found to consist of N_2 (0.95 mmole), $SF_2{=}NCl$ (0.69 mmole), SF_5Cl (1.80 mmoles), and Cl_2 (4.19 mmoles) by mass spectral analysis. Under similar reaction conditions SF_4 was completely converted to SF_5Cl by Cl_2 in the presence of CsF.

Decomposition Studies. A.—The photolytic decomposition of $O{=}SF_2{=}NCl$ was studied by irradiating a 4.08-mmole sample of $O{=}SF_2{=}NCl$ for 2 hr with a Hanovia lamp (Model 88A45). The crude mixture, 8.19 mmoles, consisted of 4.05 mmoles of SOF_2, 2.08 mmoles of N_2, and 2.06 mmoles of Cl_2. The related material, OSF_2NCOF, underwent no decomposition under these conditions.

B.—A 3.71-mmole sample of $SF_2{=}NCl$ was irradiated for 1 hr under the same conditions as above. The crude mixture contained Cl_2 (1.90 mmoles), N_2 (1.57 mmoles), SF_4 (1.32 mmoles), SOF_2 (0.61 mmole), FSN (0.32 mmole), and SiF_4 (0.36 mmole). A solid deposit was left in the reactor which was not soluble in water.

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CONTRIBUTION FROM THE GORGAS LABORATORY, ROHM AND HAAS COMPANY,
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The Reaction of Antimony(V) Fluoride with Tetrafluorohydrazine and Difluorodiazine[1]

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The reaction of SbF_5 with N_2F_4 and N_2F_2 produced the two adducts $N_2F_4 \cdot 2SbF_5$ and $N_2F_2 \cdot 2SbF_5$, respectively. Some of their reactions as well as their possible structures are discussed.

Antimony(V) fluoride is known to form complexes with a large number of fluorides. The alkali metal fluorides as well as other metallic fluorides form simple ionic salts containing the hexafluoroantimonate anion. Several nonmetal fluorides such as SF_4, SF_4O, BrF_3, and AsF_3 also form complexes with antimony(V) fluoride. Their structure, however, has not definitely been established, although some evidence for an ionic composition has been advanced.[2] A discussion of the complexes formed between antimony(V) fluoride and several nonmetal fluorides has been presented recently.[3] Since *cis*-difluorodiazine was reported to form fluorodiazonium hexafluoroarsenate when treated with arsenic(V) fluoride,[4] it was of interest to investigate the interaction of antimony(V) fluoride with difluorodiazine and tetrafluorohydrazine.

Experimental Section

Preparation of NF_2SbF_5 and $NF_2(SbF_5)_{1.5}$.—A mixture of 2.15 g of $AsF_3 \cdot SbF_5$ in 10 ml of AsF_3 was allowed to react with excess N_2F_4 (at a partial pressure of <100 mm) at −10° for 2.5 hr. After consumption of N_2F_4 ceased, the solvent and excess N_2F_4 were removed under vacuum and then fractionated. The residue was pumped on at 40–45° until it reached constant weight; mp 74–78°. The weight of product formed was 1.880 g (calculated weight based on the N_2F_4 consumed 1.870 g).

Anal. Calcd for $N_2Sb_3F_{19}$: Sb, 48.5; N, 3.71. Found: Sb, 48.1; N, 3.56; Sb/N, 1.55.

The above procedure and reaction temperature was repeated in a glass pressure vessel using 1.96 g of AsF_3SbF_5 in 10 ml of AsF_3 and excess N_2F_4 ($P_{N_2F_4} < 2$ atm). After consumption of the N_2F_4 ceased, the reaction mixture was treated as before. The residue was pumped on at 40–45° until it reached constant weight; mp 116–119°. The weight of product formed was 1.492 g (calculated weight based on the N_2F_4 consumed 1.526 g).

Anal. Calcd for NF_2SbF_5: N, 5.20; Sb, 45.4. Found: N, 5.23, 5.28; Sb, 44.9, 45.2.

A 0.612-g sample of NF_2SbF_5 was placed in a glass trap and approximately 2 ml of SbF_5 was condensed into the trap. The mixture was allowed to warm to ambient temperature and was stirred for 15 min. The excess SbF_5 was removed and the residue pumped on at 40–45° until it reached a constant weight of 0.851 g (calculated weight for $N_2F_4 \cdot 3SbF_5$ 0.860 g); mp 75–77°. A mixture melting point with a sample of $N_2F_4 \cdot 3SbF_5$ prepared as described above showed no depression. The complex $NF_2(SbF_5)_{1.5}$ could be converted to NF_2SbF_5 either by its reaction with N_2F_4 at $P_{N_2F_4} > 2$ atm as described above or by extraction with sulfur dioxide for 15 min at −65°.

