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Reaction of 1-Oxa-3-azabutatrienium Salts with Tertiary Carboxamides

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α-Chloro isocyanates **1** react with tertiary carboxamides or acylimines **3** in the presence of Lewis acids ($SbCl_5$, $FeCl_3$) to give the amino-substituted 2-azaallenium salts **5a – t** under mild conditions. With methanol the moderately strong electrophiles **5** afford the acetals **8** and **10**. According to an *X*-ray structural analysis, the $C = \overset{+}{N} = C$ unit of **5a** is bent by 128°. There exists a linear relationship between the wave number of the antisymmetric stretching vibration of 2-azaallenium salts and the $C = \overset{+}{N} = C$ bond angle. The planes defined by the sp^2 carbons of the $C = \overset{+}{N} = C$ moiety of **5a** are almost perpendicular (84°) with respect to each other.

Reaktion von 1-Oxa-3-azabutatrienium-Salzen mit tertiären Carbonsäureamiden

Die α-Chlorisocyanate **1** reagieren mit tertiären Carbonsäureamiden oder Acyliminen **3** in Gegenwart von Lewis-Säuren ($SbCl_5$, $FeCl_3$) unter milden Bedingungen zu den aminosubstituierten 2-Azaallenium-Salzen **5a – t**. Die nur mäßig starken Elektrophile **5** setzen sich mit Methanol zu den Acetalen **8** und **10** um. Eine Röntgenstrukturanalyse von **5a** zeigt, daß die $C = \overset{+}{N} = C$-Einheit dieses Moleküls gewinkelt ist (128°). Es besteht eine Proportionalität zwischen der Wellenzahl der antisymmetrischen Valenzschwingung von 2-Azaallenium-Salzen und dem $C = \overset{+}{N} = C$-Bindungswinkel. Die durch die sp^2-Kohlenstoffe der $C = \overset{+}{N} = C$-Einheit von **5a** definierten Ebenen stehen nahezu senkrecht (84°) aufeinander.

In preceding papers we reported on the preparation of the pseudocumulenes **2**[1)] and described their reactions with sulfoxides[2)], ketones, and aldehydes[3)]. It was now found that in the presence of Lewis acids the α-chloro isocyanates **1** react with tertiary carboxamides **3** to give the amino-substituted 2-azaallenium salts **5**.

Reactions of isocyanates with carbonyl groups are known since the time of *Staudinger*[4)] who kept *p*-(dimethylamino)benzaldehyde and phenyl isocyanate for 14 h at 190°C to obtain *N*-[*p*-(dimethylamino)benzylidene]aniline. Highly polarizable carbonyl compounds, e.g. tropone[5)], react with activated isocyanates like trichloroacetyl isocyanate already at room temperature. Formation of amidines from isocyanates and secondary or tertiary carboxamides has been observed by *Logemann* et al.[6–9)] and others[10–15)]. Usually, high reaction temperatures are required for nonactivated isocyanates. Phenyl isocyanate and tetramethylurea form *N,N'*-tetramethyl-*N''*-phenylguanidine[16)]. In the reaction of carbamoyldiphenylphosphane with *p*-toluenesulfonyl isocyanate the oxazetidinone **6** has been isolated in low yields[17)]. Oxazetidinones have also been obtained from catalyzed reactions of alkyl isocyanates with ketones bearing strong electron withdrawing substituents[18,19)].

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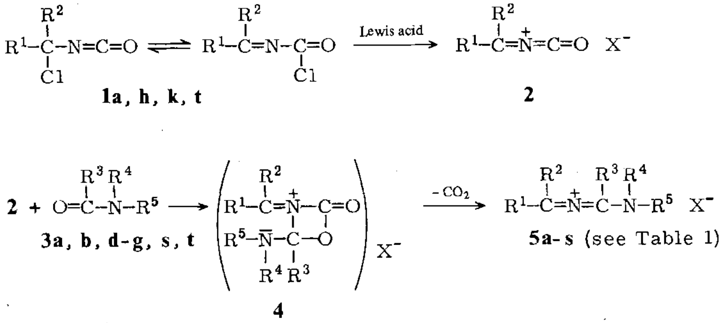
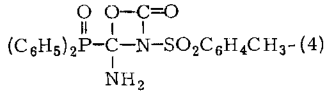
Stirring a solution of equimolecular amounts of α-chloro isocyanate **1a**, dimethylformamide, and antimony pentachloride for four hours at room temperature afforded the 2-azaallenium salt **5a** in 84% yield. The same compound was obtained from isothiocyanate **7**, antimony pentachloride, and dimethylformamide. Since the reaction products of **7** contained impurities, which we found hard to remove, and since the isocyanates **1** are more easily accessible[20,21], reactions with α-chloro isothiocyanates (e.g. **7**) were not further pursued.



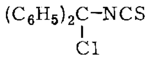
$$\underset{\mathbf{1a,\,h,\,k,\,t}}{R^1-\underset{Cl}{\overset{R^2}{C}}-N=C=O} \rightleftharpoons R^1-\overset{R^2}{C}=N-\underset{Cl}{C}=O \xrightarrow{\text{Lewis acid}} \underset{\mathbf{2}}{R^1-\overset{R^2}{C}=\overset{+}{N}=C=O\ X^-}$$

$$\mathbf{2} + \underset{\mathbf{3a,\,b,\,d\text{-}g,\,s,\,t}}{O=\overset{R^3}{C}-\overset{R^4}{N}-R^5} \longrightarrow \underset{\mathbf{4}}{\left(\begin{array}{l} R^1-\overset{R^2}{C}=\overset{+}{N}-C=O \\ \qquad\quad\ \,|\qquad\, | \\ R^5-\underset{R^4}{\bar{N}}-\underset{R^3}{C}-O \end{array}\right) X^-} \xrightarrow{-CO_2} \underset{\mathbf{5a\text{-}s}\ \text{(see Table 1)}}{R^1-\overset{R^2}{C}=\overset{+}{N}=\overset{R^3}{C}-\overset{R^4}{N}-R^5\ X^-}$$



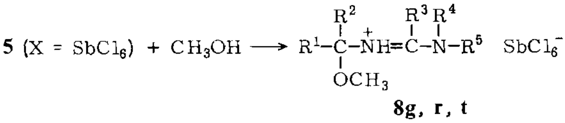
$$(C_6H_5)_2\overset{O}{\overset{\|}{P}}-\underset{NH_2}{\overset{O-C=O}{\overset{|}{C}}}\!\!-\overset{|}{N}-SO_2C_6H_4CH_3\text{-}(4)$$

