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Evaluation of parameters of intramolecular interaction from absorption and fluorescence spectra of substituted arylpolyene with poor resolved vibrational structure

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Abstract

The fluorescence and fluorescence excitation spectra (conjugated spectra) of new synthesized substituted arylpolyene (C_6H_5–[CH=CH]$_2$–C_6H_4–NH_2) in solid *n*-octane at $T = 4.2\,K$ were studied. Spectra have feebly marked vibrational structure. The method of finding vibronic interaction parameters from conjugated spectra was developed. The method is based on the modeling of spectra in the form of set of vibronic bands. Each vibronic band is represented as the sum of narrow zero-phonon line and broad phonon wing. It allows carrying out the theoretical calculation of the conjugated spectra with poor resolved vibrational structure, which can be compared with the measured spectra. The vibronic interaction parameters which determine shape of the spectra can be found from comparative analysis of experimental and model spectra. This analysis allowed us to explain the deviation from the mirror symmetry between measured conjugated spectra as a joint manifestation of Franck-Condon and Herzberg-Teller interactions. The parameters of both interactions for main normal vibrations of molecule were found.
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1. Introduction

Important information about interaction of electronic and vibronic excitations in complex organic molecules can be obtained from optical spectroscopy of doped solids at low temperatures. Luminescence

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and absorption spectra of dye molecules in solid matrices consist of the vibronic bands which correspond to the transitions of molecules to different vibronic levels. A theory shows and an experiment confirms that each vibronic band consists of narrow quasi-lorentian zero-phonon line (ZPL), which corresponds to the purely electronic transition, and broad phonon wing (PW), which is determined by creation/destruction of matrix phonons (see, for example, Ref. [1,2]). The electronic–phonon interaction resulting in the *homogeneous* broadening of vibronic bands, is stronger, the PW is more intensive. The relation between integrated intensity of ZPL and total intensity of vibronic band is represented by Debye–Waller factor, η. The η is closer to one, the electronic-phonon interaction is weaker. Thus the optical spectra of doped solids are informative only in the case of high values of Debye–Waller factor. Moreover local disorder in solid causes the additional *inhomogeneous* broadening of optical spectra [3] of impurity centers.

Both homogeneous and inhomogeneous broadening are minimal in Shpolsky matrices. Therefore many organic molecules in such matrices manifest quasi-line spectra at low temperatures even if classical light sources is used [4,5]. A vibrational analysis of such spectra enables to obtain the information about internal vibrations of chromophore molecules as well as about vibronic interaction parameters (see, for example, [6–12]).

While the same vibronic interaction determines the intensity distribution both in absorption and fluorescence spectra, the number of vibronic peaks which are used for finding the vibronic interaction parameters evaluation is doubled. According to the theory [10,13–16], if a vibronic interaction causes only the change of equilibrium positions of nuclei (Franck–Condon (FC) interaction), the absorption and fluorescence vibronic spectra should be mirror symmetric with each other. In pure form such symmetry does not usually observed. For many molecules with "rigid" frameworks of nuclei, a slight deviation from the mirror symmetry between conjugated spectra is observed. It becomes apparent in the difference of intensities of conjugated vibronic peaks (i.e. spectral lines which are mirror symmetric relative to 0–0 ZPL) and in the small difference of the corresponding vibronic frequencies. The difference of the vibronic frequencies can be explained due to a change of the steepness of FC potential curves upon electronic excitation. This difference determines a quadratic FC-interaction. The difference of the intensities of conjugated vibronic peaks can be explained as the joint manifestation of FC interaction and Herzberg–Teller (HT) interaction. The HT interaction takes into account the modulation of an electronic dipole moment by the nuclei vibrations, i.e. the dependence of the electronic matrix element $M(R)=\mathbf{E}d(R)$ on the nuclei coordinates $\boldsymbol{R}$. The interference of FC- and HT- amplitudes leads to the different intensities of conjugated vibronic peaks.

Earlier the task of the evaluation of FC- and HT-interaction parameters was solved for the fluorescence and fluorescence excitation spectra with good resolved vibrational structure (in particular, for the quasi-linear spectra of impurity chromophores in Shpolsky matrices and in cooled ultrasonic jet. A slight deviation on the mirror symmetry between conjugated spectra was explained quantitatively as a joint manifestation of FC- and HT-interactions (see, for example, [6–8]). In a set of works the quantitative analysis of the spectrum intensity distribution was performed for fluorescence only. The absence of data relating to absorption spectra creates difficulties because the number of independent experimental data becomes much less and therefore it is necessary to have very good resolved extensive fluorescence spectrum for theoretical analysis (see, for example, [16,17]).

