The entropy generation in visual-pigment system by the absorption of light

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Abstract

We first showed a general theory that reception of information from the outside in a receptor system is accompanied both by an inflow of entropy and by a generation of entropy depending on the reliability of the actual reception mechanism. Then, considering a case for the absorption of light by the visual-pigment system of visual cell, we calculated the time(t)-dependent change in the number $N_2(t)$ of excited visual-pigments to obtain the entropy increase. We thus arrived at the following four conclusions: (1) One or two photons can be detected with a reliability of at least 54%; (2) In compensation for this detection, entropy $> 1.12 \times \log 2$ is generated; (3) An incident photon of frequency ν from a light source of temperature T_s yields an entropy of $h\nu/T_s$; and (4) Depending on the characteristics of the visual-pigment system, another entropy being different from (2) is generated in proportion to $N_2(t)$.   © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

About fifty years ago, Schrödinger proposed for the first time the very important view of the inflow and outflow of entropy in biological sys-tems [1]. In chapter 6 of his book, '*What is Life? — The Physical Aspect of the Living Cell*', published in 1944, he described that 'what an organism feeds upon is negative entropy, or, to put it less paradoxically, the essential thing in metabolism is that the organism succeeds in freeing itself from all the entropy it cannot help producing while alive' (see the section titled 'It Feeds on Negative Entropy').

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As can be seen from the phrase 'to put it less paradoxically', the first sentence for 'feeding negentropy' is only a 'paradoxical' representation of the second sentence of which the physical meaning can be expressed by the inequality

$$\Delta S_i - \Delta S_o < 0 \qquad (1)$$

where ΔS_i (ΔS_o) denotes the entropy flowed into (flowed out from) a biological system. Needless to say, this inequality indicates clearly that the biological system has an excellent mechanism for entropy transmission, in which the entropy ΔS_o released to the outside of the system is larger than the entropy ΔS_i absorbed into the inside, and that the system satisfying the inequality Eq. (1) appears to be in a state of 'negentropy'. It is also well known in thermodynamics that a system in a 'negentropy state' can perform work to its outside.

This paper was devoted to achieving the following three purposes: (1) To indicate the close relation between the reception of information and the inflow and generation of entropy by phenomenologically introducing the reliability of receiving information; (2) In the case of the absorption of light by the visual-pigment system in the outer segment of visual cell, to calculate the time-dependent change in the number of excited visual pigments, and then to obtain the entropy increase in the visual-pigment system by the absorption of light; and (3) To clarify the generation of entropy without introducing the reliability of receiving information.

The exposition of this paper is as follows. In Section 2, we consider a receptor system consisting of a number of identical molecules which receives stimulus information from the outside at a certain reliability. And assuming for simplicity that each molecule takes either of the two states (i.e. the ground ψ_1 and the excited state ψ_2), we show a general theory that the reception of information in the system is accompanied both by an inflow of entropy and by a generation of entropy depending on the reliability a of the actual mechanism of receiving information. In Section 3, the time-dependences of the number N_2 of excited visual pigments and the entropy S are determined

on the basis of the following two fundamental assumptions: (1) Transition moments of the visual-pigment chromophores are on the planes perpendicular to the incident light, and show a two-dimensional random orientation; (2) The optical absorption by the visual-pigment system acts upon Lambert−Beer's law. In Section 4, the following three fundamental quantities, by which the time-dependence of N_2 is determined, are parameterized: (1) The total number N of visual pigments; (2) The probability α of the non-radiative transition from ψ_2 to ψ_1 by the photoisomerization of visual-pigment chromophore; (3) The probability β of the radiative transition from ψ_2 to ψ_1 of visual-pigment chromophore. In addition, the intensity $I_p(\nu)$ of incident light, which is necessary to formulate β, is examined, and its minimum value I_{min} is determined in a reasonable way. In Section 5, the minimum value $N_2^{(min)}$ of N_2 is first determined, and then the relation between N and α is derived using $N_2^{(min)}$ to determine the values of N and α. Then, according to this parameterization, the time-dependent changes in N_2 and S are calculated, and the physical meanings of the primary results are revealed. Finally, in Section 6, the four conclusions (see the *abstract*) at which we thus arrived are summarized, and the propriety of the parameterization is discussed.

