The *T-t* Jahn-Teller System: Solution of the Fundamental Combinatorial Problem and its Application to Moments and Zero Phonon Line

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The solution of the $T-t$ fundamental combinatorial problem is given in a purely analytic form as a sum over binomials. This simplifies other calculations considerably. For an illustration the moments of the optical absorption line shape in the strong coupling limit are derived in a closed form. Furtheron, the optical zero phonon line is calculated. An application to a $T-t$ phase transition chain is indicated.

1.) Introduction

In the treatment of triply degenerate Jahn-Teller situations ($T-t$ type) difficulties arise in the calculation of matrix elements of electronic operator products. Up to now it has not been possible to solve these integrations in a closed analytic form. The only exact description of the problem was given by Wagner [1], who calculated the matrix elements subsequently by means of a recurrence formula. But this procedure requires a great computational effort and therefore its application is restricted to a small number of problems.

The model we describe here consists of a triply degenerate high-energetic excitation (electron) and a triply degenerate low energy vibration. Both subsystems are coupled in such a way as if their degeneracy would come from a cubic point symmetry surrounding. The Hamiltonian of this problem is of the form [2]

$$H = \Omega \sum_{i=1}^{3} a_i^+ a_i + \omega \sum_{i=1}^{3} b_i^+ b_i + \varkappa \sum_{i=1}^{3} (a_i^+ a_{i+1} + a_{i+1}^+ a_i)(b_{i+2}^+ + b_{i+2}) . \quad (1)$$

The b_i^+, b_i are oscillator creation- and annihilation operators, whereas the a_i^+, a_i may either be taken as vibrational or as excitonic creation and annihilation operators.

In the present article we demonstrate the possibility to solve the integrations in the electronic subsystem using combinatorial techniques. In the same way as in the $E-e$ Jahn-Teller system we are confronted with a specific fundamental combinatorial problem, which, however, in the present case is of a much more complicated nature. Our result is compared with the numerical values derived from the recurrence formula [1]. Moreover, the result is chequed by a second and independent method. For an application the moments of the optical absorption line shape in the strong coupling limit are derived. Furtheron, the zero phonon line of the spectrum is calculated.

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2.) The Fundamental Combinatorial Problem of the *T-t* Jahn-Teller System

The fundamental combinatorial problem arises by treating matrix elements in the high-energetic subsystem. Its structural form reads

$$\langle 0\, a_j^+ \,|\, [\alpha \sum_{i=1}^{3} (a_i^+ a_{i+1} + a_{i+1}^+ a_i)]^n \,|\, a_k^+\, 0 \rangle . \quad (2)$$

We will confine ourselves to elements between equal electron states, e. g. $j=k$, and without loss of generality we set $j=k=1$. The generalisation to arbitrary j, k provides no difficulties.

We handle this problem in a similar way as given in the paper by Wagner [1]. There, the electronic integrations of the $E-e$ J.-T. system have been solved. In analogy to this work the calculation of the expectation value (2) leads to a combinatorial problem, which may be depicted in a diagram. In Fig. 1 all elements of a general diagram are drawn.

Only these combinations of operators give nonvanishing terms, in which the attached arrows form closed pathes, beginning and ending on line 1. In Fig. 1 one possible path is drawn. μ_i and $\bar{\mu}_i$ ($i=1, 2, 3$) are the numbers, how often the different elements

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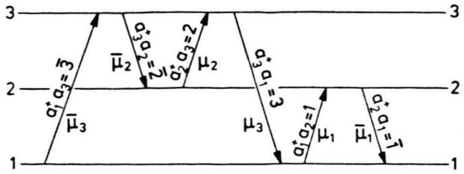

Fig. 1. All possible elements arising by the integration in the electronic subsystem of the $T-t$ J.-T. case, characterized by different arrows. μ_i and $\bar{\mu}_i$ $(i=1, 2, 3)$ count the number of appearence of the different elements in the operator product.

$$a_1^+ a_2 = 1, \quad a_2^+ a_3 = 2, \quad a_3^+ a_1 = 3,$$
$$a_2^+ a_1 = \overline{1}, \quad a_3^+ a_2 = \overline{2}, \quad a_1^+ a_3 = \overline{3}$$

appear in each single product term in Expr. (2) (Figure 1). The $\bar{\mu}_i$ count the inverse processes.

