Full Articles

A possible mechanism of image recognition by molecules and the design of receiving-and-transforming systems

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The problem of the mechanism of image recognition by a complex molecular system based on a set of independent primary signals is considered. Image recognition is interpreted as the appearance of a structural transformation in one remote receiving-and-recording center taking place when and only when all the primary signals have appeared. This principle ensures the most economical pattern for the information and energy transfer. The necessary properties of the receiving-and-transforming molecular elements are discussed. The task of designing a receiving-and-transforming molecular system is reduced to solution of either a formal logical equation or a set of kinetic differential equations of a special form. Examples of computer simulations are given.

Key words: image recognition, receiving-and-transforming molecular systems, design.

It is known that complex molecular systems, in particular, supramolecules possess the capacity for image recognition.[1] First of all, this recognition implies complementarity. The simplest complementarity requirement is based on the Fischer "key—lock" principle.[2] However, this geometric principle is only necessary but not sufficient for performing the recognition function. It is clear that an appropriate spatial arrangement of the receptor near the substrate does not by itself ensure recognition. A reaction between the receptor and the substrate (*e.g.*, addition of functional groups) that could be perceived as a signal indicating that the "key" has got into the "lock" is also required. These requirements lead to the double complementariry principle: recognition is possible only in the case where the partners forming an associate occur in the optimal information relationship, *i.e.*, they not only have appropriate geometric configurations but also contain functional groups able to react with each other to form the required signals. The term "signal" is used to mean local structural changes taking place in particular fragments of the molecular associate such as the formation or cleavage of covalent or hydrogen bonds and so on. All these issues have long been discussed in the literature and are actively used as fundamental principles in the design of supramolecular structures with specified properties.[1]

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Unfortunately, the double complementarity principle still cannot explain exhaustively the proper process of information conversion by supramolecular systems. Let us consider the principles inherent in the mathematical problem of image recognition. It is known that in the definition of this problem, a complicated object is characterized by a set of features, which ideally can be described by numbers. In this case, the object is represented by a point in a multidimensional space of features. If this point falls into a certain area, this proves the belonging of the object to a given class. Thus, "image recognition" is reduced to ascertainment of this belonging.

In the formal mathematical logic (and, in general, in obtaining a deductive inference from initial premises), a complicated object of any nature can be characterized by a set of features and actions that describe this object and that are perceived or produced by this object. In this case, image recognition does not mean some detection of the set of these actions, but implies instead obtaining of one and only one response on the basis of many premises. This single response must be uniquely related to the whole set of initial features and, hence, provide a full characterization of the object under study. It can be seen that this situation bears an analogy with the problem of image recognition based on the position of a point in a multidimensional space. Indeed, in both the former and the latter case, a single inference is deduced from numerous initial premises; thus, information is a sort of concentrated into a single inference.

In view of this fact, image recognition by supramolecular systems should imply such a response of the system to the appearance of many primary signals that would generate a single and unambiguous answer.

If the notion "image recognition by molecules" is defined on this basis as compression of the initial information, the fact of formation of the receptor—substrate associate and the appearance of local signals in different sections of the molecular space should be taken as the primary event of the whole process. In the second stage, the information on different signals should be transferred to one spatial region where it will cause the required action. We will call this receptor region the readout unit and the corresponding action will be called information readout.

In real physical systems, the collection of information from numerous sources and transfer of this information to a single center implies gathering and directed transfer of primary signals to this center. It has been shown previously[3] that in molecular structures, the transfer of signals from different sites of generation to a local section of molecular space (readout unit) is accomplished as consecutive isomerization steps, which result, under particular conditions, in the generation and ordered motion of quasi-particles called vibrons. The structural features and the motion of these quasi-particles are responsible for the direction and localization of the corresponding energy fluxes. It is clear that dissipation of these fluxes in space would be unfavorable from the energy standpoint and would decrease the efficiency of the whole process.

It follows from the foregoing that the recognizing molecular system should, first, be able to form space-separated primary signals upon a receptor—substrate interaction and, second, be able to transfer the information from these signals as directed energy pulses to the readout units (there may be several of them), in which the transferred energy induces changes, giving rise to secondary signals, and, third, it should collect all information in a single final readout unit. The chart of the recognizing system illustrating this idea is presented in Fig. 1.

