Normal Vibrations of Dimethyldimethoxysilane

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The dipole moment of dimethyldimethoxysilane was previously reported to be 1.33 D at 25°C and 1.35 D at 50°C[1]. If ∠Si–O–C in this molecule was about 130° and the rotation about two Si–O bonds was free, the observed moment agreed with that calculated from bond moments. However, the ∠Si–O–C assumed there seems to be somewhat too large compared with that in tetramethoxysilane[2]. It was suggested, therefore, that the Si–O bond would perhaps rotate under some hindrance[1]. Thereafter, the Raman and the infrared spectra of dimethyldimethoxysilane were measured, and the probable form of this molecule was discussed. As the result,

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[1] A. kotera, K. Matsumura, H. Nomura, Y. Nakano and U. Maeda, presented at the Symposium on Organosilicon Compounds, Osaka, October, 1956, and private communication.

[2] M. Yokoi, This Bulletin, 30, 106 (1957).

it was concluded that this molecule would be *GG* form in the solid state* and have no rotational isomers[3].

This paper reports the calculation of the fundamental frequencies of this molecule and their normal mode of skeletal vibrations, under the assumption that this molecule belongs to the point group C_{2v}. In the calculation, a methyl group was regarded as a point mass. The normal coordinate treatment was carried out by the Wilson *FG* matrix method[4] with a Urey-Bradley potential function. No calculation of the normal vibration for $Y_2M(XZ)_2$ type molecules has been reported as far as the author knows.

Normal Coordinate Treatment

The normal coordinate treatment has been carried out for parameters which are shown in Fig. 1. The symmetry of the molecule was assumed as C_{2v}. The interplanar angle for a Si–O–C plane and a C–Si–O plane including the same Si–O bond were also assumed to have an interplanar angle of +60° or −60°.

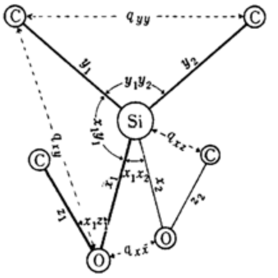

Fig. 1. Internal coordinates of the skeletal of dimethyldimethoxysilane.

According to the group theory, this molecule has fifteen skeletal vibrations, six of a_1, two of a_2, four of b_1 and three of b_2 species. Each one of the skeletal vibrations of a_2 and b_2 species is the torsional oscillation of a (Si–)OCH_3 group*. All vibrations of the a_1, b_1 and b_2 species are active both in infrared and Raman spectra, and those of the a_2 species are Raman active but infrared inactive.

From the internal coordinates indicated in Fig. 1, the following orthonormal symmetry coordinates have been found:

for the a_1 vibrations,

$$R_1=\Delta(z_1+z_2)/\sqrt{2}$$

$$R_2=\Delta(x_1+x_2)/\sqrt{2}$$

$$R_3=\Delta(y_1+y_2)/\sqrt{2}$$

$$R_4=\Delta(x_1z_1+x_2z_2)/\sqrt{2}$$

$$R_5=\Delta(x_1x_2-y_1y_2)/\sqrt{2}$$

$$R_6=\Delta(2x_1x_2+2y_1y_2-x_1y_1-x_1y_2-x_2y_1-x_2y_2)/\sqrt{12}$$

for the a_2 vibration,

$$R_7=\Delta(x_1y_1-x_1y_2-x_2y_1+x_2y_2)/2$$

for the b_1 vibrations,

$$R_9=\Delta(z_1-z_2)/\sqrt{2}$$

$$R_{10}=\Delta(x_1-x_2)/\sqrt{2}$$

$$R_{11}=\Delta(x_1z_1-x_2z_2)/\sqrt{2}$$

$$R_{12}=\Delta(x_1y_1+x_1y_2-x_2y_1-x_2y_2)/2$$

and for the b_2 vibrations,

$$R_{13}=\Delta(y_1-y_2)/\sqrt{2}$$

$$R_{14}=\Delta(x_1y_1-x_1y_2+x_2y_1-x_2y_2)/2$$

The numbering of these symmetry coordinates corresponds to the numbering of the fundamentals in Table I.

