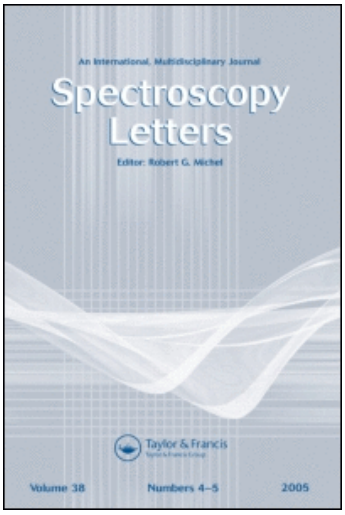

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Cartesian Dynamics of Simple Molecules VI Centrosymmetric Linear Pentatomics

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CARTESIAN DYNAMICS OF SIMPLE MOLECULES
VI CENTROSYMMETRIC LINEAR PENTATOMICS

Key Words: Molecular vibrations; infrared spectroscopy; Raman spectroscopy; lattice dynamics

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ABSTRACT

A simple spring model for molecular vibrations, which uses Cartesian co-ordinates for both longitudinal and transverse displacements, is applied to centrosymmetric linear pentatomic molecules such as carbon suboxide and carbon subsulphide. Analytical expressions for the four stretching and three bending mode frequencies are derived in terms of seven independent force constants. By substitution of Raman and infrared frequencies, values of these force constants are obtained and briefly discussed. Eigenvectors of the normal modes and eigenfrequencies of various isotopic species are also calculated.

INTRODUCTION

In previous papers[1-5], a simple spring model for molecular vibrations, which uses Cartesian co-ordinates was described and applied to diatomic, triatomic and quadratomic molecules. The rationale for this approach, which was shown to be equivalent to

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the usual one involving internal co-ordinates, is that it may be readily extended to lattice dynamics calculations[6-10], in which a uniform treatment of both intra- and inter- molecular interactions is used, leading to estimates of crystal field splittings of internal modes as well as frequencies of lattice modes. In this communication, the model is applied to the case of linear pentatomic molecules, such as carbon suboxide and carbon subsulphide.

Analytical expressions are derived for the normal mode frequencies of these molecules in terms of longitudinal and transverse force constants, atomic masses and bond lengths. Values of these force constants are obtained by substitution of the observed Raman and infrared fundamental frequencies. The form of the normal modes is verified by calculation of the eigenvectors. Finally, the eigenfrequencies of various isotopic species are calculated.

The geometry for these A_2B_3-type molecules, which have point group $D_{\infty h}$, is shown in Figure 1(a). There are 15 degrees of freedom, of which 3 correspond to pure translations and 2 to pure rotations. The remaining 10 internal degrees of freedom comprise 4 longitudinal or stretching modes which have non-degenerate representations (ω_1 and ω_2, both of σ_g^+ species and ω_3 and ω_4 of σ_u^- species) and 3 transverse or bending modes, which have doubly degenerate representations (ω_5 of π_g species and ω_6 and ω_7 of π_u species). The 3 gerade modes are Raman active and the 4 ungerade modes are infrared active. These 7 observed frequencies may be

