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The Normal Vibrations and the Isotope Effect of Molecules of the Type X_2Y_N with an N-Fold Axis of Symmetry

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In the present study it is assumed that the Y atoms are at the corners of a regular polygon and the X atoms are symmetrically situated on the N-fold axis of symmetry. The frequencies are calculated in terms of the parameters of a general quadratic potential energy form, the masses, and the geometrical constants of the figure. Frequency shifts due to isotopes are also calculated.

INTRODUCTION

THE general aspects of the theory of normal vibrations of polyatomic molecules have been treated by Brester,[1] Wigner,[2] Dennison,[3] Tisza,[4] and Wilson.[5] Other workers[6, 7, 8, 9] have given specific treatments for molecules of the form XY_2, XYZ, XY_3, XY_4 which included the derivation of algebraic expressions for the values of the frequencies and studies of the isotope effect. In the present paper this work is continued and a symmetrical molecule of the form X_2Y_N is investigated. The expression used for the potential energy is the most general consistent with the symmetry. By a series of transformations which take full advantage of the symmetry, the kinetic energy and the potential energy matrices are brought to the diagonal form. The values of the frequencies and the shifts due to isotopes are then calculated. In order to enable one to see more clearly and briefly which calculations to make, matrix notation is introduced.

GENERAL THEORY

Let there be n particles in equilibrium to which are ascribed $3n$ coordinates whose equilibrium values are $X_1, X_2, X_3, \cdots, X_{3n}$. Let the corresponding displacements $x_1, x_2, x_3, \cdots, x_{3n}$ be written in the form of a column so as to constitute a $3n$-row, 1-column matrix which will be denoted by x. Let $\mathbf{x}$ denote the transposed matrix. The usual expressions for the potential and kinetic energies

$$2V=\sum\sum v_{jk}x_jx_k, \qquad 2T=\sum\sum t_{jk}\dot{x}_j\dot{x}_k \tag{1}$$

can be written in matrix notation as follows

$$2V=\mathbf{x}v_xx, \qquad 2T=\dot{\mathbf{x}}t_x\dot{x}, \tag{2}$$

where $v_x=\|v_{jk}\|$ and $t_x=\|t_{jk}\|$. v_x and t_x will be called the potential and kinetic energy matrices respectively. When one transforms to a new set of coordinates y in accordance with the transformation $x=S_x{}^yy$ where $S_x{}^y$ is a matrix constant in time, then it can be seen that v_x, t_x, t_x^{-1} transform according to the following equations

$$v_y=\mathbf{S}_x{}^yv_xS_x{}^y, \quad t_y=\mathbf{S}_x{}^yt_xS_x{}^y, \quad t_y{}^{-1}=S_y{}^xt_x{}^{-1}\mathbf{S}_y{}^x. \tag{3}$$

According to a well-known theorem[10] the normal frequencies of vibration ν_j are given by $4\pi^2\nu_j{}^2=\lambda_j$ where the λ_j are roots of the following equation

$$|\lambda t_x-v_x|=0. \tag{4}$$

In the present paper the derived form

$$|\lambda 1-t_x{}^{-1}v_x|=0 \tag{5}$$

will be used instead of Eq. 4.

If some of the x coordinates are complex and moreover the complex coordinates occur in conjugate pairs then it can be shown that Eqs. (2), (3), (4), (5) are still valid if $\mathbf{x}$ is reinterpreted as the transposed conjugate complex to x. Under this new interpretation the previous equations for real coordinates are a special case of the more general equations for complex coordinates.

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[10] See for example reference 3, p. 284.

The Geometric Model and the Coordinates

It is assumed that the N similar Y atoms are located at the vertices of a regular polygon. The two X atoms are situated on the N-fold axis of symmetry, equidistant from the plane of the regular polygon. The Y atoms are labelled by the subscripts $0, 1, 2, \cdots, N-1$. One of the X atoms is labelled by a superscript prime, the other by a superscript double prime. The radius of the circumscribed circle of the regular polygon is denoted by R. The ratio of the distance between the two X atoms to the radius R is denoted by $2h$. The geometric model is illustrated in Fig. 1 for the case $N=3$. In this figure the atoms are labelled by their respective masses. Note the relations $m'=m''$, $m_j=m$, $j=0, 1, 2, \cdots, N-1$. A number of sets of displacement coordinates are used. Throughout subsequent calculations these displacements are considered mathematically infinitesimal and higher order terms are neglected. This is the usual approximation made in work of this type.