From the coefficients of the symmetry coordinates and the potential constants in a Urey-Bradley force field[5], the *F* matrix elements are obtained as follows:

for the a_1 vibrations,

$$F_{11}=K_z+s_{zx}{}^2F_{xz}+t_{zx}{}^2F_{xz}{}'$$

$$F_{12}=s_{xz}s_{zx}F_{xz}-t_{xz}t_{zx}F_{xz}{}'$$

$$F_{14}=(s_{zx}t_{xz}F_{xz}+s_{xz}t_{zx}F_{xz}{}')x$$

$$F_{22}=K_x+2s_{xx}{}^2F_{xx}+2s_{xy}{}^2F_{xy}+s_{xz}{}^2F_{xz}+2t_{xy}{}^2F_{xy}{}'+t_{xz}{}^2F_{xz}{}'$$

$$F_{23}=2(s_{xy}s_{yx}F_{xy}-t_{xy}t_{yx}F_{xy}{}')$$

$$F_{24}=(s_{xz}t_{zx}F_{xz}+s_{zx}t_{xz}F_{xz}{}')z$$

$$F_{25}=s_{xx}t_{xx}(F_{xx}+F_{xx}{}')x$$

$$F_{26}=\sqrt{2/3}\{s_{xx}t_{xx}(F_{xx}+F_{xx}{}')x-(s_{xy}t_{yx}F_{xy}+s_{yx}t_{xy}F_{xy}{}')y\}$$

$$F_{33}=K_y+2s_{yy}{}^2F_{yy}+2s_{yx}{}^2F_{xy}+2t_{yx}{}^2F_{xy}{}'$$

$$F_{35}=-s_{yy}t_{yy}(F_{yy}+F_{yy}{}')y$$

$$F_{36}=\sqrt{2/3}\{s_{yy}t_{yy}(F_{yy}+F_{yy}{}')y-(s_{yx}t_{xy}F_{xy}+s_{xy}t_{yx}F_{xy}{}')x\}$$

$$F_{44}=(H_{xz}+t_{xz}t_{zx}F_{xz}-s_{xz}s_{zx}F_{xz}{}')xz$$

$$F_{55}=\{(H_{xx}+t_{xx}{}^2F_{xx}-s_{xx}{}^2F_{xx}{}')x^2+(H_{yy}+t_{yy}{}^2F_{yy}-s_{yy}{}^2F_{yy}{}')y^2\}/2+3\kappa/\sqrt{8}$$

$$F_{56}=\{(H_{xx}+t_{xx}{}^2F_{xx}-s_{xx}{}^2F_{xx}{}')x^2-(H_{yy}+t_{yy}{}^2F_{yy}-s_{yy}{}^2F_{yy}{}')y^2\}/\sqrt{6}$$

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* In the liquid state, the two Si—O bonds would rotate slowly centering in the *GG* form[3].

[3] M. Hayashi, *J. Chem. Soc. Japan, Pure Chem. Sec.* (*Nippon Kagaku Zasshi*), **79**, 436 (1958).

[4] E. B. Wilson, Jr., *J. Chem. Phys.*, **7**, 1047 (1939).

* Represents the methoxyl group attached to silicon atom.

[5] T. Shimanouchi, *J. Chem. Phys.*, **17**, 245, 734, 848 (1949).

$$F_{66}=\{(H_{xx}+t_{xx}^2F_{xx}-s_{xx}^2F_{xx}')x^2$$
$$+(H_{xy}+t_{xy}t_{yx}F_{xy}-s_{xy}s_{yx}F_{xy}')xy$$
$$+(H_{yy}+t_{yy}^2F_{yy}-s_{yy}^2F_{yy}')y^2\}/3$$
$$+\kappa/\sqrt{8}$$
$$F_{13}=F_{15}=F_{16}=F_{34}=F_{45}=F_{46}=0$$

for the a$_2$ vibration,

$$F_{77}=(H_{xy}+t_{xy}t_{yx}F_{xy}-s_{xy}s_{yx}F_{ny}')xy$$
$$-\kappa/\sqrt{8}$$

