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Investigation of photochromic transformations of diarylethenes using spectrophotometry and quantum chemistry

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An analysis of the results of complex experimental and theoretical studies of photoinduced changes in the electronic spectra of photochromic diarylethenes (DAE) of various structures is presented. The electronic absorption spectra of DAE and the molecular orbitals involved in the formation of these spectra are calculated. The results of quantum chemical calculations are consistent with the experimental spectrophotometric data of the DAE. An analysis of the boundary molecular orbitals of the studied DAEs was carried out and, on its basis, their electronic properties were determined and analyzed. Features (in comparison with other considered DAE molecules) of the electronic structure of the DAE2 molecule (containing fluorine atoms) were revealed, which are also manifested in the calculated electronic parameters. The results obtained can be used to optimize the choice or targeted synthesis of photochromic diarylethenes (with required properties) used in various nanostructured systems for molecular electronics and photonics devices.

Keywords: photochromism, diarylethenes, spectrophotometry, quantum chemistry.

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Introduction

Together with other photochromic compounds (such as, for example, spiropyrans or fulgides), diarylethenes (DAE) [1] are promising for being applied in molecular electronics and photonics devices as photocontrolled switches and memory media [2]. Functionalization of the DAE molecules can vary their physical & chemical and optical (spectral) properties for various applications in the optimum way. Combination of the photochromes into nanocomposite (hydride) structures with metal or semiconductor nanoparticles [3] can produce systems for the nanophotonics with optimized tunable characteristics. It is important to theoretically investigate an electron and oscillation structure of the molecules of the photochromic compounds using quantum chemistry for implementation of targeted synthesis with required properties and for analysis of these properties [4,5]. Besides, the quantum chemical methods are also used to study various molecules of the DAE class [6–8].

For development of the previous investigations by the authors of [9–16], the present paper investigates the photoinduced modifications of the electron structures in the number of the photochromic DAEs using the methods of absorption spectrophotometry and quantum chemistry.

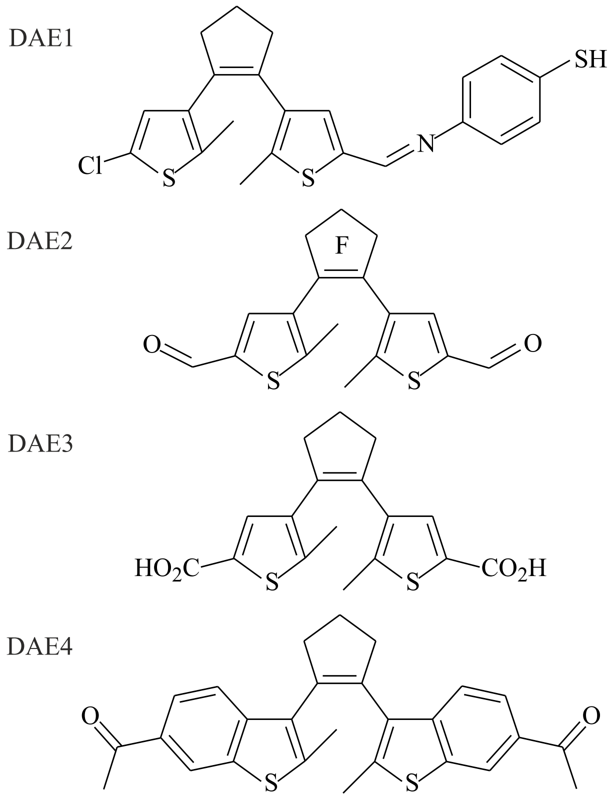
1. Experimental part

The objects of study in the present paper included previously synthesized [17–19] photochromic compounds DAE1, DAE3 and DAE4 of the class of DAE cyclopentene derivatives and DAE2 of the class of DAE perfluorocyclopentene derivatives (Diagram 1).

The DAE absorption spectra (Fig. 1) were recorded using the spectrophotometer SPECORD 200 (CarlZeiss, Germany). The samples were irradiated using LEDs with emission band maxima at 365 nm (LED365), 514 nm (LED514) and 627 nm (LED627). The half-widths of the emission bands of the light emission diodes LED365, LED514 and LED627 were 9, 32 and 16 nm, so was the optical power the 0.45, 3 and 3 W, respectively. The irradiation time required for transferring the DAE molecules from the open form A into the cyclic form B and vice versa was determined based on the adsorption spectra measured in an equilibrium state.

The quantum chemical calculations were made using the software package PC GAMESS 7.1.G (Firefly). The equilibrium geometry (using the density functional theory (DFT)) and the spectra of electron absorption (the TD-DFT method) were calculated by applying the hydride 3-parameter exchange Becke functional with the Lee-Yang-

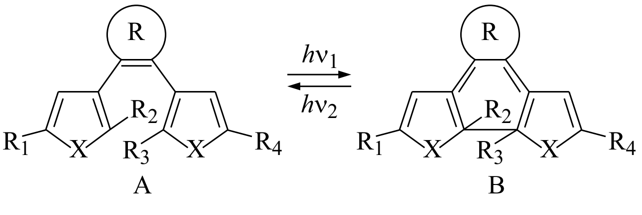
Parr correlation functional (B3LYP) in the 6-31G basis. The influence of the solvent was taken into account under the PCM.



Scheme 1.