6



$$(C_6H_5)_2\underset{Cl}{C}-NCS$$

7



$$5\ (X = SbCl_6) + CH_3OH \longrightarrow \underset{\mathbf{8g,\,r,\,t}}{R^1-\underset{OCH_3}{\overset{R^2}{C}}-\overset{+}{N}H=\overset{R^3}{C}-\overset{R^4}{N}-R^5\ SbCl_6^-}$$

The scope of the reaction of α-chloro isocyanates **1** with tertiary amides in the presence of Lewis acids to give the salts **5** seems to be broad. Amides of aromatic and aliphatic carboxylic acids, of the sterically unhindered formic acid and the sterically crowded pivalic acid react under essentially the same conditions. The Lewis acid ferric chloride is as reactive as antimony pentachloride. While the *tert*-butyl-substituted α-chloro isocyanate **1t** does not react with aldehydes or ketones[3], reaction with dimethylformamide gave the 2-azaallenium salt **5t** as an oil, which was characterized as its crystalline methanol adduct **8t**. No reaction was observed between 1-*tert*-butyl-1-chloro-2,2-dimethylpropyl isocyanate (**1**, $R^1 = R^2 =$ *tert*-butyl) and dimethylformamide, even after prolonged heating. Reaction of **1t** with *N,N*-dimethylacetamide (**3b**) led to an impure compound, which according to the NMR spectra is a complex of antimony pentachloride with **3b**. A similar complex was obtained from the reaction of **1k** with tetramethylurea and antimony pentachloride.

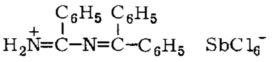
With the acylimine **3s** the 2-azaallenium salt **5s** with delocalized positive charge has been obtained. This compound bears relationship to certain diazatrimethinecyanine dyes[22].

Table 1. Residues $R^1 - R^5$ and X for **1** – **5**, **8** and Antisymmetric IR Absorptions of $C=\overset{+}{N}=C$ of Compounds **5**

1 – **5**, **8**	R^1	R^2	R^3	R^4	R^5	X	$\nu_{as}(C=\overset{+}{N}=C)$ [cm^{-1}] of **5** (CH_2Cl_2)
a	C_6H_5	C_6H_5	H	CH_3	CH_3	$SbCl_6$	1650
b	C_6H_5	C_6H_5	CH_3	CH_3	CH_3	$SbCl_6$	1630
c	C_6H_5	C_6H_5	CH_3	CH_3	CH_3	$FeCl_4$	1630
d	C_6H_5	C_6H_5	$CH(CH_3)_2$	CH_3	CH_3	$SbCl_6$	1660
e	C_6H_5	C_6H_5	$C(CH_3)_3$	CH_3	CH_3	$SbCl_6$	1630
f	C_6H_5	C_6H_5	C_6H_5	CH_3	CH_3	$SbCl_6$	1610
g	C_6H_5	C_6H_5	$-[CH_2]_3-$		CH_3	$SbCl_6$	1650
h	C_6H_5	$1\text{-}C_{10}H_7$	H	CH_3	CH_3	$SbCl_6$	1650
i	C_6H_5	$1\text{-}C_{10}H_7$	CH_3	CH_3	CH_3	$SbCl_6$	1640
j	C_6H_5	$1\text{-}C_{10}H_7$	$-[CH_2]_3-$		CH_3	$SbCl_6$	1650
k	$2\text{-}C_6H_4-C_6H_4\text{-}2'$		H	CH_3	CH_3	$SbCl_6$	1680
l	$2\text{-}C_6H_4-C_6H_4\text{-}2'$		H	CH_3	CH_3	$FeCl_4$	1680
m	$2\text{-}C_6H_4-C_6H_4\text{-}2'$		CH_3	CH_3	CH_3	$SbCl_6$	1690
n	$2\text{-}C_6H_4-C_6H_4\text{-}2'$		CH_3	CH_3	CH_3	$FeCl_4$	1690
o	$2\text{-}C_6H_4-C_6H_4\text{-}2'$		$CH(CH_3)_2$	CH_3	CH_3	$SbCl_6$	1700
p	$2\text{-}C_6H_4-C_6H_4\text{-}2'$		$C(CH_3)_3$	CH_3	CH_3	$SbCl_6$	1715
q	$2\text{-}C_6H_4-C_6H_4\text{-}2'$		C_6H_5	CH_3	CH_3	$SbCl_6$	1690
r	$2\text{-}C_6H_4-C_6H_4\text{-}2'$		$-[CH_2]_3-$		CH_3	$SbCl_6$	1695
s	C_6H_5	C_6H_5	C_6H_5	$(C_6H_5)_2C$		$SbCl_6$	1630
t	C_6H_5	$C(CH_3)_3$	H	CH_3	CH_3	$SbCl_6$	

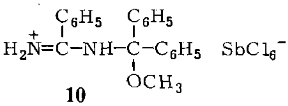
The reaction of α-chloro isocyanates **1** with tertiary carboxamides or acylimines can not be extended to carboxylic esters or carboxylic chlorides; e.g. from the reaction of **1a** or **1k** with antimony pentachloride and methyl benzoate, ethylene carbonate, or benzoyl chloride in boiling dichloromethane only starting material was recovered.

α-Chloro isocyanates **1** and Lewis acids react to give the 1-oxa-3-azabutatrienium salts **2**, which have been characterized by NMR spectroscopy and by an *X*-ray structural analysis[1]. Therefore, it seems likely that the formation of **5** from **1** and **3** in the presence of a Lewis acid proceeds via **2**. The intermediate **2** is attacked by the electron rich carbonyl group of the carboxamide to give a second intermediate **4**. In related reactions compounds of type **4** have been isolated[17-19].