for the b$_1$ vibrations,

$$F_{99}=K_z+s_{zx}^2F_{xz}+t_{zx}^2F_{xz}'$$
$$F_{910}=s_{xz}s_{zx}F_{xz}-t_{xz}t_{zx}F_{xz}'$$
$$F_{911}=(s_{zx}t_{xz}F_{xz}+s_{xz}t_{zx}F_{xz}')x$$
$$F_{1010}=K_x+2s_{xy}^2F_{xy}+s_{xz}^2F_{xz}+2t_{xx}^2F_{xx}'$$
$$+2t_{xy}^2F_{xy}'+t_{xz}^2F_{xz}'$$
$$F_{1011}=(s_{xz}t_{zx}F_{xz}+s_{zx}t_{xz}E_{xz}')z$$
$$F_{1012}=\sqrt{2}(s_{xy}t_{yx}F_{xy}+s_{yx}t_{xy}F_{xy}')y$$
$$F_{1111}=(H_{xz}+t_{xz}t_{zx}F_{xz}-s_{xz}s_{zx}F_{xz}')xz$$
$$F_{1212}=(H_{xy}+t_{xy}t_{yx}F_{xy}-s_{xy}s_{yx}F_{xy}')xy$$
$$+3\kappa/\sqrt{8}$$
$$F_{912}=F_{1112}=0$$

and for the b$_2$ vibrations,

$$F_{1313}=K_y+2s_{yx}^2F_{xy}+2t_{yx}^2F_{xy}'+2t_{yy}^2F_{yy}'$$
$$F_{1314}=\sqrt{2}(s_{yx}t_{xy}F_{xy}+s_{xy}t_{yx}F_{xy}')x$$
$$F_{1414}=(H_{xy}+t_{xy}t_{yx}F_{xy}-s_{xy}s_{yx}F_{xy}')xy$$
$$+3\kappa/\sqrt{8}$$

where the following abbreviations have been adopted:

$$s_{xx}=4x/3q_{xx} \qquad t_{xx}=\sqrt{8}x/3q_{xx}$$
$$s_{yy}=4y/3q_{yy} \qquad t_{yy}=\sqrt{8}y/3q_{yy}$$
$$s_{xy}=(3x+y)/3q_{xy} \qquad t_{xy}=\sqrt{8}y/3q_{xy}$$
$$s_{yx}=(3y+x)/3q_{xy} \qquad t_{yx}=\sqrt{8}x/3q_{xy}$$
$$s_{xz}=(x-z\cos\theta)/q_{xz} \qquad t_{xz}=z\sin\theta/q_{xz}$$
$$s_{zx}=(z-x\cos\theta)/q_{xz} \qquad t_{zx}=x\sin\theta/q_{xz}$$

and the symbols used for the equilibrium values of the interatomic distances and of the interbond angles are x=Si—O, y=Si—C, z=O—C, q_{xx}=O···O, q_{yy}=C···(Si)···C, q_{xy}=O···C, q_{xz}=Si···C and θ=∠SiOC.

The G matrix elements are obtained by the use of the table of Decius⁶⁾ and the method of Crawford⁷⁾. Using the notations ρ_x, ρ_y and ρ_z for the reciprocals of Si—O, Si—C and O—C bond lengths, and μ_{Si}, μ_{O} and μ_{C} for the reciprocals of the masses of silicon and oxygen atoms and methyl group, respectively, the following G matrix elements have been obtained:

for the a$_1$ vibrations,

$$G_{11}=\mu_{\mathrm{O}}+\mu_{\mathrm{C}}$$
$$G_{12}=\mu_{\mathrm{O}}\cos\theta$$
$$G_{14}=-\rho_x\mu_{\mathrm{O}}\sin\theta$$
$$G_{15}=-\rho_x\mu_{\mathrm{O}}\sin\theta$$
$$G_{16}=-\sqrt{3}\rho_x\mu_{\mathrm{O}}\sin\theta/\sqrt{2}$$
$$G_{22}=2\mu_{\mathrm{Si}}/3+\mu_{\mathrm{O}}$$
$$G_{23}=-2\mu_{\mathrm{Si}}/3$$
$$G_{24}=-\sqrt{8}\rho_x\mu_{\mathrm{Si}}/3$$
$$G_{25}=-\sqrt{8}(\rho_x+\rho_y)\mu_{\mathrm{Si}}/3$$
$$G_{26}=-2(\rho_x-\rho_y)\mu_{\mathrm{Si}}/\sqrt{3}$$
$$G_{33}=2\mu_{\mathrm{Si}}/3+\mu_{\mathrm{C}}$$
$$G_{34}=\sqrt{8}\rho_x\mu_{\mathrm{Si}}/3$$
$$G_{35}=\sqrt{8}(\rho_x+\rho_y)\mu_{\mathrm{Si}}/3$$
$$G_{36}=2(\rho_x-\rho_y)\mu_{\mathrm{Si}}/\sqrt{3}$$
$$G_{44}=4\rho_x^2\mu_{\mathrm{Si}}/3+\rho_z^2\mu_{\mathrm{C}}+(\rho_x^2+\rho_z^2$$
$$-2\rho_x\rho_z\cos\theta)\mu_{\mathrm{O}}$$
$$G_{45}=4\rho_x(\rho_x+\rho_y)\mu_{\mathrm{Si}}/3+\rho_x(\rho_x-\rho_z\cos\theta)\mu_{\mathrm{O}}$$
$$G_{46}=\sqrt{8}\rho_x(\rho_x-\rho_y)\mu_{\mathrm{Si}}/\sqrt{3}$$
$$+\sqrt{3}\rho_x(\rho_x-\rho_z\cos\theta)\mu_{\mathrm{O}}/\sqrt{2}$$
$$G_{55}=4(\rho_x+\rho_y)^2\mu_{\mathrm{Si}}/3+\rho_x^2\mu_{\mathrm{O}}+\rho_y^2\mu_{\mathrm{C}}$$
$$G_{56}=\sqrt{8}(\rho_x^2-\rho_y^2)\mu_{\mathrm{Si}}/\sqrt{3}$$
$$+\sqrt{3}(\rho_x^2\mu_{\mathrm{O}}-\rho_y^2\mu_{\mathrm{C}})/\sqrt{2}$$
$$G_{66}=2(\rho_x-\rho_y)^2\mu_{\mathrm{Si}}+3(\rho_x^2\mu_{\mathrm{O}}+\rho_y^2\mu_{\mathrm{C}})/2$$
$$G_{13}=0$$

for the a$_2$ vibration,

$$G_{77}=3(\rho_x^2\mu_{\mathrm{O}}+\rho_y^2\mu_{\mathrm{C}})/2$$

for the b$_1$ vibrations,

$$G_{99}=\mu_{\mathrm{O}}+\mu_{\mathrm{C}}$$
$$G_{910}=\mu_{\mathrm{O}}\cos\theta$$
$$G_{911}=-\rho_x\mu_{\mathrm{O}}\sin\theta$$
$$G_{912}=\rho_x\mu_{\mathrm{O}}\sin\theta/\sqrt{2}$$
$$G_{1010}=4\mu_{\mathrm{Si}}/3+\mu_{\mathrm{O}}$$
$$G_{1011}=\sqrt{8}\rho_x\mu_{\mathrm{Si}}/3-\rho_z\mu_{\mathrm{O}}\sin\theta$$
$$G_{1012}=-2(\rho_x+3\rho_y)\mu_{\mathrm{Si}}/3$$
$$G_{1111}=\rho_z^2(\mu_{\mathrm{O}}+\mu_{\mathrm{C}})+\rho_x^2(2\mu_{\mathrm{Si}}/3+\mu_{\mathrm{O}})$$
$$-2\rho_x\rho_y\mu_{\mathrm{O}}\cos\theta$$
$$G_{1112}=-\sqrt{2}\rho_x(\rho_x+3\rho_y)\mu_{\mathrm{Si}}/3$$
$$-\rho_x(\rho_x-\rho_z\cos\theta)\mu_{\mathrm{O}}/\sqrt{2}$$
$$G_{1212}=(\rho_x^2\mu_{\mathrm{O}}+3\rho_y^2\mu_{\mathrm{O}})/2+(\rho_x+3\rho_y)^2\mu_{\mathrm{Si}}/3$$

for the b$_2$ vibrations,

$$G_{1313}=4\mu_{\mathrm{Si}}/3+\mu_{\mathrm{C}}$$

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6) J. C. Decius, ibid., 16, 1025 (1948).

7) B. Crawford, ibid., 21, 1108 (1953).

$$G_{1314} = -2(\rho_x + \rho_y/3)\mu_{\mathrm{Si}}$$

$$G_{1414} = (3\rho_x^2\mu_{\mathrm{O}} + \rho_y^2\mu_{\mathrm{C}})/2 + (3\rho_x + \rho_y)^2\mu_{\mathrm{Si}}/3$$

The molecular and potential constants used in the calculation are listed in Table I. These values are the same as those in the preceding paper[8] except H_{yy}, F_{yy} and κ. The potential constants of $H(CH_3\text{-}Si\text{-}CH_3)$ and $F(CH_3\cdots CH_3)$ in tetramethylsilane[9] have been adopted for the H_{yy} and F_{yy} in the molecule under consideration. The κ in this molecule has been assumed to be the mean between that in tetramethylsilane[9] and that in tetramethoxysilane[10]. The values of F' have been assumed to be equal to $-F/10$ in the usual way.