The number of elements ending on one distinct line must be equal to the number of elements beginning on the same line. This leads to the relation

$$\mu_1 - \bar{\mu}_1 = \mu_2 - \bar{\mu}_2 = \bar{\mu}_3 - \mu_3 .$$

We now construct all possible, closed pathes, beginning and ending on line 1, if each of the numbers μ_i, $\bar{\mu}_i$ has a fixed value. To this end we form an arbitrary sequence of the elements 1, $\overline{1}$, 3, $\overline{3}$, as shown in the diagram of Figure 2.

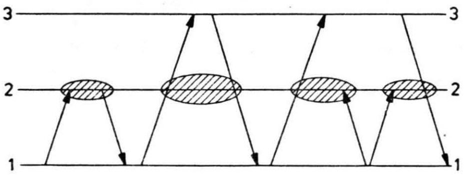

Fig. 2. Sites on line 2, where the $\mu_2 + \bar{\mu}_2$ elements $a_2^+ a_3 = 2$ and $a_3^+ a_2 = \overline{2}$ must be located.

These elements form $M = \frac{1}{2}(\mu_1 + \bar{\mu}_1 + \mu_3 + \bar{\mu}_3)$ sites on line 2, where the $\mu_2 + \bar{\mu}_2$ elements 2 and $\overline{2}$ may join the others. Since the elements 3, $\overline{3}$ connect line 1 and 3, we may think that element 3 is represented by the addition of elements 1 and 2 and element $\overline{3}$ by $\overline{1} + \overline{2}$. That means in the same way as 1, $\overline{1}$ the elements 3, $\overline{3}$ build up the sites on line 2. However, at the same time they also form a connection arrow from line 2 to line 3 like the elements 2, $\overline{2}$. So we have to distribute all the elements 2, $\overline{2}$ and 3, $\overline{3}$ onto the M sites on line 2. But not all of these elements can be placed at the sites $0, 1, 2, \ldots, M$, since at least $N = \frac{1}{2}(\mu_3 + \bar{\mu}_3)$ sites must be occupied by the elements 3, $\overline{3}$. Here we have to add a typ-distinction. $\mu_3 + \bar{\mu}_3$ is either even or odd valued. To define the quantity N unique, we set

$$\mu_3 + \bar{\mu}_3 = \text{even}: \quad N = \tfrac{1}{2}(\mu_3 + \bar{\mu}_3); \tag{3 a}$$
$$\mu_3 + \bar{\mu}_3 = \text{odd}: \quad N = \tfrac{1}{2}(\mu_3 + \bar{\mu}_3 + 1). \tag{3 b}$$

We now have to solve the first combinatorial problem, which may be stated in the following form: What is the number of possibilities to pick out $N + \eta$ sites of M ones, where η runs from 0 to $(M-N)$? The answer is well-known from combinatorial analysis (see e. g. Korn and Korn [3], p. 896); it is

$$Z_1 = \binom{M}{N+\eta}. \tag{4}$$

Onto these $N + \eta$ sites we distribute the elements 2, $\overline{2}$, 3, $\overline{3}$ in pairs. The number of elements at each site is $2, 4, \ldots \{(\mu_2 + \bar{\mu}_2 + \mu_3 + \bar{\mu}_3) - 2(N+\eta)\}$, and so the number of distinguishable arrangements is given by [3]

$$Z_2 = \binom{\frac{1}{2}(\mu_2 + \bar{\mu}_2 + \mu_3 + \bar{\mu}_3) - 1}{N + \eta - 1}. \tag{5}$$

However, we have to distinguish between the elements 2, $\overline{2}$ and 3, $\overline{3}$. For the elements 3, $\overline{3}$ there are exact $2(N+\eta)$ connecting points, and we have to count all possibilities arising, when we distribute the elements 3, $\overline{3}$ onto these points. The solution of this combinatorial problem reads [3]

$$Z_3 = \binom{2N + 2\eta}{\mu_3 + \bar{\mu}_3}. \tag{6}$$

With help of these considerations we can determine the total number of possibilities constructing closed pathes in our diagram beginning and ending on line 1. We get

$$Z_{\text{tot}} = \sum_{\eta=0}^{M-N} Z_1 Z_2 Z_3 = \sum_{\eta=0}^{M-N} \binom{M}{N+\eta} \binom{2N+2\eta}{\mu_3 + \bar{\mu}_3} \cdot \binom{\frac{1}{2}(\mu_2 + \bar{\mu}_2 + \mu_3 + \bar{\mu}_3) - 1}{N + \eta - 1}. \tag{7}$$