Note that most of dye molecules do not reveal good resolved quasi-linear spectra even in Shpolsky matrices. These spectra consist usually of small peaks on the intensive background. Such dyes are, for example, polyenes (in particular, arylpolyenes) which are widely used in modern technologies. Synthetic arylpolyenes are used in spectroscopy and photobiology (see, for example, [18]), in particular, as fluorescent membrane probes. Substituted arylpolyenes widely used in the nonsilver photography, scintillation technique etc. (see, for example, [19]). Moreover polyene-dyes can be used in dye-laser technique, which are

widely used in electronics, optics, spectroscopy, plasma physics, chemistry, biology etc. Therefore the comprehensive investigations of spectral-luminescent properties of such compounds are very actual and interesting (see, for example, [20–27]).

What spectral information can be obtained from poor resolved spectra where vibronic peaks are stayed on the intensive background? This question is discussed in the present paper.We suggest a method for analysis of such spectra, which allows to evaluate the parameters of a vibronic interaction even from the spectra with poor resolved structure.

The method is based on the modeling of the spectra in the form of a set of vibronic bands. Each vibronic band is represented as the sum of narrow Lorentian ZPL and broad Gaussian phonon wing. Such model allows to determine the intensities of vibronic peaks in the experimental spectra and to carry out vibrational analysis of conjugated spectra. We used this method for quantitative analysis of fluorescence and fluorescence excitation spectra of new synthesized substituted arylpolyene $C_6H_5–[CH=CH]_2–C_6H_4–NH_2$ in solid n-octane at $T = 4.2$ K [27].

2. Experimental results

The fluorescence and fluorescence-excitation spectra were measured at 4.2 K. Fluorescence was excited using an ultrahigh-pressure xenon lamp. The concentration of the compounds under investigation in the n-octane solution was varied from 10^{-3} to 10^{-5} mol/l. The fluorescence spectra were corrected for the setup spectral sensitivity, and the fluorescence excitation spectra were corrected for the intensity distribution over the excitation-source spectrum.

In this paper, we report the results of a study of the fluorescence and fluorescence-excitation spectra of solution of compound belonging to the class of substituted arylpolyenes [27]. The structural formula of this compound represent on Fig. 1.

We have found that the fluorescence and fluorescence-excitation spectra are independent on the excitation and detection wavelengths and not affect by variations in the solution concentrations.

The spectra have the following anomalies: a pronounced violation of bilateral symmetry between the fluorescence and fluorescence-excitation spectra (see Fig. 2). One can see that vibronic structure of spectra is poorly resolved.

Quasi-linear spectra consist of a set of vibronic ZPLs. As a result the parameters of vibronic interaction are determined from an analysis of the vibronic ZPLs intensities. In our case each vibronic band consists of both ZPL and PW, and therefore different vibronic bands are strongly overlapped. It creates difficulties in obtaining information about single ZPLs. To solve this problem we suggest to model the spectra with poor resolved structure and to compare these spectra with experimental spectra. The procedure of an
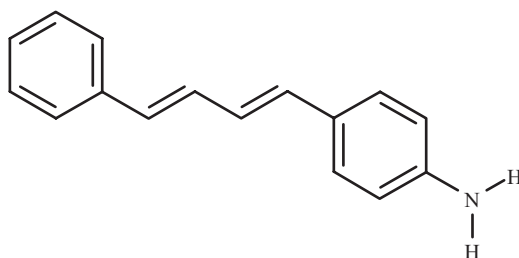

Fig. 1. Structural formula of arylpolyene under study.

experimental data processing is following:

(1) The frequencies of spectral peaks are determined from the original experimental spectra. These are the frequencies of local intensity maxima on the spectra. These frequencies are the spectral positions of ZPLs corresponding to different vibronic levels.

(2) Each vibronic band is modeled as sum of narrow Lorentian ZPL and broad Gaussian PW. PW is takes up position from the red and blue side relative to ZPL in fluorescence and fluorescence-excitation spectra correspondingly. The ZPL width is slightly increases with increasing of number of vibronic level that corresponds to the decreasing of vibronic state lifetime with increasing of their energy.