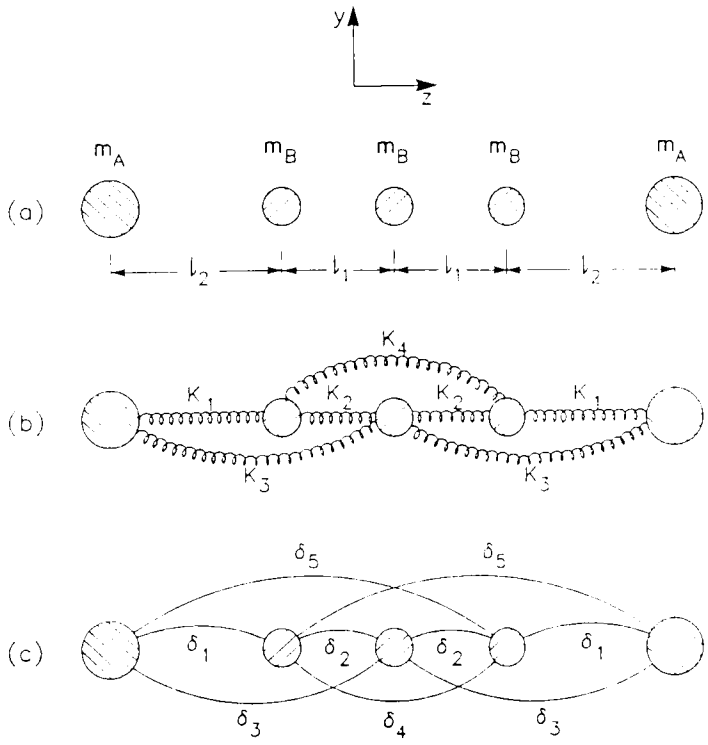

FIG. 1 Geometry and Spring Constant Model for A_2B_3 Linear Molecules

(a) Geometry, showing masses and bond lengths.

(b) Longitudinal Springs: k_1, k_2, k_3 and k_4.

(c) Transverse Springs: δ_1, δ_2, δ_3, δ_4 and δ_5.

used, in principle, to find values of 4 stretching and 3 bending force constants.

DESCRIPTION OF THE MODEL

We first consider motion along the molecular z-axis and derive expressions for ω_1, ω_2, ω_3 and ω_4. Following the

principles outlined in earlier papers in this series[1-3], we introduce longitudinal springs k_1, k_2, k_3 and k_4 as shown in Figure 1(b). The equations of motion in the z direction for the 5 atoms, according to Newton's second law and Hooke's law, are as follows:

$$m_A \ddot{z}_1 = -k_1\left(z_1 - z_2\right) - k_3\left(z_1 - z_3\right)$$

$$m_B \ddot{z}_2 = -k_1\left(z_2 - z_1\right) - k_2\left(z_2 - z_3\right) - k_4\left(z_2 - z_4\right)$$

$$m_B \ddot{z}_3 = -k_2\left(z_3 - z_2\right) - k_3\left(z_3 - z_1\right) - k_2\left(z_3 - z_4\right) - k_3\left(z_3 - z_5\right)$$

$$m_B \ddot{z}_4 = -k_2\left(z_4 - z_3\right) - k_1\left(z_4 - z_5\right) - k_4\left(z_4 - z_2\right)$$

$$m_A \ddot{z}_5 = -k_1\left(z_5 - z_4\right) - k_3\left(z_5 - z_3\right)$$

For harmonic oscillations, $z_n = z_{no} \cos \omega t$, and so

$$\ddot{z}_n = -\omega^2 z_{no} \cos \omega t = -\omega^2 z_n$$

It is now convenient to introduce new variables, to make use of the symmetry properties of the normal modes, as follows:

$q_1 = z_1 + z_5$; $q_2 = z_2 + z_4$; $q_3 = z_3$; $q_4 = z_1 - z_5$ and $q_5 = z_2 - z_4$.

When the equations of motion are re-written in terms of q_n, a 5 x 5 secular matrix is obtained, the determinant of which has the following block diagonal form:

$$\begin{vmatrix} m_A\omega^2-k_1-k_3 & k_1 & 2k_3 & 0 & 0 \\ k_1 & m_B\omega^2-k_1-k_2 & 2k_2 & 0 & 0 \\ k_3 & k_2 & m_B\omega^2-2k_2-2k_3 & 0 & 0 \\ 0 & 0 & 0 & m_A\omega^2-k_1-k_3 & k_1 \\ 0 & 0 & 0 & k_1 & m_B\omega^2-k_1-k_2-2k_4 \end{vmatrix} = 0$$