x or cartesian coordinates

A system of cartesian coordinates at rest are used for the initial expression of the kinetic energy. The z axis is the N-fold axis of symmetry of the equilibrium configuration. The x axis passes through the equilibrium position of m_0. The equilibrium values of the coordinates are given by the following equations

$$X_j=R\,\mathrm{Cos}\,j\theta, \quad Y_j=R\,\mathrm{Sin}\,j\theta, \quad \theta=2\pi/N, \qquad (6)$$
$$Z'=-Z''=hR, \quad Z_j=X'=X''=Y'=Y''=0.$$

The displacements are denoted by corresponding small letters. The whole set of displacements will be represented by the matrix x.

p coordinates

The following $3N$ interatomic distances are convenient for the initial expression of the potential energy

$P_j'+p_j'$ the distance from m_j to m',
$P_j''+p_j''$ the distance from m_j to m'',
Q_j+q_j the distance from m_j to m_{j+1}.

Again equilibrium values are denoted by capital letters and displacements by small letters. Note the relations

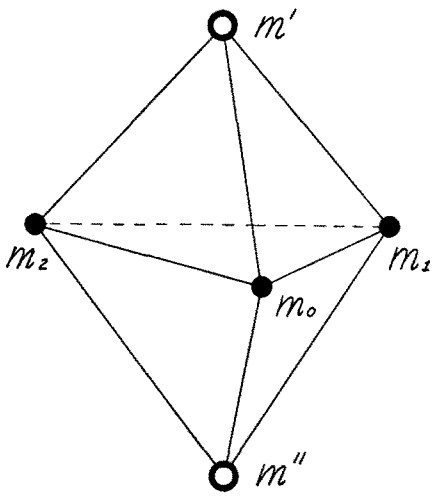

Fig. 1.

$$P_j'=P_j''=P=R(1+h^2)^{\frac{1}{2}},$$
$$Q_j=Q=2R\,\mathrm{Sin}\,\theta/2, \quad j=0, 1, 2, \cdots, N-1. \qquad (7)$$

To complete the set p the following six coordinates α_x, α_y, α_z, β_x, β_y, β_z are added. Note that the definitions of α_y, α_z, β_y, β_z are analogous to those of α_x, β_x.

$$\alpha_x=\sum_j m_j x_j+m'x'+m''x'',$$
$$\beta_x=\sum_j m_j(Y_j z_j-Z_j y_j)+m'(Y'z'-Z'y') +m''(Y''z''-Z''y''). \qquad (8)$$

The time derivatives of these six coordinates are equal to the six components of linear and angular momentum. The six coordinates are therefore constants, in particular zero.

ϕ coordinates

These symmetry coordinates were chosen because v_ϕ and t_ϕ^{-1} could be shown to have an almost diagonal form, and are defined as follows:

$$\phi_j'=\sum_k \epsilon^{jk}(p_k'-p_k'')N^{-\frac{1}{2}}P/2R,$$
$$\phi_j''=\sum_k \epsilon^{jk}(p_k'+p_k'')N^{-\frac{1}{2}}P/2R,$$
$$\phi_j'''=\sum \epsilon^{j(k+\frac{1}{2})}q_k N^{-\frac{1}{2}}Q/R. \qquad (9)$$

ϵ denotes the primitive Nth root of unity.

In order to make the transformation from x to ϕ coordinates an initial transformation from x to p coordinates is made. This initial transformation

is given by the following first order equations which may be readily derived from the geometry of the figure:

$$\begin{aligned} Pp_j' &= X_j(x_j - x') + Y_j(y_j - y') \\ &\quad - Z'(z_j - z'), \\ Pp_j'' &= X_j(x_j - x'') + Y_j(y_j - y'') \\ &\quad - Z''(z_j - z''), \\ Qq_j &= (X_{j+1} - X_j)(x_{j+1} - x_j) \\ &\quad + (Y_{j+1} - Y_j)(y_{j+1} - y_j). \end{aligned} \tag{10}$$

n or normal coordinates

These coordinates are defined by the property that the kinetic energy matrix is unity and the potential energy matrix is diagonal. Their significance is well known and their calculation is postponed pending the calculation of the frequencies.