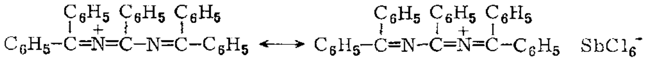
$H_2\overset{+}{N}=\overset{C_6H_5}{C}-N=\overset{C_6H_5}{C}-C_6H_5 \quad SbCl_6^-$

9



$H_2\overset{+}{N}=\overset{C_6H_5}{C}-NH-\underset{OCH_3}{\overset{C_6H_5}{C}}-C_6H_5 \quad SbCl_6^-$

10



$C_6H_5-\overset{C_6H_5}{C}=\overset{+}{N}=\overset{C_6H_5}{C}-N=\overset{C_6H_5}{C}-C_6H_5 \longleftrightarrow C_6H_5-\overset{C_6H_5}{C}=N-\overset{C_6H_5}{C}=\overset{+}{N}=\overset{C_6H_5}{C}-C_6H_5 \quad SbCl_6^-$

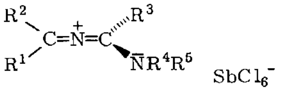
5s

The salts **5** are quite stable crystalline compounds with only moderate electrophilic properties. For instance, most compounds **5** survive short treatment with methanol at room temperature. In boiling methanol the acetals **8** are obtained. The alkylideneamino-substituted 2-azaallenium salt **5s** reacts with methanol at room temperature to give the alkylideneamidinium salt **9**, which with excess of methanol affords the acetal **10**.

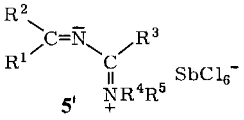
Monoamino-substituted 2-azaallenium salts **5** (R^5 = H) have been obtained from the reaction of imines with nitrilium salts[23].

$$R^1R^2C{=}NH + R^3{-}C{\equiv}\overset{+}{N}{-}R^4 \ \ SbCl_6^- \longrightarrow \mathbf{5}\ (R^5 = H)$$

In their IR spectra these compounds show a strong absorption between 1645 and 1825 cm^{-1}, which has been assigned to the antisymmetric stretching vibration of the $C=N=C$ moiety. The wave number of this band was found to be strongly dependent on the sterical requirements of the substituents R^1, R^2, R^3. The spectra were interpreted in terms of an equilibrium between two valence tautomeric forms **5** ($C=N=C$ unit with local D_{2d} symmetry) and **5'** (bent $C=N-C$ unit with local C_{2v} symmetry)[24]. Bulky substituents stabilize the allenium form **5**[25]. For the 2-azaallenium salt **11** with an IR absorption at 1825 cm^{-1} an *X*-ray structural analysis[23] revealed a molecular shape, which comes close to the allenium geometry **5**. We believed that the smaller wave number $\nu'_{as}(C=N)$ = 1650 cm^{-1} (CH_2Cl_2), which was observed for **5a**, might indicate that this compound assumes geometry **5'** in solution. Therefore, an *X*-ray structural analysis was carried out for **5a**.



5



5'

X-Ray Diffraction Analysis of 5a*)

5a, $[C_{16}H_{17}N_2]^+[SbCl_6]^-$, monoclinic, space group $P2_1/c$ (No. 14[26]), a = 978.9 (4), b = 2237 (1), c = 1680.0 (7) pm, β = 144.26 (2)°, $V = 2149 \cdot 10^6$ pm^3, $d_{calc.}$ = 1.77 gcm^{-3}, $\mu_{Mo\text{-}K_\alpha}$ = 20.4 cm^{-1}, T = 238 K, ω-scan, $\Delta\omega$ = 1.0°, $2.0 < \dot{\omega} < 29.3°$ min^{-1}, $2° \leqslant 2\theta \leqslant 42°$; 2040 independent significant reflections ($I \geqslant 2\sigma$). The cell constants and the reflections were measured on a Syntex P3-diffractometer with a graphite monochromator, $\lambda_{Mo\text{-}K_\alpha}$ = 71.069 pm. The structure was solved using the program SHEL-XTL[27] by direct methods. Hydrogen atoms, with the exception of H2, were fixed on calculated geometrically ideal positions. The partially anisotropic refinement with full matrix led to agreement factors R_1 = 0.035 and R_2 = 0.045.

A list of atomic coordinates with LS-computed standard deviations is given in Table 2. Fig. 1 shows a molecular plot with selected bond lengths, bond angles, and torsional angles for the cation of **5a**.

The crystal of **5a** consists of discrete $[C_{16}H_{17}N_2]^+$ cations and $SbCl_6^-$ anions. Comparing the structures of **5a**, **11**[23], **12**[28], and **13**[25] one observes that in all cases the planes defined by the sp^2 hybridized carbon atoms of the $C=\overset{+}{N}=C$ unit are almost perpendicular (torsional angle β) with respect to one another (allene topology). Recent *ab initio* calculations predict[25] a linear $C=N=C$ moiety of form **5** for the monoamino-substituted 2-azaallenium cation while for polyamino-substituted 2-azaallenium cations

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*) Further details and basic data concerning the *X*-ray analysis may be obtained from Fachinformationszentrum Energie Physik Mathematik, D-7514 Eggenstein-Leopoldshafen (W. Germany), by specifying the registry number CSD 50860, authors, and source.

bent C = N = C units with the symmetry of **5'** are favoured. Indeed, Gold's reagent **14**[29] has the molecular shape of **5'**[30], while the monoprotonated biguanide **15** assumes a topology intermediate to those of **5** and **5'** thus minimizing steric strain between the large amino groups[31-33]. Till now, no 2-azaallenium cation with a completely linear $C=\overset{+}{N}=C$ unit has been described[25].

Table 2. Fractional Atomic Coordinates and Temperature Parameters for **5a**[a]

atom	x/a	y/b	z/c	U11	U22	U33	U23	U13	U12
Sb	0.7561(1)	1.14392(2)	0.66328(6)	0.0288(3)	0.0285(3)	0.0257(3)	-0.0001(3)	0.0224(3)	0.0005(3)
Cl1	0.5836(4)	1.2267(1)	0.6475(3)	0.058(2)	0.043(1)	0.059(2)	0.015(1)	0.052(2)	0.019(1)
Cl2	0.8338(4)	1.1010(1)	0.8261(2)	0.061(2)	0.040(1)	0.038(1)	0.004(1)	0.041(1)	-0.001(1)
Cl3	1.1192(4)	1.1946(1)	0.8394(2)	0.035(1)	0.071(2)	0.040(1)	-0.013(1)	0.028(1)	-0.016(1)
Cl4	0.6742(4)	1.1884(1)	0.4998(2)	0.061(2)	0.051(2)	0.044(1)	0.000(1)	0.046(1)	-0.007(1)
Cl5	0.9356(6)	1.0618(1)	0.6853(3)	0.089(2)	0.058(2)	0.076(2)	0.008(2)	0.070(2)	0.028(2)
Cl6	0.3915(4)	1.0953(1)	0.4854(3)	0.051(2)	0.078(2)	0.040(1)	-0.017(1)	0.035(1)	-0.031(1)
N1	0.715(1)	0.8104(3)	0.8521(7)	0.036(4)	0.024(4)	0.035(4)	0.002(3)	0.029(4)	0.001(3)
N2	0.841(1)	0.7119(3)	0.9102(7)	0.043(5)	0.022(4)	0.040(4)	0.007(3)	0.035(4)	0.004(3)

