Lifting the hindrance of absorption in strong light fields and excitation of polyatomic molecules

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(Submitted 7 June 1978)
Zh. Eksp. Teor. Fiz. 76, 46–53 (January 1979)

It is shown that intense radiation is effectively absorbed by vibrational-rotational transitions that are forbidden in the rotational quantum number in the linear approximation. Lifting the hindrance on the transitions with Δj from ± 2 to ± 6 is due to single-photon absorption of third and fifth orders, two-photon absorption of fourth and sixth orders, and three-photon absorption of fifth order, etc. The characteristic intensities of the radiation that saturates the corresponding transitions are of the order of $(1\text{–}3)\times 10^7$ W/cm^2 for SF_6 and BCl_3. At these intensities, such processes lead to capture of a large number of rotational states of the molecules that absorb IR radiation at a frequency that does not satisfy the condition of two-photon and three-photon resonance in the Q branches of the absorption band. The influence of the lifting of the hindrances on the characteristics of multistep excitation of molecules—selectivity and long-wave shift of the absorption band—is discussed. Similar hindrance-lifting effects are possible for transitions between components of the fine structure of electronic levels, and also for Raman scattering on rotational transitions.

PACS numbers: 33.80.Kn, 33.10.Ev, 33.20.Ea

Interest in processes of absorption of strong fields on vibrational-rotational transitions of polyatomic molecules is due to the prospects of using laser radiation for multistep photodissociation of molecules, for the control of chemical reactions, and for isotope separation.[1-3] There is still much which is not clear in the physics of the absorption of high-power radiation by complex molecules. This pertains primarily to molecule transitions between low-lying vibrational states, which determine in many respects the characteristics of the photoexcitation.[1] An essential feature of these transitions in a strong IR field is the capture of many rotational states, a fact experimentally established in Ref. 3. Theoretical calculations have shown that one of the causes of the capture may be two- or three-photon resonance, which takes place for an appreciable fraction of the molecules in a rather wide frequency range.[1]

Experiments in which multistep excitation of molecules is used are presently being carried at rather appreciable radiation power densities, from 1 MW/cm^2 to 1 GW/cm^2.[3-5] Under these conditions other effects, besides ordinary two- and three-photon absorption, are possible and lead to capture of many rotational states, and influence the photoexcitation characteristics such as the absorbed power and the selectivity.

We consider in this paper one such effect, hitherto not discussed, in the literature, namely the lifting, by a strong field, of the hindrance of rotational-vibrational transitions that are forbidden in the rotational quantum number j. We shall show that the lifting of the forbiddenness of the one-photon absorption (OPA) constitutes an aggregate of odd-photon (starting with three-photon) processes, the elementary acts of which cause the absorption of one photon (see Figs. 1a and 1b). We shall therefore call these processes, respectively, one-photon absorption of third, fifth, etc. order (OPA-3, OPA-5, etc. The corresponding amplitude absorption coefficients are proportional respectively to $|\mathscr{E}|^2\mathscr{E}$, $|\mathscr{E}|^4\mathscr{E}$, etc.). The lifting of the forbiddenness of two-photon absorption (TwPA) is due to even-photon processes (Figs. 1c and 1d), in the elementary acts of which two photons are absorbed (TwPA of fourth, sixth, etc. orders; TwPA-4, TwPA-6, etc.). Analogously, besides the ordinary three-photon absorption (ThPA), it is possible to have ThPA-5, ThPA-7, etc. (Fig. 1e).

It will be shown below that even at moderate intensities, ~10^7 W/cm^2, the processes OPA-3 and OPA-5 allow vibrational-rotational transitions with $\Delta j=(\pm 2)-(\pm 5)$, TwPA-4 and TwPA-6 allow two-photon transitions with $\Delta j=(\pm 3)-(\pm 6)$, and when ThPA-5 is taken into account, three-photon transitions with $\Delta j=\pm 4, \pm 5$ become possible. It is obvious that all the indicated processes lead to a rather appreciable increase in the number of rotational states (and consequently in the number of molecules distributed among them), which are captured, when the lower vibrational states are excited by a strong IR radiation. They can therefore influence significantly the absorption of radiation and the selectivity of molecule excitation.

We investigate below the influence of OPA-3 and OPA-5, TwPA-4 and TwPA-6, as well as ThPA-5 on the vibrational-rotational absorption spectrum of polyatomic molecules, and discuss the role of these processes in the multistep excitation of molecules.