2. Results and discussion

The DAE photochromism consists in reversible valence isomerization, i.e. the photoinduced transition from the open form A into the cyclic form B [1,20,21]. Absorbing ultraviolet (UV) radiation (Diagram 2), the open colorless DAE isomer (A) is transformed into the colored (cyclic) DAE isomer (B), which, under the action of visible radiation absorbed by it, again turns into the original open isomer. The diarylethenes are thermally irreversible, since the thermal relaxation of the photoinduced cyclic DAE isomer (B) into the original open form does not occur at the room temperature.



Scheme 2.

The experimental absorption spectra of all the studied compounds exhibit the reversible photochromic modifications of the DAE molecules, which are resulted from UV and visible irradiation. This is indicated by emergence and disappearance of a band in the spectrum visible region (Fig. 1). The absorption spectra of the DAE open form in a non-polar solvent (toluene) are characterized by the absorption bands with maxima in the UV spectrum region (Fig. 1).

The UV irradiation exhibits new absorption bands in the spectrum visible region with the maxima at 525 (DAE1), 626 (DAE2), 536 (DAE3), 491 nm (DAE4), which are related to generation of the photoinduced B form of the DAE molecules.

To interpret the results obtained by the experimental methods of electron spectroscopy, as well as to analyze the mechanisms of the photoinduced transformations of the DAE molecules, the electron structure of these molecules and their properties were investigated in the quantum chemical study. The results (the calculated spectra of the electron absorption of the DAE molecules) are shown on Fig. 2–5 and in Table 1–5.

The calculated spectra also exhibit the modifications of the structure of the DAE molecules due to their photoisomerization. All the studied compounds are characterized by the calculated spectra exhibiting the electron absorption band of the close form of the DAE molecules in the region 577 nm (DAE1, Fig. 2, *c*); 654 nm (DAE2, Fig. 3, *c*); 573 nm (DAE3, Fig. 4, *c*); 522 nm (DAE4, Fig. 5, *c*).

It is clear from Fig. 2, *a* and Table 1 that for the open DAE1 isomer the most intensive absorption bands (with the maximum at 237 nm and the oscillator force of 0.20, with the maximum at 282 nm and the oscillator force of 0.22, with the maximum at 294 nm and the oscillator force of 0.16) are related to the transitions to excited singlet states of the molecule ($S_0 \to S_{18}$, $S_0 \to S_4$, $S_0 \to S_3$, respectively). The electron excitation from the 108 molecular orbital (MO) to the 114 MO mainly contributes to the absorption band at 237 nm, so does from the 109 MO to the 113 MO (LUMO) — to the absorption band at 282 nm, so does from the 110 MO to the 113 MO (LUMO) — to the absorption band at 294 nm, so does from the 112 MO to the 113 MO (LUMO) — to the absorption band at 351 nm (Fig. 2, *b*). The calculation results show that irradiation of the DAE1 molecule within the long-wave absorption band of the open form A makes the transition, $S_0 \to S_2$, which is related to charge transfer from an alken fragment and from one of the thiophen rings (including from sulphur atoms) to the C–C=N–C chain of the DAE1 molecule. The probability of this transition is not big (the oscillator force of 0.04), which is exhibited in the experimental spectra as well (Fig. 1, *a*). At the same time, the transition $S_0 \to S_1$ has a near-zero probability.

For the close (cyclic) DAE1 isomer (Fig. 2, *c* and Table 1), the most intensive absorption bands (with the maximum at 334 nm and the oscillator force of 0.18, with the maximum at 418 nm and the oscillator force of 0.15, with the maximum at 577 nm and the oscillator force of 0.55) are related to the transitions to the excited