atom	x/a	y/b	z/c	U	atom	x/a	y/b	z/c	U
C1	0.746(1)	0.8635(4)	0.8378(8)	0.026(2)	C2	0.875(1)	0.7679(4)	0.9415(8)	0.031(2)
H2	1.02(1)	0.776(3)	1.049(7)	0.04(2)	C3	0.572(1)	0.9094(4)	0.7784(8)	0.027(2)
C4	0.509(1)	0.9554(4)	0.6965(8)	0.032(2)	C5	0.349(2)	0.9977(4)	0.6446(9)	0.036(2)
C6	0.250(2)	0.9950(4)	0.6721(9)	0.037(2)	C7	0.307(1)	0.9495(4)	0.7510(8)	0.034(2)
C8	0.468(1)	0.9066(4)	0.8039(8)	0.031(2)	C9	0.944(1)	0.8786(4)	0.8787(8)	0.025(2)
C10	1.043(2)	0.8357(4)	0.8766(9)	0.036(2)	C11	1.239(2)	0.8483(4)	0.9244(9)	0.045(2)
C12	1.336(2)	0.9055(4)	0.975(1)	0.048(3)	C13	1.237(2)	0.9483(4)	0.9736(9)	0.044(2)
C14	1.039(1)	0.9364(4)	0.9251(8)	0.031(2)	C15	0.647(2)	0.6902(4)	0.7717(9)	0.046(3)
C16	0.995(2)	0.6666(4)	1.017(1)	0.047(3)					

[a] The anisotropic thermal parameters are defined by the equation: $T = \exp(-2\pi^2[U_{11}h^2a^{*2} + U_{22}k^2b^{*2} + U_{33}l^2c^{*2} + 2\,U_{12}hka^*b^* + 2\,U_{13}hla^*c^* + 2\,U_{23}klb^*c^*])$.

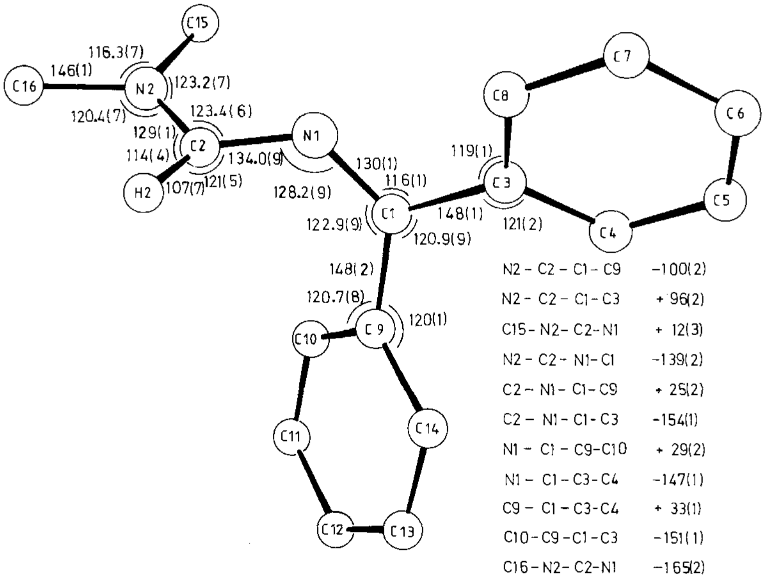
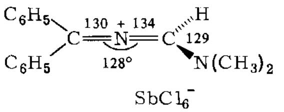
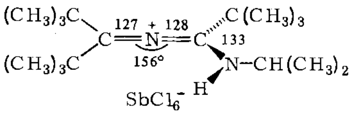


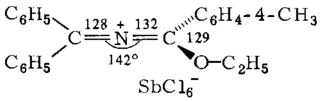
Fig. 1. Molecular Plot with Selected Bond Lengths [pm], Bond Angles, and Torsional Angles [°] for the Cation of **5a**



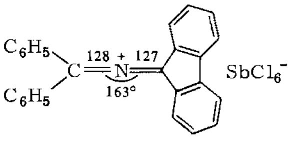
5a, $\beta \approx 84°$, IR(CH_2Cl_2): 1650 cm^{-1}



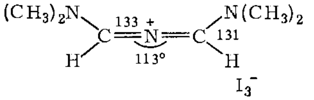
11, $\beta \approx 87°$, IR(CH_2Cl_2): 1825 cm^{-1}



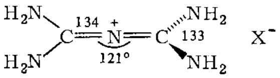
12, $\beta \approx 85°$, IR(nujol): 1695 cm^{-1}



13, $\beta \approx 85°$, IR(CH_2Cl_2): 1880 cm^{-1}



14, $\beta = 0°$, IR: 1600 cm^{-1}



15, β: 40-50°

Bond lengths in pm

There seems to exist a rough linear proportionality between the C = N = C bond angle (but not between the torsional angle β) and the wave number (1600 – 1910 cm^{-1}) of the antisymmetric stretching vibration of this unit (Fig. 2).

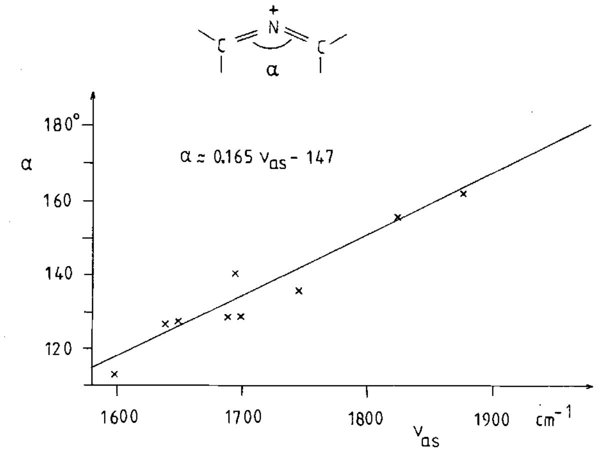


Fig. 2. Linear Relationship Between the C = N = C Bond Angle α and the Antisymmetric Stretching Frequency ν_{as} for 2-Azaallenium Salts

With increasing C = N = C bond angle the C = N distances decrease enhancing the force constant for the stretching vibration. Contrary to other cumulenes, 2-azaallenium salts do not show a positional constant C$\overset{+}{N}$C group frequency in the IR. Substituted

2-azaallenium cations are flexible assuming conformations which are mainly determined by steric and electronic demands of the substituents.