Previously,[4] we pointed out the fact that any intramolecular processes can be described in terms of logical propositions and logical equations (by means of Boolean algebra[5]).

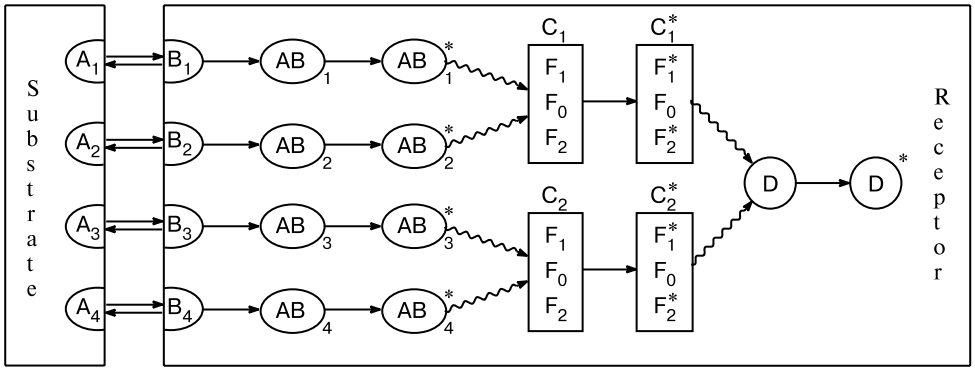

Fig. 1. Principle of image recognition by a molecular structure; A_1, A_2, A_3, and A_4 are functional groups of the substrate; B_1, B_2, B_3, and B_4 are the complementary groups of the receptor; C_1 and C_2 are the intermediate readout units with substructures F_1, F_0, and F_2 (the broken arrows show the paths of information (energy) transfer); D is the final readout unit. The characters $(AB)_i^*$, C_k^*, and D^* are used to designate intramolecular structures isomerized upon reception of the initial signals.

In particular, the set of logical equations corresponding to Fig. 1 would have the form shown below.

$$\left.\begin{aligned}\varphi_1 &= \{(A_i \wedge B_i) \rightarrow (AB)_i\}\\ \varphi_2 &= \{(AB)_i = (AB)_i^*\}\\ \varphi_3 &= \{(AB)_i = A_i\}\end{aligned}\right\}\ i = 1,\ \ldots,\ 4$$

$$\varphi_4 = \{[(AB)_1^* \wedge (AB)_2^*] \rightarrow C_1\}$$

$$\varphi_5 = \{C_1 = C_1^*\}$$

$$\varphi_6 = \{[(AB)_3^* \wedge (AB)_4^*] \rightarrow C_2\}$$

$$\varphi_7 = \{C_2 = C_2^*\}$$

$$\varphi_8 = \{(C_1^* \wedge C_2^*) \rightarrow D\}$$

$$\varphi_9 = \{D = D^*\}$$

The sign "$\wedge$" corresponds to conjunction (logical multiplication) and in the "natural language", it reproduces the proposition "A_i and B_i, or A_i as well as B_i". The sign "=" transmits the statement "$(AB)_i$ if and only if $(AB)_i^*$, and *vice versa*". The sign "$\rightarrow$" (implication) characterizes the cause-and-effect relations: "if $(A_i \wedge B_i)$, then $(AB)_i$". Altogether, this is outlined as follows.

If the substrate and the receptor contain groups A_i and B_i, the addition of the substrate to the receptor gives initially groups $(AB)_i$ and further their isomeric states $(AB)_i^*$. The presence of the groups $(AB)_i$ indicates unambiguously the presence of groups A_i. The presence of states $(AB)_1$ and $(AB)_2$ gives rise to the structure C_1 and the isomeric state C_1^*, while the presence of states $(AB)_3$ and $(AB)_4$ entails the presence of structure C_2 and the isomeric state C_2^*. The appearance of states C_1^* and C_2^* dictates the appearance of structure D and isomeric structure D^*.

All the elementary statements are combined to form one Boolean function $T = (\varphi_1 \wedge \varphi_2 \wedge \varphi_3 \ldots \wedge \varphi_9)$, in which all notions included in the consideration are interrelated.