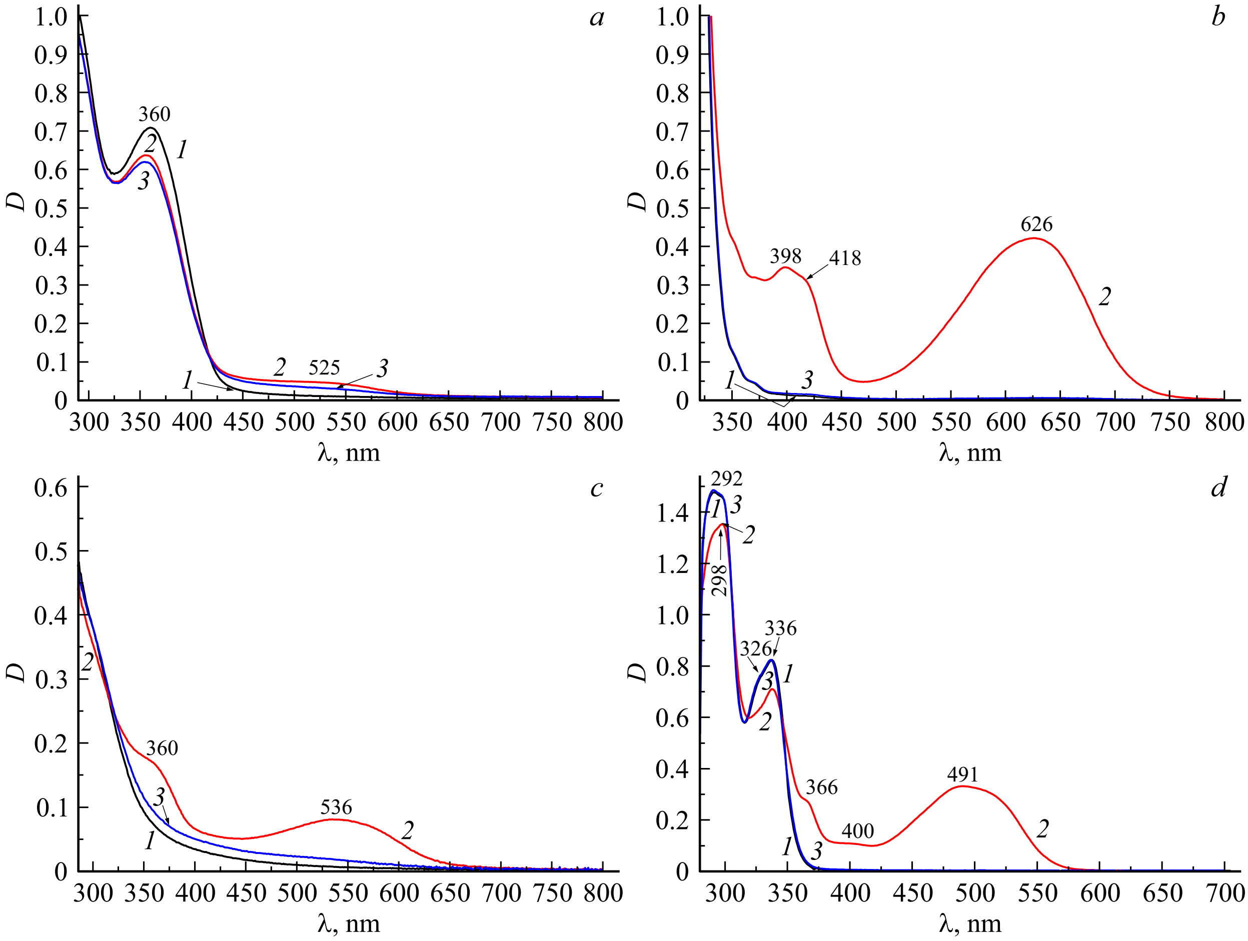


Figure 1. Absorption spectra of DAE1 (*a*), DAE2 (*b*), DAE3 (*c*), DAE4 (*d*) in toluene before (*1*) and after subsequent UV (*2*) ($\lambda = 365$ nm, $t = 30$ s) and visible light (*3*) ($\lambda = 514$ nm (*a, c, d*), $\lambda = 627$ nm (*b*), $t = 60$ s) irradiation, the DAE concentration in the solution $2 \cdot 10^{-4}$ mol/dm^3, the cuvette thickness 0.2 cm.

Table 1. Electronic structure of the open and cyclic forms of the DAE1 molecule as calculated relative to the main peaks

Configuration	State	Wave length, nm	Transition energy, eV	Decomposition of wave functions along singly excited configuration	Oscillator strength f
Open	$S_0 \to S_{18}$	237	5.23	$0.57(108 \to 114) + 0.45(109 \to 114) - 0.33(112 \to 117) - 0.28(112 \to 115)$	0.20
	$S_0 \to S_4$	282	4.40	$-0.88(109 \to 113) - 0.26(110 \to 113) - 0.24(108 - 113)$	0.22
	$S_0 \to S_3$	294	4.21	$0.88(110 \to 113) - 0.31(108 \to 113) - 0.25(109 \to 113)$	0.16
	$S_0 \to S_2$	351	3.53	$-0.98(112 \to 113) - 0.16(110 \to 113)$	0.04
Cyclic	$S_0 \to S_6$	334	3.71	$0.83(109 \to 113) + 0.27(111 \to 113) -0.25(112 \to 115) - 0.20(107 \to 113)$	0.18
	$S_0 \to S_2$	418	2.96	$-0.92(111 - 113) - 0.32(112 \to 114) + 0.18(109 \to 113)$	0.15
	$S_0 \to S_1$	577	2.15	$-0.99(112 \to 113) - 0.13(113 \to 112)$	0.55