It is easy to verify that only the combinations $\mu_i + \bar{\mu}_i$ = even for all i, and $\mu_i + \bar{\mu}_i$ = odd for all i, can occur. If we introduce the abbreviations

$$\mu_i + \bar{\mu}_i = \text{even} = 2x_i \quad (i = 1, 2, 3) \tag{8 a}$$
$$\mu_i + \bar{\mu}_i = \text{odd} = 2x_i + 1 \tag{8 b}$$

and use the relations (3 a, b) we get by separating into even and odd terms the result

$$Z_{\text{tot}}^e(x_1, x_2, x_3) = \sum_{\eta=0}^{K} \binom{x_1 + x_3}{x_3 + \eta} \binom{x_2 + x_3 - 1}{x_3 + \eta - 1} \cdot \binom{2x_3 + 2\eta}{2x_3} \tag{9 a}$$

Table 1. Values of the combinatorial solution $Z^{e}_{\text{tot}}(x_1, x_2, x_3)$ for some different arguments. Explanation see text.

$n = 2(x_1+x_2+x_3)$	x_1	x_2	x_3	$Z^{e}_{\text{tot}}(x_1, x_2, x_3)$
2	1	0	0	1
	0	1	0	0
	0	0	1	1
4	2	0	0	1
	0	2	0	0
	0	0	2	1
	1	1	0	1
	1	0	1	2
	0	1	1	1
6	3	0	0	1
	0	3	0	0
	0	0	3	1
	2	1	0	2
	2	0	1	3
	1	2	0	1
	1	0	2	3
	0	2	1	1
	0	1	2	2
	1	1	1	8
8	4	0	0	1
	0	4	4	0
	0	0	4	1
	3	1	0	3
	3	0	1	4
	1	3	0	1
	1	0	3	4
	0	3	1	1
	0	1	3	3
	2	2	0	3
	2	0	2	6
	0	2	2	3
	2	1	1	21
	1	2	1	14
	1	1	2	21
10	5	0	0	1
	0	5	0	0
	0	0	5	1
	4	1	0	4
	4	0	1	5
	1	4	0	1
	1	0	4	5
	0	4	1	1
	0	1	4	4
	3	2	0	6
	3	0	2	10
	2	3	0	4
	2	0	3	10
	0	3	2	4
	0	2	3	6
	3	1	1	40
	1	3	1	20
	1	1	3	40
	2	2	1	54
	2	1	2	72
	1	2	2	54

for the even part and

$$Z^{0}_{\text{tot}}(x_1, x_2, x_3) = \sum_{\eta=0}^{K} \binom{x_1+x_3+1}{x_3+\eta+1} \binom{x_2+x_3}{x_3+\eta} \cdot \binom{2x_3+2\eta+2}{2x_3+1} \tag{9 b}$$

for the odd part. K is the upper border of η and defined by:

$$K = \begin{cases} x_1 & \text{for } x_1 < x_2 \\ x_2 & \text{for } x_2 > x_1 \end{cases} . \tag{10}$$

In Table 1 values of $Z^{e}_{\text{tot}}(x_1, x_2, x_3)$ are given for some different arguments. They coincide with the results of Wagner [1] derived by successive calculations of the recurrence formula.

These combinatorial results allow for the quantummechanical calculation of nontrivial three-level problems in an analytic way. Because of the complicated nature of the considered system the final solution is given in the form of a sum over binomial coefficients, which up to now we have not been able to sum up. Nevertheless, the combinatorial result simplifies the calculations considerably and cuts down the computational effort.

3.) Test of the Combinatorial Result

The result of Sect. 2 allows the calculation of arbitrary and complicated matrix elements in a three level system. Since there are also some cases where direct calculations are possible, the derived results can be chequed.

To do so we transform the Hamiltonian (1) by an exponential transformation

$$\tilde{H} = e^{-S} H e^{+S} = H + [H, S] + \tfrac{1}{2}[[H, S], S] + \dots,$$

where the exponent should be given by

$$S = \{\sum_{i=1}^{3} (a_i^{+} a_{i+1} + a_{i+1}^{+} a_i)\} B . \tag{11}$$

B is an imaginary coefficient, the exact structure of which is of no relevance in this context. Since S is antihermitian, the transformation is unitary.