(3) The homogeneous width of ZPL, the value of inhomogeneous broadening, the value of Debye–Waller factor in a vibronic band are chosen so that modeled spectra would fit experimental spectra the best (see Figs. 2, 3.and Table 1). The best agreement between experimental and modeled spectra was obtained for the following parameters values: the Debye–Waller factor $\eta = (S_{ZPL}/S_{ZPL} + S_{PW}) = 0.29$ (here S_{ZPL} and S_{PW} are the integrated intensities of ZPL and PW, correspondingly); the widths of ZPL and PW are $\Gamma_{ZPL} = 40\,cm^{-1}$ and $\Gamma_{PW} = 480\,cm^{-1}$, correspondingly; the frequency distance between ZPL and PW
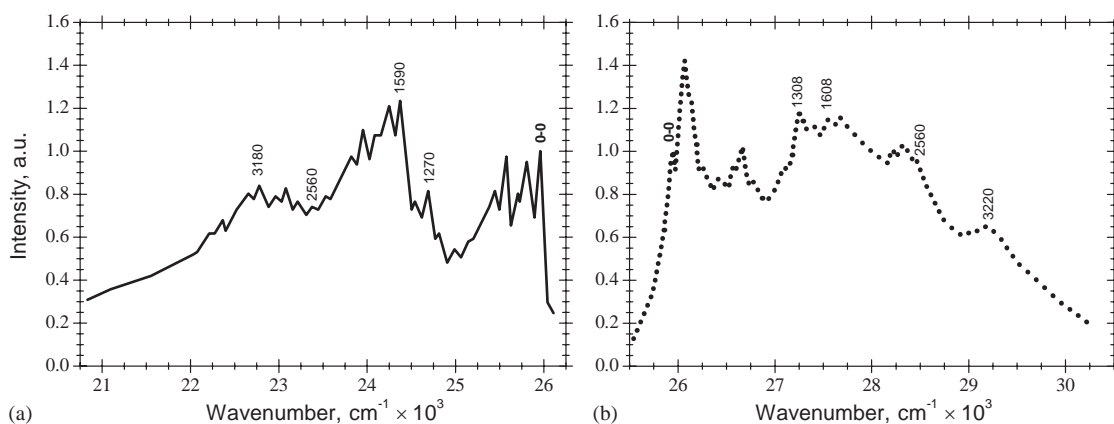

Fig. 2. Experimental fluorescence (*solid line*, $\lambda_{exc} = 360$ nm; concentration 10^{-5} mol/l) and fluorescence excitation (*dotted line*, $\lambda_{det} = 414$ nm; concentration 10^{-4} mol/l) spectra for the studied compound which is dissolved in *n*-octane at temperature 4.2 K.
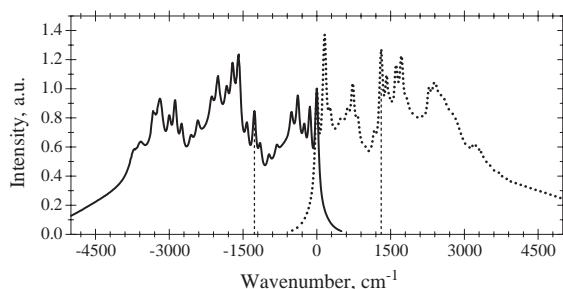
Fig. 3. Modeled fluorescence (*solid line*) and fluorescence excitation (*dotted line*) spectra (see text for details). The parameters of FC- and HT-interactions were fitted by the analysis of vibronic peaks inside of the region between vertical dashed lines (from $-1270\,cm^{-1}$ up to $1308\,cm^{-1}$ excluding the vibration with frequency $\sim 1600\,cm^{-1}$). Vibronic spectra outside of the selected interval were calculated on the base of obtained parameters of FC- and HT-interactions. The border between these two regions corresponds to the bold line in Table 1.

Table 1

Calculated and experimental intensities of vibronic peaks in the fluorescence and fluorescence excitation spectra for studied arylpolyene C_6H_5–[CH=CH]$_2$–C_6H_4–NH_2