TABLE I. MOLECULAR AND POTENTIAL CONSTANTS OF DIMETHYLDIMETHOXYSILANE

Bond distance and angle	Potential constant	
	Type	md./Å
x=Si—O	K_x(Si—O)	3.25
=1.64 Å a)	K_y(Si—CH_3)	3.03
y=Si—C	K_z(O—CH_3)	4.88
=1.888 Å b)	H_{xx}(O—Si—O)	0.13
z=O—C	H_{xy}(O—Si—CH_3)	0.12
=1.42 Å a)	H_{yy}(CH_3—Si—CH_3)	0.11
∠OSiC	H_{xz}(Si—O—CH_3)	0.22
=∠OSiC	F_{xx}(O···O)	0.13
=∠CSiC	F_{xy}(O···CH_3)	0.08
=109°28	F_{yy}(CH_3···CH_3)	0.034
=∠SiOC	F_{xz}(Si···CH_3)	0.21
=113° a)	κ	0.185 Å²

a) Values in $Si(OCH_3)_4$; M. Yokoi, This Bulletin, **30**, 100 (1957).

b) Value in $Si(CH_3)_4$; W. F. Sheehan, Jr. and V. Schomaker, *J. Am. Chem. Soc.*, **74**, 3956 (1952).

Assignments of the Observed Frequencies

From the F and G matrix elements given above, the fundamental frequencies have been calculated by the use of the molecular and the potential constants listed in Table I, and the results are shown in Table II, together with the observed wave numbers[3] of the Raman and the infrared bands in the region of the skeletal vibrations. The skeletal vibrations have been assigned by the calculation of the potential energy distribution in the symmetry coordinates.

In the present calculation, the wave number of the Si-O symmetrical stretching vibration(a_1) is higher than that of the Si-O nonsymmetrical stretching vibration(b_1). Nevertheless, the observed frequencies near 690 and 735 cm^{-1} have been assigned as $\nu_2(a_1)$ and $\nu_{10}(b_1)$, respectively (Table II), considering the intensities of Raman and infrared bands.

As is shown in Table II, the calculated values of ν_2 and ν_{10} are considerably higher than the observed ones, respectively. In order to get a better agreement between the calculated wave number and the observed, it is necessary

TABLE II. RAMAN AND INFRARED DATA, CALCULATED WAVE NUMBERS AND THE ASSIGNMENT FOR DIMETHYLDIMETHOXYSILANE

ν. Obs.		ν. Calcd.	Assignment	Mode of vibration
Raman a)	Infrared a)			
98 (1b)		136	ν_6 (a_1)	$Si(CH_3)_2+SiO_2$ sym. def.
		190	ν_{12} (b_1)	SiO_2 rocking
218 (4)		208	ν_7 (a_2)	$Si(CH_3)_2$ twisting
		238	ν_5 (a_1)	$Si(CH_3)_2+SiO_2$ nonsym. def.
260 (2)		288	ν_4 (a_1)	SiOC sym. def.
355 (0)		302	ν_{14} (b_2)	$Si(CH_3)_2$ rocking
380 (4bb)		400	ν_{11} (b_1)	SiOC nonsym. def.
618 (106)	613 (w)	618	ν_3 (a_1)	Si—C sym. str.
	673 (w)			
691 (3)	690 (w)	813	ν_2 (a_1)	Si—O sym. str.
736 (2)	730 (s)	779	ν_{10} (b_1)	Si—O nonsym. str.
804 (5)	797 (s)	791	ν_{13} (b_2)	Si—C nonsym. str.
845 (1)	849 (vs)			(Si—)CH_3 rocking
865 (2)				
995 (1)	998 (vw)			
1080 (2)	1073 (vs)	1069	ν_9 (b_1)	C—O nonsym. str.
	1088 (sh)	1071	ν_1 (a_1)	C—O sym. str.

a) Values in the liquid state

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[8] T. Tanaka, This Bulletin, **33**, 446 (1960).

[9] T. Shimanouchi, *J. Chem, Soc. Japan, Pure Chem. Sec.* (*Nippon Kagaku Zasshi*), **74**, 266 (1953).

[10] H. Murata, ibid., **77**, 893 (1956).