In the following we treat the matrix element of the transformed operator $\tilde{a}_1^{+} = e^{-S} a_1^{+} e^{S}$ between the states $|a_1^{+} 0\rangle$ and $|0\rangle$. We get

$$\langle 0\, a_1^{+} | \tilde{a}_1^{+} | 0 \rangle = \langle 0\, a_1^{+} | e^{-S} a_1^{+} e^{S} | 0 \rangle , \tag{12}$$

and the relation $e^{S}|0\rangle = |0\rangle$ leads us to the identity

$$\langle 0\, a_1^{+} | \tilde{a}_1^{+} | 0 \rangle \equiv \langle 0\, a_1^{+} | e^{-S} | a_1^{+} 0 \rangle . \tag{13}$$

This matrix element can be solved in two different ways.

a) First we use the result of Section 2. Therefore we expand the exponentialoperator on the right-hand side of Eq. (13) and get

$$\langle 0\, a_1^+ | e^{-S} | a_1^+ 0 \rangle = \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \langle 0\, a_1^+ | S^n | a_1^+ 0 \rangle\,, \tag{14}$$

and with definition (11):

$$\sum_{n=0}^{\infty} \frac{1}{n!} B^n \langle 0\, a_1^+ | [\sum_{i=1}^{3} (a_i^+ a_{i+1} + a_{i+1}^+ a_i)]^n | a_1^+ 0 \rangle\,. \tag{15}$$

With the result of Sect. 2 we arrive at ($x_1 + x_2 + x_3 = n$)

$$\langle 0\, a_1^+ | e^{-S} | a_1^+ 0 \rangle = \sum_{n=0}^{\infty} \frac{B^n}{n!} \sum_{x_1, x_2, x_3}^{n} Z_{\text{tot}}(x_1, x_2, x_3)\,. \tag{16}$$

b) Second we solve the matrix element by calculating the transformed operator $\tilde{a}_1^+$. For an arbitrary $\tilde{a}_i^+$ ($i = 1, 2, 3$) we get the expansion

$$\tilde{a}_i^+ = e^{-S} a_i^+ e^S = a_i^+ + [a_i^+, S] + \frac{1}{2!} [[a_i^+, S], S] + \dots \tag{17}$$

The first commutator reads

$$[a_i^+, S] = (a_{i+2}^+ + a_{i+1}^+) B\,. \tag{18}$$

For an arbitrary order n one can write

$$[\dots[a_i^+, S], S \dots]_n = a_i^+ C_i^{(n)} + a_{i+1}^+ C_{i+1}^{(n)} + a_{i+2}^+ C_{i+2}^{(n)} \tag{19}$$

and for the next order

$$[[\dots]_n, S] = a_i^+ (C_{i+1}^{(n)} + C_{i+2}^{(n)}) B + a_{i+1}^+ (C_i^{(n)} + C_{i+2}^{(n)}) B + a_{i+2}^+ (C_i^{(n)} + C_{i+1}^{(n)}) B\,. \tag{20}$$

This leads to the recurrence formulas:

$$C_i^{(n+1)} = (C_{i+1}^{(n)} + C_{i+2}^{(n)}) B\,; \tag{21 a}$$

$$C_{i+1}^{(n+1)} = (C_i^{(n)} + C_{i+2}^{(n)}) B\,; \tag{21 b}$$

$$C_{i+2}^{(n+1)} = (C_i^{(n)} + C_{i+1}^{(n)}) B\,. \tag{21 c}$$

Combining these expressions we get the following difference equation

$$C_j^{(n+2)} - B\, C_j^{(n+1)} - 2 B^2 C_j^{(n)} = 0\,,\ j = i, i+1, i+2\,. \tag{22}$$

The solution of this equation is found to be

$$C_j = \alpha_j e^{2B} + \beta_j e^{-B}\,, \tag{23}$$

where α_j and β_j are arbitrary constants. Then the transformed operator (17) reads

$$\tilde{a}_i^+ = a_i^+ \{\alpha_i e^{2B} + \beta_i e^{-B}\} + a_{i+1}^+ \{\alpha_{i+1} e^{2B} + \beta_{i+1} e^{-B}\} + a_{i+2}^+ \{\alpha_{i+2} e^{2B} + \beta_{i+2} e^{-B}\}\,. \tag{24}$$