The lower block when expanded gives two roots, both non-zero, which correspond to the Raman active modes, ω_1 and ω_2. A quadratic equation is obtained of the form $A\omega^4 + B\omega^2 + C = 0$ and the following relations are obtained:

$$\omega_1^2 + \omega_2^2 = -B/A = \left(k_1 + k_2\right)/m_A + \left(k_1 + k_2 + 2k_4\right)/m_B \qquad (1)$$

$$\omega_1^2\,\omega_2^2 = C/A = \left(k_1k_2 + k_1k_3 + k_2k_3 + 2k_1k_4 + 2k_3k_4\right)/m_Am_B \qquad (2)$$

The exact form of the eigenvectors for these modes depends on the values of the force constants, but the higher frequency mode involves the symmetric stretching of both A-B and B-B bonds, whereas the lower frequency mode is primarily the symmetric B-B stretch, with the central B atom stationary in both cases. The approximate form of these modes is shown in Figure 2(a).

The upper block of the determinant when expanded gives three roots, one of which is the pure translational mode, ω_0, with zero frequency. The other two correspond to the infrared active modes ω_3 and ω_4. A cubic equation in ω^2 is obtained of the form

$$A\omega^6 + B\omega^4 + C\omega^2 + D = 0$$

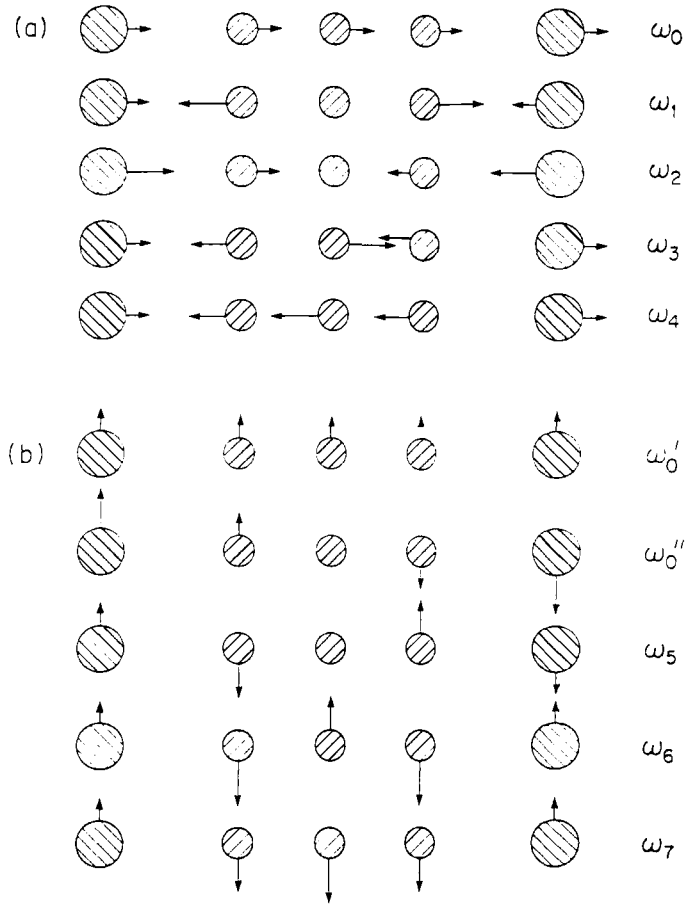

FIG. 2 Normal Modes of A_2B_3 molecules

(a) Longitudinal (Stretching) Modes.

(b) Transverse (Bending) Modes.

