Free and Coordinated Iminosilanes: Synthesis and Structures

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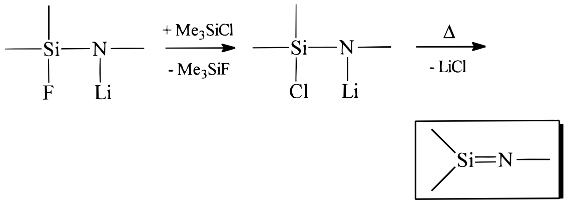
The uncoordinated and thermally stable iminosilane $(Me_3C)_2Si{=}NSi(CMe_3)_2Ph$ (**1**) is formed quantitatively in the reaction of the lithiated aminofluorosilane and Me_3SiCl with subsequent elimination of Me_3SiF and LiCl. Di-*tert*-butyldifluorosilane and diisopropyldifluorosilane react with lithiated amino-*tert*-butyldiisopropylsilane and amino-di-*tert*-butylphenylsilane to give the aminofluorosilanes $(Me_3C)_2FSiNHSi(CHMe_2)_2CMe_3$ (**2**) and $(Me_2HC)_2FSiNHSi(CMe_3)_2Ph$ (**3**). **2** and **3** are converted into their lithium derivatives $(Me_3C)_2FSiNLi(THF)_2Si(CHMe_2)_2CMe_3$ (**4**) and $(Me_2HC)_2FSiNLi(THF)_2Si(CMe_3)_2Ph$ (**5**) with ^{n}BuLi. **4** reacts with Me_3SiCl to give Me_3SiF and the lithiated aminochlorosilane. Thermal LiCl elimination leads to the formation of the iminosilane $(Me_3C)_2Si{=}NSi(CHMe_2)_2CMe_3$ (**6**). A fluorine−bromine exchange occurs when **5** is treated with Me_3SiBr in THF/toluene. In a subsequent LiBr elimination $(Me_2HC)_2(C_4H_7O)SiNHSi(CMe_3)_2Ph$ (**7**) is obtained. **1** and **6** form the THF adducts $(Me_3C)_2(THF)Si{=}NR$ (**8**, R = $Si(CMe_3)_2Ph$; and **9**, R = $Si(CHMe_2)_2CMe_3$). The adducts $(Me_3C)_2(py)Si{=}NR$ (**10**, R = $Si(CMe_3)_2Ph$; and **11**, R = $Si(CHMe_2)_2CMe_3$) are formed when pyridine is used as a donor. At 50 °C in toluene **10** and **11** decompose under Stevens migration into $(Me_3C)_2(2\text{-py})SiNHR$ (**12**, R = $Si(CMe_3)_2Ph$; **13**, R = $Si(CHMe_2)_2CMe_3$). In the reaction of **1** with 2-cyclohexen-1-one the ene reaction product $(Me_3C)_2(C_6H_7O)SiNHSi(CMe_3)_2Ph$ (**14**) is formed. X-ray crystal structure determinations are presented for **1**, **9**, **10**, and **11**.

Introduction

Compounds with double bonds to second-row elements, once thought to be nonexistent, are now being synthesized. Stability can often be achieved by the use of bulky substituents, which prevent further reactions. This experimental success is one of the most fruitful recent innovations of organometallic chemistry. Silenes and disilenes have been isolated as stable compounds at the beginning of the 1980s, and the first iminosilanes were reported in the mid-1980s.[1−3]

We found that lithium derivatives of bulky aminofluorosilanes crystallize as LiF adducts of iminosilanes and react with $ClSiMe_3$ with formation of lithiated aminochlorosilanes.[3,4] LiCl elimination gives iminosilanes, which can be distilled in vacuo without decomposition (eq 1).

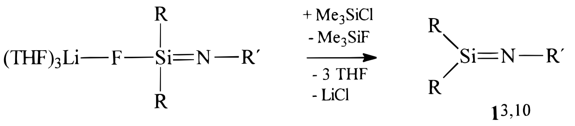


(1)

Although the Si═N double bond is sterically shielded, the high reactivity of iminosilanes allows the synthesis of a number of interesting compounds.[3,5−9] In this paper the crystal structure of the uncoordinated iminosilane $(Me_3C)_2Si{=}N{-}Si(CMe_3)_2Ph$ (**1**)[3] is reported and the synthesis of a new stable iminosilane and the formation of THF and pyridine adducts are described.

Results and Discussion

The uncoordinated and thermally stable iminosilane $(Me_3C)_2Si{=}NSi(CMe_3)_2Ph$ (**1**) is formed in quantitative yield in the reaction between the lithiated fluorosilylamine compound and Me_3SiCl (eq 2).[3,10]



(2)

1[3,10]

R = CMe_3
R′ = $Si(CMe_3)_2Ph$

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(1) Brook, A. G.; Brook, M. A. *Adv. Organomet. Chem.* **1996**, *39*, 71.

(2) Okazaki, R.; West, R. *Adv. Organomet Chem.*. **1996**, *39*, 232.

(3) Hemme, I.; Klingebiel, U. *Adv. Organomet. Chem.* **1996**, *39*, 159.

(4) Walter, S.; Klingebiel, U. *Coord. Chem. Rev.* **1994**, *130*, 481.

(5) Wiberg, N.; Schurz, K. *Chem. Ber.* **1988**, *121*, 581.

(6) Grosskopf, D.; Klingebiel, U.; Belgardt, T.; Noltemeyer, M. *Phosphorus, Sulfur Silicon Relat. Elem.* **1994**, *91*, 241.

(7) Niesmann, J.; Klingebiel, U.; Rudolph, S.; Herbst-Irmer, R.; Noltemeyer, M. *J. Organomet. Chem.* **1996**, *515*, 43.

(8) Niesmann, J.; Klingebiel, U.; Noltemeyer, M. *J. Organomet. Chem.* **1996**, *521*, 191.

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Table 1. Selected Bond Lengths (Å) and Angles (deg) for 1

molecule 1		molecule 2	
Si(1)−N(1)	1.695(2)	Si(1A)−N(1A)	1.690(3)
Si(2)−N(1)	1.573(2)	Si(2A)−N(1A)	1.573(3)
N(1)−Si(2)−C(19)	119.47(13)	N(1A)−Si(2A)−C(19A)	119.60(15)
N(1)−Si(2)−C(15)	123.10(13)	N(1A)−Si(2A)−C(15A)	122.19(14)
C(19)−Si(2)−C(15)	117.26(13)	C(19A)−Si(2A)−C(15A)	118.16(15)
Si(2)−N(1)−Si(1)	168.34(16)	Si(2A)−N(1A)−Si(1A)	172.95(18)

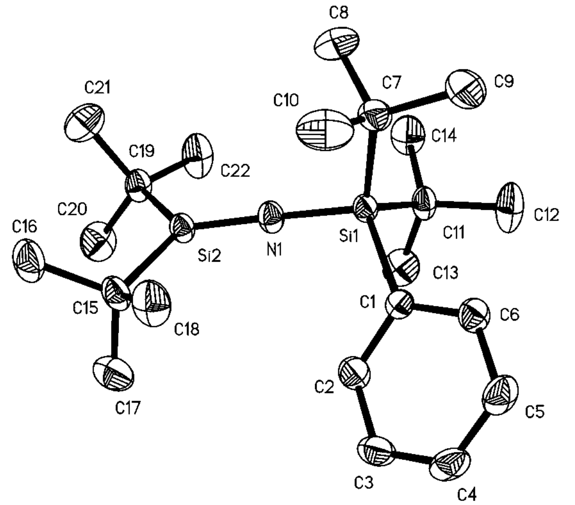
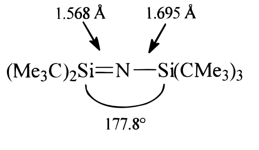
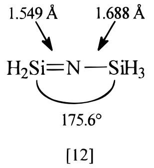


Figure 1. Crystal structure of **1**, with anisotropic displacement parameters depicting 50% probability. The hydrogen atoms have been omitted for clarity.