2. General theory for the inflow and generation of entropy accompanied by receiving information

What the term 'information transmission' means is no more than the phenomenon without reference to its underlying mechanism, and the most fundamental physical quantity that controls this phenomenon is entropy. Brillouin noted the importance of the concept 'negentropy' proposed by Schrödinger, and established a unique theory of information by investigating its relation with 'amount of information' [2]. In this section, to understand the relation between information and entropy, we will make it clear that the nature of the phenomenon of 'receiving information' is the 'inflow and generation of entropy'.

Let us consider a receptor system consisting of N identical molecules which obtains stimulus in-

formation from the outside with reliability a, under the assumption that each molecule takes either of the two states (the ground ψ_1 and the excited state ψ_2). And let the number of molecules in the state ψ_j be N_j in the receptor system at thermal equilibrium in the presence of external stimuli. Then, according to Boltzmann's principle

$$S = k\log[N!/(N_1!N_2!)] \tag{2}$$

and using Stirling's formula

$$\log(N!) \doteqdot N(\log N - 1), N \gg 1 \tag{3}$$

the entropy is expressed as follows:

$$S \doteqdot k\left(N\log N - \sum_{j=1}^{2} N_j \log N_j\right) \tag{4}$$

where k is Boltzmann's constant.

In the receptor systems at thermal equilibrium, the probability of finding a molecule in the state ψ_j is given by

$$p_j = N_j/N, \; j = 1,2 \tag{5}$$

With this definition, the probability that m molecules are found in ψ_1 and the remaining $N - m$ in ψ_2 is expressed as

$$p(m, N-m) \equiv \{N!/[m!(N-m)!]\}p_1^m p_2^{N-m} \tag{6}$$

which satisfies the equation

$$\sum_{m=0}^{N} p(m, N-m) = (p_1 + p_2)^N = 1 \tag{7}$$

Therefore, the probability that one or more molecules are found in the excited state ψ_2 is obtained as follows:

$$\sum_{m=0}^{N-1} p(m, N-m) = 1 - p_1^N \tag{8}$$

The condition for obtaining the stimulus infor-

mation at the reliability a can then be expressed as $1 - p_1^N = a$, from which p_1 is solved to be

$$p_1 = (1 - a)^{1/N} \tag{9}$$

Here, $a = 0.5$ means that the reliability of obtaining the information is 50%. Therefore, we have

$$N_1 = N(1 - a)^{1/N}, \quad N_2 = N - N_1 \tag{10}$$

based on the definition Eq. (5), and we can rewrite the entropy Eq. (4) in the form

$$S_a = S_a^{(1)} + S_a^{(2)};$$
$$S_a^{(1)} \equiv kN_2\log(N_1/N_2),$$
$$S_a^{(2)} \equiv k\log[1/(1-a)] \tag{11}$$

To understand the meaning of the first term $S_a^{(1)}$, we assume here that the receptor system obtains stimulus information mediated by certain substances to reach a thermal equilibrium at temperature T_e. Then, N_j is proportional to exp $(-E_j/kT_e)$, E_j being the energy of the state ψ_j, and $S_a^{(1)}$ is rewritten as follows:

$$S_a^{(1)} = N_2(\Delta E/T_e), \quad \Delta E \equiv E_2 - E_1 \tag{12}$$

If the volume of the receptor system remains constant, $\Delta E/T_e$ represents the increase in entropy resulting from the increase ΔE in internal energy caused by the external stimuli. Namely, the $S_a^{(1)}$ described above indicates that the entropy flowed from the outside into the receptor system is $\Delta E/T_e$ per excited molecule.

To examine whether the result Eq. (12) is adequate, let us assume that the carrier of stimulus information is photon derived from the thermal radiation field at temperature T_e, and that the receptor system is thermally equilibrated with the thermal radiation field. In this case, photoreceptive molecules having only two energy states selectively absorb photons of frequency $\Delta E/h \equiv \nu$

(h being Planck's constant), so that $S_a^{(1)}$ of Eq. (12) is rewritten as

$$S_a^{(1)} = N_2(h\nu/T_e) \tag{13}$$

This result completely agrees with that obtained in our previous study [3].

On the other hand, the second term $S_a^{(2)}$ in Eq. (11) indicates that another entropy generates depending on the reliability a of the means whereby stimulus information is obtained. At $a = 0.5$ (i.e. when the reliability is 50%), for example, $S_a^{(2)}$ is calculated to be

$$S_{0.5}^{(2)} = k \log 2, \quad k = 1.381 \times 10^{-16} \text{ [erg/K]} \tag{14}$$

which agrees with the result by Szilard [4] and Brillouin [2,5]. Generally, $S_a^{(2)}$ ensures that the material systems which obtain information do not contradict the second law of thermodynamics. Indeed, the generation of entropy Eq. (14) was proposed to solve the famous paradox 'Maxwell's Demon' [4].