Fluorescence				Fluorescence excitation				Parameters	
ω_n, cm^{-1}	Combination of ω_n, (cm^{-1})	J_n^{fluo}		ω_n, cm^{-1}	Combination of ω_n, (cm^{-1})	J_n^{abs}		FC	HT
		Experiment	Calculation			Experiment	Calculation	$a/2$	α
0		1		0		1			
140		0.6	0.59	130		0.3	0.3	0.66	−0.11
160		0	0	160		1.1	1.08	0.52	0.52
254		0.28	0.28	270		0.29	0.29	0.53	0.005
386		0.49	0.49	386		0	0	0.35	−0.35
516		0.27	0.25	480		0.07	0.07	0.38	−0.12
620		0	0	620		0.15	0.14	0.19	0.19
730		0	0	730		0.49	0.49	0.35	0.35
817		0.11	0.11	835		0.21	0.21	0.39	0.06
975		0.14	0.14	975		0.05	0.05	0.3	−0.07
1150		0.3	0.3	1165		0.19	0.18	0.49	−0.06
1270		0.67	0.67	1308		0.93	0.92	0.89	0.07
1420	1270 + 140	0.39	0.64	1420	1308 + 130	0.52	0.58		
1457	730 × 2	0	0	1457	730 × 2	0.05	0.07		
1590		1.18	1.17	1608		0.52	0.52	0.9	−0.18
1713	1590 + 140	0.78	0.66	1713	1608 + 130	0.17	0.14		
1722		0		1722		0.38			
1835	1590 + 254	0.36	0.33	1830	1608 + 270	0.09	0.15		
2010	1590 + 386	0.55	0.48	2010	1608 + 386	0	0		
2140	1590 + 516	0.28	0.28	2140	1608 + 480	0.04	0.03		
2280	1590 + 516 + 160	0	0.01	2280	1608 + 480 + 160	0.29	0.07		
	1270 + 817 + 160		0.01		1308 + 835 + 160		0.2		
2410	1590 + 620 + 160	0	0	2390	1608 + 620 + 160	0.32	0.07		
	1270 + 620 + 480		0		1308 + 620 + 480		0.02		
2422	1270 + 1150	0.27	0.21	2430	1308 + 1165	0.12	0.17		
2560	1270 × 2	0.18	0.22	2560	1308 × 2	0.39	0.41		
2748	1590 + 1150	0.44	0.48	2765	1608 + 1165	0.12	0.16		
2882	1590 + 1270	0.8	0.81	2840	1608 + 1308	0.18	0.49		
3000	1270 × 2 + 386	0.49	0.13	2950	1308 × 2 + 386	0.05	0		
	1590 + 1150 + 254		0.1		1608 + 1165 + 270		0.02		
3182	1590 × 2	0.76	0.64	3220	1608 × 2	0.14	0.12		
3306	1590 × 2 + 140	0.52	0.69	3306	1608 × 2 + 130	0.07	0.05		
3590	1590 × 2 + 386	0.31	0.23	3590	1608 × 2 + 386	0	0		
3750	1590 × 2 + 516	0.22	0.14	3750	1608 × 2 + 480	0	0		

Estimated parameters of vibronic interactions a and α for generating vibronic bands.

maxima is $\Delta\omega_{ZPL-PW} = 380\,cm^{-1}$. All parameters are determined taking into account the inhomogeneous broadening.

The modeled spectra are obviously most sensitive to the values of vibronic peaks intensities, hence the shapes of the modeled spectra are rather critical with respect to the determined intensities values. It facilitates the fitting of above parameters. At the same time the values of parameters η, Γ_{ZPL}, Γ_{PW} and $\Delta\omega_{ZPL-PW}$ are influenced by the presence of inhomogeneous broadening and by the restriction imposed on the PW shape. Particularly, it is known [28], that the value of $\Delta\omega_{ZPL-PW}$ is approximately equal to the

Debye frequency of a system under study whose typical values amount $50–150\,\mathrm{cm}^{-1}$. We suppose, that the obtained difference between theoretical and experimental spectra can be related to the fact that the form of PW in real systems can be more complicated than the Gauss function. However, this is not the principal because the determination of above parameters with high accuracy is beyond the present paper.

Thus we obtain the value of intensities of vibronic peaks which can be used in vibrational analysis.

3. Vibrational analysis of conjugated spectra

In this section we shortly restate the main equations of the theory vibronic spectra of complex impurity molecules in solids (see for details Ref. [15]), which we used for the analysis of experimental data. Since the studied molecule has no symmetry elements, all its normal vibrations are totally symmetrical (see Fig. 1). Vibronic interaction is manifested as the change of an adiabatic potential at the electronic excitation

$$\sum_{n=1}^{N}\frac{\eta\Omega_n}{2}(R_n - a_n)^2 - \sum_{n=1}^{N}\frac{\eta\Omega_n}{2}R_n^2 \tag{1}$$

and as the modulation of electronic matrix element by the nuclei vibrations

$$M(R) = M(0)\left(1 + \sum_{n=1}^{N}\alpha_n R_n + ...\right), \tag{2}$$

where R_n are dimensionless nuclear displacement.