Crystals of **1** were obtained by zone refining with two independent molecules in the asymmetric unit. Both molecules have fairly similar conformations. The rms deviation of the fitted atoms (all non-hydrogen atoms except the disordered carbon atoms) was 0.147 Å. The crystal structure of one molecule of **1** (Figure 1, Tables 1 and 5) shows a trigonal arrangement of the $C_2Si{=}N$ unit around the silicon atom. In the solid state the iminosilane is monomeric and the Si=N−Si skeleton is nearly linear (168.3°/173.0°). The Si−N bond lengths are drastically different. The Si−N single bond is 1.695/1.690 Å, and the Si=N double bond is 1.573/1.573 Å. The results are consistent with Wiberg's experimental[11] and von Ragué-Schleyer's calculated[12] results.



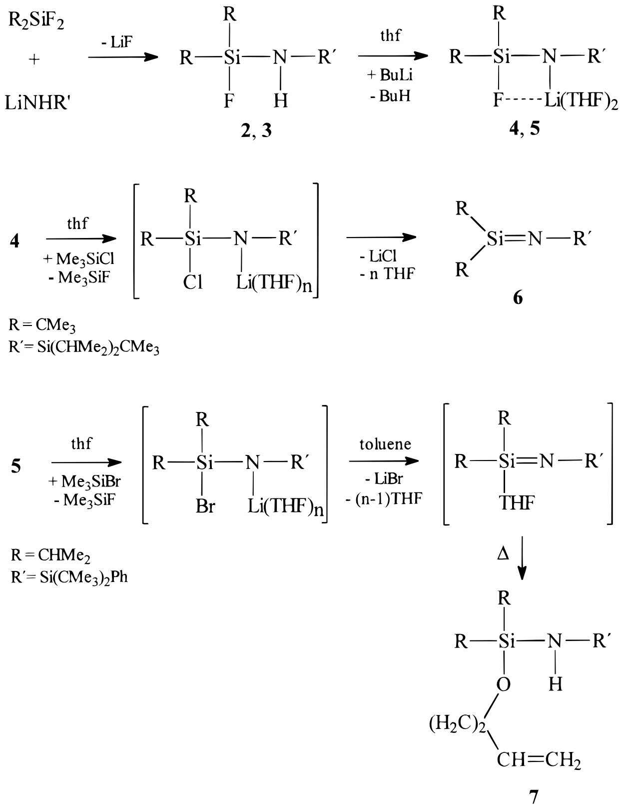
[11]



[12]

Scheme 1 shows the stepwise synthesis of the iminosilane **6** and the silyl ether **7**. Di-*tert*-butyldifluorosilane and diisopropyldifluorosilane react with lithiated amino-*tert*-butyldiisopropylsilane and lithiated amino-di-*tert*-butyl-phenylsilane to give the aminofluorosilanes **2** and **3**.

Scheme 1. Synthesis of 6 and 7



In the reaction with ^{n}BuLi the lithium derivatives **4** and **5** are formed, which yielded colorless crystals from hexane/THF. The crystals were taken up in C_6D_6, and their NMR spectra revealed the presence of two molecules of THF coordinated to the lithium atoms. The ^{19}F NMR spectrum of **4** shows a significant downfield shift of the fluorine signal compared with that of **2** (difference nearly 13 ppm) and an upfield shift of the signals in the ^{29}Si NMR spectrum (difference nearly 7 ppm). The covalent part of the LiF contact is shown by the presence of a LiF coupling constant of $^{1}J_{\mathrm{LiF}} = 5.9$ Hz.

As mentioned above, addition of Me_3SiCl to **4** immediately gives Me_3SiF, as revealed by ^{19}F NMR spectroscopic monitoring. The characterization of the lithium-aminochlorosilane in solution shows an upfield shift of the signals in the ^{29}Si NMR spectrum (δ (^{29}Si) −14.1/−19.7 ppm) compared with that of **4**. Thermal LiCl elimination leads to the formation of the iminosilane **6**, which can be distilled in vacuo without decomposition (bp 91 °C/0.01 mbar). The ^{29}Si NMR spectrum of **6** shows the typical downfield shift (δ (^{29}Si) 76.6 ppm) of the unsaturated silicon atom.

As shown in Scheme 1, the treatment of **5** with Me_3SiBr in THF leads to elimination of Me_3SiF, shown by ^{19}F NMR, under formation of a lithium aminobromosi-

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(10) Grosskopf, D.; Marcus, L.; Klingebiel, U.; Noltemeyer, M., *Phosphorus, Sulfur Silicon Relat. Elem.* **1994**, *97*, 113.

(11) Wiberg, N.; Schurz, K.; Reber, G.; Müller, G. *J. Chem. Soc., Chem. Commun.* **1986**, 591.

(12) Schleyer, P. v. R.; Stout, P. O. *J. Chem. Soc., Chem. Commun.* **1986**, 1373.

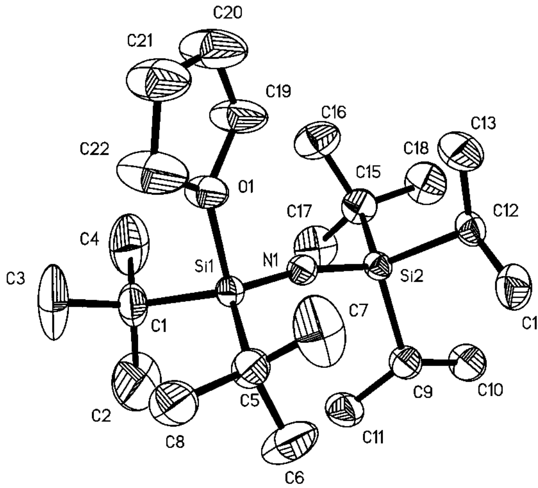


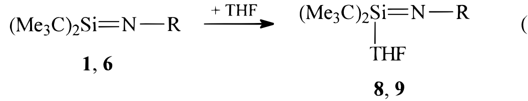
Figure 2. Crystal structure of **9**, with anisotropic displacement parameters depicting 50% probability. The hydrogen atoms have been omitted for clarity.