The Potential Energy Matrix

Since the potential energy is independent of the six degrees of freedom corresponding to rotation and translation, it may be expressed in terms of the $3N$ coordinates p_j', p_j'', q_j or ϕ_j', ϕ_j'', ϕ_j'''. The forms of the matrices v_p and v_ϕ were investigated by performing the following three symmetrical operations:

(1) Reflection with respect to the x, y plane

$p_j' \to p_j''$, $p_j'' \to p_j'$, $q_j \to q_j$,
$\phi_j' \to -\phi_j'$, $\phi_j'' \to \phi_j''$, $\phi_j''' \to \phi_j'''$.

(2) Rotation about the z axis by an angle $\theta = 2\pi/N$

$p_j' \to p_{j+1}'$, $p_j'' \to p_{j+1}''$, $q_j \to q_{j+1}$,
$\phi_j' \to \epsilon^{-j}\phi_j'$, $\phi_j'' \to \epsilon^{-j}\phi_j''$, $\phi_j''' \to \epsilon^{-j}\phi_j'''$.

(3) Reflection with respect to the x, z plane

$p_j' \to p_{N-j}'$, $p_j'' \to p_{N-j}''$, $q_j \to q_{N-j-1}$,
$\phi_j' \to \phi_{N-j}'$, $\phi_j'' \to \phi_{N-j}''$, $\phi_j''' \to -\phi_{N-j}'''$.

The action of the symmetrical operations upon the coordinates p_j', p_j'', q_j and ϕ_j', ϕ_j'', ϕ_j''' is indicated in order to show how one takes into account symmetry. It is clear that after any one of the three operations the geometric figure and the potential and kinetic energy matrices will retain their original form. Consider the following examples of this method applied to the investigation of v_ϕ.

As a consequence of operation (1) $\phi_j'\phi_k'' \to -\phi_j'\phi_k''$. The coefficient of $\phi_j'\phi_k''$ must be zero in order for v_ϕ to remain unaltered after this symmetrical operation.

As a consequence of operation (2) $\phi_j'\phi_k' \to \epsilon^{-j-k}\phi_j'\phi_k'$ so that only the coefficient of $\phi_j'\phi_{N-j}'$ may be non-vanishing and v_ϕ retain its original form.

As a consequence of operation (3) $\phi_j''\phi_{N-j}''' \to -\phi_{N-j}''\phi_j'''$ and $\phi_{N-j}''\phi_j''' \to -\phi_j''\phi_{N-j}'''$. In order for v_ϕ to remain unaltered the coefficients of the terms $\phi_j''\phi_{N-j}'''$, $\phi_{N-j}''\phi_j'''$ must be opposite in sign.

In order to present the results of the above considerations it is convenient to group the ϕ coordinates into the submatrices ϕ', ϕ'', ϕ''', α, β. They are defined as follows:

$$\phi' = \begin{pmatrix} \phi_0' \\ \phi_1' \\ \cdot \\ \cdot \\ \cdot \\ \phi_{N-1}' \end{pmatrix}, \quad \phi'' = \begin{pmatrix} \phi_0'' \\ \phi_1'' \\ \cdot \\ \cdot \\ \cdot \\ \phi_{N-1}'' \end{pmatrix}, \quad \phi''' = \begin{pmatrix} \phi_0''' \\ \phi_1''' \\ \cdot \\ \cdot \\ \cdot \\ \phi_{N-1}''' \end{pmatrix},$$

$$\alpha = \begin{pmatrix} \alpha_x \\ \alpha_y \\ \alpha_z \end{pmatrix}, \quad \beta = \begin{pmatrix} \beta_x \\ \beta_y \\ \beta_z \end{pmatrix}.$$

The submatrices of the p coordinates, namely p', p'', q, α, β are defined analogously.