Shift of the equilibrium positions of normal vibrations, a_n, and coefficients α_n are the dimensionless parameters of FC- and HT- interactions, correspondingly.

In Ref. [10] the common formula for calculation of integral reduced intensity, $J^g(\omega)$, of vibronic band at arbitrary dependence of electronic matrix element on the nuclear coordinates was derived (see for review Ref. [15]):

$$J^{g,e}(\omega) = J_0\Bigg\{\delta(\omega - \omega_0) + \sum_n\left(\frac{\pm a_n}{2} + \alpha_n\right)^2\delta(\omega - \omega_0 \mp v_n)$$

$$+ \frac{1}{2!}\sum_{n_1 n_2}\left(\frac{a_{n_1}}{2}\frac{a_{n_2}}{2} \pm \frac{a_{n_1}}{2}\alpha_{n_2} \pm \frac{a_{n_2}}{2}\alpha_{n_1} + \alpha_{n_1 n_2}\right)^2\delta(\omega - \omega_0 \mp v_{n_1} \mp v_{n_2}) + ...\Bigg\}, \tag{3}$$

where upper signs is used for absorption (fluorescence excitation) spectrum and lower signs is used for fluorescence spectrum; a_n is parameter of FC interaction representing shift of equilibrium position of normal coordinate, R_n; α_n is parameter of HT interaction, which is given by:

$$\alpha_n = \frac{1}{M(a/2)}\left(\frac{\partial M}{\partial R_n}\right)_{R_n = a/2} \tag{4}$$

if both FC- and HT-interactions are allowed for.

Eq. (3) contains the summation over all intra- and intermolecular vibrations of an impurity center. Assuming that HT-interaction for intermolecular modes is negligible, and taking into account that quadratic FC-interaction broadens ZPL (i.e. it transforms each delta-function $\delta(\omega)$ in Eq. (3) into
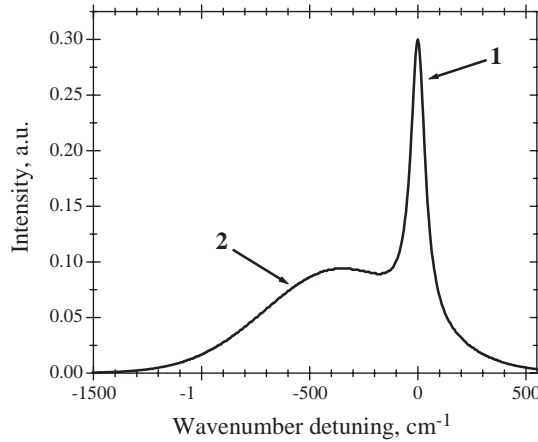
Fig. 4. The model of vibronic band in a fluorescence spectrum, which consists of narrow ZPL (1) and broad PW (2) (see text for details).

Lorentian $L(\omega)$), the Eq. (3) can be rewritten as follows:

$$J(\omega) = J_0 \left\{ I(\omega - \omega_0) + \sum_n \left(\pm \frac{a_n}{2} + \alpha_n \right)^2 I(\omega - \omega_0 \mp \Omega_n) \right. $$
$$\left. + \frac{1}{2!} \sum_{n_1 n_2} \left(\frac{a_{n_1}}{2} \frac{a_{n_2}}{2} \pm \frac{a_{n_1}}{2} \alpha_{n_2} \pm \frac{a_{n_2}}{2} \alpha_{n_1} + \alpha_{n_1 n_2} \right)^2 I(\omega - \omega_0 \mp \Omega_{n_1} \mp \Omega_{n_2}) + \ldots \right\}, \tag{5}$$

where

$$I(\omega) = \eta L(\omega) + (1 - \eta) \Phi(\omega) \tag{6}$$

describes the vibronic band. Each vibronic band consists of Lorentian-like ZPL $L(\omega)$ and PW $\Phi(\omega)$ which was modeled by Gauss function (see Fig. 4).

Eq. (6) takes into account electron–phonon interaction. Therefore indexes n in Eq. (5) relate solely to intramolecular normal vibrations.