Table 2. Selected Bond Lengths (Å) and Angles (deg) for 9

Si(1)−N(1)	1.599(2)	Si(2)−N(1)	1.665(2)
Si(1)−O(1)	1.902(2)		
N(1)−Si(1)−C(5)	116.07(10)	C(5)−Si(1)−C(1)	115.09(12)
N(1)−Si(1)−C(1)	116.61(11)	Si(1)−N(1)−Si(2)	168.88(13)

lane. This fluorine−bromine exchange leads to a significant upfield shift of the signals (δ (^{29}Si) −11.5/−22.4 ppm) in the ^{29}Si NMR spectrum of the solution. After partial removal of the solvent in vacuo, heating of the residue in toluene leads to the formation of **7** by LiBr elimination and decomposition of the metastable adduct $(Me_2HC)_2(THF)Si{=}N{-}Si(CMe_3)_2Ph$.

In comparison to **7**, stable THF adducts of iminosilanes are formed in the reactions of **1**[3] and **6** with THF by complexation of the unsaturated silicon atoms (eq 3).



$$(Me_3C)_2Si{=}N{-}R \xrightarrow{+\,THF} (Me_3C)_2\underset{\substack{|\\ THF}}{Si}{=}N{-}R \qquad (3)$$

1, **6** **8**, **9**

1, **8**: R = $Si(CMe_3)_2Ph$
6, **9**: R = $Si(CHMe_2)_2CMe_3$

The adduct formation shows a remarkable upfield shift of the unsaturated silicon atoms in **8** (δ (^{29}Si) 2.68 ppm) and **9** (δ (^{29}Si) 3.13 ppm) compared to the free iminosilanes **1** (δ (^{29}Si) 80.4 ppm) and **6** (δ (^{29}Si) 76.6 ppm). In the ^{13}C NMR spectrum of **8** and **9**, the C_2O signals of the THF molecules appear at 73.3 ppm (**8**) and 72.9 ppm (**9**)—a downfield shift of nearly 6 ppm compared with that of free THF. This reflects the strong Lewis acid character of three-coordinated silicon in iminosilanes.

An X-ray structural analysis was carried out on single crystals of **9** (Figure 2, Tables 2 and 5), recrystallized from toluene.

This THF adduct (Si(1)−O(1) = 1.902 Å) of an iminosilane has a nearly linear geometry on the nitrogen atom (168.9°), with two significantly different Si−N bond lengths. These bond lengths (Si(1)−N(1) = 1.599 Å, Si(2)−N(1) = 1.665 Å) reflect the difference between a double and a single Si−N bond. The unsaturated Si(1) atom has a distorted-tetrahedral geometry; the two *tert*-butyl groups and the nitrogen atom tend to form a trigonal-planar environment around Si(1) (bond angle sum 347.8°).

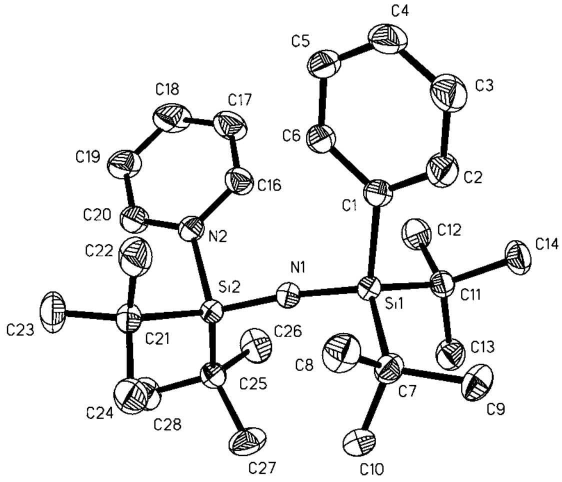


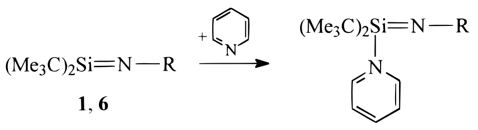
Figure 3. Crystal structure of **10**, with anisotropic displacement parameters depicting 50% probability. The hydrogen atoms have been omitted for clarity.

Table 3. Selected Bond Lengths (Å) and Angles (deg) for 10

Si(1)−N(1)	1.654(2)	Si(2)−N(2)	1.949(2)
Si(2)−N(1)	1.611(2)		
N(1)−Si(2)−C(21)	115.46(10)	C(21)−Si(2)−C(25)	115.48(11)
N(1)−Si(2)−C(25)	116.03(10)	Si(2)−N(1)−Si(1)	172.47(13)

Several other THF adducts have already been prepared by Wiberg[5,11,13] and our group.[3,4]

For the first time, pyridine adducts of iminosilanes which are stable under normal conditions are formed in toluene at room temperature in the reaction of the iminosilanes **1**[3] and **6** with pyridine to give **10** and **11** (eq 4).



(4)

1, **6** **10**, **11**

1, **10**: R = $Si(CMe_3)_2Ph$
6, **11**: R = $Si(CHMe_2)_2CMe_3$

The unsaturated silicon atoms form five bonds because of the donor; both are four-coordinated and have therefore a tetrahedral environment. The ^{29}Si NMR spectrum of the pyridine compounds **10** (δ (^{29}Si) −12.6 ppm) and **11** (δ (^{29}Si) −12.2 ppm) show a strong upfield shift of the signals of the unsaturated silicon atoms compared to the free iminosilanes.

Pale yellow crystals of **10** and **11** are obtained by crystallization from toluene. The crystal structures of **10** (Figure 3, Tables 3 and 5) and **11** (Figure 4, Tables 4 and 5) show a nearly trigonal arrangement of the C_2-Si=N units around the silicon atoms without the pyridine bond (**10**, 347.0°; **11**, 346.6°). The Si=N−Si skeletons are linear (**10**, 172.5°; **11**, 176.0°), and the Si−N bond lengths are different (**10**, 1.611, 1.654 Å; **11**, 1.606, 1.661 Å). This reflects the difference between double and single Si−N bonds.

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[13] Wiberg, N.; Schurz, K. *J. Organomet. Chem.* **1988**, *341*, 145.

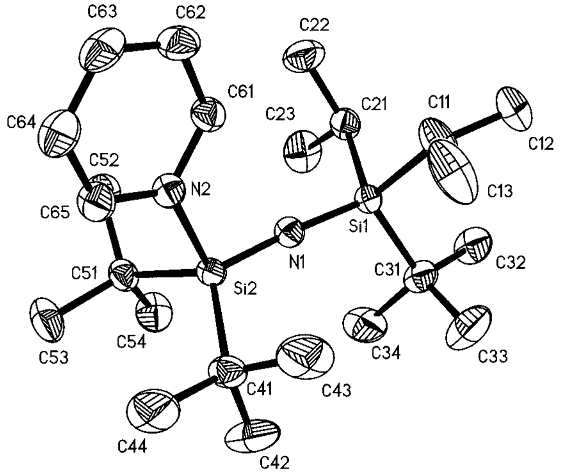
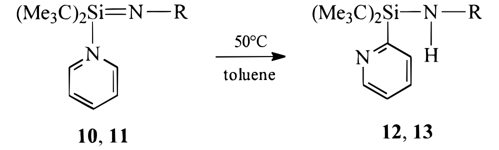


Figure 4. Crystal structure of **11**, with anisotropic displacement parameters depicting 50% probability. The hydrogen atoms have been omitted for clarity.