By means of the considerations previously illustrated it can be shown that v_p and v_ϕ have the following forms:

v_p	$\mathfrak{p}'$	$\mathfrak{p}''$	$\mathfrak{q}$	$\mathfrak{\alpha}$	$\mathfrak{\beta}$
p'	a'	b'	c'	0	0
p''	$\mathfrak{b}'$	a'	c'	0	0
q	$\mathfrak{c}'$	$\mathfrak{c}'$	d'	0	0
α	0	0	0	0	0
β	0	0	0	0	0

v_ϕ	$\mathfrak{\phi}'$	$\mathfrak{\phi}''$	$\mathfrak{\phi}'''$	$\mathfrak{\alpha}$	$\mathfrak{\beta}$
ϕ'	a	0	0	0	0
ϕ''	0	b	c	0	0
ϕ'''	0	c	d	0	0
α	0	0	0	0	0
β	0	0	0	0	0

a' b' c' d' are non-diagonal submatrices while a, b, c, d are diagonal submatrices. The following equations relate the elements of v_p and v_ϕ.

$$\begin{aligned} a_{jj} &= 2R^2/P^2 \sum_k (a_{0k}' - b_{0k}') \cos jk\theta, \\ b_{jj} &= 2R^2/P^2 \sum_k (a_{0k}' + b_{0k}') \cos jk\theta, \\ c_{jj} &= 2R^2/PQ \sum_k c_{0k}' \cos j(k+\tfrac{1}{2})\theta, \\ d_{jj} &= R^2/Q^2 \sum_k d_{0k}' \cos jk\theta. \end{aligned} \tag{11}$$

The Kinetic Energy Matrix

It is to be expected that the kinetic energy matrix and its reciprocal will have simple forms

for the ϕ coordinates. t_ϕ^{-1} can be calculated by using Eq. (3); namely $t_\phi^{-1}=S_\phi{}^x t_x^{-1}\mathbf{S}_\phi{}^x$ and is found to have the following form:

t_ϕ^{-1}	ϕ'	ϕ''	ϕ'''	α	β
ϕ'	A	0	0	0	0
ϕ''	0	B	C	0	0
ϕ'''	0	C	D	0	0
α	0	0	0	M_α	0
β	0	0	0	0	I_β

The submatrices A, B, C, D, M_α, M_β are all diagonal. In particular

$$M_\alpha=\begin{Vmatrix} M & 0 & 0 \\ 0 & M & 0 \\ 0 & 0 & M \end{Vmatrix}, \quad I_\beta=\begin{Vmatrix} I_x & 0 & 0 \\ 0 & I_y & 0 \\ 0 & 0 & I_z \end{Vmatrix},$$

where M is the total mass of all the atoms and I_x, I_y, I_z are the moments of inertia about the three axes. The following are the values of the diagonal terms of the other submatrices. The reciprocals of the masses m, m' are denoted by μ, μ', respectively.

$$\begin{aligned} &A_0=h^2\mu+h^2\mu' N/2, \quad A_1=A_{N-1}=h^2\mu+\mu' N/4, \\ &\qquad A_j=A_{N-j}=h^2\mu. \quad 1<j\leq N/2, \\ &B_0=\mu+h^2\mu' N/2, \quad B_1=B_{N-1}=\mu+\mu' N/4, \\ &\qquad B_j=B_{N-j}=\mu, \quad 1<j\leq N/2, \quad (12) \\ &C_j=2(1-\cos\theta)\ \mathrm{Cos}\,[j\theta/2]\mu, \\ &D_j=4(1-\cos\theta)^2\ \mathrm{Cos}^2\,[j\theta/2]\mu \\ &\qquad +4\ \mathrm{Sin}^2\,\theta\ \mathrm{Sin}^2\,[j\theta/2]\mu. \end{aligned}$$

The Values of the Frequencies

The values of λ can be obtained by solving $|\lambda 1-t_\phi^{-1}v_\phi|=0$ after utilizing the values just obtained for t_ϕ^{-1}, v_ϕ. The following linear and quadratic equations give the values of λ:

$$\begin{aligned} &\lambda_j'=A_j a_j, \quad \lambda_j''+\lambda_j'''=B_j b_j+2C_j c_j+D_j d_j, \\ &\qquad \lambda''\lambda'''=[B_jD_j-C_j^2][b_jd_j-c_j^2]. \quad (13) \end{aligned}$$

Most of the frequencies are double as shown by the relations:

$$\lambda_j'=\lambda_{N-j}', \quad \lambda_j''=\lambda_{N-j}'', \quad \lambda_j'''=\lambda_{N-j}''', \quad 0<j<N.$$

The Normal Coordinates and the Normal Vibrations

The normal coordinates n may be determined from the following equations:

$$\lambda=v_n=\mathbf{S}_\phi{}^n v_\phi S_\phi{}^n, \quad 1=t_n{}^{-1}=S_n{}^\phi t_\phi^{-1}\mathbf{S}_n{}^\phi. \quad (14)$$