1. LIFTING OF HINDRANCES ON ONE-PHOTON ABSORPTION

Assume that the resonance condition $\omega_1=\omega_{21}+\Delta$ is satisfied for a certain transition 1–2. Then, obviously, the conditions

$$2\omega_1-\omega_1=\omega_{21}+\Delta, \quad 3\omega_1-2\omega_1=\omega_{21}+\Delta \tag{1}$$

etc. are satisfied for the same transitions, and are the conditions for the resonances of third, fifth, etc. order.

In accordance with the procedure developed in Ref. 6 for the description of multiphoton processes, the matrix element of the energy of interaction of the field with the resonant transition 1–2 can be obtained in a given concrete case in the form¹⁾

$$V_{12}=\sum_q V_{12}^{(q)},$$

where q is the order of the resonance,

$$V_{12}^{(q)}=\sum_a \varkappa^{12}_{a_1a_2\ldots a_q}|\mathscr{E}_a|^{q-1}\mathscr{E}_a, \tag{2}$$

where $\mathscr{E}_a$ are the components of the field amplitude, $\varkappa^{12}_{a1a2\ldots a_q}=\varkappa_{(q)\mathrm{OPA}}$ is the polarizability tensor of q-th order for the resonant transition:

$$\varkappa_{(q)\mathrm{OPA}}=\hbar^{(1-q)}\sum_{k_1k_2\ldots k_{q-1}}\hat{C}\begin{pmatrix}1,-1,1\ldots,1\\ a,\ a,\ a,\ldots,a\end{pmatrix}$$
$$\times\frac{(d_a)_{1k_1}(d_a^*)_{k_1k_2}\ldots(d_a)_{k_{q-1}2}}{(\omega_{1k_1}+\omega_1)(\omega_{1k_2}+\omega_1+\omega_{-1})\ldots(\omega_{1k_{q-1}}+\omega_1+\omega_{-1}+\ldots+\omega_1)}, \tag{3}$$

and $\varkappa_a\equiv d_{12}$ is the dipole moment of the transition 1–2. In (3),

$$\hat{C}\begin{pmatrix}\alpha,\ \beta,\ \gamma,\ldots\\ a_\alpha,a_\beta,a_\gamma\ldots\end{pmatrix}$$

is the operator of all the possible simultaneous permutations of the frequency indices ²⁾ [in this case −1 and 1, inasmuch as only the frequencies ω_1 and $-\omega_1=\omega_{-1}$ take part in the resonances (1)] and of the coordinates a.

We shall show that for vibrational-rotational transitions the polarizabilities (3) have terms with small denominators, of the order of the rotational frequencies, as a results of which these terms increase sharply. By way of example we consider the transitions $|v=0,j\rangle\rightarrow|v=1,j\pm3\rangle$, which are forbidden in the linear approximation. We recognize that the frequencies of the P and R branches of the vibrational-rotational transitions are equal to $\nu_{01}-v\nu_a\pm2Bj$, where B is the rotational constant (in cm^{-1}) and ν_a is the anharmonicity constant, and assume that there is no two-photon resonance between the radiation frequency and the $|0,j\rangle\rightarrow|2,j\pm2\rangle$ transition. We can then obtain from (3)

$$\varkappa_{(3)\mathrm{OPA}}\approx\frac{d_{0,j}^{1,j\pm1}}{4Bj(2\pi c\hbar)^2}\left\{\frac{d_{1,j\pm1}^{0,j\pm2}d_{0,j\pm2}^{1,j\pm3}}{4Bj}+\frac{d_{1,j\pm1}^{2,j\pm2}d_{2,j\pm2}^{1,j\pm3}}{\nu_a\pm8Bj}\right\}, \tag{4}$$

where $d_{v_1,j_1}^{v_2,j_2}=\langle v_1,j_1|d_a|v_2,j_2\rangle$; we have left out of the curly brackets in (4) the terms with denominators of the order $\nu_1=(\omega_1/2\pi)\gg2Bj$.