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Experimental Part

IR spectra: Perkin-Elmer IR 229, solutions in dichloromethane. – ^{1}H and ^{13}C NMR spectra: JEOL JNM-MH-100 and Bruker WM-250 spectrometer, δ-scale, internal reference tetramethylsilane. – The melting points are uncorrected.

3-(Dimethylamino)-1,1-diphenyl-2-azaallenium Hexachloroantimonate (**5a**): To **1a**[21] (1.22 g, 5.00 mmol) and **3a** (0.37 g, 5.06 mmol) in absol. acetonitrile (25 ml) antimony pentachloride (1.50 g, 5.00 mmol) was added at –40°C with stirring. The reaction mixture was kept for 20 min at –40°C, and was then stirred for 4 h at +22°C. After evaporation of the solvent under reduced pressure the residue was dissolved in dry dichloromethane (10 ml). Dropwise addition of dry pentane (30 ml) afforded a pale yellow powder (2.40 g, 84%) (including work-up of the mother liquids), which was recrystallized from dichloromethane/pentane; m.p. 171 – 175°C. – IR: 1650, 1590, 1530 cm^{-1}. – ^{1}H NMR (CD_3CN): CH_3 δ = 3.51, 3.52, NCH 8.31. – ^{13}C NMR (CD_3CN): CH_3 δ = 39.7, 45.4, C=N 185.1, 164.3, *ipso,p*-C 136.2, 135.2, *o,m*-C 132.4, 129.9.

$[C_{16}H_{17}N_2]SbCl_6$ (571.8) Calcd. C 33.61 H 3.00 N 4.90 Found C 33.70 H 3.16 N 4.85

With **7**[20] instead of **1a** an impure product with an IR spectrum similar to that of **5a** was obtained.

1-(Dimethylamino)-1-methyl-3,3-diphenyl-2-azaallenium Hexachloroantimonate (**5b**): From **3b** (0.44 g, 5.05 mmol) as described for **5a**. Stirring the oily product in methanol(20 ml)/pentane (20 ml) afforded a yellow powder, which was recrystallized from hot acetonitrile (10 ml) giving pale yellow cubes (2.31 g, 79%); m.p. 188 – 192°C (dec.). – IR: 1630, 1590 cm^{-1}. – ^{1}H NMR (CD_3CN): CH_3 δ = 2.18, 3.36, 3.40. – ^{13}C NMR (CD_3CN): CH_3 δ = 21.3, 42.5, 42.7, *ipso,p*-C 135.8, 134.2, *o,m*-C 130.7, 130.0, C=N 175.4, 173.5.

$[C_{17}H_{19}N_2]SbCl_6$ (585.8) Calcd. C 34.85 H 3.27 N 4.78 Found C 34.92 H 3.33 N 4.73

1-(Dimethylamino)-1-methyl-3,3-diphenyl-2-azaallenium Tetrachloroferrate (**5c**): From **3b** (0.44 g, 5.05 mmol), **1a** (1.22 g, 5.00 mmol) and anhydrous iron trichloride (0.81 g, 5.00 mmol) as described for **5a**. The oily product crystallized from dichloromethane (10 ml)/methanol (10 ml) at –25°C after addition of pentane (20 ml) affording light green needles (1.79 g, 80%); m.p. 94 – 95°C. – IR: 1630, 1590 cm^{-1}.

$[C_{17}H_{19}N_2]FeCl_4$ (449.0) Calcd. C 45.47 H 4.26 N 6.24 Found C 45.51 H 4.28 N 6.21

1-(Dimethylamino)-1-isopropyl-3,3-diphenyl-2-azaallenium Hexachloroantimonate (**5d**): From **3d** (0.59 g, 5.08 mmol) as described for **5a**. The oily product crystallized from dichloromethane (10 ml)/methanol (10 ml) on slow addition of pentane (20 ml). Recrystallization from dichloromethane/pentane afforded pale yellow needles (1.81 g, 59%); m.p. 175 – 179°C. – IR: 1660, 1590 cm^{-1}. – ^{1}H NMR (CD_2Cl_2): CH_3 δ = 0.97 (d, J = 7 Hz), 3.44, 3.63, CH 3.30 (sept, J = 7 Hz). – ^{13}C NMR (CD_2Cl_2): CH_3 δ = 19.5, 41.7, 42.9, CH 33.8, *ipso,p*-C 134.7, 134.2, *o,m*-C 130.3, 129.8, C=N 177.8, 170.4.

$[C_{19}H_{23}N_2]SbCl_6$ (613.9) Calcd. C 37.17 H 3.78 N 4.56 Found C 36.96 H 3.75 N 4.64

1-tert-Butyl-1-(dimethylamino)-3,3-diphenyl-2-azaallenium Hexachloroantimonate (**5e**): From **3e** (0.65 g, 5.03 mmol) as described for **5c**. Yield 1.66 g (53%) of pale yellow needles; m.p. 177 – 179 °C (dec.). – IR: 1630, 1590 cm^{-1} (shoulder 1580). – ^1H NMR (CD_2Cl_2): CH_3 δ = 1.29, 3.40, 3.73. – ^{13}C NMR (CD_2Cl_2): CH_3 δ = 28.7, 43.6, 45.1, C 40.6, *ipso,p*-C 134.6, 134.2, *o,m*-C 130.1, 129.9, C=N 179.7, 167.6.

$[C_{20}H_{25}N_2]SbCl_6$ (627.9) Calcd. C 38.26 H 4.01 N 4.46 Found C 38.37 H 4.12 N 4.35

1-(Dimethylamino)-1,3,3-triphenyl-2-azaallenium Hexachloroantimonate (**5f**): From **3f** (0.75 g, 5.03 mmol) as described for **5a**. The oily product crystallized from dichloromethane (10 ml)/methanol (10 ml) at –25 °C after addition of pentane (20 ml) affording a nearly colourless powder (1.36 g, 42%); m.p. 144 – 145 °C (dec.). – IR: 1610, 1590, 1570 cm^{-1}. – ^1H NMR (CD_3CN): CH_3 δ = 3.44, 3.57. – ^{13}C NMR (CD_3CN): CH_3 δ = 42.6, 44.7, C=N 177.7, 173.6.

