_5^- attachment spectrum as well as the VE spectra in Fig. 2 support a different interpretation: at electron energies up to at least 1 eV, SF_6^- and SF_5^- anion formation via electron attachment to SF_6 occurs through a single SF_6^- scattering state of octahedral symmetry in which the symmetric stretch vibration v_1 plays the central role in the primary attachment process. This state is probably a virtual state [14,15] which enhances the amplitude of the s-wave electron at the molecule. Coupling of this ‘doorway’ state to other vibrational modes by intramolecular vibrational redistribution (IVR) leads to the surprisingly long lifetime of the metastable SF_6^- anion [29], but also provides the symmetry-breaking path towards dissociation into the $SF_5^- + F$ fragments. The branching ratio for the final channels into which the SF_6^- complex evolves (SF_6^- survival, slow autodetachment to yield SF_6 plus a free electron, and SF_5^- formation via dissociation of the complex; this latter process is endothermic by about 0.15 eV [16,18]) depends on the internal energy of the molecule prior to attachment and on the electron energy E, as discussed in some detail in [18]. It could also be strongly influenced by a barrier of the SF_6^- potential surface along the dissociation path towards $SF_5^- + F$ formation.

It appears that at gas temperatures around 300 K and low electron energies ($E < 0.1$ eV), autodetachment is very slow [12,15] (although the lifetime of SF_6^- anions is still a debated issue, see , e.g. [12,29]). At a gas temperature of 300 K and near-zero electron energies only a very small fraction (about 10^{-3}) of these highly vibrationally excited SF_6^- ions (the adiabatic electron affinity of SF_6 is about 1 eV [1]) have sufficient energy to dissociate into $SF_5^- + F$ fragments, i.e. the branching ratio $R = I(SF_5^-)/[I(SF_6^-) + I(SF_5^-)]$ is close to zero. At the same gas temperature, but electron energies above about 0.3 eV the total energy of the primary SF_6^- ion is already so high that the branching ratio R approaches unity [23]. Since the total attachment cross-section is essentially independent of gas temperature [1,17], the changes of the partial cross-section for SF_5^- formation with electron energy and initial rovibrational energy of the SF_6 molecule reflect the variation of the dissociation efficiency of the SF_6^- complex with its total energy (with the *caveat* that towards higher temperatures the effects of autodetachment may progressively play a role and reduce the anion yield in a way which depends on the anion detection time).

In Fig. 3, we demonstrate that the vibrational structure in the SF_5^- yield persists even at elevated gas temperatures. The spectrum was measured with a seeded supersonic beam target (12.5% SF_6 in helium carrier gas, stagnation pressure $p_0 = 1$ bar, nozzle temperature $T_0 = 550$ K). As discussed in [12] these expansion conditions lead to a vibrational energy distribution in the SF_6 molecules which corresponds to that of a thermal target gas at a somewhat lower temperature (about $T_G = 450$ K). Spectra similar to that in Fig. 3 were reported in [12] for $T_0 = 500$ and 600 K, but due to improved counting statistics vibrational structure is now clearly visible. The structure again correlates well with the thresholds for vibrational excitation of the v_1 mode in the neutral SF_6 molecule. As a result of the enhanced primary vibrational excitation, the SF_5^- yield is now largest at zero electron energy, and the minimum at intermediate energies is lost. We note that at these expansion conditions and near-zero electron energies, the SF_6^- yield is still substantially higher than the SF_5^- yield [12], but the dissociation probability of the SF_6^- complex towards SF_5^- has strongly grown at low electron energies (the ratio $R(E \approx 0)$ is now about 0.04 [23]). In summary, the present results indicate – in agreement with the postulate in [17] (see Section 1) – that a single SF_6^- doorway state dominates

SF_6^- and SF_5^- formation in low-energy electron collisions with SF_6 molecules.

We should not conclude without noting that vibrational structure in dissociative attachment spectra may in principle have at least three different origins. Apart from the mechanism invoked here, for which the diatomic Cl^-/HCl and Br^-/HBr cases [30–32] may serve as prototypes, structure has also been observed due to vibrational (or boomerang) structure of the intermediate negative ion resonance, or due to thresholds for vibrational excitation of a molecular fragment. A prototype for both latter cases is the O^-/CO_2 process (see [33,34] and references herein). On the one hand, the vibrational structure of a resonant anion state has been observed with energy spacings different from those of the neutral molecule under study [33,34]. The fact that the vibrational spacings and positions observed in the present SF_5^- spectra coincide with those of neutral SF_6 thus speak against this explanation; moreover, the structures in the lower curve of Fig. 1 are clearly unsymmetrical (steep drops followed by rather slow rises): they are downward steps and not peaks. On the other hand, structure due to thresholds for vibrational excitation of molecular fragments has also been observed, for example due to CO in the O^-/CO_2 process. This structure appears only in an experiment measuring selectively threshold processes by detecting only fragment ions with near zero kinetic energy [33]. The structure observed here for SF_5^- formation cannot be of this origin because the present experiments do not impose energy selection for the detected anions. Additional reason speaking against this interpretation is that the vibrational spacing observed in this work fits better the vibrational spacing of the target SF_6 than that of the fragment SF_5^-. The molecular fragment SF_5^- (C_{4v} symmetry) possesses a vibrational mode of a_1 symmetry with an energy spacing (98.6 meV [35]), not far, but distinguishable from that of the $SF_6(v_1)$ mode (96.0 meV [24]). Moreover SF_5^- has, in contrast to CO in the O^-/CO_2 case, many more vibrational modes which would smear out all structure.

4. Conclusions

In a high resolution electron attachment experiment involving SF_6 molecules, vibrational structure has been observed in the cross-section for SF_5^- formation. At the thresholds for vibrational excitation of the symmetric stretch mode ($v_1 \geqslant 1$) of SF_6 the cross-section exhibits clear downward cusps due to interaction of the attachment channel with the vibrational excitation channels. While such structure was theoretically predicted [14] and observed [3] at high resolution in the cross-section for SF_6^- at the $v_1 = 1$ threshold (see also [7,15]), it is clearly seen in the SF_5^- cross-section up to the onset for $v_1 = 10$, i.e. into the higher energy region, where a peak is observed around 0.55 eV at $T_G = 300$ K. This peak has been previously attributed to attachment into a repulsive potential surface and subsequent direct dissociation. The newly observed vibrational structure as well as VE energy loss spectra (which show dominant excitation of the symmetric stretch mode v_1 up to high quantum numbers) indicate, however, that the attachment process, yielding long-lived SF_6^- anions and dissociated $SF_5^- + F$ products, as well as the vibrational excitation process are mediated by the same SF_6^- scattering state throughout the low-energy range from zero up to at least 1 eV. Following s-wave electron capture via this anion state, the system evolves along three competing channels, namely elastic scattering, vibrational excitation of the v_1 mode (responsible for cusp structure in other channels) and non-adiabatic couplings with the v_1 mode leading to intramolecular vibrational redistribution of the available energy and the formation of a long-lived SF_6^- complex. This intermediate state dissociates into $SF_5^- + F$ with a probability depending on the initial thermal energy in the neutral SF_6 molecule, on the electron energy, and on the details of the SF_6^- potential surface along the dissociation path.

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