We can analogously calculate the polarizability $\varkappa_{(5)\mathrm{OPA}}$, which is responsible for the one-photon absorption of fifth order, etc. The complete expressions for the polarizabilities of higher order are cumbersome. We present therefore order-of-magnitude estimating expressions. For one-photon absorption of fifth order one can use

$$\varkappa_{(5)\mathrm{OPA}}\approx10^{-2}(d_{0,1}^{1,j\pm1})^5/(2\pi c\hbar Bj)^4. \tag{5}$$

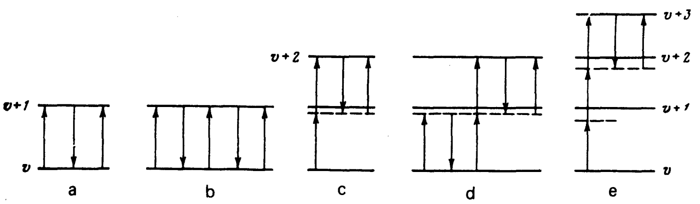

FIG. 1. Typical schemes of OPA, TwPa, and ThPA elementary acts of higher order.

2. LIFTING THE HINDRANCES OF TwPA and ThPA

If two-photon resonance $2\omega_1=\omega_{21}+\Delta$ obtains for some transition 1–2, then resonances of fourth, sixth, etc. orders exist for the same transition (Figs. 1c and 1d):

$$3\omega_1-\omega_1=\omega_{21}+\Delta,\quad 4\omega_1-2\omega_1=\omega_{21}+\Delta;$$

as a result of the elementary acts of the corresponding multiphoton processes, absorption of two photons takes place and the transitions with $\Delta j=\pm3,\pm4$ and $\Delta j=(\pm3)-(\pm6)$ respectively are no longer forbidden. The total energy of the interaction of the field with the resonant transition, in analogy with OPA, is $V_{12}=\sum_q V_{12}{}^{(q)}$, where

$$V_{12}^{(q)}=\sum_a\varkappa_{(q)\mathrm{TwPA}}|\mathscr{E}_a|^{(q-2)}\mathscr{E}_a^2. \tag{6}$$

The expressions for the polarizabilities of the TwPA of higher order, $\varkappa_{(q)\mathrm{TwPA}}$, can be obtained in analogy with formula (3) with the aid of the general expressions (2.20) of Ch. I of Ref. 6 (with account taken of footnote 2). We shall not present them here, and write out by way of example only that part of $\varkappa_{(4)\mathrm{TwPA}}$ which is responsible for allowing the transition $|0j\rangle\rightarrow|2,j\pm4\rangle$:

$$\varkappa_{(4)\mathrm{TwPA}}\approx\frac{2d_{0,j}^{1,j\pm1}}{(2\pi c\hbar)^3Bj[16(Bj)^2-\nu_a^2]}$$
$$\times[d_{1,j\pm1}^{2,j\pm2}d_{2,j\pm2}^{3,j\pm3}d_{3,j\pm3}^{2,j\pm4}+3(d_{1,j\pm1}^{0,j\pm2}d_{0,j\pm2}^{1,j\pm3}d_{1,j\pm3}^{2,j\pm4}$$
$$-d_{1,j\pm1}^{2,j\pm2}d_{2,j\pm2}^{1,j\pm3}d_{1,j\pm3}^{2,j\pm4})]. \tag{7}$$

In the derivation of (7), just as before [see (4)], it was assumed that there are no additional resonances with intermediate levels, meaning in this case no three-photon resonance for the transition $|0,j\rangle\rightarrow|3,j\pm3\rangle$.

We present an estimating formula for $\varkappa_{(6)\mathrm{TwPA}}$

$$\varkappa_{(6)\mathrm{TWPA}}\approx\frac{5(d_{0,j}^{1,j\pm1})^6}{(2\pi c\hbar)^5(Bj)^2\nu_a[-\nu_a^2+16(Bj)^2]}. \tag{8}$$

The hindrances on ThPA are lifted when account is taken of three-photon resonances of fifth, seventh, etc. orders. The corresponding matrix elements of the interactions energy are

$$V_{12}^{(q)}=\sum_a\varkappa_{(q)\mathrm{ThPA}}|\mathscr{E}_a|^{q-3}\mathscr{E}_a^3. \tag{9}$$