$[C_{22}H_{21}N_2]SbCl_6$ (647.9) Calcd. C 40.78 H 3.27 N 4.32 Found C 40.62 H 3.40 N 4.29

(Diphenylmethylene)(1-methyl-2-pyrrolidinylidene)ammonium Hexachloroantimonate (**5g**): From **3g** (0.50 g, 5.04 mmol) as described for **5a**. The oily product crystallized when rubbed under methanol (20 ml). Recrystallization from hot acetonitrile (5 ml) afforded nearly colourless crystals (2.12 g, 71%); m.p. 182 – 185 °C (dec.). – IR: 1650, 1590 (shoulder 1580) cm^{-1}. – ^1H NMR (CD_3CN): CH_3 δ = 3.24, CH_2 2.12 (m), 2.69 (m), 3.96 (m). – ^{13}C NMR (CD_3CN): CH_3, CH_2 δ = 19.9, 34.6, 35.6, 58.4, C=N 178.3, 177.2, *ipso,p*-C 135.9, 134.4, *o,m*-C 130.9, 130.0.

$[C_{18}H_{19}N_2]SbCl_6$ (597.8) Calcd. C 36.16 H 3.20 N 4.69 Found C 36.49 H 3.02 N 4.72

3-(Dimethylamino)-1-(1-naphthyl)-1-phenyl-2-azaallenium Hexachloroantimonate (**5h**): From **1h**[21] (1.47 g, 5.00 mmol) and **3a** (0.37 g, 5.06 mmol) as described for **5a**. The oily product crystallized from methanol (10 ml) on addition of pentane (20 ml). Recrystallization from dichloromethane/pentane afforded orange prisms (2.02 g, 65%); m.p. 155 – 156 °C. – IR: 1650, 1530 (shoulder 1580) cm^{-1}. – ^1H NMR (CD_3CN): CH_3 δ = 3.37, 3.61 (d, J = 1.2 Hz), NCH 8.28 (m). – ^{13}C NMR (CD_3CN): CH_3 δ = 40.1, 45.6, C=N 186.6, 165.0, 14 aromatic C.

$[C_{20}H_{19}N_2]SbCl_6$ (621.8) Calcd. C 38.63 H 3.08 N 4.51 Found C 38.72 H 3.06 N 4.40

1-(Dimethylamino)-1-methyl-3-(1-naphthyl)-3-phenyl-2-azaallenium Hexachloroantimonate (**5i**): From **3b** (0.44 g, 5.05 mmol) as described for **5h**. The oily product crystallized at –25 °C from dichloromethane (10 ml)/methanol (10 ml) after addition of pentane (20 ml). Recrystallization from dichloromethane/pentane gave yellow needles (1.65 g, 52%); m.p. 129 – 130 °C. – IR: 1640, 1590, 1575 cm^{-1}. – ^1H NMR (CD_3CN): CH_3 δ = 2.12, 3.27, 3.40. – ^{13}C NMR (CD_3CN): CH_3 δ = 21.4, 42.9, 43.0, C=N 176.0, 173.7, 14 aromatic C.

$[C_{21}H_{21}N_2]SbCl_6$ (635.9) Calcd. C 39.66 H 3.33 N 4.41 Found C 39.39 H 3.10 N 4.14

(1-Methyl-2-pyrrolidinylidene)(1-naphthylphenylmethylene)ammonium Hexachloroantimonate (**5j**): From **3g** (0.50 g, 5.04 mmol) as described for **5h**. The oily product crystallized from methanol (10 ml)/pentane (20 ml) affording dark yellow prisms (1.81 g, 56%); m.p. 174 – 176 °C (dec.). – IR: 1650, 1590, 1575 cm^{-1}. – ^1H NMR (CD_2Cl_2): CH_3 δ = 3.43, CH_2 2.06 (m), 2.60 (m), 3.97 (m). – ^{13}C NMR (CD_2Cl_2): CH_3, CH_2 δ = 19.4, 34.5, 36.0, 58.1, C=N 178.22, 178.19, 14 aromatic C.

$[C_{22}H_{21}N_2]SbCl_6$ (647.9) Calcd. C 40.78 H 3.27 N 4.32 Found C 40.58 H 3.29 N 4.44

[(Dimethylamino)methylene]-9-fluorenylideneammonium Hexachloroantimonate (**5k**): From **1k**[25] (1.21 g, 5.00 mmol) and **3a** (0.37 g, 5.06 mmol) as described for **5a**. The product was stirred for 10 min in methanol (10 ml)/dichloromethane (10 ml). Addition of pentane (30 ml) afforded an orange powder, which was recrystallized from acetonitrile (5 ml)/pentane giving orange-red prisms (1.91 g, 67%); m.p. 169 – 172 °C. – IR: 1650 (shoulder 1670, 1700), 1590 (shoulder 1610)

cm^{-1}. – ^{1}H NMR (CD_3CN, 273 K): CH_3 δ = 3.36, 3.67, CH 9.18. – ^{13}C NMR (CD_3CN, 273 K): CH_3 δ = 45.7, 39.6, C=N 172.6, 163.5, aromatic C 144.7, 137.3, 133.9, 130.3, 127.7, 122.7.

$[C_{16}H_{15}N_2]SbCl_6$ (569.8) Calcd. C 33.73 H 2.65 N 4.92 Found C 33.54 H 2.51 N 4.73

[(Dimethylamino)methylene]-9-fluorenylideneammonium Tetrachloroferrate (**5l**): To iron trichloride (0.81 g, 5.00 mmol) a solution of **1k** (1.21 g, 5.00 mmol) and **3a** (0.37 g, 5.60 mmol) in absol. acetonitrile (25 ml) was given at –10°C. After stirring for 2 h at +22°C the solvent was evaporated under reduced pressure. The oily residue was dissolved in absol. dichloromethane (10ml). Slow addition of absol. ether (20 ml) afforded a precipitate, which was recrystallized from dry dichloromethane/ether giving orange needles (1.58 g, 73%); m.p. 126–127°C. – IR: 1640 (shoulder 1680), 1590 cm^{-1}.