The α_j and β_j ($j = i, i+1, i+2$) can be determined, if we expand Expr. (24) in powers of B:

$$\tilde{a}_i^+ = a_i^+ \{\alpha_i + \beta_i + B(2\alpha_i - \beta_i) + \dots\} + a_{i+1}^+ \{\alpha_{i+1} + \beta_{i+1} + B(2\alpha_{i+1} - \beta_{i+1}) + \dots\} + a_{i+2}^+ \{\alpha_{i+2} + \beta_{i+2} + B(2\alpha_{i+2} - \beta_{i+2}) + \dots\}\,. \tag{25}$$

This result must be equal to the direct expansion of the commutator series (17):

$$\tilde{a}_i^+ = a_i^+ \{1 + O(B^2)\} + a_{i+1}^+ \{B + O(B^2)\} + a_{i+2}^+ \{B + O(B^2)\}\,. \tag{26}$$

Comparing equal orders of B in both expansions we get for the constants

$$\alpha_i = \tfrac{1}{3}\,,\quad \alpha_{i+1} = \tfrac{1}{3}\,,\quad \alpha_{i+2} = \tfrac{1}{3}\,,$$
$$\beta_i = \tfrac{2}{3}\,,\quad \beta_{i+1} = -\tfrac{1}{3}\,,\quad \beta_{i+2} = -\tfrac{1}{3}\,. \tag{27}$$

Thus the transformed operator $\tilde{a}_i^+$ is determined uniquely. For $i = 1$ it has the form

$$\tilde{a}_1^+ = \tfrac{1}{3}(a_1^+ + a_2^+ + a_3^+) e^{2B} + \tfrac{1}{3}(2 a_1^+ - a_2^+ - a_3^+) e^{-B}\,, \tag{28}$$

and the matrix element (14) is given by

$$\langle 0\, a_1^+ | \tilde{a}_1^+ | 0 \rangle = \langle 0\, a_1^+ | \tfrac{1}{3}(a_1^+ + a_2^+ + a_3^+) e^{2B} + \tfrac{1}{3}(2 a_1^+ - a_2^+ - a_3^+) e^{-B} | 0 \rangle = \tfrac{1}{3} e^{2B} + \tfrac{2}{3} e^{-B}\,. \tag{29}$$

Both solutions [Eqs. (16) and (29)] must be equal,

$$\tfrac{1}{3} e^{2B} + \tfrac{2}{3} e^{-B} = \sum_{n=0}^{\infty} \frac{1}{n!} B^n \{\tfrac{1}{3}[2^n + 2(-1)^n]\} = \sum_{n=0}^{\infty} \frac{1}{n!} B^n \sum_{x_1, x_2, x_3}^{n} Z_{\text{tot}}(x_1, x_2, x_3)\,, \tag{30}$$

and we get the result ($x_1 + x_2 + x_3 = n$)

$$\sum_{x_1, x_2, x_3}^{n} Z_{\text{tot}}(x_1, x_2, x_3) = \tfrac{1}{3} \{2^n + 2(-1)^n\}\,. \tag{31}$$

Table 2. Result of the summed combinatorial solution ($T-t$ case) as derived by an independent method (see text).

n	$\frac{1}{3}\{2^n+2(-1)^n\}$	n	$\frac{1}{3}\{2^n+2(-1)^n\}$
0	1	1	0
2	2	3	2
4	6	5	10
6	22	7	63
8	86	9	170
10	342	11	682

In Table 2 we have written down the result for the terms up to $n = 11$.

To compare the results of the preceeding section with these of Table 2, the numbers in the 3. and 6. column of Table 1 have to be summed up for each distinct n. The results coincide.

4.) Moments of the Optical Absorption Spectrum in the Strong Coupling Limit

The optical absorption line shape can be characterised by its moments. Following the notation in the paper of Wagner [1] in the strong coupling limit they are defined by

$$M_m = (1-\lambda)^3 \sum_{n_1, n_2, n_3}^{\infty} \lambda^{n_1+n_2+n_3} \langle n_1 n_2 n_3 a_1^+ | [\varkappa \sum_{i=1}^{3} (a_i^+ a_{i+1} + a_{i+1}^+ a_i)(b_{i+2}^+ + b_{i+2})]^m | a_1^+ n_1 n_2 n_3 \rangle . \quad (32)$$