Subsequently, this function is used to solve the logical equation for the A_i groups. It is convenient to use the method of image numbers for this purpose.[6]

According to the above definition of recognition of a complicated object as concentration of the information, the fact of appearance of structure D^* means that the recognition has taken place. If the spatial arrangement of groups $A_1, \ldots, A_4$ is specified by the geometric complementarity principle or their sequence is known, one can restore the complete chemical structure of the substrate.

Now we will discuss some conditions of principle under which the above-described mechanism of recognition of the substrate structure by the receptor can be effective.

1. It is clear that the interactions of functional groups A_i and B_i of the receptor and the substrate should be independent of one another. This means that the formation of the associated group $(AB)_i$ should not affect the occurrence of reaction $A_j + B_j = (AB)_j$ ($j \neq i$). Since the intramolecular effects (in the first approximation) are relatively weak, the reactions in large molecular entities are known to be mainly determined by the structure and the properties of local reaction centers. Such reaction centers (if there are several of them) can act independently if they are remote from one another. Moreover, under such conditions, the reactions $A_i + B_i = (AB)_i$ will occur at different points in time depending on how the geometric shape of the receptor is adjusted to the substrate shape.

2. For the recognition event to take place, the primary signals should not only be generated (this means the formation of groups $(AB)_i$ or changes in groups B_i, *e.g.*, under the action of light) but they should also independently impact on centers C_k at different instants of time. This impact requires directed transfer of energy from the region of generation of the primary signal to the readout unit, *i.e.*, from $(AB)_i$ to C_k. It was shown[3] that this transfer can take place *via* a directed migration of a bond or a side group along a nonuniform one-dimensional molecular chain as a result of an appropriate sequence of local isomer—isomer transformations. This directed migration is possible where upon the $A_i + B_i = (AB)_i$ addition event, the $(AB)_i$ group becomes excited and can transfer this excitation to the chain (transition of $(AB)_i$ into $(AB)_i^*$) and the structure $(AB)_i^*R$ is able to undergo successive isomerization steps. Here the character R stands for the structure (the chain + C_k). The conditions under which each isomerization step is most probable have been elucidated previously.[7] It was also shown[3] that the process of signal transfer should end in the step of "falling of the $(AB)_i^*R$ structure into the deepest potential well."

3. The process described above (see point 2) ensures the transfer of energy to center C_k. The perception of information from the primary fragment $(AB)_i$ means that the structure of this center also changes. This would ensure recording of the obtained information. This record should not be erased or distorted when the same C_k center receives a signal from a different primary source. Thus it follows that the structure of center C_k should be complex, so that changes in one its part would not affect the properties of the other part. In addition, the C_k center should be able to switch to the final state C_k^* after receiving two signals. This final state should always arise after the action of both signals but not after either of the signals alone.

From the standpoint of mathematical logic, this means that the C_k center operates as a unit performing logical multiplication. As applied to chemical structures, this looks as follows. Let the C_k center consist of three groups: F_1, F_0, and F_2 (to simplify the designations, we do not

specify whether or not the F_i groups are the same for different C_k).

The F_1 and F_2 groups independently perceive the signals from groups $(AB)_1$ and $(AB)_2$ and are thus converted into isomeric states F_1^* and F_2^*.

If the structure of groups F_1^*, F_0, and F_2^* is again capable of isomerization, for example, *via* the formation of the bond between F_1^* and F_2^*, this will give the desired logical element.

In the logical language, the first step in the sequence of recognition should correspond to the $(A_i \wedge B_i) \rightarrow (AB)_i$ operation (if there is a pair of partners A_i and B_i, then an associated group $(AB)_i$ is formed). The next step corresponds to the $(AB)_1 \rightarrow F_1^*$ and $(AB)_2 \rightarrow F_2^*$ operations.

The operation of C_k transformation into C_k^*, *i.e.*, the appearance of a readout signal, can also be written as a logical relation: $(F_1^* \wedge F_2^*) \rightarrow C_1^*$. If we continue this consideration, we will conclude that signals from the C_1^* and C_2^* centers can also be "combined" due to operation of the D center.

Thus, the whole mechanism of "initial image recognition" is described as a sequence of appropriate isomer—isomer transformations.