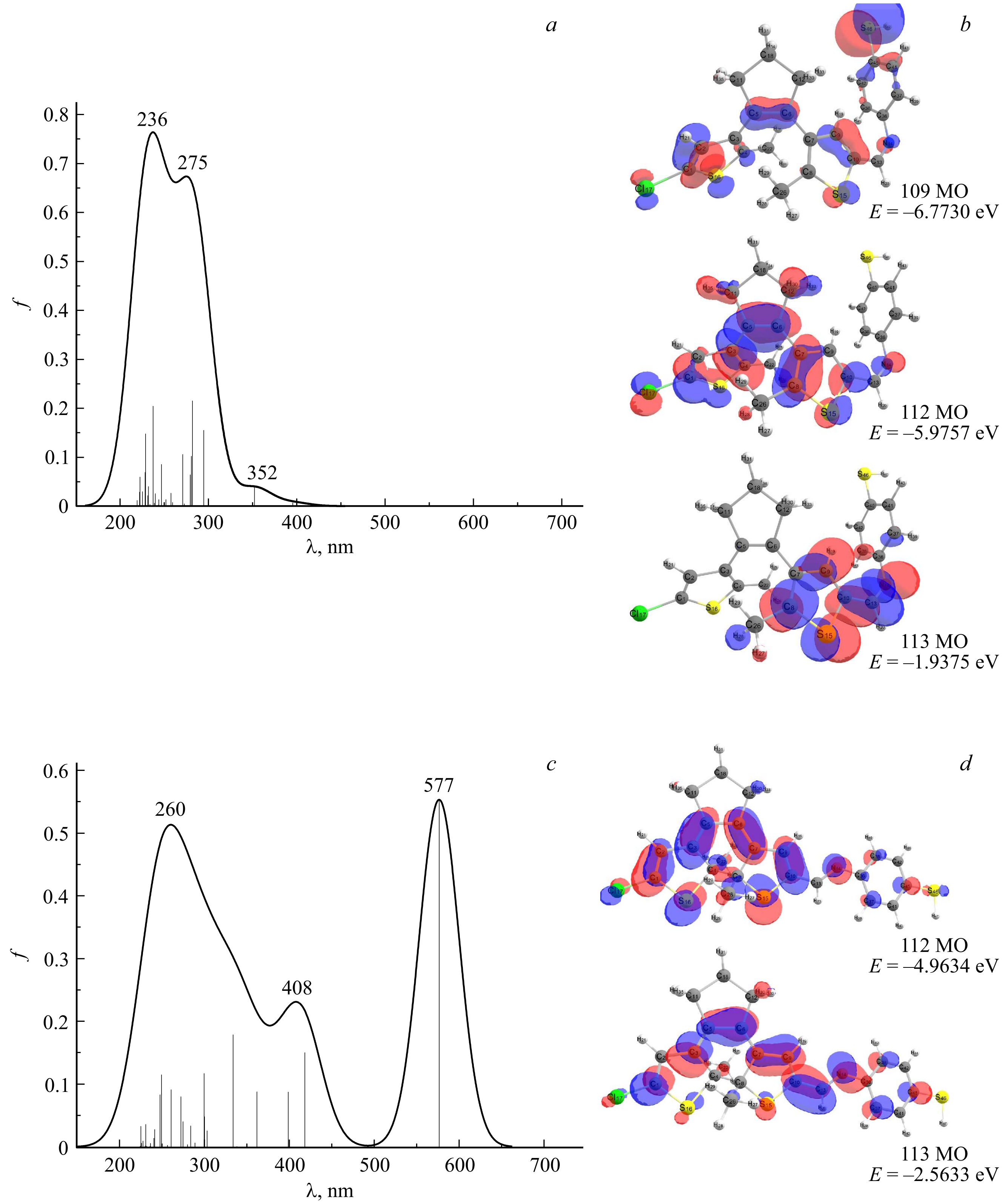


Figure 2. Calculated spectra of electronic absorption (*a, c*), shapes of boundary MOs involved in formation of DAE1 absorption spectrum (*b, d*), of the open (*a, b*) and cyclic (*c, d*) isomers of the DAE1 molecule.

singlet states of the molecule ($S_0 \rightarrow S_6$, $S_0 \rightarrow S_2$, $S_0 \rightarrow S_1$, respectively). The electron excitation from the 109 MO to the 113 mainly contributes to the absorption band at 334 nm, so does from the 111 MO to the 113 MO —