It is readily shown after some algebraic manipulation that D = 0, so that $\omega^2 = 0$ is a root. In addition, the following relations are obtained for the other two roots:

$$\omega_3^2 + \omega_4^2 = -B/A = \left(k_1 + k_3\right)/m_A + \left(k_1 + 3k_2 + 2k_3\right)/m_B \qquad (3)$$

$$\omega_3^2\ \omega_4^2 = C/A = \left(k_1k_2 + k_1k_3 + k_2k_3\right)\ \left(3m_B + 2m_A\right)/m_B^2\ m_A \qquad (4)$$

As for the Raman active modes, the eigenvectors for ω_3 and ω_4 depend on the force constants, but the higher frequency mode, ω_3, is the asymmetric combined A-B and B-B stretches and the lower frequency mode primarily the asymmetric B-B stretch as shown in Figure 2(a). It is noted that since the two non-central B atoms are in phase for these modes, the force constant k_4 does not appear in equations (3) and (4).

From the four stretching mode frequencies (two Raman and two infrared), it should be possible to find optimum values for the four force constants k_1, k_2, k_3 and k_4 using equations (1), (2), (3) and (4). It is expected that those representing springs between adjacent (bonded) atoms, k_1 and k_2, will be much larger than those between more distant (non-bonded) atoms, k_3 and k_4.

We next consider motion perpendicular to the molecular axis. Five transverse springs, δ_1, δ_2, δ_3, δ_4 and δ_5 are introduced. These are displayed as bow symbols in Figure 1(c). It will be shown later that only three of these transverse force constants are independent. The equations of motion in the transverse (x or y) direction have the following form:

$$m_A\ddot{y}_1 = -\delta_1(y_1 - y_2) - \delta_3(y_1 - y_3) - \delta_5(y_1 - y_4)$$

$$m_B\ddot{y}_2 = -\delta_1(y_2 - y_1) - \delta_2(y_2 - y_3) - \delta_4(y_2 - y_4) - \delta_5(y_2 - y_5)$$

$$m_B\ddot{y}_3 = -\delta_2(y_3 - y_2) - \delta_2(y_3 - y_4) - \delta_3(y_3 - y_1) - \delta_3(y_1 - y_5)$$

$$m_B\ddot{y}_4 = -\delta_1(y_4 - y_5) - \delta_2(y_4 - y_3) - \delta_4(y_4 - y_2) - \delta_5(y_4 - y_1)$$

$$m_A\ddot{y}_5 = -\delta_1(y_5 - y_4) - \delta_3(y_5 - y_3) - \delta_5(y_5 - y_2)$$

We now make the usual substitution for harmonic oscillations,

$$\ddot{y}_n = -\omega^2 y_{no} \cos \omega t = -\omega^2 y_n$$

and introduce the following symmetry co-ordinates, similar to the longitudinal case:

$p_1 = y_1 + y_5$, $p_2 = y_2 + y_4$, $p_3 = y_3$, $p_4 = y_1 - y_5$ and $p_5 = y_2 - y_4$.

The equations of motion are then re-written in terms of p_n, and the following secular determinant in block diagonal form is obtained:

$$\begin{vmatrix} m_A\omega^2-\delta_1-\delta_3-\delta_5 & \delta_1+\delta_5 & 2\delta_3 & 0 & 0 \\ \delta_1+\delta_5 & m_B^2\omega-\delta_1-\delta_2-\delta_5 & 2\delta_2 & 0 & 0 \\ \delta_3 & \delta_2 & m_B\omega^2-2\delta_2-2\delta_3 & 0 & 0 \\ 0 & 0 & 0 & m_A\omega^2-\delta_1-\delta_3-\delta_5 & \delta_1-\delta_5 \\ 0 & 0 & 0 & \delta_1-\delta_5 & m_B\omega^2-\delta_1-\delta_2-2\delta_4-\delta_5 \end{vmatrix} = 0$$

This determinant is very similar to that obtained for the longitudinal modes, with δ's replacing k's and with $\delta_5 = 0$.