Table 4. Selected Bond Lengths (Å) and Angles (deg) for 11

Si(1)–N(1)	1.661(2)	Si(2)–N(2)	1.955(2)
Si(2)–N(1)	1.606(2)		
N(1)–Si(2)–C(41)	116.05(9)	C(41)–Si(2)–C(51)	115.53(10)
N(1)–Si(2)–C(51)	115.00(9)	Si(2)–N(1)–Si(1)	176.01(11)

To prove the thermal stability of these pyridine adducts, the compounds **10** and **11** were slowly heated in toluene. At 50 °C the adducts decompose under Stevens migration into **12** and **13** (eq 5), as revealed by ^{29}Si NMR spectroscopic monitoring. Similar stabilization reactions are known from metastable stanna- and germaimines using pyridine as trapping agent.[14]

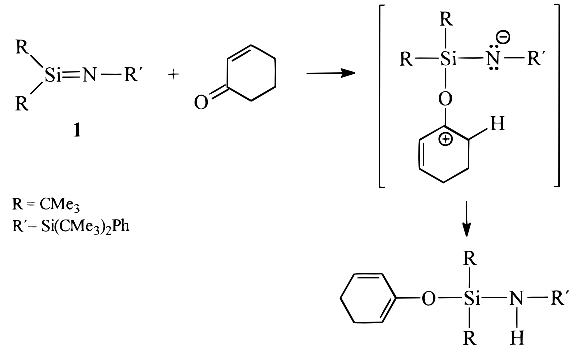


(5)

10, 11 **12, 13**

10, **12**: R = Si(CMe$_3$)$_2$Ph
11, **13**: R = Si(CHMe$_2$)$_2$CMe$_3$

Using cyclohexenone as the donor the Lewis base adduct is much less stable than the pyridine adducts. In an intramolecular ene reaction the (cyclohexa-1,5-dien-1-yloxy)silane **14** is formed (eq 6). Other compounds, like acetone or dimethylbutadiene react in a comparable way.[9]



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(6)

14

Experimental Section

All experiments were performed in oven-dried glassware under purified nitrogen or argon using standard inert atmosphere and vacuum-line techniques. The solvents were dried over Na and distilled under nitrogen prior to use. All NMR spectra were obtained on either a Bruker AM-250 or MSL-400 spectrometer and were recorded in C_6D_6 or $CDCl_3$ (**2**, **3**, **14**) with $SiMe_4$ as internal and C_6F_6 or LiCl as external references. Mass spectra were obtained on a Varian CH-5- and Finnigan MAT 8200- or 9500-spectrometer. The data are reported in mass to charge units (*m*/*z*) with their relative intensities in parentheses. The progress of the reactions was monitored by ^{1}H-, ^{19}F– or ^{29}Si NMR spectroscopy. The purity of the compounds was measured by NMR spectroscopy, C, H analyses (**2**–**8**, **12**–**14**) and if possible analyzed by gas chromatography.

Di-*tert*-butyl[(di-*tert*-butylphenylsilyl)imino]silane (1). The compound was prepared according to the literature.[10] Single crystals of **1** were obtained by zone refining.

(Di-*tert*-butylfluorosilyl)(*tert*-butyldiisopropylsilyl)amine (2). To a solution of 18.7 g of amino-*tert*-butyldiisopropylsilane (0.1 mol) in 100 mL of n-hexane and 50 mL of THF was slowly added at ambient temperature 62.5 mL of ^{n}BuLi (1.6 M solution in *n*-hexane, 0.1 mol). The reaction mixture was refluxed for 1 h and cooled to room temperature. After 18.0 g of di-*tert*-butyldifluorosilane (0.1 mol) had been added, the mixture was heated under reflux for 24 h and for a further 48 h at 90 °C, after distilling of excess solvent. **2** was separated from LiF by condensing the product and solvents into a cooled trap in vacuo. **2** (21.6 g, 62 mmol, 62% yield) was distilled (bp 97 °C/0.01 mbar) in vacuo to give a pure sample. ^{1}H NMR: δ 0.02 (br, s, 1H, NH), 1.03 (s, 9H, CMe$_3$), 1.04 (d, 18H, $^4J_{SiF}$ = 1.0 Hz, CMe$_3$), 1.15 (br, s, 14H, CHMe$_2$). ^{13}C NMR: δ 14.26 (SiCC$_2$), 19.07 (d, $^5J_{CF}$ = 1.8 Hz, SiC*C*$_2$), 19.20 (d, $^5J_{CF}$ = 1.7 Hz, SiC*C*$_2$), 20.94 (d, $^2J_{CF}$ = 15.4 Hz, FSiCC$_3$), 21.17 (SiCC$_3$), 27.78 (d, $^3J_{CF}$ = 0.6 Hz, FSiC*C*$_3$), 28.88 (d, $^5J_{CF}$ = 0.7 Hz, SiC*C*$_3$). ^{19}F NMR: δ −1.51. ^{29}Si NMR: δ 3.05 (d, $^1J_{SiF}$ = 300.1 Hz, SiF), 6.85 (d, $^3J_{SiF}$ = 1.2 Hz, SiC$_3$). MS (EI; 70 eV) for $C_{18}H_{42}FNSi_2$ (347.71): *m*/*z* (%) 338 (5) [M − CH$_3$]$^+$, 304 (20) [M − C$_3$H$_7$]$^+$, 290 (50) [M − C$_4$H$_9$]$^+$. Anal. Calcd: C, 62.22; H, 12.17. Found: C, 62.98; H, 12.87.

(Fluorodiisopropylsilyl)(di-*tert*-butylphenylsilyl)amine (3). A 23.5 g amount of amino-di-*tert*-butylphenylsilane (0.1 mol) was reacted in a mixture 100 mL of *n*-hexane and 50 mL of THF with 62.5 mL of ^{n}BuLi (1.6 M solution in *n*-hexane, 0.1 mol) and heated under reflux for 1 h to complete the lithiation. A 15.2 g amount of difluorodiisopropylsilane (0.1 mol) was added at −10 °C, and the reaction mixture was heated under reflux for 3 h. The product (25.4 g, 69 mmol, 69% yield) was separated from LiF in vacuo and purified by distillation (bp 121 °C/0.01 mbar). ^{1}H NMR: δ 0.78 (br, s, 1H, NH), 1.16 (s, 18H, CMe$_3$), 1.13–1.19 (m, 14H, CHMe$_2$), 7.38–7.42 (m, 3H, 3-H/4-H/5-H (Ph)), 7.81–7.88 (m, 2H, 2-H/6-H (Ph)). ^{13}C NMR: δ 13.44 (d, $^2J_{CF}$ = 17.1 Hz, FSiCC$_2$), 17.47 (d, $^3J_{CF}$ = 2.0 Hz, FSiC*C*$_2$), 17.72 (d, $^3J_{CF}$ = 1.0 Hz, FSiC*C*$_2$), 20.55 (SiCC$_3$), 28.80 (d, $^5J_{CF}$ = 0.9, SiC*C*$_3$), 127.11 (C-3/C-5 (Ph)), 128.90 (C-4 (Ph)), 135.47 (d, $^5J_{CF}$ = 1.6 Hz, C-2/C-6 (Ph)), 136.08 (d, $^4J_{CF}$ = 0.7 Hz, C-1 (Ph)). ^{19}F NMR: δ 13.22 (d, $^3J_{HF}$ = 2.2 Hz). ^{29}Si NMR: δ 1.09 (d, $^3J_{SiF}$ = 1.2 Hz, SiC$_3$), 3.32 (d, $^1J_{SiF}$ = 297.4 Hz, SiF). MS (EI, 70 eV) for $C_{20}H_{38}FNSi_2$ (367.70): *m*/*z* (%) 310 (100) [M − C$_4$H$_9$]$^+$. Anal. Calcd: C, 65.33; H, 10.42. Found: C, 65.74; H, 10.83.