One can derive from Eq. (5) the following equations for the intensities of vibration transitions and their combinations (see, for example Ref. [15,16]), which we will use for vibrational analysis of conjugated spectra:

$$J_n = \left[\pm \frac{a_n}{2} + \alpha_n \right]^2, \tag{7.1}$$

$$J_{2n} = \frac{1}{2} \left[\left(\frac{a_n}{2} \right)^2 \pm 2 \frac{a_n}{2} \alpha_n \right]^2, \tag{7.2}$$

$$J_{n_1 + n_2} = \left[\frac{a_{n_1}}{2} \frac{a_{n_2}}{2} \pm \frac{a_{n_1}}{2} \alpha_{n_2} \pm \frac{a_{n_2}}{2} \alpha_{n_1} \right]^2, \tag{7.3}$$

$$J_{n_1 + 2n_2} = \frac{1}{2} \left[\pm \frac{a_{n_1}}{2} \left(\frac{a_{n_2}}{2} \right)^2 + 2 \frac{a_{n_1}}{2} \frac{a_{n_2}}{2} \alpha_{n_2} + \left(\frac{a_{n_2}}{2} \right)^2 \alpha_{n_1} \right]^2, \tag{7.4}$$

$$J_{n_1 + n_2 + n_3} = \left[\pm \frac{a_{n_1}}{2} \frac{a_{n_2}}{2} \frac{a_{n_3}}{2} + \frac{a_{n_1}}{2} \frac{a_{n_2}}{2} \alpha_{n_3} + \frac{a_{n_1}}{2} \frac{a_{n_3}}{2} \alpha_{n_2} + \frac{a_{n_2}}{2} \frac{a_{n_3}}{2} \alpha_{n_1} \right]^2. \tag{7.5}$$

We employed these formulae for the intensities, because only these combinations of vibrations are manifested in our spectra.

Vibronic peak intensities, *I*, measured in experiment are connected with reduced intensities, *J*, by means of frequency factor, that can be taken into account as follows (see details, for example, in Ref. [15]):

$$J_n^{\text{fluo}} = \frac{I_n^{\text{fluo}}}{I_0^{\text{fluo}}} \left(\frac{\omega_0}{\omega_n}\right)^3, \quad J_n^{\text{abs}} = \frac{I_n^{\text{abs}}}{I_0^{\text{abs}}} \frac{\omega_0}{\omega_n}, \tag{8}$$

The first and the second formulae are used for fluorescence and absorption spectra, respectively.

The theoretical absorption and fluorescence spectra were calculated using Eq. (5) and (6) with amplitudes (7.1)–(7.5). Using Eq. (7.1) for one-quant vibrations, we chose pair of parameters a_n and α_n for each vibronic peak the way the calculated intensities of one-quant peaks should coincide with experimental one. After such fit of parameters a_n and α_n values, the Eqs. (7.2)–(7.5) do not contain unknown parameters, hence the intensities of the vibrations combinations can be calculated.

Table 1 presents the reduced intensities of fluorescence and fluorescence excitation spectra which were calculated from experimental data as described above. These data were used in vibronic analysis and they permit us to find values of the parameters of FC- and HT-interactions for 12 main normal frequencies. These parameters are shown in the upper part of Table 1 (above bold line, splitted the Table, plus generating vibration with frequency ̃1600 cm^{-1}), as well as corresponding intensities of vibronic peaks calculated with these parameters.

The lower part of Table 1 presents the intensities of vibronic peaks, calculated on the base of parameters fitted from one-quant peaks. One can see rather good agreement between experiment and calculation. Using of the evaluated parameters we described 15 vibronic peaks (combinations) in conjugated spectra. The errors in the parameters evaluated can be estimated as 15–20%, that in our case (for the spectra with poor resolved vibrational structure) seems to be acceptable value. To support the 12 generating vibrations obtained the analysis of resonance Raman spectra can be used as well as a normal mode analysis of the molecular ground state.

4. Concluding remarks

(1) The fluorescence and fluorescence excitation spectra of new synthesized substituted arylpolyene (C_6H_5–[CH=CH]$_2$–C_6H_4–NH$_2$) in solid *n*-octane at $T = 4.2$ K have been measured. The marked deviation from the mirror symmetry between spectra was found. The spectra are poor resolved despite the fact that the solvent is a typical Shpolsky matrix.
(2) The modeling of experimental spectra as a set of vibronic bands which consist of ZPL and PW allows us to determine the intensities of vibronic peaks in poor resolved spectra in more adequate fashion as compared to the case when background is subtracted. The vibrational analysis of conjugated spectra of system under study has been performed. The parameters of FC- and HT-interactions were determined for normal vibrations of the molecule.
(3) The deviation from the mirror symmetry in conjugated fluorescence and fluorescence excitation spectra can be quantitatively explained by joint influence of Franck–Condon and Herzberg–Teller interactions.

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