With the result of Sect. 2 the integration over the electronic part can be done ($x_1+x_2+x_3=m$)

$$M_m = (1-\lambda)^3 \sum_{n_1, n_2, n_3} \lambda^{n_1+n_2+n_3} \varkappa^m \sum_{x_1, x_2, x_3}^{m} Z_{\text{tot}}^e(x_1, x_2, x_3) \prod_{i=1}^{3} \langle n_i | (b_i^+ + b_i)^{2x_i} | n_i \rangle . \quad (33)$$

For odd values of m this expression vanishes, because the matrix elements with respect to the phonon coordinates vanish, whereas for even values of m they get the form

$$\langle n | (b^+ + b)^{2m} | n \rangle = (-1)^m \binom{2m}{m} \sum_{\nu=0}^{m} \binom{m}{\nu} \nu! (\tfrac{1}{2})^\nu n(n-1) \ldots [n-(m-\nu+1)] . \quad (34)$$

This expression is derived in Refs. [1] and [4]. The sums over n_1, n_2, n_3 may be performed by use of the identity

$$\sum_{n=0}^{\infty} \lambda^n n(n-1) [n-((\mu-\nu)-1)] = \lambda^{(\mu-\nu)} \left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^{(\mu-\nu)} \sum_{n=0}^{\infty} \lambda^n = \frac{1}{1-\lambda} \left(\frac{\lambda}{1-\lambda}\right)^{\mu-\nu} (\mu-\nu)! . \quad (35)$$

Combining Eqs. (34) and (35) we arrive at

$$(1-\lambda) \sum_{n=0}^{\infty} \lambda^n \langle n | (b^+ + b)^{2\mu} | n \rangle = \left(\frac{1+\lambda}{2(1-\lambda)}\right)^{\mu} \frac{(2\mu)!}{\mu!} . \quad (36)$$

If we insert this result into formula (33) and use the explicite form of the combinatorial relation (9 a) the moments in the strong coupling limit may be written as

$$M_{2m} = \varkappa^{2m} \left(\frac{1+\lambda}{2(1-\lambda)}\right)^m \sum_{x_1 x_2 x_3}^{m} \frac{(2x_1)!}{x_1!} \frac{(2x_2)!}{x_2!} \frac{(2x_3)!}{x_3!} \sum_{\eta} \binom{x_2+x_3}{x_3+\eta} \binom{x_1+x_3-1}{x_3+\eta-1} \binom{2x_3+2\eta}{2x_3} \quad (37\text{ a})$$

$$= \varkappa^{2m} \left(\frac{1+\lambda}{2(1-\lambda)}\right)^m \sum_{x_1 x_2 x_3}^{m} \frac{(2x_1)!}{x_1!} \frac{(2x_2)!}{x_2!} \frac{(2x_3)!}{x_3!} Z_{\text{tot}}^e(x_1, x_2, x_3)$$

and

$$M_{2m+1} = 0 . \quad (37\text{ b})$$

For simplicity we introduce the abbreviation

$$\overline{M}_m = M_m \left[\frac{1+\lambda}{1-\lambda} \varkappa^2\right]^{-m/2} . \quad (38)$$

Table 3. The first 21 values of $\overline{M}_m$, which determine the exact moments of the optical $T-t$ J.-T. system in the strong coupling limit (see text).

m	$\overline{M}_m$	m	$\overline{M}_m$
0	1	11	0
1	0	12	$1.31706 \cdot 10^5$
2	2	13	0
3	0	14	$2.28249 \cdot 10^6$
4	$1.0 \cdot 10^1$	15	0
5	0	16	$4.54456 \cdot 10^7$
6	$7.4 \cdot 10^1$	17	0
7	0	18	$1.02328 \cdot 10^9$
8	$7.26 \cdot 10^2$	19	0
9	0	20	$5.728 \cdot 10^{10}$
10	$8.91 \cdot 10^3$	21	0

In Table 3 the quantity $\overline{M}_m$ for the first 20 exact moments in the strong coupling limit is tabulated.

5.) The Zero Phonon Line of the Optical Absorption Spectrum

As a further example of the applicability of our combinatorial results we calculate the zero phonon line of the optical absorption function. This will be done by the exponential transformation method. In the case of the $T-t$ J.-T. system the exponential operator reads

$$S = -\left(\frac{\varkappa}{\omega}\right) \sum_{i=1}^{3} (a_i^+ a_{i+1} + a_{i+1}^+ a_i)(b_{i+2}^+ - b_{i+2}) . \quad (39)$$

This operator is derived and discussed in Refs. [2] and [5]. In Ref. [2] the zero and one phonon lines are calculated by solving the combinatorial problems using the recurrence formula of Wagner [1], but the calculations lead to very laborious numerical treatments.