Expansion of the lower 2 x 2 block leads to the following equation:

$$A\omega^4 + B\omega^2 + C = 0$$

where

$$A = m_1 m_2, \quad B = -\left(\delta_1 + \delta_3 + \delta_5\right)m_1 - \left(\delta_1 + \delta_2 + 2\delta_4 + \delta_5\right)m_2$$

and

$$C = \left(\delta_1 + \delta_3 + \delta_5\right)\left(\delta_1 + \delta_2 + 2\delta_4 + \delta_5\right) - \left(\delta_1 - \delta_5\right)^2$$

In order for a zero frequency root to exist, C must be equal to zero. Physically this results from a pure rotation about the centre of mass, which for these molecules coincides with the central atom. This mode is shown as ω_o'' in Figure 2(b). For this motion, if $y_1 = y$, then $y_2 = \alpha y$, $y_3 = 0$, $y_4 = -\alpha y$ and $y_5 = -y$ with $\alpha = \ell_1 / \left(\ell_1 + \ell_2\right)$. If these relations are substituted into the first two equations of motion, together with $\ddot{y}_1 = \ddot{y}_2 = 0$, corresponding to no restoring forces, the following two equations are obtained after some algebraic re-arrangement:

$$\delta_3 = -\delta_1\left(1 - \alpha\right) - \delta_5\left(1 + \alpha\right)$$

$$\delta_4 = -\delta_2 / 2 + \delta_1\left(1 - \alpha\right) / 2\alpha - \delta_5\left(1 + \alpha\right) / 2\alpha$$

Substitution of these expressions for δ_3 and δ_4 into the equation for the coefficient C confirms that it is indeed equal to zero.

It is then easily shown that the non-zero root of the quadratic equation has the following form:

$$\omega_5^2 = \left(\delta_1 - \delta_5\right)\left(\alpha/m_2 + 1/\alpha m_1\right) \tag{5}$$

This corresponds to the Raman active π_g bending mode, the approximate form of which is shown in Figure 2(b). The absence of δ_2 in the equation for ω_5 is explained physically by noting that the central three B atoms maintain their linearity for this mode, so that this bending force is not activated.

Expansion of the upper 3 x 3 block of the determinant leads to a cubic equation in ω_2 of the following form:

$$A\omega^6 + B\omega^4 + C\omega^2 + D = 0$$

After some routine algebra, it is confirmed that D = 0, so that one root has zero frequency. This corresponds to a pure translation of the whole molecule in the transverse direction, shown as ω_0' in figure 2(b). Further manipulation of the resulting quadratic equation leads to the following relations for the two non-zero roots:

$$\omega_6^2 + \omega_7^2 = -B/A \tag{6}$$

$$= \alpha\left(\delta_1 - \delta_5\right)/m_A + \left[3\delta_2 + \delta_1\left(2\alpha - 1\right) - \delta_5\left(2\alpha + 1\right)\right]/m_B$$

$$\omega_6^2\,\omega_7^2 = C/A \tag{7}$$

$$= \left(3m_B + 2m_A\right)\left(\alpha\delta_1^2 - \delta_1^2 - 2\delta_1\delta_5 + \alpha\delta_1\delta_2 - \alpha\delta_2\delta_5 - \delta_5^2 - \alpha\delta_5^2\right)/m_B^2 m_A$$

These roots correspond to the infrared active π_u modes, in which similar atoms move in phase. The higher frequency ω_6 mode has adjacent carbon atoms moving out of phase, the lower frequency ω_5 has them in phase, as shown in Figure 2(b). From the three bending mode frequencies, ω_5 (Raman), ω_6 and ω_7 (infrared), it should be possible to derive optimum values for δ_1, δ_2 and δ_5 using equations (5), (6) and (7), and hence values of the dependent transverse force constants, δ_3 and δ_4. As for the longitudinal case, based on atomic proximities, it is expected that δ_1 and δ_2 will be much larger than δ_5.