TABLE III. THE L MATRICES OF THE SKELETAL NORMAL VIBRATIONS OF DIMETHYLDIMETHOXYSILANE

a$_1$ vibrations

	Q_1	Q_2	Q_3	Q_4	Q_5	Q_6
R_1 (C—O sym. str.)	0.36	−0.03	0.02	−0.02	0.00	0.01
R_2 (Si—O sym. str.)	−0.10	−0.23	0.14	−0.04	0.00	0.00
R_3 (Si—C sym. str.)	0.01	0.22	0.20	0.01	0.01	0.00
R_4 (SiOC sym. def.)	−0.07	0.23	−0.15	−0.18	0.10	0.06
R_5 ($Si(CH_3)_2+SiO_2$ nonsym. def.)	−0.08	0.21	−0.02	−0.16	−0.16	−0.04
R_6 ($Si(CH_3)_2+SiO_2$ sym. def.)	−0.12	0.05	−0.04	−0.06	0.15	−0.14

b$_1$ vibrations

	Q_9	Q_{10}	Q_{11}	Q_{12}
R_9 (C—O nonsym. str.)	0.35	−0.07	0.01	0.00
R_{10} (Si—O nonsym. str.)	−0.13	−0.30	−0.01	−0.01
R_{11} (SiOC nonsym. def.)	−0.08	0.09	0.30	−0.10
R_{12} (SiO_2 rocking)	0.10	0.14	−0.21	−0.14

b$_2$ vibrations

	Q_{13}	Q_{14}
R_{13} (Si—C nonsym. str.)	−0.33	−0.03
R_{14} ($Si(CH_3)_2$ rocking)	0.18	−0.28

TABLE IV. POTENTIAL ENERGY DISTRIBUTION $F_{ii}L_{ia}^2/\lambda_a$ FOR THE NORMAL VIBRATION OF SKELETAL IN DIMETHYLDIMETHOXYSILANE

a$_1$ vibrations

	ν_1	ν_2	ν_3	ν_4	ν_5	ν_6
R_1 (C—O sym. str.)	0.94	0.01	0.01	0.05	0.00	0.01
R_2 (Si—O sym. str.)	0.05	0.50	0.33	0.11	0.00	0.00
R_3 (Si—C sym. str.)	0.00	0.39	0.59	0.01	0.01	0.00
R_4 (SiOC sym. def.)	0.01	0.09	0.07	0.42	0.20	0.22
R_5 ($Si(CH_3)_2+SiO_2$ nonsym. def.)	0.01	0.07	0.00	0.32	0.53	0.08
R_6 ($Si(CH_3)_2+SiO_2$ sym. def.)	0.01	0.00	0.00	0.03	0.26	0.69

b$_1$ vibrations

	ν_9	ν_{10}	ν_{11}	ν_{12}
R_9 (C—O nonsym. str.)	0.91	0.08	0.01	0.00
R_{10} (Si—O nonsym. str.)	0.09	0.89	0.01	0.02
R_{11} (SiOC nonsym. def.)	0.00	0.01	0.66	0.33
R_{12} (SiO_2 rocking)	0.01	0.03	0.32	0.64

b$_2$ vibrations

	ν_{13}	ν_{14}
R_{13} (Si—C nonsym. str.)	0.96	0.04
R_{14} ($Si(CH_3)_2$ rocking)	0.06	0.95

to adopt a lower value of K_x(Si—O), than the 3.25 md./Å of methyltrimethoxysilane[8]. Accordingly, it seems that the value of K_x(Si—O) in dimethyldimethoxysilane is rather lower than that in methyltrimethoxysilane. This may correspond to the tendency for the bond energy of Si–O bond which decreases as the number of oxygen atoms attached to silicon decreases; $-E_{Si-O}$ in SiO_4 unit $> -E_{Si-O}$ in $CSiO_3$ unit $> -E_{Si-O}$ in C_2SiO_2 unit, as reported by the present author[11].

The L matrices and the distribution of potential energy in symmetry coordinates have been calculated for the skeletal normal modes; the results are shown in Tables III and IV, respectively. As shown in Table IV, some of the symmetry coordinates of totally symmetric species are related to several normal modes to a considerable extent; the R_2 and R_3 coordinates to both modes of ν_2 and ν_3, and the R_4 and R_5 coordinates to a mode of ν_4.

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[11] T. Tanaka, *J. Inorg. Nucl. Chem.*, **13**, 225 (1961).