The allowed three-photon transitions are those with $\Delta j=\pm4,\pm5$ at $q=5$ and $\Delta j=\pm4,\pm5,\pm6,\pm7$ at $q=7$. An estimating formula for the polarizability $\Delta\varkappa_{(5)\mathrm{ThPA}}$ of the transition $|0,j\rangle\rightarrow|3,j\pm5\rangle$ can be obtained in the form

$$\varkappa_{(5)\mathrm{ThPA}}\approx\frac{10^2(d_{0,j}^{1,j\pm1})^5}{(2\pi c\hbar)^4Bj(9\nu_a^2-16B^2j^2)(\pm8Bj-3\nu_a)}. \tag{10}$$

3. RADIATION ABSORPTION DUE TO OPA, TwPA, AND ThPA OF HIGHER ORDERS

The field at which changes of level populations due to the processes considered above become substantial can be estimated from the condition for saturation of the level population difference of the corresponding transition (for example, $|0,j\rangle \to |1,j-3\rangle$), neglecting further absorption from the excited state. We note first that in all experiments on photoexcitation and photodissociation of the molecules SF_6, BCl_3, and others, the pulse durations employed are $\tau_p \geqslant T_1$ (T_1 is the collision-dominated lifetime of the excited state, $T_1 = 10^{-7}$ sec for SF_6 at a pressure ~1 Torr, Ref. 4). The saturating field for any of the considered processes under these conditions can be estimated with the aid of (2) and the relation (see Ref. 6, p. 42)

$$|\mathscr{E}_{\text{sat}}|^2 = \left(\frac{\hbar^2(1+\Delta^2 T_2^{\,2})}{4\varkappa(q)^2 T_1 T_2}\right)^{1/q}. \tag{11}$$

The line width T_2^{-1} in (11), for molecules of the type SF_6, BCl_3, etc., should apparently be chosen to be the width of the "Coriolis" splitting $\Delta\nu_{\text{Cor}}$ ($\Delta\nu_{\text{Cor}} \approx 0.0037j$ [cm^{-1}] for SF_6, Ref. 8).

We have chosen for the estimates the molecule SF_6, with the following known spectroscopic data 3): $\nu_{01} = 948$ cm^{-1}, $d_{01} = 3\cdot 10^{-19}$ cgs esu, $B \approx 0.0278$ cm^{-1}, $\nu_a = 7$ cm^{-1} (Ref. 8). For this molecule, assuming that $T_2 \approx (\Delta\nu_{\text{Cor}})^{-1}$, the expression for the fields that saturate the populations can be written in the form

$$|\mathscr{E}_{\text{sat}}|^2 \approx (10^{-39} j/4\varkappa(q)^2)^{1/q} \text{cgs esu}. \tag{12}$$

We assume that the molecule is acted upon by a CO_2 laser of frequency $\nu_1 = 938$ cm^{-1}. We consider now the contributions made by the processes investigated above to the absorption at this frequency.

a) **OPA-3, OPA-5.** Taking into account the Coriolis splitting, the condition for the resonance of the frequency ν_1 with the transitions $|0,j\rangle \to |1,j-3\rangle$, $|0,j\rangle \to |1,j-2\rangle$ is satisfied for the levels with $j = 58$–61 and $j = 88$–92 (the exact resonances are at $j = 60$ and 89), whose populations are appreciable. 4) Molecules with these values of j participate in one-photon absorption of third and fifth order. The condition for OPA of fifth order only is satisfied at $j = 35$–37 and $j = 44$–46 because of the resonances of ν_1 with the transitions $|0,j\rangle \to |1,j-5\rangle$, $|0,j\rangle \to |1,j-4\rangle$.

It follows from (4) that at $j = 6$ we have 5) $\varkappa_{(3)\text{OPA}} \approx 7.3\cdot 10^{-26}$ cgs esu; the population saturation because of OPA-3 under quasistationary excitation sets in at a radiation intensity $I_{\text{sat}} \approx 7$ MW/cm^2. The quantity $\varkappa_{(5)\text{OPA}}$ calculated from the exact formulas at $j = 36$ is 6×10^{-32} cgs esu. The saturating intensity $I_{\text{sat}} \approx 35$ MW/cm^2.