Preparation of Compounds 4 and 5. A 3.5 g amount of **2** (0.01 mol) or 3.7 g of **3** (0.01 mol) was lithiated in 30 mL of *n*-hexane/THF with 6.3 mL of ^{n}BuLi (1.6 M solution in *n*-hexane, 0.01 mol) and stirred for 5 h at ambient temperature. **4** (4.7 g, 9.5 mmol, 95% yield) and **5** (4.8 g, 9.2 mmol, 92% yield) crystallized at 0 °C from the reaction mixtures and were purified by recrystallization from *n*-hexane.

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(14) Ossig, G.; Meller, A.; Freitag, S.; Herbst-Irmer, R.; Sheldrick, G. M. *Chem. Ber.* **1993**, *126*, 2247.

Table 5. Crystal Data and Structure Refinement Details for 1, 9, 10, and 11

	1	**9**	**10**	**11**
formula	$C_{22}H_{41}NSi_2$	$C_{22}H_{49}NOSi_2$	$C_{27}H_{46}N_2Si_2$	$C_{23}H_{46}N_2Si_2$
M_r	375.74	399.80	454.84	406.80
cryst syst	triclinic	monoclinic	monoclinic	monoclinic
space group	$P\bar{1}$	$P2_1/n$	$P2_1/c$	$P2_1/c$
a (Å)	11.265(2)	11.331(1)	17.398(4)	8.577(1)
b (Å)	14.213(2)	16.804(2)	11.461(3)	17.736(2)
c (Å)	16.975(3)	13.408(2)	15.577(4)	16.618(2)
α (deg)	111.26(1)	90	90	90
β (deg)	97.31(1)	95.81(1)	114.20(1)	91.66(1)
γ (deg)	102.13(1)	90	90	90
V (Å^3)	2413.6(7)	2539.9(5)	2832.9(12)	2526.9(5)
Z	4	4	4	4
D_{exptl} (Mg/m^3)	1.034	1.046	1.066	1.069
μ (mm^{-1})	0.152	0.151	0.141	0.151
F(000)	832	896	1000	904
cryst size (mm)	0.24 × 0.21 × 0.13	0.80 × 0.80 × 0.60	0.60 × 0.40 × 0.10	0.80 × 0.60 × 0.50
2θ range (deg)	3.2−48	7−45	4.4−52.8	7−45
range of hkl	$-12 \leq h \leq 10$	$-12 \leq h \leq 12$	$-21 \leq h \leq 19$	$-9 \leq h \leq 9$
	$-16 \leq k \leq 15$	$-12 \leq k \leq 18$	$0 \leq k \leq 14$	$0 \leq k \leq 19$
	$0 \leq l \leq 19$	$-14 \leq l \leq 14$	$0 \leq l \leq 19$	$-16 \leq l \leq 17$
no. of rflns collected	7320	6178	34961	3341
no. of indep rflns	6824	3323	5749	3313
R(int)	0.0348	0.0346	0.0657	0.0332
$T_{\max}$	0.9805	0.9151	0.9888	0.9284
$T_{\min}$	0.9644	0.8891	0.9586	0.8889
no. of data	6824	3323	5749	3313
no. of params	537	248	293	257
no. of restraints	458	0	0	0
S	1.013	1.028	1.151	1.058
R1 ($I > 2\sigma(I)$)	0.0447	0.0457	0.0574	0.040
wR2 (all data)	0.1154	0.1231	0.1222	0.1045
extinction coeff			0.0050(5)	
largest diff peak (e/Å^3)	0.314	0.348	0.379	0.363
largest diff hole (e/Å^{-3})	−0.335	−0.298	−0.332	−0.273

Lithium−(Di-*tert*-butylfluorosilyl)(*tert*-butyldiisopropylsilyl)amide−THF Adduct (4): $C_{26}H_{57}FLiNO_2Si_2$ (497.86). ^{1}H NMR: δ 1.24 (s, 9H, CMe_3), 1.26−1.41 (m, 14H, $CHMe_2$),1.30 (s, 8H, $O(CH_2CH_2)_2$), 1.34 (d, 18H, $^4J_{\mathrm{SiF}} = 1.0$ Hz, $FSiCMe_3$), 3.42 (s, 8H, $O(CH_2CH_2)_2$). ^{7}Li NMR: δ −0.01 (d, $^1J_{\mathrm{LiF}} = 5.9$ Hz). ^{13}C NMR: δ 16.64 (d, $^4J_{\mathrm{CF}} = 1.5$ Hz, $SiCC_2$), 20.72 ($SiCC_2$), 20.91 ($SiCC_2$), 22.34 ($SiCC_3$), 22.86 (d, $^2J_{\mathrm{CF}} = 20.7$ Hz, $FSiCC_3$), 25.33 (OCC), 29.73 ($FSiCC_3$), 30.71 ($SiCC_3$) 68.30 (OCC). ^{19}F NMR: δ 11.78 (q, $^1J_{\mathrm{LiF}} = 5.9$ Hz). ^{29}Si NMR: δ −12.30 (d, $^3J_{\mathrm{SiF}} = 9.1$ Hz, SiC_3), −3.95 (d, $^1J_{\mathrm{SiF}} = 254.6$ Hz, SiF). Anal. Calcd: C, 62.73; H, 11.54. Found: C, 63.11; H, 11.97.

Lithium−(Diisopropylfluorosilyl)(di-*tert*-butylphenylsilyl)amide−THF Adduct (5): $C_{28}H_{53}FLiNSi_2$ (517.85). ^{1}H NMR: δ 1.27 (s, 8H, $O(CH_2CH_2)_2$), 1.34 (s, 18H, CMe_3), 1.36−1.41 (m, 14H, $CHMe_2$), 3.37 (s, 8H, $O(CH_2CH_2)_2$), 7.24−7.35 (m, 3H, 3-H/4-H/5-H (Ph)), 8.12−8.19 (m, 2H, 2-H/6-H (Ph)). ^{13}C NMR: δ 16.85 (d, $^2J_{\mathrm{CF}} = 21.4$ Hz, $FSiCC_2$), 18.99 ($FSiCC_2$), 19.01 (d, $^3J_{\mathrm{CF}} = 1.5$ Hz, $FSiCC_2$), 22.05 (d, $^4J_{\mathrm{CF}} = 1.3$ Hz, $SiCC_3$), 25.34 (OCC), 30.54 ($SiCC_3$), 68.09 (OCC), 126.83 (C-3/C-5 (Ph)), 127.32 (C-4 (Ph)), 136.18 (C-2/C-6 (Ph)), 145.29 (C-1 (Ph)). ^{19}F NMR: δ 10.82. ^{29}Si NMR: δ −16.20 (d, $^3J_{\mathrm{SiF}} = 10.9$ Hz, SiC_3), −4.94 (d, $^1J_{\mathrm{SiF}} = 248.9$ Hz, SiF). Anal. Calcd: C, 64.49; H, 10.32. Found: C, 65.01; H, 10.81.