From these equations the following expressions for n may be derived:

$$\begin{aligned} &0<j<N/2, \quad n_j'+in_{N-j}''=\gamma_{11,\,j}\phi_j'\sqrt{2}, \\ &n_j''+in_{N-j}''=[\gamma_{22,\,j}\phi_j''+\gamma_{23,\,j}\phi_j''']\sqrt{2}, \\ &n_j'''+in_{N-j}'''=[\gamma_{32,\,j}\phi_j''+\gamma_{33,\,j}\phi_j''']\sqrt{2}. \quad (15) \end{aligned}$$

It is to be noted that these expressions are not accurate for the cases $j=0$, $j=N/2$. In the latter cases the complex term is to be omitted from the left-hand side of the equation and the factor $\sqrt{2}$ from the right side of the equation. The constants introduced in Eqs. (15) have the following values:

$$\begin{aligned} &\gamma_j=[B_jD_j-C_j^2], \quad \gamma_{11,\,j}=[A_j]^{-\frac{1}{2}}, \\ &\gamma_{22,\,j}=[\gamma_j b_j-D_j\lambda_j''']^{\frac{1}{2}}[\gamma_j(\lambda_j''-\lambda_j''')]^{-\frac{1}{2}}, \\ &\gamma_{23,\,j}=[\gamma_j d_j-B_j\lambda_j''']^{\frac{1}{2}}[\gamma_j(\lambda_j'''-\lambda_j'')]^{-\frac{1}{2}}, \\ &\gamma_{32,\,j}=[\gamma_j b_j-D_j\lambda_j'']^{\frac{1}{2}}[\gamma_j(\lambda_j''-\lambda_j''')]^{-\frac{1}{2}}, \\ &\gamma_{33,\,j}=[\gamma_j d_j-B_j\lambda_j'']^{\frac{1}{2}}[\gamma_j(\lambda_j'''-\lambda_j'')]^{-\frac{1}{2}}. \quad (16) \end{aligned}$$

Now that the normal coordinates have been obtained one can solve for the complete transformation $S_x{}^n$ from n to x coordinates. $S_x{}^n$ gives the complete geometrical representation of the normal vibrations. Its explicit form is rather long and complex and will be omitted. One may derive from it the following qualitative results.

The vibration λ_0' is active with electrical moment parallel to the axis of symmetry; the

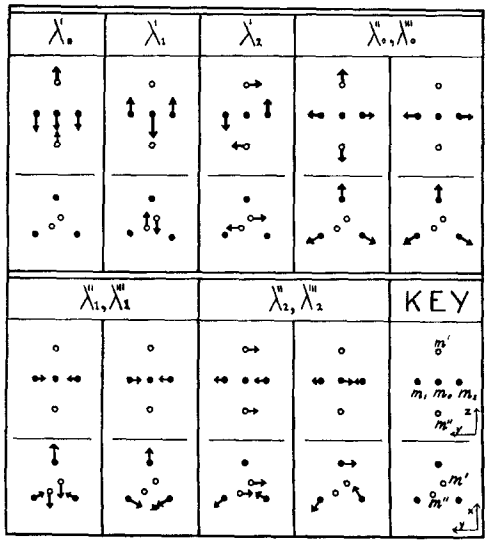

FIG. 2.

vibrations λ_1'', λ_1''', λ_{N-1}'', λ_{N-1}''' are active with electrical moment perpendicular to the axis of symmetry; the remaining vibrations are inactive.

The complete results are graphically illustrated for the molecule X_2Y_3 by Fig. 2.

The Isotope Effect

The effects due to the presence of an isotope of either one X atom or one Y atom or the simultaneous presence of both will now be considered. Let the symbol corresponding to the variation in any particular quantity be denoted by a prefixed δ and in particular let the variations of the reciprocals of the masses of the two isotopic atoms be $(\delta\mu_0)$, $(\delta\mu')$. The usual assumption will be made that v_p is not affected by the presence of isotopes. Calculation of t_ϕ^{-1} and careful examination of its subsequent usage indicates that only the variations in the elements that were previously nonzero affect the final results. The values of the following variations in the elements of t_ϕ^{-1} were obtained by recalculating t_ϕ^{-1} according to the method already presented.