In this section, we showed a general theory that reception of information from the outside in a receptor system is accompanied both by an inflow of entropy and by a generation of entropy depending on the reliability of the actual mechanism of receiving information (see the Eqs. (11)–(14)).

3. Formulation for a case of the visual-pigment system

3.1. Transitions between the states of visual-pigment chromophore

We consider the system consisting of N identical visual-pigments, assuming for simplicity that the electron system of visual-pigment chromophore takes either of the ground ψ_1 or the excited state ψ_2 of which the eigenvalues for energy are given by E_1 and E_2, respectively. Let $N_1(t)$ be the number of visual-pigment chromophores in ψ_1 at time t. Then, since each chromophore in ψ_1 can give rise to the transition from ψ_1 to ψ_2 by absorbing a photon of frequency $\nu = (E_2 - E_1)/h$, the number $N_{21}(t)$ of the chromophores of which the radiative transition $\psi_1 \rightarrow \psi_2$, occurring in a unit time, can be expressed as follows, in terms of the radiative transition probability β:

$$N_{21}(t) = \beta N_1(t) \tag{15}$$

The transition moments of visual-pigment chromophores are assumed to be on the planes perpendicular to the direction of incident light, and to show a two-dimensional random orientation. Then, β in Eq. (15) can be expressed as

$$\beta = (4\pi^3/h^2 c)|\mathbf{P}_{21}|^2 I_p(\nu),$$

$$\mathbf{P}_{21} \equiv \langle \psi_2 | \mathbf{P} | \psi_1 \rangle \tag{16}$$

Here c represents the velocity of light; $\mathbf{P}_{21}$ does the transition moment; $\mathbf{P}$ denotes the total electric dipole-moment of the chromophore; $I_p(\nu)$ is the intensity distribution of the radiant energy incident into the visual-pigment system.

The propriety of expression Eq. (16) is confirmed as follows: When the transition moments show a two- or three-dimensional random orientation to the direction of incident light, we have to take account of the orientational factor of $1/2$ or $1/3$, respectively; Accordingly, if Eq. (16) is multiplied by $2/3$, the probability of radiative transition in the case of a three-dimensional random orientation must be obtained; Eq. (16) satisfies this requirement.

On the other hand, the chromophore in the excited state ψ_2 can transfer to the ground state ψ_1, by emitting a photon of frequency ν or by the photoisomerization from the 11-*cis* to the all-*trans* form. Since $|\mathbf{P}_{21}| = |\mathbf{P}_{12}|$, the transition probability from ψ_2 to ψ_1 by the emission of light is the same as β in Eq. (16). Therefore, using the transition probability α by the photoisomerization and denoting by $N_2(t)$ the number of the chromophores in ψ_2 at time t, the number $N_{12}(t)$ of the chromophores which give rise to the transition $\psi_2 \rightarrow \psi_1$ in a unit time can be expressed as

$$N_{12}(t) = (\alpha + \beta)N_2(t) \tag{17}$$

3.2. The dependence of N_2 on time t

When a photoreceptor system consisting of identical molecules absorbs light according to Lambert–Beer's law, the absorption depends only on the total number of molecules in the system, but not on their distribution. Assuming that the above law is realized in the visual-pigment system, we express the equation for $N_2(t)$ as follows:

$$dN_2/dt = N_{21} - N_{12} = \beta N - (\alpha + 2\beta)N_2,$$
$$N_1 + N_2 = N \tag{18}$$

Let the visual-pigment system be in a thermal equilibrium state of temperature T_0 in the region $t < 0$, and start receiving light at $t = 0$. Then, the values of N_1 and N_2 at $t = 0$ can be taken to be

$$N_1(0) = N, \; N_2(0) = 0 \tag{19a}$$

respectively, because when we choose

$$\lambda = c/\nu = 500[\text{nm}], \; T = 300[\text{K}] \tag{19b}$$

we obtain $h\nu/kT_0 \doteq 96.1$, and $N_2(0)/N_1(0) = \exp(-h\nu/kT_0)$ shows an extremely small value.