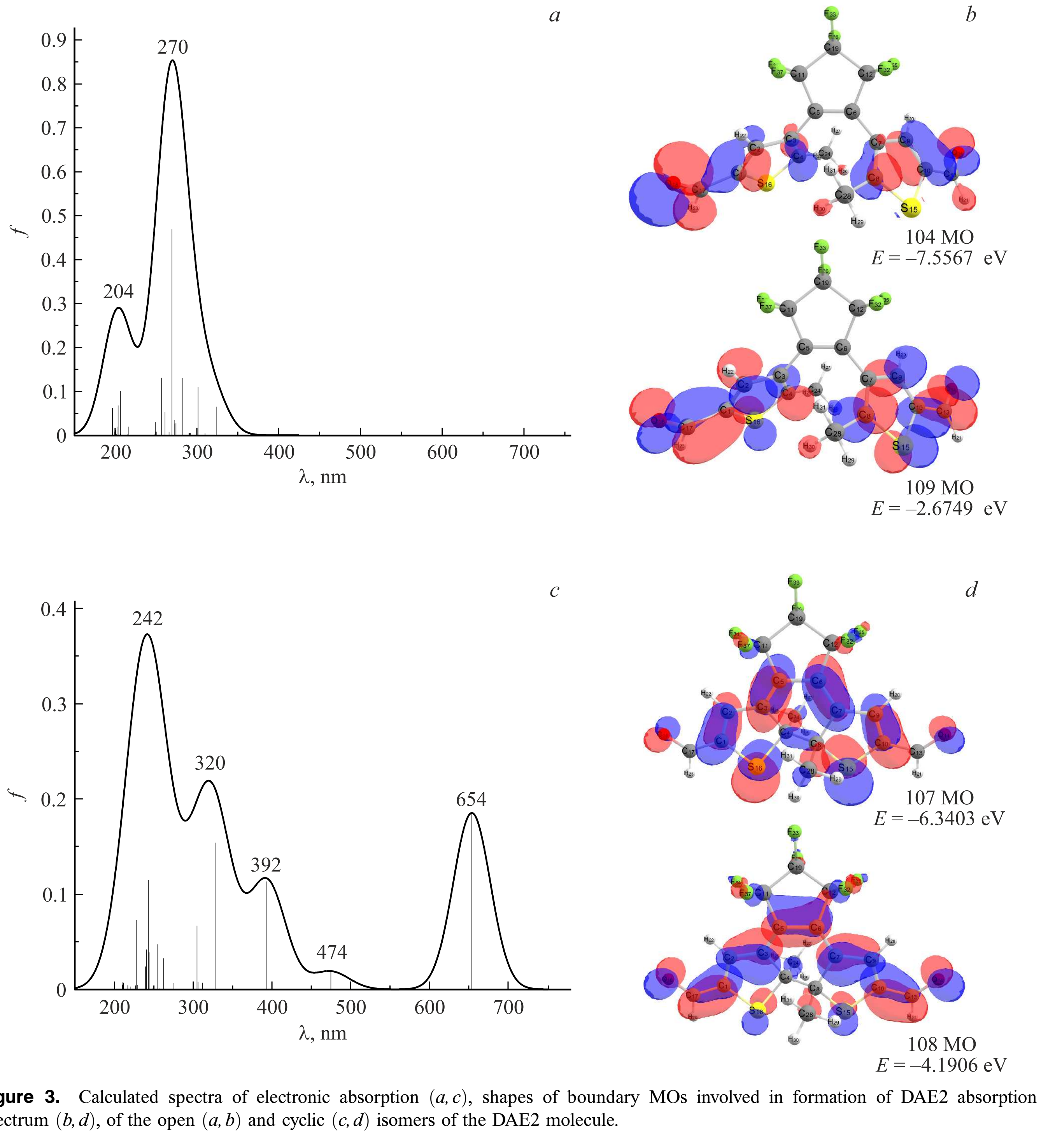


Figure 3. Calculated spectra of electronic absorption (a, c), shapes of boundary MOs involved in formation of DAE2 absorption spectrum (b, d), of the open (a, b) and cyclic (c, d) isomers of the DAE2 molecule.

to the absorption band at 418 nm, so does from the 112 MO (HOMO) to the 113 MO (LUMO) — to the absorption band at 577 nm (Fig. 2, *d*). Irradiation of the DAE1 molecule within the long-wave absorption band of the cyclic form B results in the $\pi - \pi^*$-transition $S_0 \rightarrow S_1$, localized in the alken fragment and in the thiophen rings and the rings coupled thereto, which is related to char redistribution in these fragments (at the same time the charge is transferred from the sulphur atoms). It significantly improves electrical conductivity of the DAE1 molecule (by generating a developed π-electron system of the coupled cycles), thereby allowing this compound being promising for use in the photocontrolled electric-signal switches.

The quantum chemical calculations have demonstrated similarity of the electron structure of the studied compounds DAE1-DAE4 and its modifications as a result of isomerization. The above analysis of the calculation results for the DAE1 compound is true for other compounds DAE2-

Table 2. Electronic structure of the open and cyclic forms of the DAE2 molecule as calculated relative to the main peaks

Configuration	State	Wave length, nm	Transition energy, eV	Decomposition of wave functions along singly excited configuration	Oscillator strength f
Open	$S_0 \rightarrow S_{13}$	269	4.60	$-0.67(104 \rightarrow 109) - 0.36(105 \rightarrow 109) +$ $0.32(103 \rightarrow 110) - 0.31(106 \rightarrow 109)$	0.47
	$S_0 \rightarrow S_9$	282	4.40	$0.88(103 \rightarrow 108) + 0.23(103 \rightarrow 110) +$ $0.22(107 \rightarrow 110) + 0.21(104 \rightarrow 109)$	0.13
	$S_0 \rightarrow S_5$	301	4.12	$-0.81(107 \rightarrow 110) + 0.41(107 \rightarrow 108) +$ $0.29(103 \rightarrow 108)$	0.11
Cyclic	$S_0 \rightarrow S_6$	328	3.78	$-0.89(103 \rightarrow 108) - 0.39(107 \rightarrow 110) -$ $0.17(106 \rightarrow 109)$	0.15
	$S_0 \rightarrow S_5$	393	3.15	$0.91(107 \rightarrow 109) - 0.34(106 \rightarrow 108) -$ $0.18(102 \rightarrow 108)$	0.11
	$S_0 \rightarrow S_1$	653	1.90	$1(107 \rightarrow 108) - 0.16(108 \rightarrow 107)$	0.19

Table 3. Electronic structure of the open and cyclic forms of the DAE3 molecule as calculated relative to the main peaks