RESULTS

Bond lengths and observed fundamental vibrational frequencies for C_3O_2 and C_3S_2 are listed in Table 1. These values have been used with equations (1) to (7) to obtain the force constants given in Table 2. These coupled equations are sufficiently complex that it was not possible to transform them to give the force constants directly as analytical expressions in terms of the frequencies. Instead, a numerical optimization routine known as SIMPLEX[13] was used to approximatethe force constants. For both molecules, frequencies calculated from the optimized force constants are within 1 cm^{-1} of the observed values in all cases. In Table 3, values of the normal mode frequencies for various isotopic species of these molecules are listed. These have been calculated using the force constants of Table 2. Unfortunately, experimental values of these frequencies for gas phase samples are unavailable for comparison.

TABLE 1

Bond Lengths and Normal Mode Frequencies for A_2B_2 Linear Molecules

Bond Lengths* (Å)	Molecule C_3O_2	Molecule C_3S_2
ℓ_1 (C-C)	1.28	1.28
ℓ_2 (C-X)	1.16	1.54
Frequencies† (cm^{-1})		
ω_1 (σ_g^+)	2196.9	1663
ω_2 (σ_g^+)	787.7	489.8
ω_3 (σ_u^-)	2289.8	2100.0
ω_4 (σ_u^-)	1587.4	1030.2
ω_5 (π_g)	580.2	470
ω_6 (π_u)	540.2	502
ω_7 (π_u)	18.3	93.7

*From Ref 11 †From Ref 12

Calculated eigenvectors for the seven normal modes of these two molecules are listed in Table 4.

DISCUSSION

Inspection of Table 2 shows the expected dominance of the longitudinal valence springs, k_1 and k_2, for both molecules. The longitudinal springs between non-bonded atoms, represented by k_3 and k_4, are much weaker. Similarly, the transverse force constants between adjacent atoms, δ_1 and δ_2, are much larger than those between non-bonded atoms, δ_5 and the dependent constants δ_3 and δ_4.

TABLE 2

Optimized Force Constants of A_2B_3 Linear Molecules

Force Constants* (ucm^{-2})	Molecule C_3O_2	Molecule C_3S_2
k_1	2.5800×10^7	1.2569×10^7
k_2	1.4035×10^7	1.5560×10^7
k_3	2.142×10^6	7.09×10^5
k_4	2.288×10^6	1.478×10^6
δ_1	1.594×10^6	1.018×10^6
δ_2	8.01×10^5	9.48×10^5
δ_5	-1.61×10^5	-9.9×10^4
(δ_3)	-5.13×10^5	-4.12×10^5
(δ_4)	5.56×10^5	2.97×10^5

*Units from $k = \mu\omega^2$ where μ is in atomic mass units (u) and ω in wavenumbers (cm^{-1}). Multiply table entries by 5.90×10^{-5} to convert to N/m or by 5.90×10^{-2} for dyne/cm.

As for the previous molecules studied[1-5], the principal longitudinal force constants, which are mainly elastic in nature, are greater than their transverse counterparts, which result from electrostatic interactions, as discussed in the first paper in this series[1]. The model described in this paper allows a direct comparison between the transverse and longitinal force constants, as they are expressed in the same units of u cm^{-2}, in contrast to the more conventional approach which uses bending constants expressed in angular co-ordinates.