Thus, at moderate power densities, substantial absorption on transitions that are forbidden in the linear approximation takes place both on account of OPA-3 and on account of OPA-5.

b) **TwPA-4 and TwPA-6.** We now determine the radiation intensity at which these processes make a substantial contribution to the transitions of the SF_6 molecules from the ground vibrational state $v = 0$ to the excited state $v = 2$. We note first that the condition for ordinary two-photon resonance is satisfied for weakly populated levels $j = 118$, $\sigma(j = 118)/\sigma(j_{\text{max}}) \approx 5\cdot 10^{-2}$. On the other hand, the condition of two-photon resonance of fourth and sixth order, with allowance for the Coriolis broadening, takes place for levels whose population is appreciable: for $j = 58$–60 (the transitions $|0,j\rangle \to |2,j-4\rangle$) and $j = 76$–80 (transitions $|0,j\rangle \to |2,j-3\rangle$).

TwPA-6 takes place for $j = 38$–41 and $j = 46$–48 (the respective transitions $|0,j\rangle \to |2,j-6\rangle$, $|0,j\rangle \to |2,j-5\rangle$). At $j = 59$ (the exact resonance of TwPA-4) we have $\varkappa_{\text{TwPA}} \approx 1.5\cdot 10^{-29}$ cgs esu; it follows from (12) that the populations of the resonance levels become equalized at $I_{\text{sat}} \approx 40$ MW/cm^2. Analogous estimates in the case of TwPA-6 yield $\varkappa_{(6)} \approx 2.64\cdot 10^{-34}$ cgs esu and $I_{\text{sat}} \approx 35$ MW/cm^2.

c) **ThPA-5.** An example of transition with ThPA-5 in SF_6 are the transitions $|0,j\rangle \to |3,j-5\rangle$, $|0,j\rangle \to |3,j-4\rangle$; the resonance condition is satisfied for molecules with $j = 31$–33 and $j = 40$–42, respectively. Calculation of $\varkappa_{(5)}$ from the exact formulas yields (at $j = 32$) $\varkappa_{(5)\text{ThPA}} \approx 5.5\cdot 10^{-32}$ cgs esu; $I_{\text{sat}} \approx 38$ MW/cm^2.

Thus, at intensities $\geqslant 10$ MW/cm^2 the absorption of radiation by the SF_6 molecule becomes substantially influenced by the processes considered above, that lift the hindrances on the transitions with $\Delta j = \pm 3; \pm 4; \pm 5; \pm 6$. It is easy to estimate that these are threshold intensities for OPA, TwPA, and ThPA of higher order also in the BCl_3 molecule.

To conclude, this section, we indicate the field values within which expressions (2), (6), and (9), as well as the foregoing analysis of multiphon absorption processes, are valid. The applicability condition is the relation $V_{\text{sat}} \ll 4\pi c\hbar Bj$, from which it follows that we must have

$$\mathscr{E}_{\text{sat}} \ll \mathscr{E}_{\text{crit}} \approx (4\pi c\hbar Bj/\varkappa_{(q)})^{1/q}.$$

For example, for OPA-3 we have $\mathscr{E}^2_{\text{crit}} \approx 8.8\cdot 10^6$ cgs esu, corresponding to an intensity $I = 4$ GW/cm^2.

4. INFLUENCE OF OPA, TwPA, AND ThPA OF HIGHER ORDERS ON MULTISTEP EXCITATION OF MOLECULES

In accordance with the presently assumed hypothesis, photodissociation of a molecule calls for excitation of its vibrational state $v = 3$–4. It is known that such an excitation of the molecules SF_6 and BCl_3 has been observed, including also at wavelengths not satisfying the conditions for two- and three-photon resonance in the Q branches of the ν_3 mode. Moreover, the dependence of the absorbed energy on the frequency has no resonance peaks.[3,5] Recognizing that the experiments described in Refs. 3 and 5 were made at room temperature, we can attribute this fact to the presence of "hot" absorption bands.[1] Another cause of the absorption are the sideband modes of the ordinary TwPA and ThPA.[1] Estimates presented above indicate that besides the mechanisms listed above, an appreciable contribution to the radiation absorption at a frequency that does not satisfy the conditions for resonances in the Q branches is made by the processes OPA-3 and OPA-5, TwPA-4 and TwPA-5, ThPA-5, etc.