$[C_{16}H_{15}N_2]FeCl_4$ (433.0) Calcd. C 44.38 H 3.49 N 6.47 Found C 44.40 H 3.32 N 6.51

[1-(Dimethylamino)ethylidene]-9-fluorenylideneammonium Hexachloroantimonate (**5m**): From **3b** (0.44 g, 5.05 mmol) as described for **5k**. The oily product was crystallized from methanol (10 ml)/dichloromethane (5 ml)/pentane (20 ml) at –25°C. Recrystallization from dichloromethane/pentane gave orange crystals (2.13 g, 73%, after work-up of the mother liquids); m.p. 194–195°C. – IR: 1690, 1625, 1600 cm^{-1}. – ^{1}H NMR (CD_3CN): CH_3 δ = 2.71, 3.26 (d, J = 0.6 Hz), 3.58. – ^{13}C NMR (CD_3CN): CH_3 δ = 20.9, 42.5, 43.2, C=N 175.7, 165.2, aromatic C 144.5, 136.6, 133.4, 130.3, 126.7, 122.5.

$[C_{17}H_{17}N_2]SbCl_6$ (583.8) Calcd. C 34.97 H 2.94 N 4.80 Found C 35.08 H 2.78 N 4.66

[1-(Dimethylamino)ethylidene]-9-fluorenylideneammonium Tetrachloroferrate (**5n**): From **3b** (0.44 g, 5.05 mmol) as described for **5l**. The crude oily product was dissolved in absol. dichloromethane (10 ml). Slow addition of absol. ether afforded an oily precipitate, which after drying gave a foam (1.97 g, 88%). From a solution of this foam in dry dichloromethane (5 ml) a small amount of an orange powder crystallized at –20°C; m.p. 99–101°C. – IR: 1690, 1630, 1610, 1600 cm^{-1}.

$[C_{17}H_{17}N_2]FeCl_4$ (447.0) Calcd. C 45.68 H 3.83 N 6.27 Found C 45.42 H 4.00 N 6.10

[1-(Dimethylamino)-2-methylpropylidene]-9-fluorenylideneammonium Hexachloroantimonate (**5o**): From **3d** (0.59 g, 5.08 mmol) as described for **5k**. The product was crystallized from dichloromethane (10 ml)/methanol (20 ml)/pentane (20 ml). Recrystallization from dichloromethane/pentane afforded orange-yellow leaflets (2.35 g, 77%); m.p. 187–189°C (dec.). – IR: 1700, 1600 cm^{-1}. – ^{1}H NMR (CD_2Cl_2, 263 K): CH_3 δ = 1.35 (d, J = 6.7 Hz), 3.28, 3.79, CH 3.59 (sept, J = 6.7 Hz). – ^{13}C NMR (CD_2Cl_2, 263 K): CH_3 δ = 19.8, 42.2, 42.4, CH 33.8, C=N 180.7, 164.4, aromatic C 144.0, 136.5, 132.4, 130.1, 125.6, 122.0.

$[C_{19}H_{21}N_2]SbCl_6$ (611.9) Calcd. C 37.30 H 3.46 N 4.58 Found C 37.23 H 3.43 N 4.58

[1-(Dimethylamino)-2,2-dimethylpropylidene]-9-fluorenylideneammonium Hexachloroantimonate (**5p**): From **3e** (0.65 g, 5.03 mmol) as described for **5k**. The product was crystallized from dichloromethane (10 ml)/methanol (10 ml)/pentane (20 ml) at –25°C giving a yellow powder (1.97 g, 63%); m.p. 220–225°C (dec.). – IR: 1715, 1690 cm^{-1}. – ^{1}H NMR (CD_2Cl_2): CH_3 δ = 1.58, 3.28, 3.85. – ^{13}C NMR (CD_2Cl_2): CH_3 δ = 29.3, 44.0, 44.5, C 41.1, C=N 181.1, 162.3, aromatic C 144.1, 136.8, 132.6, 130.3, 125.4, 122.2.

$[C_{20}H_{23}N_2]SbCl_6$ (625.9) Calcd. C 38.38 H 3.70 N 4.48 Found C 38.18 H 3.83 N 4.39

[(Dimethylamino)phenylmethylene]-9-fluorenylideneammonium Hexachloroantimonate (**5q**): From **3f** (0.75 g, 5.03 mmol) as described for **5k**. Crystallization at –25°C from dichloromethane (10 ml)/methanol (10 ml)/pentane (20 ml) and recrystallization from dichloromethane/pentane afforded dark yellow crystals (2.26 g, 70%); m.p. 215–218°C (dec.). – IR:

1690, 1600 cm^{-1}. – ^{1}H NMR (CD_3CN): CH_3 δ = 3.39, 3.64. – ^{13}C NMR (CD_3CN): CH_3 δ = 42.4, 45.1, C=N 172.9, 166.0.

$[C_{22}H_{19}N_2]SbCl_6$ (645.9) Calcd. C 40.91 H 2.97 N 4.34 Found C 40.97 H 3.03 N 4.31

9-Fluorenylidene(1-methyl-2-pyrrolidinylidene)ammonium Hexachloroantimonate (**5r**): From **3g** (0.50 g, 5.04 mmol) as described for **5k**. Crystallization from methanol (10 ml)/pentane (20 ml) and recrystallization from hot acetonitrile afforded orange prisms (2.38 g, 80%); m.p. 191 – 195 °C (dec.). – IR: 1695, 1675, 1600 cm^{-1}. – ^{1}H NMR (CD_3CN): CH_3 δ = 3.16, CH_2 2.48 (m), 3.36 (m), 4.18 (m). – ^{13}C NMR (CD_3CN): CH_3, CH_2 δ = 19.9, 35.0, 35.5, 59.1, C=N 178.3, 167.7, aromatic C 144.6, 136.8, 133.5, 130.4, 126.7, 122.6.

$[C_{18}H_{17}N_2]SbCl_6$ (595.8) Calcd. C 36.28 H 2.88 N 4.70 Found C 36.46 H 2.88 N 4.74

1-[(Diphenylmethylen)amino]-1,3,3-triphenyl-2-azaallenium Hexachloroantimonate (**5s**): To **1a** (1.22 g, 5.00 mmol) and **3s**[34] (1.44 g, 5.05 mmol) in absol. dichloromethane (20 ml) a solution of antimony pentachloride (1.50 g, 5.00 mmol) in absol. dichloromethane (10 ml) was added dropwise with stirring at –78 °C. The reaction mixture was warmed to +22 °C within 1 h and stirred for additional 3 h at this temperature. Dropwise addition of absol. ether (60 ml) and pentane (20 ml) afforded a yellow precipitate, which was recrystallized from dichloromethane/ether giving yellow prisms (2.74 g, 70%); m.p. 172 – 175 °C. – IR: 1780 (broad), 1630, 1590 cm^{-1}. – ^{13}C NMR (CD_3CN): C=N δ = 173.3 (2 C), 166.7 (1 C), gem. phenyl: *ipso,p*-C 135.4, 133.2, *o,m*-C 131.8, 130.3, 1-phenyl: *ipso,p*-C 138.1, 128.9, *o,m*-C 131.9, 131.2.