Refering to Ref. [2] the intensity of the zero phonon line is given by ($T=0$ °K)

$$I_0 = |\langle 0\, a_1^+ | e^{-S} | a_1^+ 0 \rangle|^2 . \tag{40}$$

Inserting the operator S we get the matrix element

$$\langle 0\, a_1^+ | e^{-S} | a_1^+ 0 \rangle = \sum_{n=0}^{\infty} \frac{1}{(2n)!} \left(\frac{\varkappa}{\omega}\right)^{2n} \langle 0\, a_1^+ | [\sum_{i=1}^{3} (a_i^+ a_{i+1} + a_{i+1}^+ a_i)(b_{i+2}^+ - b_{i+2})]^{2n} | a_1^+ 0 \rangle , \tag{41}$$

which have nearly the same form as expression (32). In a completely analogous way as in Sect. 4 matrix element (41) leads to ($x_1 + x_2 + x_3 = n$)

$$\sum_{n=0}^{\infty} \frac{1}{(2n)!} \left(\frac{\varkappa}{\omega}\right)^{2n} (-1)^n \left(\frac{1}{2}\right)^n \sum_{x_1 x_2 x_3}^{n} \frac{(2x_1)!}{x_1!} \frac{(2x_2)!}{x_2!} \frac{(2x_3)!}{x_3!} Z_{\text{tot}}^e(x_1, x_2, x_3) . \tag{42}$$

Using this result and the expression for the moments of the optical absorption function in the strong coupling limit ($T=0$ °K) the intensity of the zero phonon line can be written as

$$I_0 = \left| \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n)!} \left(\frac{\varkappa}{\omega}\right)^{2n} \overline{M}_{2n} \right|^2 . \tag{43}$$

In this way the zero phonon line of the intermediate and small coupling region is related to the strong coupling momenta. In Fig. 3 the zero phonon line is drawn as a function of the coupling parameter.

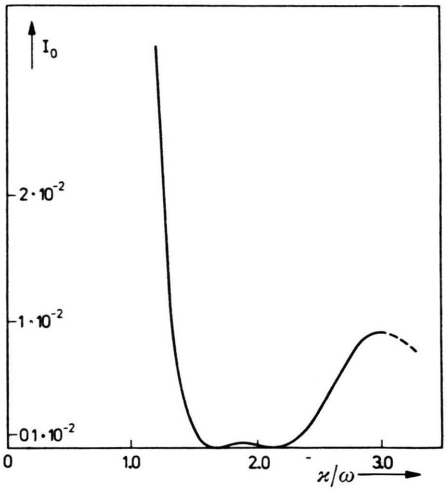

Fig. 3. Dynamical resonance effect of the zero-phonon line intensity in the dynamical $T-t$ J.-T. system. I_0 is the intensity of the zero-phonon line and $\varkappa/\omega$ is a measure for the coupling strength.

It shows the characteristics of resonant systems. If the coupling strength is varied, the line undergoes two resonant dips. For great interaction values the intensity asymptotically approaches zero. The same result was derived in Ref. [4] by a much greater computational effort.

6.) Summary and Discussion

In the quantum mechanical treatment of a high-energetic three level system interacting with a sequence of low-energetic oscillators the calculation of matrix elements in the high-energetic subsystem in all non-trivial cases is very difficult. One example of such systems is that of a Jahn-Teller system of species $T-t$, which we have discussed in some detail. After a step by step combinatorial argumentation we are able to derive a general solution for all different matrix elements. The result is given in the form of a sum over three binomial coefficients. Up to now we could not remove the remaining single summation, because of the complicated structure of the binomials. Nevertheless, the solution of the combinatorial problem simplifies our treatments considerably, for we now are able to do the calculations with considerably reduced effort.

As an illustration we have calculated the moments of the optical absorption line shape in the strong coupling limit. The exact moments are given in an analytic form of two combined sums. Furtheron, the optical zero phonon line is derived and compared with earlier results.

The solution of the combinatorial treatment is also applicable to the calculation of the phase transition behaviour of a chain of $T-t$ Jahn-Teller molecules. As will be shown in the near future, it allows a first step of an approximative transfer matrix description, which can be summed in a closed form. In a similar way, extended cluster approximations can be given.

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