Since supramolecular structures possess an enormous potential in this respect, the above-described mechanism of information perception and processing does not appear unique. The probability of this process would actually be determined only by the probabilities of local isomer—isomer transitions. A method for calculating these probabilities has been developed.[7] A model description of the probabilities of nonradiative isomer—isomer transitions taking into account the quantum beats has been proposed.[8]

Now we pass to the problem of designing the receiving-and-transforming supramolecular systems for image recognition. It should be borne in mind that elementary molecular processes (chemical addition or decomposition reactions, light absorption or emission, phototransformation, *etc.*) can be associated with elementary statements and logical equations and the relations between the events can be expressed as implications.

If an "image" has been characterized by a combination of features (for example, functional groups or the spectral composition upon irradiation with light), the primary interaction events between two complex systems can be described by a set of elementary statements and implications. The next task is to select groups and intramolecular rearrangements such that they would compress the initial information until a single response is formed.

The whole sequence of operations will be described by a Boolean function, and the operation of recognition, by a logical equation. By solving this equation with respect to the initial structural groups, one can ascertain, first, whether the maximum "compression" of the information resulting in one final signal would produce the desired response (meaning that the initial object has been fully recognized), second, whether or not this response would be unambiguous, third, wouldn´t this make the designed system self-contradictory (meaning that the logical elements of the whole system have been chosen improperly), and, fourth, wouldn´t the designed system become redundant.

These questions can be easily answered by using computer simulation (mathematical modeling) techniques. As noted above, for this purpose, one should properly describe the action of each elementary operation using logical terms, then generate a logical equation, and solve it.

Let us consider a simple example. Let there be a chromophore group (B) able to absorb electromagnetic radiation with frequencies ω_1, ω_2, and ω_3. We assume that after an absorption event, due to spontaneous radiation and transitions from upper to lower levels, the chromophore group passes into an excited state from which a nonradiative transition giving another isomeric form (B^*) can take place. We take that this form is rather stable and can be detected in an appropriate experiment. Then the formation of this form can be regarded as a way of recording the initial information (the action of an external electromagnetic field).

Since isomer B^* can appear, according to the assumption, upon separate absorption of waves with frequencies ω_1, ω_2, and ω_3 and the initial spectrum was not defined (*i.e.*, one can suspect the presence of either waves with only one frequency, ω_1, ω_2 or ω_3, or with any combination of these frequencies), the corresponding logical equation will have the form

$$(\omega_1 \vee \omega_2 \vee \omega_3) \rightarrow B^*.$$

The sign "$\vee$" designates the logical addition. The verbal notation is as follows: "if there is radiation with frequency ω_1, or ω_2, or ω_3, or with any combination of these frequencies, a signal corresponding to the appearance of isomer B^* is generated."

This logical correlation corresponds to the following reverse correlation:

$$B^* \rightarrow \Big[(\omega_1 \wedge \overline{\omega}_2 \wedge \overline{\omega}_3) \vee (\overline{\omega}_1 \wedge \omega_2 \wedge \overline{\omega}_3) \vee (\overline{\omega}_1 \wedge \overline{\omega}_2 \wedge \omega_3) \vee \vee (\omega_1 \wedge \omega_2 \wedge \overline{\omega}_3) \vee (\omega_1 \wedge \overline{\omega}_2 \wedge \omega_3) \vee \vee (\overline{\omega}_1 \wedge \omega_2 \wedge \omega_3) \vee (\omega_1 \wedge \omega_2 \wedge \omega_3)\Big].$$

The bar above the character ω implies a negation operation.

The answer is ambiguous and is read as follows: "the appearance of form B^* means that the irradiating light could have contained waves with any of the frequencies ω_1, ω_2, or ω_3 as either a single frequency, or a pair combination, or all three of them."

It can be seen that the presence of one chromophore is inadequate for solving the problem of recognition of the

spectral composition of an external electromagnetic field. In order to determine the composition of the incident light, it is necessary to complicate the receiving system.

Let us assume that the chosen chromophore is able to isomerize along two channels (B passes into either B^* or B^{**}), the additional channel being activated only upon absorption of light with the frequency ω_1. The logical operations corresponding to the action of this chromophore can be written as follows:

$$\omega_1 \rightarrow B^{**}$$

$$(\omega_1 \vee \omega_2 \vee \omega_3) \rightarrow \left[(B^* \wedge \overline{B}^{**}) \vee (\overline{B}^* \wedge B^{**})\right].$$

It is taken into account that deexcitation of the chromophore after absorption of electromagnetic energy can furnish either isomer B^{**} or isomer B^* but not both of them.