Based on the initial condition Eq. (19a), the solution of Eq. (18) can be found to be

$$N_2(t) = [N\beta/(\alpha + 2\beta)]\{1 - \exp[-(\alpha + 2\beta)t]\} \tag{20}$$

3.3. The t-dependence of S caused by the absorption of light

When $(N, N_1, N_2) \gg 1$, the entropy increase $S(t)$ of the visual-pigment system by the absorption of light can be expressed as follows, according to Boltzmann's theorem and using Stirling's formula

$$\log(N!) \doteq N(\log N - 1): \tag{21}$$
$$S(t) = k\log[N!/(N_1!N_2!)]$$
$$\doteq kN_2(t)\log[N_1(t)/N_2(t)]$$
$$\quad + kN\log[N/N_1(t)] \tag{22}$$

On the other hand, when $(N, N_1) \gg 1$ and $N_2/N \ll 1$, $S(t)$ is to be expressed in the form

$$S(t) \doteq kN_2(t)\log[N_1(t)/N_2(t)]$$
$$\quad + k\{N_2(t)\log N_2(t) - \log[N_2(t)!]\} \tag{23}$$

neglecting the contributions of the order of $(N_2/N)^2$. The first and the second term of the right side in Eq. (22) or Eq. (23) are the entropies corresponding to $S_a^{(1)}$ and $S_a^{(2)}$ in Eq. (11), respectively (see Appendix for the detailed derivation of Eq. (23)).

In short, we could determine in this section the time(t)-dependences of $N_2(t)$ (see Eq. (20)) and $S(t)$ (see Eq. (23)) on the basis of the following two fundamental assumptions: (1) Transition moments of the visual-pigment chromophores are on the planes perpendicular to the incident light, and show a two-dimensional random orientation; and (2) The optical absorption by the visual-pigment system acts upon Lambert–Beer's law.

4. Parameterization

4.1. The total number N of visual pigments

Hecht et al. [6] performed a psychological experiment on the threshold of photosensitivity (i.e. on the minimum number of photons necessary to cause photosensitivity) by radiating a flashlight of wavelength 510 nm for 0.001 s upon the surround of retina where about 500 rod cells exist [6]. They found that subjects became consciously aware of the light when the number of photons reaching their retinas was between 54 and 148. According to their calculation, this range of 54–148 photons means that 5–10 photons were actually absorbed by the rod cells. Moreover, at this threshold, the probability that one rod cell absorbed two photons was calculated by them to be only 5%, and hence they concluded that each of the 5–10 rod cells absorbed one photon at the threshold of photosensitivity.

One outer segment in a rod cell contains several thousand disks, and a disk has several million visual-pigments [7]. Therefore, it seems that the 500 rod cells have visual pigments of the order

$$O(N) = 10^{12} \tag{24}$$

4.2. The transition probability α by the photoisomerization

When the visual pigment absorbs the visible light, its chromophore is isomerized from the 11-*cis* to the all-*trans* form. According to the calculations by Kikuchi et al., the period T of this photoisomerization is in the range of 1 ps $\leq T <$ 10 ps [8]. As the time being several times longer than the period T is necessary to give rise to the entropy increase in the opsin (being the protein moiety of visual pigment), the value of α^{-1} is considered to be about 50 ps:

$$O(\alpha) = 10^{10}[\mathrm{s}^{-1}] \tag{25}$$

4.3. The probability β of the radiative transition

Using the oscillator strength f for the transition between ψ_1 and ψ_2, β in Eq. (16) is rewritten as

$$\beta = (3\pi e^2 f / 2mch\nu) I_p(\nu) \tag{26}$$

Calculating the oscillator strength f for the visible light of $\lambda = 500$ nm, based both on the model of Sugimoto et al. for the visual-pigment chromophore and on the INDO-CI method [9], we obtain the result

$$f = 1.389 \tag{27}$$

Also, for this model, the Einstein coefficient of spontaneous emission, i.e.

$$A = (64\pi^4 / 3hc^3)\nu^3 |\mathbf{P}_{12}|^2 = (8\pi^2 e^2 / mc)\lambda^{-2} f \tag{28a}$$

is calculated to be

$$A = 3.707 \times 10^8 [\mathrm{s}^{-1}] \tag{28b}$$

That is, if the chromophore is isolated, the lifetime of its excited state is about 3×10^{-9} s.