Configuration	State	Wave length, nm	Transition energy, eV	Decomposition of wave functions along singly excited configuration	Oscillator strength, f f
Open	$S_0 \rightarrow S_{11}$	247	5.01	$-0.83(88 \rightarrow 93) - 0.31(88 \rightarrow 92) +$ $0.28(91 \rightarrow 94) - 0.19(89 \rightarrow 92)$	0.16
	$S_0 \rightarrow S_9$	256	4.84	$0.85(91 \rightarrow 94) - 0.27(89 \rightarrow 93) -$ $0.26(88 \rightarrow 93) - 0.21(91 \rightarrow 95)$	0.14
	$S_0 \rightarrow S_7$	262	4.74	$0.72(89 \rightarrow 92) + 0.60(90 \rightarrow 93) -$ $0.21(88 \rightarrow 93) + 0.15(91 \rightarrow 92)$	0.55
	$S_0 \rightarrow S_1$	333	3.72	$-0.98(91 \rightarrow 92) + 0.12(89 \rightarrow 92) + 0.11(90 \rightarrow 93)$	0.03
Cyclic	$S_0 \rightarrow S_7$	312	3.97	$0.91(89 \rightarrow 92) - 0.26(91 \rightarrow 95) + 0.24(88 \rightarrow 92)$	0.15
	$S_0 \rightarrow S_3$	358	3.47	$-0.89(91 \rightarrow 93) + 0.32(90 \rightarrow 92) +$ $0.20(91 \rightarrow 94) + 0.19(86 \rightarrow 92)$	0.11
	$S_0 \rightarrow S_1$	572	2.17	$-0.99(91 \rightarrow 92) - 0.14(92 \rightarrow 91) - 0.11(89 \rightarrow 92)$	0.22

Table 4. Electron structure of the open and cyclic forms of the DAE4 molecule as calculated relative to the main peaks

Configuration	State	Wave length, nm	Transition energy, eV	Decomposition of wave functions along singly excited configuration	Oscillator strength f
Open	$S_0 \rightarrow S_{11}$	270	4.59	$-0.95(111 \rightarrow 118) - 0.21(114 \rightarrow 119)$	0.23
	$S_0 \rightarrow S_7$	285	4.35	$-0.78(115 \rightarrow 118) + 0.32(116 \rightarrow 119) -$ $0.30(114 \rightarrow 119) + 0.25(115 \rightarrow 119)$	0.47
	$S_0 \rightarrow S_3$	336	3.69	$-0.94(117 \rightarrow 118) - 0.28(117 \rightarrow 119)$	0.08
Cyclic	$S_0 \rightarrow S_{27}$	233	5.33	$-0.57(116 \rightarrow 121) + 0.48(112 \rightarrow 119) -$ $0.46(116 \rightarrow 123)$	0.70
	$S_0 \rightarrow S_6$	362	3.43	$-0.97(115 \rightarrow 118) + 0.15(117 \rightarrow 118)$	0.39
	$S_0 \rightarrow S_1$	522	2.38	$0.99(117 \rightarrow 118) + 0.15(115 \rightarrow 118)$	0.40