Preparation of $NFSbF_5$.—Approximately a 2-g sample of SbF_5 was condensed onto the walls of a U tube. Then the system was pressurized to 300–400 mm of N_2F_2 (either the *trans* or the *cis* isomer could be used). The SbF_5 was allowed to melt and then a 40–50° water bath was placed around the U tube. When the consumption of N_2F_2 was nearly complete, the excess was removed and its amount determined. The residue was pumped on until it reached a constant weight of 1.592 g (calculated weight based on the N_2F_2 consumed 1.567 g); mp 82–84°.

Anal. Calcd for $NFSbF_5$: N, 5.60; Sb, 48.8. Found: N, 5.51; Sb, 48.6, 48.7.

Thermal Decomposition of NF_2SbF_5 and $NFSbF_5$.—A 0.6275-g sample of NF_2SbF_5 was placed in a U tube in a drybox. The U tube was then attached to a fractionation train on a standard vacuum line and evacuated. A 2.516-g sample of KF was placed in an adjacent U tube and this was cooled to −78°. A −196° bath was placed around the next U tube. While the system was pumped on, a 200° oil bath was raised around the U tube containing the sample and kept there for 15 min. The contents of the cold traps were then analyzed. Tetrafluorohydrazine, 1.12 mmoles of 97% purity, was obtained in the

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[1] This work was carried out under the sponsorship of the U. S. Army Missile Command, Redstone Arsenal, Ala., under Contract No. DA-01-021 ORD-11878. Preliminary results of this investigation were reported in *J. Am. Chem. Soc.*, **87**, 1140 (1965).

[2] A. A. Banks, H. J. Emeléus, and A. A. Woolf, *J. Chem. Soc.*, 2861 (1949).

[3] F. Seel and O. Detmer, *Z. Anorg. Allgem. Chem.*, **301**, 8 (1959).

[4] D. Moy and A. R. Young, *J. Am. Chem. Soc.*, **87**, 1889 (1965).

$-196°$ trap. The $-78°$ trap gained 0.472 g in weight and infrared spectroscopy showed the presence of the SbF_6^- ion.

A similar procedure was used when a 0.521-g sample of $NFSbF_5$ was decomposed at 200°. The $-196°$ trap contained 1.72 mmoles of material which analyzed for 38% *cis*-N_2F_2, 20% SiF_4, and 41% N_2O.

Reaction of NF_2SbF_5 and $NFSbF_5$ with Ferrocene.—To a 0.501-g sample of NF_2SbF_5 in 5 ml of AsF_3 at $-8°$, ferrocene, 0.405 g, was added. Gas was evolved immediately and the solution turned dark blue. After stirring for 10 min the reaction mixture was fractionated through -78 and $-196°$ cold traps. *trans*-Difluorodiazine, 0.87 mmole of 98% purity, was obtained in the $-196°$ trap. The $-78°$ trap contained arsenic trifluoride and small amounts of unreacted ferrocene. Analysis of the dark blue residue showed it to be $(C_5H_5)_2FeSbF_6$.

Anal. Calcd for $C_{10}H_{10}FeSbF_6$: C, 28.47; H, 2.39. Found: C, 28.62; H, 2.49.

The reaction of 0.371 g of $NFSbF_5$ in 5 ml of SO_2 at $-45°$ with a 0.274-g sample of ferrocene was performed in the manner described above. A 0.72-mmole sample of noncondensable gas was formed. It was removed by means of a Toepler pump and the amount determined in a calibrated bulb system. It was identified as nitrogen by mass spectroscopy. Analysis of the dark blue residue showed it to be $(C_5H_5)_2FeSbF_6$.

Anal. Calcd for $C_{10}H_{10}FeSbF_6$: C, 28.47; H, 2.39. Found: C, 28.25; H, 2.30.

Reaction of NF_2SbF_5 with NOCl and I_2.—A 1.80-mmole sample of NOCl was condensed into a mixture of 0.487 g of NF_2SbF_5 in 10 ml of AsF_3. The system was warmed to $-8°$ and stirred for 0.5 hr. Fractionation of the reaction mixture through -78, -145, and $-196°$ cold traps yielded 0.83 mmole of *trans*-N_2F_2 and 0.08 mmole of N_2O in the $-196°$ trap and 0.85 mmole of chlorine in the $-145°$ trap. Analysis of the solid residue showed that it was $NOSbF_6$.