Di-*tert*-butyl[(*tert*-butyldiisopropylsilyl)imino]silane (6). A solution of 5.0 g of **4** (0.01 mol) in 50 mL of *n*-hexane/THF was treated with 1.3 mL of Me_3SiCl (0.01 mol) and the mixture stirred for 16 h. The formation of Me_3SiF was monitored by ^{19}F NMR spectroscopy. **6** (2.9 g, 8.7 mmol, 87% yield) was separated from LiCl and purified by distillation (bp 91 °C/0.01 mbar) in vacuo. ^{1}H NMR: δ 1.10 (s, 18H, $SiCMe_3$), 1.24 (s, 9H, CMe_3), 1.28−1.36 (m, 14H, $CHMe_2$). ^{13}C NMR: δ 14.95 ($SiCC_2$), 20.02 ($SiCC_2$), 20.31 ($N{=}SiCC_3$), 20.88 ($SiCC_3$), 29.07 ($N{=}SiCC_3$), 29.67 ($SiCC_3$). ^{29}Si NMR: δ −9.57 (SiC_3), 76.63 ($C_2Si{=}N$). MS (EI, 70 eV) for $C_{18}H_{41}NSi_2$ (327.71): *m/z* (%) 284 (40) $[M - C_3H_7]^+$, 270 (100) $[M - C_4H_9]^+$. Anal. Calcd: C, 65.97; H, 12.60; N, 4.27. Found: C, 66.14; H, 12.80; N, 4.05.

[Diisopropyl(but-3-en-1-yloxy)silyl](di-*tert*-butylphenylsilyl)amine (7). A solution of 5.2 g of **5** (0.01 mol) in 50 mL of *n*-hexane/THF was treated with 1.3 mL of Me_3SiBr (0.01 mol) and the mixture stirred for 16 h. The formation of Me_3SiF was monitored by ^{19}F NMR spectroscopy.

After 50 mL of toluene had been added, the mixture was heated under reflux for 5 h, after distilling of excess *n*-hexane/THF. **7** was separated from LiBr by condensing the product and solvents into a cooled trap in vacuo. **7** (2.6 g, 6.3 mmol, 63% yield) was distilled (bp 154 °C/0.01 mbar, mp 13 °C) in vacuo to give a pure sample. ^{1}H NMR: δ 0.38 (s, 1H, NH), 1.12 (s, 18H, CMe_3), 1.01−1.14 (m, 14H, $SiCHMe_2$), 2.15 (dddt, 2H, $^3J_{\mathrm{HH}} = 6.8$ Hz, $^3J_{\mathrm{HH}} = 6.8$ Hz, $^4J_{\mathrm{cis}} = 1.5$ Hz, $^4J_{\mathrm{trans}} = 1.1$ Hz, C_2CH_2), 3.63 (t, 2H, $^3J_{\mathrm{HH}} = 6.8$ Hz, OCH_2), 4.96 (ddt, 1H, $^3J_{\mathrm{cis}} = 10.3$ Hz, $^2J_{\mathrm{gem}} = 2.2$ Hz, $^4J_{\mathrm{trans}} = 1.1$ Hz, $C{=}CH_{\mathrm{trans}}$), 4.97 (ddt, 1H, $^3J_{\mathrm{trans}} = 17.1$ Hz, $^2J_{\mathrm{gem}} = 2.2$ Hz, $^4J_{\mathrm{cis}} = 1.5$ Hz, $C{=}CH_{\mathrm{cis}}$), 5.74 (ddt, 1H, $^3J_{\mathrm{trans}} = 17.1$ Hz, $^3J_{\mathrm{cis}} = 10.3$ Hz, $^3J_{\mathrm{HH}} = 6.8$ Hz, $C{=}CHC$), 7.18−7.26 (m, 3H, 3-H/4-H/5-H (Ph)), 7.82−7.88 (m, 2H, 2-H/6-H (Ph)). ^{13}C NMR: δ 14.62 ($SiCC_2$), 18.62 ($SiCC_2$), 18.71 ($SiCC_2$), 21.10 ($SiCC_3$), 29.40 ($SiCC_3$), 37.57 (C_2CH_2), 62.90 (OCH_2), 116.39 ($C{=}CH_2$), 127.31 (C-3/C-5 (Ph)), 129.08 (C-4 (Ph)), 135.75 ($C{=}CHC$), 136.02 (C-2/C-6 (Ph)), 136.75 (C-1 (Ph)). ^{29}Si NMR: δ −5.12 (C_2SiO), 0.54 (SiC_3). MS (EI, 70 eV) for $C_{24}H_{45}NOSi_2$ (419.80): *m/z* (%) 362 (100) $[M - C_4H_9]^+$. Anal. Calcd: C, 68.67; H, 10.08. Found: C, 68.43; H, 9.92.

Preparation of Compounds 8 and 9. To a 3.8 g amount of **1** (0.01 mol) or 3.3 g of **6** (0.01 mol) in 30 mL of toluene at ambient temperature was added an equimolar amount of THF (0.8 mL) with stirring. **8** (4.1 g, 9.1 mmol, 91% yield) and **9** (3.8 g, 9.4 mmol, 94% yield) crystallized at 0 °C from the reaction mixtures and were purified by recrystallization from toluene (mp 142 °C (**8**), 121 °C (**9**)).

Di-*tert*-butyl[(di-*tert*-butylphenylsilyl)imino]silane−THF Adduct (8). ^{1}H NMR: δ 1.09 (s, 4H, O(CH$_2$C*H*$_2$)$_2$), 1.21 (s, 18H, CMe$_3$), 1.42 (s, 18H, CMe$_3$), 3.86 (s, 4H, O(C*H*$_2$CH$_2$)$_2$), 7.28−7.37 (m, 3H, 3-H/4-H/5-H(Ph)), 8.17−8.20 (m, 2H, 2-H/6-H(Ph)). ^{13}C NMR: δ 22.86 (Si*C*C$_3$), 24.39 (OC*C*), 25.03 (Si*C*C$_3$), 30.23 (SiC*C*$_3$), 30.91 (SiC*C*$_3$), 73.32 (O*C*C), 126.57 (C-3/C-5 (Ph)), 127.19 (C-4 (Ph)), 136.87 (C-2/C-6 (Ph)), 144.71 (C-1 (Ph)). ^{29}Si NMR: δ −20.26 (SiC$_3$), 2.68 (C$_2$OSi=N). MS (EI, 70 eV) for $C_{26}H_{49}NOSi_2$ (447.86): *m/z* (%) 318 (100) [M − C$_4$H$_8$O − C$_4$H$_9$]$^+$. Anal. Calcd: C, 69.73; H, 11.03. Found: C, 70.05; H, 11.38.