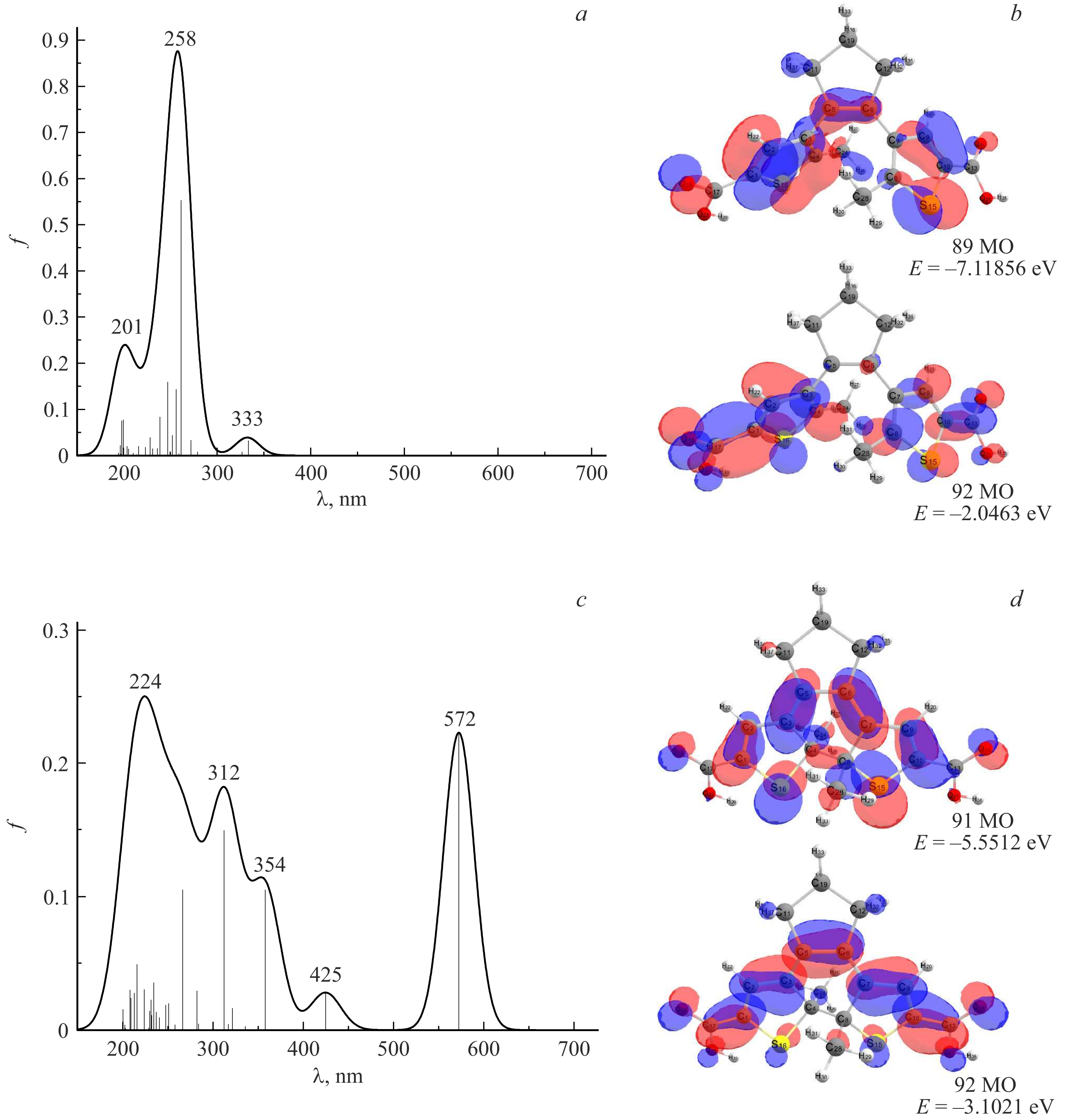


Figure 4. Calculated spectra of electronic absorption (*a, c*), shapes of boundary MOs involved in formation of DAE3 absorption spectrum (*b, d*), of the open (*a, b*) and cyclic (*c, d*) isomers of the DAE3 molecule.

DAE4 as well (Fig. 3–5, Table 2–4). At the same time, the form of the boundary molecular orbitals (Fig. 2–5) has been analyzed to show that the open isomer of the DAE2 compound is somewhat differs in its electron properties from the other studied compounds. The DAE2 molecule (unlike DAE1, DAE3, DAE4) in its original (open) form has no charge localized on the alken fragment. UV irradiation of the DAE2 molecule within the long-wave absorption band of the open isomer A makes the transition $S_0 \rightarrow S_{13}$, which is related to charge redistribution in the thiophen rings and the fragments couple thereto. The other transitions of the open form of the DAE2 molecule to the excited states have lower probabilities (the oscillator forces) due to symmetry of the DAE2 molecule. In the open isomers of the other DAE compounds studied in the paper, the charge is transferred from the alken