Anal. Calcd for $NOSbF_6$: N, 5.26; F, 42.9; Sb, 45.8. Found: N, 5.11; F, 42.1; Sb, 45.3.

Iodine, 0.432 g, was added to a mixture of 1.348 g of NF_2SbF_5 in 7 ml of AsF_3 at $-78°$. The mixture was warmed to $-8°$ and stirred for 0.5 hr. The mixture was fractionated through -78 and $-196°$ cold traps. The $-196°$ trap yielded 2.41 mmoles of *trans*-N_2F_2 contaminated with traces of N_2O and SiF_4. No attempt to identify the residue was made.

Infrared Spectra.—The infrared spectra of NF_2SF_5 and $NFSbF_5$ were obtained under several conditions. Antimony(V) fluoride was used as a mulling agent and silver chloride plates were employed. The spectra were taken with a Perkin-Elmer Model 21 spectrometer. Equally good spectra were obtained in the absence of any mulling agent. A summary of the spectra of $NFSbF_5$ and NF_2SbF_5 is presented in Table I.

Nmr Spectrum.—The F^{19} nmr spectrum of NF_2SbF_5 was obtained using a Varian Model V4310A spectrometer operating at 40 Mc. Trifluoroacetonitrile was used as an internal standard. It has a chemical shift of ϕ +55.8 in liquid SO_2 at $-70°$ (ϕ is defined as ppm measured from CCl_3F).

TABLE I

INFRARED SPECTRA OF NF_2SbF_5 AND $NFSbF_5$ (CM^{-1})[a]

NF_2SbF_5	$NFSbF_5$	$CF_2{=}NF$
1517 m	1054 m	1733 s
1340 sh	704 vs, br	1376 s
1300 ms	673 vs	1017 s
1124 ms	656 vs, br	930 m
997 vw	516 m, vbr	
926 ms		
698 w, sh		
666 vs, br		
615 w, sh		
498 m, br		

[a] s, strong; v, very; m, medium; w, weak; br, broad; sh, shoulder.

The F^{19} nmr spectrum of NF_2SbF_5 was obtained in sulfur dioxide at $-70°$. No change in this spectrum was observed after several hours and the complex recovered after the run showed no change in melting point. It is concluded that no reaction of the complex with the solvent occurred under these conditions. The complex NF_2SbF_5 was found to be extremely soluble in sulfur dioxide and solution occurred at $-70°$ when 2.5–3 moles of sulfur dioxide per mole of NF_2SbF_5 had been added. On warming slightly, a solid precipitate formed in the mixture. It is possible that a very soluble or even a liquid solvate is formed which is stable only at low temperatures. The spectrum was also obtained in liquid SO_2 at 25° and on a melt of the compound at 120°. The F^{19} nmr spectrum obtained at low temperature is shown in Figure 1.

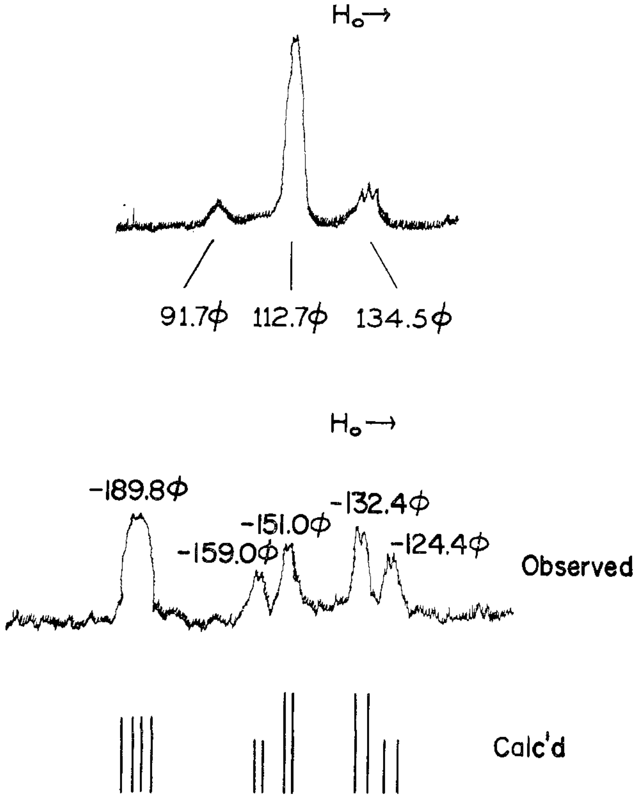

Figure 1.—F^{19} nmr spectrum of NF_2SbF_5.