If both isomers B^* and B^{**} are rather stable, either state B^* or state B^{**} being observed, we actually have two separate independent problems. This can be detected by the appearance of logical incompatibility. This contradiction is eliminated by assuming that the chromophore is complex, isomerizations of B^* and B^{**} take place in different groups, and the next isomerization step (which no longer requires light) giving rise to structure B^{***} is possible. The logical relation describing this step can be written as $(B^* \wedge B^{**}) \rightarrow B^{***}$.

Thus we obtain

$$B^{***} \rightarrow \left[(\omega_1 \wedge \overline{\omega}_2 \wedge \overline{\omega}_3) \vee (\omega_1 \wedge \omega_2 \wedge \overline{\omega}_3) \vee \vee (\omega_1 \wedge \overline{\omega}_2 \wedge \omega_3) \vee (\omega_1 \wedge \omega_2 \wedge \omega_3)\right].$$

The degree of ambiguity in image recognition has diminished relative to the previous case, although this still does not provide a unique response. The mathematical simulation that we carried out for complicated objects shows that with the knowledge of the logical relations corresponding to real elementary processes in molecules (which can be seen to be very simple), it is possible to represent all the relations between the initial and resulting values (notions) by Boolean functions.

If $T(\alpha,\beta)$ is the Boolean function describing all the relations between the notions being introduced, $R(\alpha)$ is the Boolean function corresponding to the "final" values or notions, and $f(\beta)$ is the Boolean function describing the initial image, the logical analysis can be expressed symbolically in the form

$$T(\alpha,\beta) \rightarrow \{R(\alpha) \rightarrow f(\beta)\}.$$

This is the logical equation with respect to $f(\beta)$.

Thus it follows that the first problem of designing a supramolecular system as a logical element for image recognition can be solved by means of Boolean algebra *via* a rather primitive operation.

Analysis of the possibility of creating a receiving-and-transforming supramolecular system on the basis of formal logical correlations suffers from two drawbacks. It does not take into account, first, the factors of time and, second, the decrease in the efficiency due to the dissipation of energy and information in the molecular space. These drawbacks can, however, be eliminated by connecting the process of image recognition to the processes of transfer of state populations, both initial and intermediate ones, to form a single receiving-and-recording center.

This approach can be plainly explained by using a simplified pattern where information is perceived only from two initial receivers and transferred subsequently to a single center. The receipt of the initial information, *i.e.*, the formation of group $(AB)_1$ (or transfer of group B to the excited state B^* upon light absorption in the example given above) can be attributed to the initial population of state **1**. The initial state **2** will be described in a similar way. It is assumed that these states can be populated independently of each other at different points in time.

Let us take that state **1** is connected to state **3** and can transfer its population to this state with a particular probability α_{13}. This means that optical or nonradiative $(AB)_1 \rightarrow (AB)_1^*$ transition can take place. The probability of reverse transition is neglected, as it does not affect the generality of the subsequent inferences. We assume that states **2** and **4** are interrelated in a similar way.

As noted above, "compression" of the initial information coming from two sources and its transfer to a single center requires the existence of structural element C_1 able to be transformed (*e.g.*, able to pass into a different isomer C_1^*) within its two independent sections (F_1 and F_2). Isomerization taken place in two centers enables the transition into the new stable isomer (C_1^*). This would imply recording of information from two initial signal receivers, which takes place only in the case where both receivers has functioned.

This corresponds to the logical statement $(F_1^* \wedge F_2^*) \rightarrow C_2^*$.

Let us denote the resultant state as **7** (states **5** and **6** are described below). In terms of the populations (n_i) and the probabilities (α_{ij}) of transitions between states **3** and **4** and the resultant state **7** and taking into account the structure and the meaning of logical multiplication, we must consider that the population of state **7** is determined by the product of the populations of states **3** and **4**.

Now we introduce states **5** and **6**, the population outflow to which simulates the energy dissipation in the molecular space, which is useless as regards the transfer of information into a single center.