By way of example we consider the excitation of SF_6

molecules to the state $v=3$ under the influence of radiation with $\nu_1=938$ cm^{-1}. At this frequency, besides the three-photon resonances in the sidebands (which are satisfied only for molecules with $j=54-56$ and $j=80-82$), because of the transition to the third vibrationally excited state can be, in particular, one of the following processes:

a) OPA-3 or OPA-5 during the first stage and resonant one-photon absorption in the succeeding two stages. In fact, it is easy to verify that for all j at which OPA-3 takes place,[6] the conditions for one-photon resonance in the next steps is satisfied, such as $|1,j-3\rangle \to |2,j-4\rangle$ (frequency deviation $\Delta_2=0.33$ cm^{-1}) followed by $|2,j-4\rangle \to |3,j-3\rangle$ ($\Delta_3 \approx 0.66$ cm^{-1}). Similar conditions are satisfied for all molecules that take part in OPA-5: the frequency deviations in the second and third stages are ~1 cm^{-1}.

b) TwPA-4 or TwPA-6 during the first stage followed by one-photon absorption in the second state (e.g., $|2,j-4\rangle \to |3,j-3\rangle$ with $\Delta_2 \approx 0.5$ cm^{-1}, $|2,j-5\rangle \to |3,j-4\rangle$ with $\Delta_2 \approx 0.6$ cm^{-1}).

c) ThPA-5. It is easy to show that the intensities at which the equalization of the populations of all three vibrational states takes place on account of stepwise excitations of type a) and b) is of the order I_{sat} for OPA-3 and OPA-5, and ThPA-5 and ThPA-6, respectively.

When the intensity is increased to 50–100 MW/cm^2, the absorption is strongly influenced by the processes OPA-7, ThPA-7, and TwPA-8. At these intensities, the dependence of the absorbed power on the frequency should have no resonant peaks in the Q branches and at low temperatures, since many molecules with different j participate in the absorption process. Thus, the processes OPA-3 and OPA-5, TwPA-4 and TwPA-6, ThPA-7, etc. decrease the selectivity of the excitation of the molecules, and consequently decrease the efficiency of isotope separation. It is obvious that in addition to the use of low temperatures, to attain high selectivity of the excitation of the molecules it is necessary to use radiation with intensity ≤ 10 MW/cm^2.

We note that another aspect of the problem of the absorption of high-power IR radiation by the molecules SF_6, BCl_3, SiF_4, C_2H_4—the long-wave shift of the absorption maximum[2,3,5,9]—also agrees well with the excitation mechanism due to the OPA, TwPA, and ThPA processes of higher order. According to formulas (4)-(10), transitions with decreasing j are more effectively allowed than those with increasing j; in addition, the frequency detuning for single-photon transitions is smaller during the last stages of excitation in the case of transitions with decreased j. With increasing intensity, the long-wave shifts increase, inasmuch as OPA, TwPA, and ThPA of ever increasing order begin to take part in the radiation-absorption process.

We call attention to the fact that an increase of the long-wave shift of the absorption maximum with increasing radiation intensity was observed in SF_6 in Ref. 9.

We note in conclusion that the processes of lifting the hindrance on absorption in strong fields can be observed not only on vibrational-rotational transitions of polyatomic molecules, but also on transitions between components between the fine structure of electronic transitions in atoms. It is obvious that at high intensities it is also possible to observe spontaneous Raman scattering on forbidden rotational transitions in molecules with $\Delta j = \pm 3$ and ± 4.

The authors thank the participants of the seminar of the Physics Institute of the Academy of Sciences under the direction of A. M. Prokhorov for a discussion of the result of the work and A. N. Oraevskiĭ, S. S. Alimpiev, and V. T. Galochkin for stimulating discussions.

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[1] In light of this relation, the term "allowing the transition" is not quite exact; however, its use in the case of optical fields is justified to the same extent that the concept "allowance by application of a constant field" is justified.

[2] It is erroneously indicated in the definition of the polarizability tensor on p. 28 of Ref. 6 that $\hat{C}$ is the operator of the cyclic permutations only.

[3] By B we mean the quantity $B'(1-\zeta)$, where ζ is the Coriolis constant.[7]

[4] At 300 K the levels with maximum population are $j_{\max} \approx 50$; the level population ratio is $\sigma(j=60)/\sigma(j=50) \approx 0.94$.

[5] The polarizabilities $\varkappa_{(4)}$, $\varkappa_{(7)}$, etc. were calculated under the assumption that $d_v^{v+1} = v^{1/2} d_0^{1}$.

[6] Examples of the corresponding values of j are given in preceding section.

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Translated by J. G. Adashko