Molecular Weight Determination.—The equipment used was described previously.[5] Arsenic trifluoride was distilled over NaF and stored in polyethylene bottles. All preparation of solutions was done in a nitrogen-filled drybox. The value of the freezing point depression constant used was taken from the literature.[6] A typical determination is described below: the addition of 2.192 g of NF_2SbF_5 to 83.612 g of AsF_3 resulted in a depression of the freezing point of 0.76°. This gives a value of 255 for the molecular weight of NF_2SbF_5.

Analysis.—Nitrogen analyses were performed by the Dumas method. Antimony was determined by the bromate method. Analyses of gaseous mixtures were performed with the aid of a mass spectrometer which had been calibrated with the known components of the mixture.

Results

N_2F_4–SbF_5 System.—Tetrafluorohydrazine was found to react with antimony(V) fluoride to produce a complex having the composition NF_2SbF_2. However, the system is complicated in that evidence for the formation of

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[5] J. K. Ruff and M. F. Hawthorne, *J. Am. Chem. Soc.*, **83**, 535 (1961).

[6] L. Kolditz and S. Engels, *Z. Anorg. Allgem. Chem.*, **302**, 88 (1959).

another complex with the approximate composition $NF_2(SbF_5)_{1.5}$ was found. When an excess of tetrafluorohydrazine, at high pressure, was allowed to react with the complex AsF_3SbF_5 in arsenic(III) fluoride solution, the material NF_2SbF_5 (I) was obtained. If, however, the partial pressure of tetrafluorohydrazine was below 100 mm, even though an excess was employed, another material approaching the composition $NF_2(SbF_5)_{1.5}$ (II) was formed. This later material could also be obtained by the reaction of tetrafluorohydrazine with excess antimony(V) fluoride regardless of the partial pressure of the former. It was possible to convert I to II by treatment with excess antimony(V) fluoride. The reverse transformation could be accomplished by the reaction of II with excess N_2F_4 at high pressure in AsF_3 solution or by extraction of II with sulfur dioxide at low temperature. It was not possible to prepare I in liquid sulfur dioxide although the adduct appears to be stable in this solvent at low temperatures.

Both NF_2SbF_5 and $NF_2(SbF_5)_{1.5}$ are white solids, mp 116–119 and 74–78°, respectively. The melting points of these materials seem to be dependent on the heating rate, although little decomposition was noted at this temperature. The thermal decomposition of NF_2SbF_5, which was only slight at 150°, was complete at 200° and obeyed the equation

$$2NF_2SbF_5 \xrightarrow{200^\circ} N_2F_4 + 2SbF_5$$

A similar reaction was noted when I was heated to 125° in the presence of potassium fluoride

$$2KF + 2NF_2SbF_5 \xrightarrow{125^\circ} N_2F_4 + 2KSbF_6$$

A 98% recovery of the N_2F_4 originally charged was achieved in both of the above reactions. The complex NF_2SbF_5 was found to be extremely reactive toward a wide variety of materials. It ignited acetone, ethanol, and other simple organic materials, and decomposition was noted in CF_3COOH or $POCl_3$. It reacted with water explosively to produce NF_3, N_2F_2, oxides of nitrogen, SiF_4, and oxygen.

The reaction of NF_2SbF_5 with materials capable of being oxidized to form cations was found to produce N_2F_2. Several examples of this reaction are listed below

$$2(C_5H_5)_2Fe + 2NF_2SbF_5 \xrightarrow[AsF_3]{-8^\circ} 2(C_5H_5)_2FeSbF_6 + N_2F_2$$

$$2NOCl + 2NF_2SbF_5 \xrightarrow[AsF_3]{-8^\circ} 2NOSbF_6 + Cl_2 + N_2F_2$$

Treatment of NF_2SbF_5 with 0.5 equiv of iodine also produced N_2F_2 in similar yields. A similar reaction occurred with bromine, but chlorine was unreactive. The halogen species were not characterized. Only *trans*-N_2F_2 was obtained in these reactions, so this appears to be a good method of preparation for this isomer of N_2F_2.