$$
\begin{aligned}
&(\delta A_0)=(\delta\mu_0)h^2/N+(\delta\mu')h^2N/4,\ (\delta A_{N/2})=(\delta\mu_0)h^2/N,\ (\delta A_j)=(\delta\mu_0)2h^2/N,\ (\delta A_{N-j})=0,\ 1<j<N/2,\\
&(\delta A_1)=(\delta\mu_0)2h^2/N+(\delta\mu')N/8,\ (\delta A_{N-1})=(\delta\mu')N/8,\ (\delta B_1)=(\delta\mu_0)2/N+(\delta\mu')N/8,\ (\delta B_{N-1})=(\delta\mu')N/8,\\
&(\delta B_0)=(\delta\mu_0)/N+(\delta\mu')h^2N/4,\ (\delta B_{N/2})=(\delta\mu_0)/N,\ (\delta B_j)=(\delta\mu_0)2/N,\ \delta B_{N-j}=0,\ 1<j<N/2,\\
&(\delta C_0)=(\delta\mu_0)2(1-\cos\theta)/N,\ (\delta C_{N/2})=0,\ (\delta C_j)=(\delta\mu_0)4(1-\cos\theta)\cos[j\theta/2]/N,\ (\delta C_{N-j})=0,\ 0<j<N/2,\\
&(\delta D_0)=(\delta\mu_0)4(1-\cos\theta)^2/N,\quad (\delta D_{N/2})=(\delta\mu_0)4\sin^2\theta/N,\\
&(\delta D_j)=(\delta\mu_0)8(1-\cos\theta)^2\cos^2[j\theta/2]/N,\quad \delta D_{N-j}=(\delta\mu_0)8\sin^2\theta\sin^2[j\theta/2]/N,\quad 0<j<N/2. \qquad (17)
\end{aligned}
$$

The values of $(\delta\lambda)$ may be derived by observing the variations in the expressions for λ given by Eqs. (13). The following results are obtained:

$$
\begin{aligned}
&(\delta\lambda_j')/\lambda_j'=(\delta A_j)/A_j,\\
&(\delta\lambda_j'')+(\delta\lambda_j''')=b_j(\delta B_j)+2c_j(\delta C_j)+d_j(\delta D_j),\\
&(\delta\lambda_j'')/\lambda_j''+(\delta\lambda_j''')/\lambda_j'''=[D_j(\delta B_j)\\
&\qquad +B_j(\delta D_j)-2C_j(\delta C_j)]/[B_jD_j-C_j^2]. \qquad (18)
\end{aligned}
$$

Due to the presence of isotopes a new set of normal coordinates n' will be required. The following are the fundamental equations for the new coordinates:

$$
\begin{aligned}
&v_{n'}=\lambda+(\delta\lambda)=\mathbf{S}_n{}^{n'}v_nS_n{}^{n'},\\
&1=t_{n'}{}^{-1}=S_{n'}{}^x[t_x{}^{-1}+(\delta t_x{}^{-1})]\mathbf{S}_{n'}{}^x\\
&\qquad =S_{n'}{}^nS_n{}^x[t_x{}^{-1}+(\delta t_x{}^{-1})]\mathbf{S}_n{}^x\mathbf{S}_{n'}{}^n. \qquad (19)
\end{aligned}
$$

By making first order approximations the transformation $S_{n'}{}^n$ which expresses the new normal coordinates in terms of the old may be calculated. By utilizing this result one obtains the complete qualitative description of the isotope effect now given.

Effects due to isotope of X atom

All frequencies that were double remain so. The following frequencies shift in value. They are also the only active frequencies. λ_0', λ_0'', λ_0''' have ($\|$) moments. λ_1', λ_1'', λ_1''', λ_{N-1}', λ_{N-1}'', λ_{N-1}''' have ($\perp$) moments.

Effects due to isotope of Y atom

All frequencies become single. All frequencies with the exception of λ_j' $j>N/2$ are active and shift in value. λ_j', $j\leq N/2$ have ($\|$) moments. λ_j'' and λ_j''' have ($\perp$) moments.

Concluding Remarks

The coordinates used lend themselves readily to the introduction of various force models. Although preliminary calculations of a number of force models have been carried out, the author felt that such calculations should go hand in hand with the examination of experimental data, and accordingly has omitted these calculations, inasmuch as experimental data have not been treated in the present paper. The writer wishes to express sincere thanks to Professor E. E. Witmer for his guidance in solving the problem presented in this paper.