4.4. The minimum value of intensity distribution $I_p(\nu)$ of incident light

With respect to the psychological experiment by Hecht et al. [6], let us now assume that the occupied area of about 500 rod cells is 1.0×10^{-4} cm^2 [10], and that the number of photons of $\lambda = 510$ nm incident into this area within 0.001 s is 10. Then, the minimum value of the total radiant energy that is incident upon the rod–cell system can be calculated as follows:

$$E_{\min} = 3.894 \times 10^{-4} [\mathrm{erg}/(\mathrm{cm}^2 \cdot \mathrm{s})] \tag{29}$$

For the case of $\lambda = 510$ nm, the Einstein coefficient $[A]_{510}$ of spontaneous emission can be obtained as

$$[A]_{510} = 3.563 \times 10^8 [\mathrm{s}^{-1}] \tag{30a}$$

on the basis of Eq. (28a) and Eq. (28b), and the spectral curve in frequency expression for the flashlight of $\lambda = 510$ nm is estimated to have at least the same half-value width as

$$\gamma = [A]_{510} / (2\pi) = 0.5670 \times 10^8 [\mathrm{s}^{-1}] \tag{30b}$$

Therefore, by assuming the spectral curve of absorption to be the Lorentz type, and by using the relation

$$I_{\min} = (2/\pi\gamma) E_{\min} \tag{31a}$$

the minimum value $I_{\min}$ of $I_p(\nu)$ can be calculated as follows:

$$I_{\min} = 4.371 \times 10^{-12} [\mathrm{erg}/\mathrm{cm}^2] \tag{31b}$$

If we assume $I_{\max} = 1.2 \times 10^4 \times I_{\min}$ for the reason that the maximum intensity $I_{\max}$ of radiant energy which can be incident into human eyes is about ten thousand times larger than the minimum intensity $I_{\min}$, this $I_{\max}$ can well explain the maximum luminance directly visible by the unaided human eye (see the last section).

Using the black-body radiation for unit volume and unit frequency, i.e.

$$u(\nu, T_s) = (8\pi h/c^3)\nu^3/[\exp(h\nu/kT_s) - 1] \tag{32a}$$

the intensity $I_1(\nu)$ of radiant energy at the surface of the sun for unit area, unit time and unit frequency can be expressed as follows:

$$I_1(\nu) = (c/4)u(\nu, T_s) \tag{32b}$$

where T_s is the effective temperature of the sun. Based on Wien's displacement law

$$\lambda_m T_s = 0.2898[\text{cm} \cdot \text{K}] \tag{33a}$$

and taking $\lambda_m = 500$ nm, the value of T_s can be obtained

$$T_s = 5796[\text{K}] \tag{33b}$$

Therefore, the value $[I_1]_{510}$ of $I_1(\nu)$ for $\lambda = 510$ nm is calculated from Eq. (32a) and Eq. (32b) as

$$[I_1]_{510} = 0.7303 \times 10^{-4}[\text{erg/cm}^2] \tag{34a}$$

and by comparing this value with I_{min} of Eq. (31b), we find

$$I_{min}/[I_1]_{510} = 0.5985 \times 10^{-7} \tag{34b}$$

In Section 4, we could determine the following three fundamental quantities: (1) The total number N of visual pigments, $O(N) = 10^{12}$; (2) The probability α of the non-radiative transition from ψ_2 to ψ_1 by the photoisomerization of visual-pigment chromophore, $O(\alpha) = 10^{10}$ [s^{-1}]; and (3) The probability β of the radiative transition from ψ_2 to ψ_1 of visual-pigment chromophore. In addition, the intensity $I_p(\nu)$ of incident light, which is necessary to formulate β, was examined, and its minimum value I_{min} was determined as 4.371×10^{-12} [erg/cm^2].

5. Calculations

5.1. The minimum value of N_2 *and the relation between* N *and* α

Using I_{min} of Eq. (31b), the minimum value β_{min} of β of Eq. (26) for $\lambda = 510$ nm is first calculated as

$$\beta_{min} = 6.206 \times 10^{-2}[\text{s}^{-1}] \tag{35}$$

Then, taking the respective values of β and t in Eq. (20) equal to β_{min} and α^{-1}, and paying attention to the condition $\beta_{min} \ll \alpha$, the relation between the minimum value $N_2^{(min)}$ of N_2 and N/α is expressed as follows:

$$N_2^{(min)} \doteqdot 3.923 \times 10^{-2} \times (N/\alpha) \tag{36}$$

Furthermore, with regard to the second term of the right side of Eq. (23), we assume that

$$N_2^{(min)}! \doteqdot N_2^{(min)}(N_2^{(min)} - 1) = 1 \tag{37a}$$

and we determine the value of $N_2^{(min)}$ as

$$N_2^{(min)} \doteqdot 1.618 \tag{37b}$$

In this case, we have

$$N_2^{(min)}\log N_2^{(min)} \doteqdot 0.7786 = 1.123 \times \log 2 \tag{37c}$$

indicating that the visual-pigment system can detect a photon with the reliability of 54.09% (see $S_a^{(2)}$ in Eq. (11)).