N_2F_2–SbF_5 System.—Contrary to the results reported when AsF_5 was employed as a Lewis acid,[4] both the *cis* and *trans* isomers of N_2F_2 were found to react with antimony(V) fluoride to produce a solid complex having the composition $NFSbF_6$ (III). This material was much more reactive than either I or II. It was impossible to find a solvent in which it could be prepared, so the neat reaction between the components was employed. The complex was found to react rapidly with AsF_3 to produce nitrogen and AsF_5. Sulfur dioxide, when allowed to stand with $NFSbF_6$, yielded N_2O, N_2, SOF_2, SO_2SbF_5, and SO_2F_2. No nonvolatile residue was obtained, which indicates that no $NOSbF_6$ was formed.

The thermal decomposition of $NFSbF_6$ proceeded readily at 200°. However, the decomposition was not quantitative and only about 40% of the charged N_2F_2 could be recovered. Some N_2O and SiF_4 were also formed (presumably by reaction of N_2F_2 with glass). The interesting feature of the decomposition is that essentially pure *cis*-N_2F_2 is recovered, regardless of which isomer of N_2F_2 was used in the preparation of the complex. The reaction of III with KF also yielded only *cis*-N_2F_2, but again the recovery was not complete. The reaction of $N_2F_2 \cdot 2SbF_5$ with ferrocene produced only nitrogen and $(C_5H_5)_2FeSbF_6$.

Discussion

Antimony(V) fluoride is a strong Lewis acid capable of forming stable complexes with nitrogen bases. In addition it forms the very stable hexafluoroantimonate with fluorine ion donors and forms fluoride bridges with itself.[7] Thus a discussion of the composition of the adducts $NFSbF_6$ and NF_2SbF_5 must consider these three types of complexes. Although the characterization of these materials by standard methods was made complicated by their high reactivity, infrared and F^{19} nmr spectroscopy in the case of NF_2SbF_5 yielded significant information.

The F^{19} nmr spectrum of NF_2SbF_5 was obtained at three different temperatures: −70° in (SO_2), 25° (in SO_2), and 120° (as a melt). The spectra obtained at the two higher temperatures were practically identical. A single band was observed in the SbF region and three broad peaks having relative areas of 1.0:1.05:0.97 were observed at lower field. Much better resolution was achieved at −70°, and the spectrum obtained is reproduced in Figure 1. The appearance of the bands at low field is typical of an ABX group in which all eight bands of the AB group are observed. The X portion of the spectrum is less well defined. The values of the coupling constants and the chemical shifts were obtained by the method outlined by Pople.[8] Since there are two ways of choosing the two quartets of the AB group, four sets of the coupling constants, J_{AX} and J_{BX}, were obtained.[9] Two of these could be eliminated by a comparison of the calculated and observed intensity of the bands in the X portion of the spectra. The fit of

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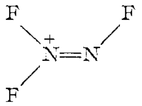
(7) C. H. Hoffman, B. E. Holder, and W. L. Jolly, *J. Phys. Chem.*, **62**, 364 (1958).

(8) J. H. Pople, W. G. Schneider, and H. J. Bernstein, "High-Resolution Nuclear Magnetic Resonance," McGraw-Hill Book Co., Inc., New York, N. Y., 1959.

(9) Numbering the AB peaks from left to right the two sets of quartets are: 1, 3, 6, 8 and 2, 4, 5, 7 or 1, 3, 5, 7 and 2, 4, 6, 8.

the calculated and observed spectra was very good using either of the two remaining sets of values (see Figure 1). The values thus obtained are $J_{AB} = 379$ cps, $J_{AX} = 81$ cps ($J_{AX} = 82$ cps), and $J_{BX} = 45$ cps ($J_{BX} = -50$ cps) and the chemical shifts are δ_X, ϕ -189.8; δ_A, ϕ -128.8; and δ_B, ϕ -154.0. If this ABX group is attributed to the fluorines bound to nitrogen, then a F_2NNF group (or less likely a N_4F_6 group) which contains three (or three sets of) nonequivalent fluorine must be present in the adduct. The nonequivalency of the fluorine can be readily explained if there is hindered rotation around the N–N bond in the N_2F_3 group. This can occur either because of steric or electronic repulsion factors, as has been observed for N_2F_4 at low temperatures,[10] or because of π bonding between the two nitrogens. Since the hindered rotation is still evident at 120°, the latter explanation appears more reasonable. The extent of π bonding would be greatly enhanced in a cationic species over that in a neutral or anionic species. Evidence for increased π bonding in the N-fluorodiazonium cation over that in N_2F_2 has been found.[4] No evidence for coupling between fluorine and nitrogen was observed at −70°, which is likely due to an increase in the quadrupole relaxation at low temperatures. The loss of resolution at higher temperatures may be a result of increased nitrogen–fluorine coupling.