Di-*tert*-butyl[(*tert*-butyldiisopropylsilyl)imino]silane−THF Adduct (9). ^{1}H NMR: δ 1.16 (s, 18H, SiCMe$_3$), 1.24 (s, 4H, O(CH$_2$C*H*$_2$)$_2$), 1.40 (s, 9H, CMe$_3$), 1.45 (d, 6H,$^3J_{HH}$ = 3.7 Hz, CHMe$_2$), 1.47 (d, 6H,$^3J_{HH}$ = 3.7 Hz, CHMe$_2$), 3.79 (s, 4H, O(C*H*$_2$CH$_2$)$_2$). ^{13}C NMR: δ 16.34 (Si*C*C$_2$), 20.68 (SiC*C*$_2$), 20.94 (SiC*C*$_2$), 22.70 (Si*C*C$_3$), 24.42 (N=Si*C*C$_3$), 24.66 (OC*C*), 29.97 (N=SiC*C*$_3$), 30.83 (SiC*C*$_3$) 72.86 (O*C*C). ^{29}Si NMR: δ −15.55 (SiC$_3$), 3.13 (C$_2$OSi=N). MS (EI, 70 eV) for $C_{22}H_{49}NOSi_2$ (399.81): *m/z* (%) 284 (50) [M − C$_4$H$_8$O − C$_3$H$_7$]$^+$, 270 (100) [M − C$_4$H$_8$O − C$_4$H$_9$]$^+$.

Preparation of Compounds 10 and 11. To a 3.8 g amount of **1** (0.01 mol) or 3.3 g of **6** (0.01 mol) in 30 mL of toluene at 0 °C was added an equimolar amount of pyridine (0.8 mL) =with stirring for 1 h. **10** (4.4 g, 9.6 mmol, 96% yield) and **11** (3.9 g, 9.5 mmol, 95% yield) crystallized at 0 °C from the reaction mixtures and were purified by recrystallization from toluene (mp: 122 °C (**10**), 96 °C (**11**)).

Di-*tert*-butyl[(di-*tert*-butylphenylsilyl)imino]silane−Pyridine Adduct (10). ^{1}H NMR: δ 1.22 (s, 18H, SiCMe$_3$), 1.49 (s, 18H, SiCMe$_3$), 6.29−6.38 (m, 2H, 3-H/5-H(Py)), 6.61−6.72 (m, 1H, 4-H (Py)), 7.18−7.40 (m, 3H, 3-H/4-H/5-H (Ph)), 8.21−8.28 (m, 2H, 2-H/6-H (Ph)), 9.06−9.14 (m, 2H, 2-H/6-H (Py)). ^{13}C NMR: δ 23.20 (Si*C*C$_3$), 23.77 (Si*C*C$_3$), 30.26 (SiC*C*$_3$), 31.10 (SiC*C*$_3$), 124.10 (C-3/C-5 (Py)), 126.65 (C-3/C-5 (Ph)), 127.13 (C-4 (Ph)), 136.85 (C-2/C-6 (Ph)), 141.78 (C-4(Py)), 145.28 (C-1 (Ph)), 147.53 (C-2/C-6 (Py)). ^{29}Si NMR: δ −20.20 (SiC$_3$), −12.59 (C$_2$NSi=N). MS (EI, 70 eV) for $C_{27}H_{46}N_2Si_2$ (454.85): (%) 397 (30) [M − C$_4$H$_9$]$^+$, 318 (100) [M − C$_5$H$_5$N − C$_4$H$_9$]$^+$.

Di-*tert*-butyl[(*tert*-butyldiisopropylsilyl)imino]silane−Pyridine Adduct (11). ^{1}H NMR: δ 1.17 (s, 18H, SiCMe$_3$), 1.43 (s, 9H, CMe$_3$), 1.48 (br, s, 14H, CHMe$_2$), 6.45 (dd, 2H, $^3J_{HH}$ = 7.6 Hz, $^3J_{HH}$ = 6.0 Hz, 3-H/5-H (Py)), 6.77 (tt, 1H, $^3J_{HH}$ = 7.6 Hz, $^4J_{HH}$ = 1.6 Hz, 4-H (Py)), 9.12 (d, 2H, $^3J_{HH}$ = 6.0 Hz, 2-H/6-H (Py)). ^{13}C NMR: δ 16.61 (Si*C*C$_2$), 20.80 (SiC*C*$_2$), 21.21 (SiC*C*$_2$), 22.94 (Si*C*C$_3$), 23.24 (N=Si*C*C$_3$), 30.03 (N=SiC*C*$_3$), 31.09 (SiC*C*$_3$), 123.93 (C-3/C-5(Py)), 141.58 (C-4 (Py)), 147.43 (C-2/C-6 (Py)). ^{29}Si NMR: δ −15.42 (SiC$_3$), −12.20 (C$_2$NSi=N). MS (EI, 70 eV) for $C_{23}H_{46}N_2Si_2$ (406.81): *m/z* (%) 391 (1) [M − CH$_3$]$^+$, 363 (40) [M − C$_3$H$_7$]$^+$, 349 (100) [M − C$_4$H$_9$]$^+$.

Preparation of Compounds 12 and 13. A 2.3 g amount of **10** (5.0 mmol) or 2.0 g of **11** (5.0 mmol) in 20 mL of toluene was slowly heated. Decomposition into **12** and **13** started at 50 °C. The progress of the reactions was monitored by ^{29}Si NMR spectroscopy. **10** (2.1 g, 4.6 mmol, 92% yield) and **11** (1.8 g, 4.5 mmol, 89% yield) crystallized at 0 °C from the reaction mixtures and were purified by recrystallization from toluene (mp 171 °C (**12**), 143 °C (**13**)).

[(Di-*tert*-butylphenylsilyl)amino]di-*tert*-butyl-2-pyridylsilane (12). ^{1}H NMR: δ 1.28 (s, 18H, SiCMe$_3$), 1.29 (s, 18H, SiCMe$_3$), 6.59 (ddd, 1H, $^3J_{HH}$ = 7.7 Hz, $^3J_{HH}$ = 4.8 Hz, $^4J_{HH}$ = 1.3 Hz, 5-H (Py)), 6.82 (dt, 1H,$^3J_{HH}$ = 7.7 Hz, $^4J_{HH}$ = 1.8 Hz, 4-H (Py)), 7.01−7.09 (m, 3H, 3-H/4-H/5-H (Ph)), 7.59 (ddd, 1H, $^3J_{HH}$ = 7.7 Hz, $^4J_{HH}$ = 1.3 Hz, $^5J_{HH}$ = 1.1 Hz, 3-H (Py)), 7.78−7.86 (m, 2H, 2-H/6-H (Ph)), 8.53 (ddd, 1H, $^3J_{HH}$ = 4.8 Hz, $^4J_{HH}$ = 1.8 Hz, $^5J_{HH}$ = 1.1 Hz, 6-H (Py)). ^{13}C NMR: δ 22.03 (Si*C*C$_3$), 22.08 (Si*C*C$_3$), 29.96 (SiC*C*$_3$), 30.01 (SiC*C*$_3$), 122.48 (C-5 (Py)), 126.92 (C-3/C-5 (Ph)), 128.92 (C-4 (Ph)), 131.62 (C-3 (Py)), 132.68 (C-4 (Py)), 136.35 (C-1 (Ph)), 136.49 (C-2/C-6 (Ph)), 148.98 (C-6 (Py)), 165.65 (C-2 (Py)). ^{29}Si NMR: δ −4.09 (C$_2$(Py)SiN), 0.69 (SiC$_3$). MS (EI, 70 eV) for $C_{27}H_{46}N_2Si_2$ (454.85): *m/z* (%) 454 (2) [M]$^+$, 439 (1) [M − CH$_3$]$^+$, 397 (100) [M − C$_4$H$_9$]$^+$. Anal. Calcd: C, 71.30; H, 10.19. Found: C, 71.62; H, 10.43.