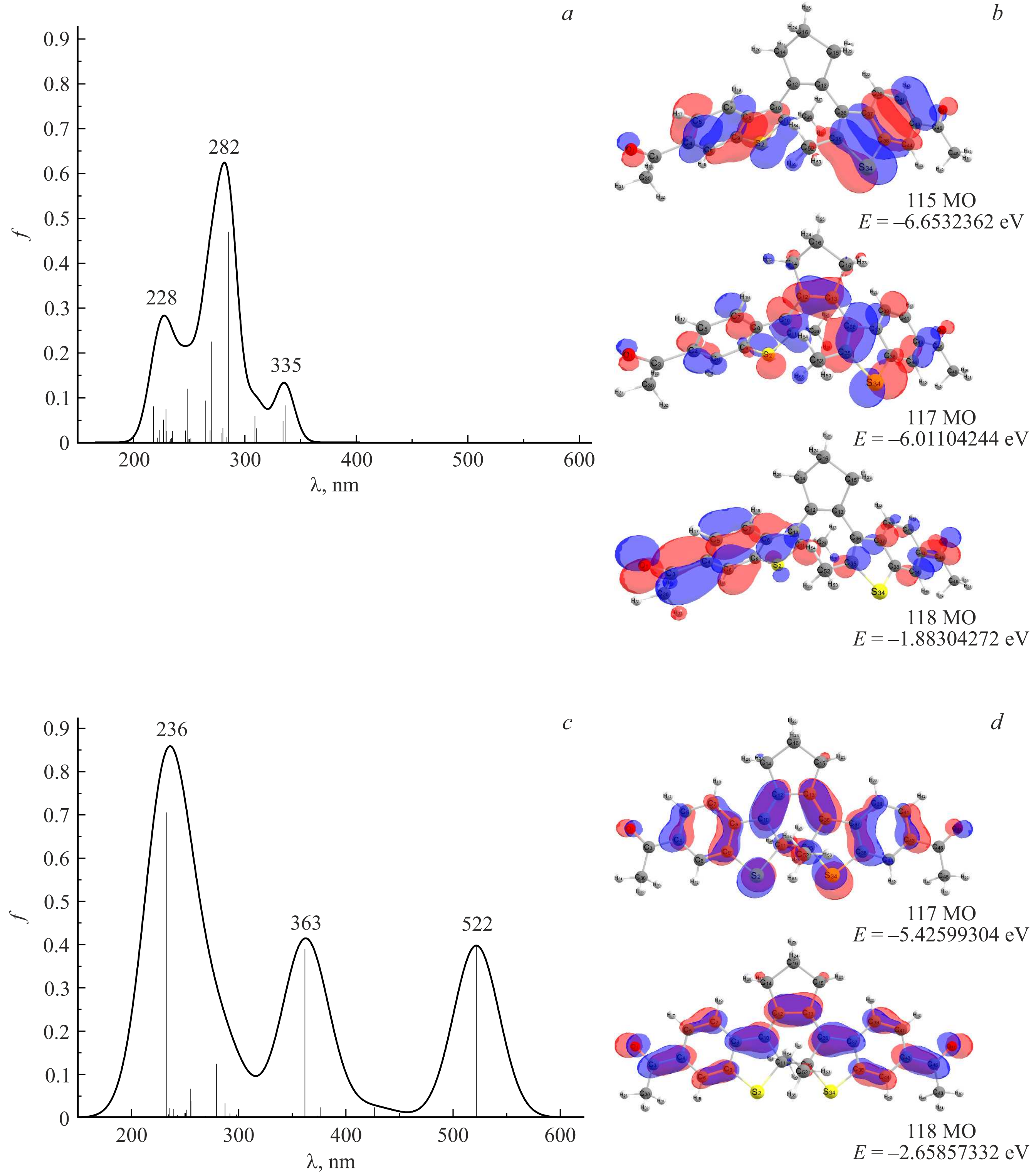


Figure 5. Calculated spectra of electronic absorption (a, c), shapes of boundary MOs involved in formation of DAE4 absorption spectrum (b, d), of the open (a, b) and cyclic (c, d) isomers of the DAE4 molecule.

fragment to the thiophen rings and the chains coupled thereto.

The analysis of the boundary molecular orbitals of the compounds DAE1-DAE4 included calculation of ionization energy $IP = -E_{\text{HOMO}}$, electron affinity $E_A = -E_{\text{LUMO}}$, total hardness $\eta = (IP - EA)/2$, band gap width $E_g = E_{\text{LUMO}} - E_{\text{HOMO}}$, electrophilicity $\omega = \chi^2/2\eta$, electronegativity $\chi = (IP + EA)/2$, electron chemical po-

Table 5. Calculated electronic properties of the open and cyclic isomers of the DAE molecules

DAE	Isomer	E_{HOMO}, eV	E_{LUMO}, eV	IP, eV	EA, eV	E_g, eV	μ, eV	η, eV	S, eV	χ, eV	ω, eV
DAE1	Open	−5.97	−1.94	5.97	1.94	4.03	−3.96	2.02	0.50	3.96	3.88
	Cyclic	−4.96	−2.56	4.96	2.56	2.40	−3.76	1.20	0.83	3.76	5.89
DAE2	Open	−7.23	−2.84	7.23	2.84	4.39	−5.04	2.20	0.46	5.04	5.77
	Cyclic	−6.34	−4.19	6.34	4.19	2.15	−5.27	1.08	0.93	5.27	12.89
DAE3	Open	−6.30	−2.05	6.30	2.05	4.25	−4.18	2.13	0.47	4.18	4.10
	Cyclic	−5.55	−3.10	5.55	3.10	2.45	−4.33	1.23	0.82	4.33	7.63
DAE4	Open	−6.01	−1.88	6.01	1.88	4.13	−3.95	2.07	0.48	3.95	3.77
	Cyclic	−5.43	−2.66	5.43	2.66	2.77	−4.05	1.39	0.72	4.05	5.91

tential $\mu = -(IP + EA)/2$ and chemical softness $S = 1/\eta$. The results of the analysis of the boundary molecular orbitals of all the DAE compounds studied in the paper are shown in Table 5.

The parameters shown in Table 5 define the electron and optical properties of the compounds. The orbitals HOMO and LUMO act as electron donors and electron acceptors, respectively. The ionization energy (IP) is defined as the quantity of energy required to remove the electron out of the molecule. The electron affinity (EA) is defined as the energy release or absorbed when the electron is added into the system. The electronegativity is defined as capability of the atom or the atom group to attract electrons to itself. The hardness may be defined as a property, which shows both stability and reactivity. The absolute hardness corresponds to the energy disruption of HOMO-LUMO. The molecule with low energy disruption has high chemical reactivity, low kinetic stability and is a soft molecule, whereas the hard molecule has a higher energy disruption. The chemical softness (S) — a measure of a capability of the atom or the atom group to accept electrons. The electrophilicity index (ω) defines a capability of chemical particles to accept electrons. The strong nucleophile is characterized by a lower value of μ, ω, so is the strong electrophile — by a higher value of μ, ω.

It is obvious that the differences of the molecular structures of the compounds exhibit in the differences of the electron properties. The biggest difference of the properties are in the DAE2 compound (as compared to the other compounds). It is largely due to the presence of fluorine atoms. As a result, the DAE2 molecule has high electrophile properties, so has its cyclic isomer B, especially.The calculation results shown in Table 5 can be used for optimizing selection of the functionalized photochromic diarylethenes or the targeted synthesis thereof, which are used in the molecular electronics and photonics devices (including the nanocomposite organico-inorganic ones).

Conclusions

Using spectrophotometry and quantum chemistry methods, photoinduced changes in the electronic spectra of photochromic DAEs of various structures were studied and analyzed. The electronic absorption spectra of DAE and molecular orbitals involved in the formation of these spectra were calculated. The results of quantum chemical calculations are consistent with the experimental spectrophotometric data of DAE. An analysis of the boundary molecular orbitals of the studied DAEs was carried out and, on its basis, their electronic properties were determined and analyzed. Significant features of the electronic structure of the DAE2 molecule (in comparison with other compounds) were discovered, which is also manifested in the calculated electronic parameters (in particular, an increased value of electrophilicity). The results obtained can be used to optimize the selection or targeted synthesis of photochromic DAEs (with specified properties) used in various nanostructured systems for molecular electronics and photonics devices.

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Conflict of interest

The authors declare that they have no conflict of interest.

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