Unambiguous assignment of the observed coupling constants to particular interactions cannot be made. However, in the isoelectronic compound $CF_2{=}NF$,[11] which contains three nonequivalent fluorines, the *trans* coupling constant is larger than either the geminal or *cis* coupling constants. This is also true in a number of substituted fluorocarbon olefins.[8] Furthermore the J_{FF} coupling constant in *trans*-N_2F_2 is close to the value obtained for J_{AB}, and the J_{FF} coupling constant in the *cis* isomer is close to the value for J_{AX}.[12] On the other hand, the only reported value of a coupling constant for nonequivalent fluorines in a NF_2 group is approximately 600 cps.[10] This value would be expected to decrease upon π-bond formation due to a decrease in the electron density on the nitrogen containing the two fluorines.

The single band observed in the SbF region of the spectrum at 25° split into three peaks upon cooling to −70°. Their relative areas are 1.0 (ϕ 91.7); 8.3 (ϕ 112.7); 1.9 (ϕ 134.5). Fine structure was observed on the two larger peaks. The smaller of these is a quintuplet $J_{FF} = 93$ cps and the other appears to consist of two overlapping doublets although the resolution is not very good ($J_{FF} = 60$ cps and $J_{FF} = 93$ cps). This spectrum is consistent with the antimony species $Sb_2F_{11}^-$, which has recently been identified by Gillespie in anhydrous hydrogen fluoride.[13] The anion consists of two octahedra joined by a common apex and thus contains three types of fluorine: one bridging (b), two apical (a), and eight equatorial (e). The values of the coupling constants J_{ae} and J_{be} observed by Gillespie were 100 and 53 cps, in fair agreement with those obtained here. The observed collapse of these peaks into a single broad peak at 25° and above probably arises from an exchange process involving cleavage of the fluorine bridge. In anhydrous HF solution, a similar temperature effect on the spectrum of the $Sb_2F_{11}^-$ ion was observed.

The epr spectrum of NF_2SbF_5 showed no evidence for the presence of an unpaired electron under three different conditions: the solid state, in solution of SO_2 at −70°, and in arsenic trifluoride solution at −5°. Thus, it is concluded that the N–N bond in tetrafluorohydrazine either is intact in the complex or has undergone heterolytic cleavage. An effort was made to obtain a molecular weight of NF_2SbF_5 in AsF_3 solution. The average value obtained was 260 ± 26; however, the material recovered after the run had undergone partial decomposition as evidenced by a melting point depression of approximately 7°. Therefore, some error may be in this value. The lack of an epr signal and the observed molecular weight are in agreement with the ionic formation of the adduct, $N_2F_3^+Sb_2F_{11}^-$, assuming that complete dissociation occurs in solution.

The infrared spectra of the two adducts, NF_2SbF_5 and $NFSbF_5$, are presented in Table I. Also included are the data for $CF_2{=}NF$.[11] The three strong bands in the spectrum of NF_2SbF_5 at 1300, 1124, and 926 cm^{-1} probably arise from NF stretching modes in the $N_2F_3^+$ ion. The fair correspondence with the CF and NF stretching modes in the isoelectronic compound $CF_2{=}NF$ (*e.g.*, 1376, 1017, and 930 cm^{-1}) lends support to this assignment. The band of medium to weak intensity at 1517 cm^{-1} is in the region attributed to the N=N stretching mode of *cis*-N_2F_2[14] and several perfluoroazoalkanes.[15,16] The strong band at 666 cm^{-1} arises from Sb–F stretching modes.[17] The spectrum of the other adduct has only a single band, at 1054 cm^{-1}, in the NF region. This is very close to the band attributed to the NF stretching mode in the N_2F^+ ion.[4] Furthermore, the liberation of only the *cis* isomer of N_2F_2 upon treatment of the adduct with KF is identical with the observed behavior of N_2FAsF_6. Therefore, it is felt that this adduct contains the N_2F^+ cation. The coordination site of the second mole of SbF_5 is not known definitely, but it is possible that the $Sb_2F_{11}^-$ ion is again present. Thus, this material is probably $N_2F^+Sb_2F_{11}^-$.

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