TABLE 3

Calculated Frequencies for Isotopic A_2B_3 Linear Molecules*

Molecule	Frequency (cm^{-1})						
	ω_1	ω_2	ω_3	ω_4	ω_5	ω_6	ω_7
$^{16}O^{12}C^{12}C^{12}C^{16}O$	2196.9	787.7	2289.8	1587.4	580.2	540.2	18.3
$^{18}O^{12}C^{12}C^{12}C^{16}O$	2179.5	770.8	2282.0	1569.6	577.4	536.8	17.8
$^{16}O^{13}C^{12}C^{12}C^{16}O$	2157.4	783.7	2270.6	1586.1	572.2	531.6	18.0
$^{16}O^{12}C^{13}C^{12}C^{16}O$	2196.9	787.7	2270.6	1548.9	580.2	536.6	17.5
$^{18}O^{12}C^{12}C^{12}C^{18}O$	2164.4	754.0	2271.3	1552.2	574.4	534.0	17.7
$^{32}S^{12}C^{12}C^{12}C^{32}S$	1663.0	489.8	2100.0	1030.2	470.0	502.0	93.7
$^{34}S^{12}C^{12}C^{12}C^{32}S$	1661.3	483.1	2099.7	1024.7	469.5	501.6	93.6
$^{32}S^{13}C^{12}C^{12}C^{32}S$	1631.7	489.3	2082.5	1023.5	460.0	497.4	93.7
$^{32}S^{12}C^{13}C^{12}C^{32}S$	1633.0	489.8	2056.9	1015.2	470.0	495.5	91.9
$^{34}S^{12}C^{12}C^{12}C^{34}S$	1659.6	476.4	2099.4	1019.3	469.0	501.2	93.1

*Force constants listed in Table 2 have been used.

Also of interest are the differences between the principal interactions involving C-O and C-S bonds. The force constant k_1 for C_3O_2 is more than twice the value for C_3S_2 and δ_1 for C_3O_2 is more than 50% larger than the value for C_3S_2. On the other hand, the principal C-C interactions, k_2 and δ_2, are somewhat less for C_3O_2 than their counterparts for C_3S_2. In particular, δ_2 is about 15% smaller for C_3O_2. This force constant is primarily involved in the ν_7 mode (approximately a CCC bend) which has an anomalously low value of 18 cm^{-1} in C_3O_2. However, other force constants (δ_1 and δ_5) are involved, and there appears to be a partial cancellation of

TABLE 4

Eigenvectors for Normal Modes of A_2B_3 Linear Molecules

Mode	Molecule			
	C_3O_2		C_3S_2	
	Relative Atomic Displacements*			
	O	C	S	C
ω_1	0.524	0	0.167	0
ω_2	1.431	0	2.246	0
ω_3	0.428	0.858	0.090	1.521
ω_4	3.248	6.654	0.662	1.523
ω_5	0.394	0	0.170	0
ω_6	0.481	0.717	0.179	1.047
ω_7	5.426	12.47	0.883	2.711

*Displacements of the non-central carbon atoms have been normalized to unity in all cases. C above refers to central carbon atom. See Figure 2 for phase relationships between all five atoms.

their various contributions, so that the net restoring force for this mode is very small. Unfortunately, the analytical expressions for the coupled modes ν_6 and ν_7 (equations 6 and 7) are sufficiently complex that the specific origin of this "softening" is not obvious. Because of the low resistance to bending, C_3O_2 is often described as a "quasi-linear" molecule.

In previous papers[1-5], the validity of the models was verified by the agreement between calculated and observed frequencies of

isotopically substituted molecules. However, very few observed gas phase frequencies of isotopic species of C_3O_2 and C_3S_2 are available to compare with our calculated values (Table 3). An alternative verification was adopted involving a comparison of calculated and observed frequencies for these molecules in condensed phases. These frequencies were previously calculated by Smith and co-workers[14-16] with a model using 8 adjustable parameters instead of our 7. We have repeated the exercise using their observed frequencies for solid samples and our Cartesian model. Our calculated values are the same as theirs within ±2 cm^{-1}, and this, together with the similar values obtained for the principal force constants, suggests that our model is essentially equivalent to the one described in their papers. For these condensed phases, however, intermolecular perturbations may be appreciable and change the effective values of the intramolecular constants. For example, ν_7 of C_3O_2 has its gas phase value of 18 cm^{-1} raised to 72 cm^{-1} in the liquid. The absence of definitive crystal structure determinations for these molecules precludes the possibility of lattice dynamics calculations at the present time. In addition, the lattice spectra are incomplete, although evidence for a solid state phase transition in C_3O_2 has been observed[14]. The next paper in this series will deal with A_2BC planar molecules with C_{2v} symmetry.

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