Finally, from Eq. (36) and Eq. (37b), the relation between N and α is obtained

$$N = 41.24[\alpha], \quad \text{or} \quad [\alpha] = 0.02425N \tag{38}$$

where $[\alpha]$ represents the value of α in s^{-1} units.

5.2. The self-consistent determination of the values of N *and* α

The number N_0 of visual pigments per rod cell had once been reported to be about 1.6×10^9 [7]. Accordingly, we take the value of N_0 as

$$N_0 = 1.64 \times 10^9. \tag{39a}$$

On the other hand, with regard to the number n of rod cells existing in the surround of retina, Hecht et al. [6] assumed that $n \doteqdot 500$, and hence we take

$$n = 540. \tag{39b}$$

N can thus be calculated as

$$N = nN_0 = 0.8856 \times 10^{12}, \tag{39c}$$

of which the order agrees with Eq. (24). Using this value and the relation Eq. (38), we obtain

$$\alpha = 2.148 \times 10^{10} [\text{s}^{-1}], \tag{40a}$$

of which the order also agrees with Eq. (25). That is, the value of α^{-1} is calculated as follows:

$$\alpha^{-1} = 46.55 [\text{ps}] = 6.650 \times 7 [\text{ps}]. \tag{40b}$$

This α^{-1} is acceptable for the following two reasons: (1) As mentioned previously, Kikuchi et al. [8] have shown that the period T of the photoisomerization of the visual-pigment chromophore is in the range of 1 ps $\leq T < 10$ ps, and that $T = 7$ ps is the most desirable value [8]; (2) The period $T = 7$ ps is indispensable for the transfer of vibrational energy from the visual-pigment chromophore to the neighboring moiety of protein. It is thus reasonable to assume that a period of about $6T$ is needed for the macroscopic increase of entropy in the opsin. Needless to say, the value of N of Eq. (39c) indicates the minimum number of visual pigments in the visual-pig-

ment system because when the visual-pigment system can detect a photon with a reliability of more than 54.09%, the coefficient of $[\alpha]$ of the first formula in Eq. (38) must be more than 41.24, and hence, as far as the value of α of Eq. (40a) is reasonable, the value of N becomes greater than that of Eq. (39c).

5.3. Calculations of $N_2(t)$ and $S(t)$

Based on Eq. (32a), Eq. (32b) and Eq. (33b), the value $[I_1]_{500}$ of I_1 for $\lambda = 500$ nm can be calculated as

$$[I_1]_{500} = 0.7026 \times 10^{-4} [\text{erg/cm}^2]. \tag{41a}$$

Using this value, we here define

$$I \equiv [I_p]_{500} / [I_1]_{500} \tag{41b}$$

and we represent the β (in s^{-1} units) of Eq. (26) for $\lambda = 500$ nm as

$$\beta_{500} = 0.9777 \times 10^6 \times I. \tag{41c}$$

With these preliminaries, $N_2(t)$ of Eq. (20) is first calculated by choosing four values (10^{-7}, 10^{-6}, 10^{-5}, 10^{-4}) for the I in β_{500} and five values (10^{-14}, 10^{-13}, 10^{-12}, 10^{-11}, 10^{-10}) for time t (in s units) and by using the values of N of Eq. (39c) and α of Eq. (40a). Then, noticing the difference between Eq. (22) and Eq. (23), $S(t)/k$ is calculated. These results for $N_2(t)$ and $S(t)/k$ are presented in Tables 1 and 2, respectively.

$S(t)$ of Eq. (22) is divided into the two parts:

Table 1
Number of $N_2(t)$ of chromophores in the excited state ψ_2 at time t. The values $N_2(t)$ of Eq. (20) were calculated by choosing four values for I in β_{500} and five values for time t and by using the values of N of Eq. (39c) and α of Eq. (40a)

t \ I	10^{-14} s	10^{-13} s	10^{-12} s	10^{-11} s	10^{-10} s
10^{-4}	0.8658	0.8649×10	0.8566×10^2	0.7792×10^3	0.3560×10^4
10^{-5}	0.8658×10^{-1}	0.8649	0.8566×10^1	0.7792×10^2	0.3560×10^3
10^{-6}	0.8658×10^{-2}	0.8649×10^{-1}	0.8566	0.7792×10	0.3560×10^2
10^{-7}	0.8658×10^{-3}	0.8649×10^{-2}	0.8566×10^{-1}	0.7792	0.3560×10