$[C_{33}H_{25}N_2]SbCl_6$ (784.0) Calcd. C 50.55 H 3.21 N 3.57 Found C 50.53 H 3.11 N 3.60

(Methoxydiphenylmethyl)(1-methyl-2-pyrrolidinylidene)ammonium Hexachloroantimonate (**8g**): A solution of **5g** (2.99 g, 5.00 mmol) in dry methanol (20 ml) plus acetonitrile (5 ml) was boiled under reflux for 8 h. Evaporation of the solvent under reduced pressure and crystallization of the residue from dichloromethane (5 ml)/ether (30 ml) afforded a pale yellow powder (2.46 g, 78%); m.p. 124 – 129 °C. – IR: 1650 cm^{-1}. – ^{1}H NMR (CD_3CN, 263 K): NCH_3 δ = 3.25, 3.29 (ca 1:5), OCH_3 3.36, NH 7.87. – ^{13}C NMR (CD_3CN, 263 K): CH_3, CH_2 δ = 19.7 (large), 19.9 (small), 33.0, 34.3, 34.8, 35.6, 51.8 (OCH_3), 56.9, 58.2, OCN 93.2, C=N 170.3, 8 aromatic C.

$[C_{19}H_{23}N_2O]SbCl_6$ (629.9) Calcd. C 36.23 H 3.68 N 4.45 Found C 36.51 H 3.77 N 4.45

(9-Methoxy-9-fluorenyl)(1-methyl-2-pyrrolidinylidene)ammonium Hexachloroantimonate (**8r**): A solution of **5r** (2.98 g, 5.00 mmol) in absol. methanol (20 ml) was boiled under reflux for 4 h. Evaporation of the solvent under reduced pressure gave an orange powder (2.48 g, 79%), which was washed with pentane (60 ml); m.p. 144 – 145 °C. – IR: 1660 cm^{-1}. – ^{1}H NMR (CD_2Cl_2, 250 K): NCH_3 δ = 2.90, OCH_3 3.29, CH_2 1.88 (m), 1.96 (m), 3.76 (m), NH 6.94. – ^{13}C NMR (CD_2Cl_2, 250 K): CH_3, CH_2 δ = 18.6, 29.6, 33.8, 51.7 (OCH_3), 56.0, OCN 96.1, C=N 165.2, aromatic C 140.7, 138.9, 132.3, 129.9, 125.1, 121.3.

$[C_{19}H_{21}N_2O]SbCl_6$ (627.9) Calcd. C 36.35 H 3.37 N 4.46 Found C 36.57 H 3.47 N 4.50

1-(1-Methoxy-2,2-dimethyl-1-phenylpropyl)-3,3-dimethylformamidinium Hexachloroantimonate (**8t**): To **1t**[21] (1.12 g, 5.00 mmol) and **3a** (0.37 g, 5.06 mmol) in absol. acetonitrile (25 ml) was added at –40 °C antimony pentachloride (1.50 g, 5.00 mmol). The reaction mixture was stirred for 5 h at +22 °C and refluxed for additional 2 h. Evaporation of the solvent provided a dark orange oil, which could not be crystallized. The oil was dissolved in absol. methanol (10 ml)/dichloromethane (10 ml). After addition of pentane (30 ml) red-brown crystals were formed at –25 °C (1.34 g, 46%); m.p. 137 – 139 °C. – IR: 1690 cm^{-1}. – ^{1}H NMR (CD_3CN, 263 K): CH_3 δ = 0.94, 3.15, 3.23, 3.30, CH 7.83 (d, J = 13.4 Hz), NH 6.69 (d, J = 13.4 Hz). – ^{13}C NMR (CD_3CN, 263 K): CH_3 δ = 25.3, C 38.1, NCH_3 41.5, 45.2, OCH_3 52.5, OCN 98.6, C=N 155.5, *ipso,p*-C 133.9, 129.8, *o,m*-C 129.8, 128.3.

$[C_{15}H_{25}N_2O]SbCl_6$ (583.8) Calcd. C 30.86 H 4.32 N 4.80 Found C 31.15 H 4.04 N 4.81

N-(Diphenylmethylene)benzamidinium Hexachloroantimonate (**9**): A solution of **5s** (2.35 g, 3.00 mmol) and absol. methanol (0.91 g, 6.00 mmol) in absol. dichloromethane (20 ml) was boiled under reflux for 4 h. Slow addition of pentane (30 ml) and ether (50 ml) yielded an oil, which soon solidified. Recrystallization from dichloromethane/ether afforded colourless needles (1.51 g, 81%); m.p. 153 – 156°C (dec.). – IR: 1640 cm^{-1}. – ^{13}C NMR (CD_2Cl_2): C = N δ = 175.2, 173.6, gem. phenyl 134.4, 131.0, 129.6, 129.3, phenyl 137.2, 133.8, 130.7, 127.0.

$[C_{20}H_{17}N_2]SbCl_6$ (619.8) Calcd. C 38.75 H 2.77 N 4.52 Found C 39.04 H 3.05 N 4.54

N-(Methoxydiphenylmethyl)benzamidinium Hexachloroantimonate (**10**): A solution of **5s** (2.35 g, 3.00 mmol) in a mixture of dry methanol (15 ml) and dichloromethane (5 ml) was stirred at 22°C for 4 h. After evaporation of the solvent the residue was dissolved in dichloromethane (10 ml)/acetonitrile (1 ml). Dropwise addition of ether (30 ml) afforded yellow needles (1.84 g, 94%); m.p. 136 – 138°C (dec.). – IR: C = N 1650, NH 3400, 3320, 3270 cm^{-1}. – ^1H NMR (CD_3CN): OCH_3 δ = 3.40. – ^{13}C NMR (CD_3CN): C = N δ = 166.8, NCO 94.0, OCH_3 52.3, 8 phenyl C.

$[C_{21}H_{21}N_2O]SbCl_6$ (651.9) Calcd. C 38.69 H 3.25 N 4.30 Found C 38.99 H 3.10 N 4.27

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