[(*tert*-butyldiisopropylsilyl)amino]di-*tert*-butyl-2-pyridylsilane (13). ^{1}H NMR: δ 0.98 (s, 1H, NH), 1.15 (d, 6H, $^3J_{HH}$ = 7.4 Hz, CMe$_2$), 1.20 (d, 2H, $^3J_{HH}$ = 7.4 Hz, CMe$_2$), 1.21 (s, 9H, CMe$_3$), 1.24 (s, 18H, SiCMe$_3$), 1.36 (sept, 2H, $^3J_{HH}$ = 7.4 Hz, CH), 6.65 (ddd, 1H, $^3J_{HH}$ = 7.7 Hz, $^3J_{HH}$ = 4.8 Hz, $^4J_{HH}$ = 1.3 Hz, 5-H (Py)), 7.06 (dt, 1H, $^3J_{HH}$ = 7.7 Hz, $^4J_{HH}$ = 1.8 Hz, 4-H (Py)), 7.66 (ddd, 1H, $^3J_{HH}$ = 7.7 Hz, $^4J_{HH}$ = 1.3 Hz, $^5J_{HH}$ = 1.1 Hz, 3-H (Py)), 8.57 (ddd, 1H, $^3J_{HH}$ = 4.8 Hz, $^4J_{HH}$ = 1.8 Hz, $^5J_{HH}$ = 1.1 Hz, 6-H (Py)). ^{13}C NMR: δ 15.29 (Si*C*C$_2$), 19.81 (SiC*C*$_2$), 19.91 (SiC*C*$_2$), 21.70 (PySi*C*C$_3$), 22.25 (Si*C*C$_3$), 29.65 (PySiC*C*$_3$), 30.04 (SiC*C*$_3$), 122.60 (C-5 (Py)), 131.03 (C-3 (Py)), 132.83 (C-4 (Py)), 149.20 (C-6 (Py)), 166.07 (C-2 (Py)). ^{29}Si NMR: δ −4.09 (C$_2$PySiN), 6.03 (SiC$_3$). MS (EI, 70 eV) for $C_{23}H_{46}N_2Si_2$ (406.81): *m/z* (%) 406 (1) [M]$^+$, 349 (100) [M − C$_4$H$_9$]$^+$. Anal. Calcd: C, 67.91; H, 11.40; N, 3.44. Found: C, 68.14; H, 11.63; N, 3.51.

[(Di-*tert*-butylphenylsilyl)amino]di-*tert*-butyl(cyclohexa-1,5-dien-1-yloxy)silane (14). To a 3.8 g amount of **1** (0.01 mol) in 30 mL of toluene at ambient temperature was added an equimolar amount of 2-cyclohexen-1-one (1.0 mL), and the mixture was stirred for 1 h. **14** (3.4 g, 7.3 mmol, 73% yield) crystallized at 0 °C from the reaction mixture and was purified by recrystallization from toluene (mp 69 °C). ^{1}H NMR: δ 0.38 (br, s, 1H, NH), 1.12 (br, s, 36H, SiCMe$_3$), 2.00−2.24 (m, 4H, 4-H/5-H(Cy)), 5.09−5.17 (m, 1H, 3-H (Cy)), 5.88−5.95 (m, 2H, 2-H/6-H (Cy)), 7.23−7.38 (m, 3H, 3-H/4-H/5-H (Ph)), 7.87−7.94 (m, 2H, 2-H/6-H (Ph)). ^{13}C NMR: δ 21.36 (Si*C*C$_3$), 21.92 (C-4/C-5 (Cy)), 22.47 (Si*C*C$_3$), 22.55 (C-4/C-5 (Cy)), 29.05 (SiC*C*$_3$), 29.24 (SiC*C*$_3$), 103.01 (C-3 (Cy)), 126.67 (C-3/C-5 (Ph)), 126.77 (C-2 (Cy)), 128.53 (C-4 (Ph)), 128.62 (C-6 (Cy)), 135.72 (C-1 (Ph)), 136.23 (C-2/C-6 (Ph)), 148.31 (C-1 (Cy)). ^{29}Si NMR: δ −8.25 (C$_2$OSiN), 0.84 (SiC$_3$). MS (EI, 70 eV) for $C_{28}H_{49}NOSi_2$ (471.88): *m/z* (%) 414 (100) [M − C$_4$H$_9$]$^+$. Anal. Calcd: C, 71.27; H, 10.47. Found: C, 71.49; H, 10.73.

X-ray Structure Determinations for 1 and 9−11. Data for **1** were collected at −143 °C on a four-circle diffractometer, data for **9** and **11** were collected on a Stoe four-circle diffractometer at −60 °C and −73 °C, respectively, and data for **10** were collected at −140 °C on a Stoe-Siemens-Huber diffractometer using a SMART-CCD area detector. For all experiments monochromated Mo Kα radiation was used (λ = 0.710 73 Å). For the integration of intensities of the area detector data, the program SAINT[15] was used. For **10** a semiempirical absorption correction was employed. All structures were solved by direct methods using SHELXS-90.[16] All non-hydrogen atoms were refined anisotropically. For the hydrogen atoms the riding model was used. The structure was refined against F^2 with a weighting scheme of $w^{-1} = \sigma^2(F_o^2) + (g_1P)_2 + g_2P$ with $P = (F_o^2 + 2F_c^2)/3$, using SHELXL−97.[17] The R values are defined as $R1 = \sum||F_o| - |F_c||/\sum|F_o|$ and $wR2 = [\sum w(|F_o^2 - F_c^2)^2/\sum w(F_o^4)]^{0.5}$. Figures 1−4 (hydrogens are omitted) show 50% probability displacement ellipsoids. Crystal data and structure refinement details are listed in Table 5.

In one of the two independent molecules of structure **1** the three carbon atoms of the *tert*-butyl group (C(16A), C(17A) and C(18A)) and the *tert*-butyl group of C(20A), C(21A), and C(22A) are disordered over two positions (occupancies 0.6:0.4 and 0.7:0.3, respectively). These parts of the structure are refined using distance restraints and restraints for the anistropic

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[15] SAINT, Program for Reduction and Integration of Diffraction Data, version 4.050; AXS, 1996.

[16] Sheldrick, G. M. *Acta Crystallogr.* **1990**, *A46*, 467.

[17] Sheldrick, G. M. SHELXL-97, Program for Crystal Structure Refinement; University of Göttingen, Göttingen, Germany, 1997.

displacement parameters. The remaining structures were refined without the need of any restraints.

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Supporting Information Available: For **1** and **9**−**11** tables of data collection parameters, displacement parameters, non-hydrogen atomic coordinates, bond distances and angles, and hydrogen coordinates and *U* values (28 pages). Ordering information is given on any current masthead page. Further details of the crystal structure investigations are available on request from the Fachinformationszentrum Karlsruhe, Gesellschaft für Wissenschaftlich-Technische Information mbH, D-7514 Eggenstein-Leopoldshafen 2, on quoting the depository number CSD-408220 (**1**), -408221 (**9**), -408222 (**10**), and -408223 (**11**), the names of the authors, and the journal citation.

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