The whole process of reception and transfer of information can thus be written as a set of differential equations differing from a usual kinetic set of equations by the

fact that the state populations can appear in these equations as the products

$$dn_1/dt = -\alpha_{13}n_1, \qquad dn_2/dt = -\alpha_{24}n_2,$$

$$dn_3/dt = \alpha_{13}n_1 - \alpha_{35}n_3 - \alpha_{37}n_3,$$

$$dn_4/dt = \alpha_{24}n_2 - \alpha_{46}n_4 - \alpha_{47}n_4,$$

$$dn_5/dt = \alpha_{35}n_3, \qquad dn_6/dt = \alpha_{46}n_4,$$

$$dn_7/dt = (\alpha_{37} + \alpha_{47})n_3n_4.$$

Writing down such equations allows one to perform model mathematical experiments. Their results are shown in Fig. 2. Usually, it is considered that state **2** is populated independently of state **1** with some time lag. As was to be expected, this influences the beginning of population of state **7**, *i.e.*, recording the information indicating that both primary receiving centers, **1** and **2**, have operated.

Figure 2, *a* corresponds to the situation where the probabilities of transitions from states **3** and **4** to states **5** and **6**, respectively, are rather high and, hence, the loss of energy or useful signals is possible. The populations of states **5** and **6** are very high in this case, while the population of state **7** is low. The efficiency of the receiving-and-transforming unit is insufficient.

As expected, a decrease in the probabilities of $\mathbf{3} \rightarrow \mathbf{5}$ and $\mathbf{4} \rightarrow \mathbf{6}$ transitions increases substantially the desired signal (population of state **7**) (see Fig. 2, *b*).

Figure 2, *c* corresponds to a model in which the probability of the $\mathbf{2} \rightarrow \mathbf{4}$ transition is lower than the probability of the $\mathbf{1} \rightarrow \mathbf{3}$ transition. Since the population of state **3** during the early period of time (0—5 arbitrary units of the time scale) does not depend at all on the action of the second channel, initially, the population curves of state **3** are totally identical; they become different only after the onset of operation of the second channel of the primary signal. Since the information outflow to states **5** and **6** is still relatively low, the population of the resultant state **7** is again high, although it has decreased with respect to that presented in Fig. 2, *b*. This is due to the decrease in the maximum population of state **4** caused by the fact that the signal transfer along the $\mathbf{2} \rightarrow \mathbf{4}$ channel is slower than that along the $\mathbf{1} \rightarrow \mathbf{3}$ channel.

The results of the experiment in which the probability of signal transfer along the first channel is five times lower than that along the second channel are presented in Fig. 3. The population of the final state still remains high!

We considered the situation where the signal is accumulated from two sources.

It is clear that one can consider both the action of the system presented in Fig. 1 and a more complicated system. This will increase the number of differential equations; in addition, it will be more difficult to take into account the time diversity in the action of the primary signal receivers. Generally, the problem remains rather

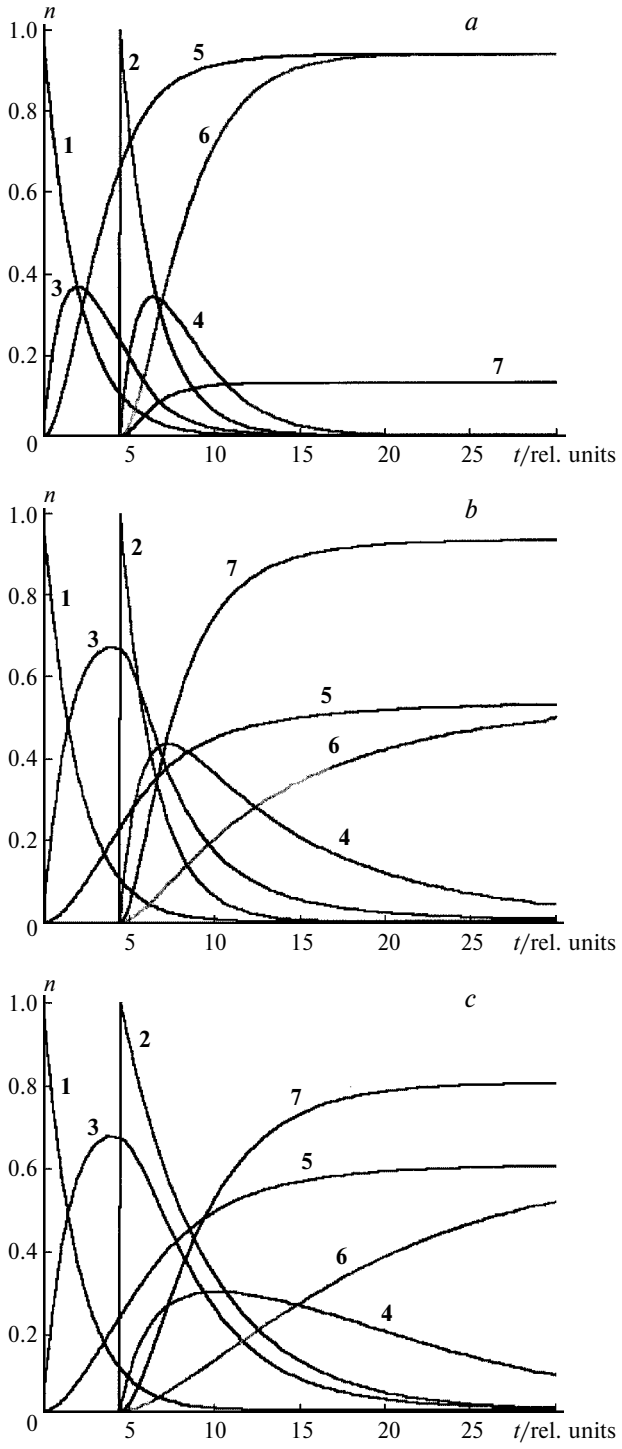

Fig. 2. Time variation of the populations (n) of states **1—7** under conditions of substantial dissipation in space (*a*) and where the probabilities of transitions α_{35} and α_{46} have been decreased fivefold (*b*, *c*); (*c*) the result of an experiment in which the probability of signal transfer along the second channel (the α_{24} coefficient) was decreased by 60% with respect to the probability of signal transfer along the first channel (the α_{13} coefficient). The vertical line marks the onset of operation of the second channel of reception of the primary information.

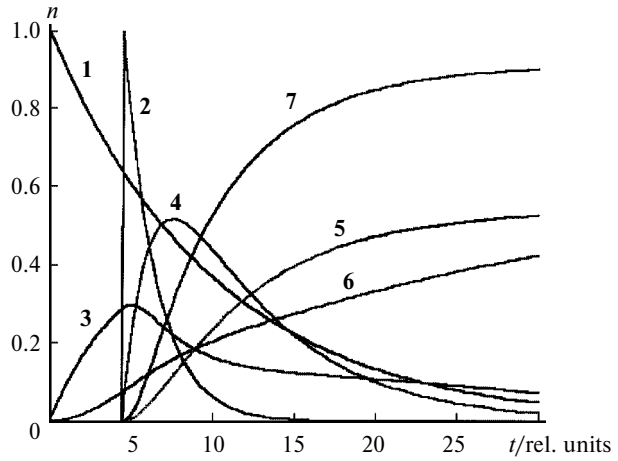

Fig. 3. Result of an experiment in which the probability of signal transfer along the first channel (α_{13}) is 5 times lower than that for the second channel (α_{24}).

clear and can be easily solved. Moreover, using the described rules, one can compile a general set of equations from blocks corresponding to separate, partial receiving-and-transforming units.

The next important and complex step is to pass to real chemical structures. This requires the knowledge of the properties of particular structural groups, the capabitity of describing them, and, most important, in-depth research into intramolecular isomerization processes and the transfer of signals during chain isomer—isomer transformations. Fortunately, one can rely in this field on the enormous chemical experience and the potential of theoretical estimates, in particular, on the basis of methods proposed previously.[7]

Thus, there is every reason for concluding that the idea of computer design of supramolecular systems for image recognition is not fantastic but it relies on the stable foundation already created in modern theoretical chemistry.

Of course, practical implementation of the approach described here would require a lot of effort, in particular, for developing the software. However, here one can also rely on many algorithms proposed previously and on the existing software.

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