Table 2
Time (t)-dependence of $S(t)/k$

I	$t = 10^{-14}$ s	$t = 10^{-13}$ s	$t = 10^{-12}$ s	$t = 10^{-11}$ s	$t = 10^{-10}$ s
A. Time (t)-dependence of S(t)/k, *in which* S(t) *is the entropy increase by the absorption of light*[a]					
10^{-4}	2.481×10	2.279×10^2	2.061×10^3	1.703×10^4	7.239×10^4
10^{-5}	2.680	2.478×10	2.258×10^2	1.882×10^3	8.059×10^3
10^{-6}	2.879×10^{-1}	2.678	2.455×10	2.061×10^2	8.879×10^2
10^{-7}	3.071×10^{-2}	2.877×10^{-1}	2.653	2.241×10	9.699×10
B. Time (t)-dependence of S(t)/k *with neglecting the order of* $(N_2/N)^2$ [b]					
10^{-4}	2.382×10	2.258×10^2	2.058×10^3	1.702×10^4	7.239×10^4
10^{-5}	2.382	2.379×10	2.237×10^2	1.879×10^3	8.055×10^3
10^{-6}	2.382×10^{-1}	2.379	2.357×10	2.041×10^2	8.851×10^2
10^{-7}	2.382×10^{-2}	2.379×10^{-1}	2.357	2.143×10	9.529×10

[a] The values $S(t)/k$ of Eq. (22) were calculated by using the value of N of Eq. (39c) and those of $N_2(t)$ in Table 1.
[b] The values of $S(t)/k$ of Eq. (23), neglecting the contributions of the order of $(N_2/N)^2$, were calculated by using the value of N of Eq. (39c) and those of $N_2(t)$ in Table 1.

$$S(t) = S_1(t) + S_2(t);$$

$$S_1(t)/k = N_2(t)\log[N_1(t)/N_2(t)],$$

$$S_2(t)/k = N\log[N/N_1(t)] \tag{42a}$$

In the case of Eq. (23), $S_2(t)/k$ must be taken as

$$S_2(t)/k = N_2(t)\log N_2(t) - \log[N_2(t!)] \tag{42b}$$

The relation between $\log_{10}[S_1(t)/k]$ or $\log_{10}[N_2(t)]$ and $\log_{10}t$ is thus shown in Fig. 1 for the case of $I = 10^{-7}$, and the relation between $\log_{10}[S_2(t)]$ and $\log_{10}t$ is shown in Fig. 2 for the same case.

5.4. A few comments on the results of our calculations

Fig. 1 shows the comparison between $S_1(t)/k$ and $N_2(t)$ for the case of $I = 10^{-7}$ in Table 2, and we find that $S_1(t)/k$ is almost proportional to $N_2(t)$, and that both curves reach the respective equilibrium values at $t = 10^{-10}$ s. With respect to the difference between these two equilibrium values, their physical meanings can be understood as follows. On the basis of Eq. (20) and Eq. (42a), $S_1(\infty)$ for $t = \infty$ can be rewritten as

$$S_1(\infty)/N_2(\infty) = k\log(1 + \alpha/\beta) \tag{43a}$$

Then, by defining $I_p(\nu) = I_1(\nu) \times I$ according to the definition of Eq. (41b) and by rewriting β of Eq. (16) in terms of I_1 of Eq. (32b), we have

$$\beta = (3AI/8)[\exp(h\nu/kT_s)]^{-1},$$

$$A = (64\pi^4/3hc^3)\nu^3|\mathbf{P}_{21}|^2 \tag{43b}$$

where A represents the Einstein coefficient of induced emission. Therefore, in the case of $\lambda = 500$ nm, $\log(1 + \alpha/\beta)$ can be calculated as follows:

$$\log(1 + \alpha/\beta) \fallingdotseq h\nu/(kT_s) + \log[(8\alpha/3A)I^{-1}]$$

$$\fallingdotseq 26.1 \tag{43c}$$

The first term in the right side of the above equation shows that the radiant energy being incident into the visual-pigment system carries the entropy of $h\nu/T_s$ per photon. On the other hand, the second term shows such an entropy generation that depends on the characteristics of both the absorption and the emission mechanism for the radiant energy in the visual-pigment system. That is, the factor of $8/3$ is due to the interaction between the two-dimensionally isotropic system of visual-pigments and the photons incident from the three-dimensionally isotropic black-body field. Moreover, the factor of α/A indicates that the transfer of energy from
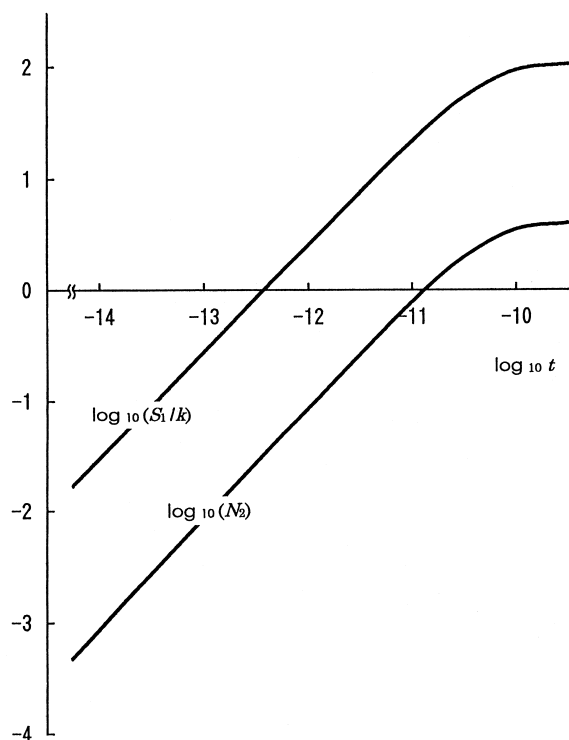
Fig. 1. Time (t)-dependence of $S_1(t)/k$, in which $S_1(t)$ is the flowed entropy into the visual-pigment system by the absorption of light, and that of the number N_2 of the excited chromophores. The first term S_1 in Eq. (42a) and the number of N_2 of visual-pigments were calculated as respective functions of t, for the case of $I = 10^{-7}$ in Table 2B, and $\log_{10}(S_2/k)$ was compared with $\log_{10}(N_2)$.
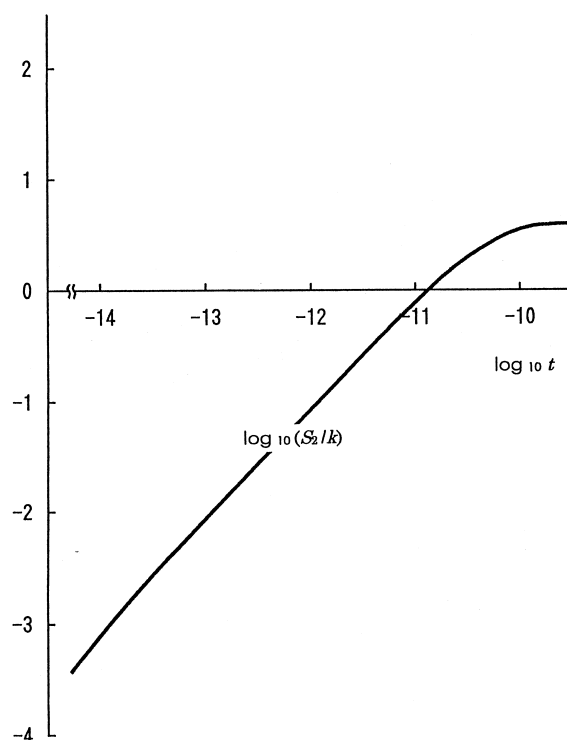
Fig. 2. Time (t)-dependence of $S_2(t)/k$, in which $S_2(t)$ is the generated entropy at the visual-pigment system by the absorption of light. The second term S_2 in Eq. (42a) was calculated as a function of t, for the same case as Fig. 1.

the visual-pigment chromophore to the surrounding protein moiety is caused by the *cis-trans* isomerization of the chromophore, and hence that this transfer is not concerned at all with such a spontaneous emission of photon, as in the case of black-body radiation.

Fig. 2 compares log2 with the time-dependence of S_2/k for $I = 10^{-7}$ in Table 2. By using Eq. (20), Eq. (39c), Eq. (40a), and Eq. (41c), the value $N_{2\alpha}$ of N_2 at $t = \alpha^{-1}$ is calculated as 2.549, and the value $S_{2\alpha}$ of S_2 is found to be

$$S_{2\alpha}/k \fallingdotseq (N_{2\alpha} - 1)\log N_{2\alpha} - \log(N_{2\alpha} - 1)$$
$$\fallingdotseq 1.01 \fallingdotseq 1.46 \times \log 2$$

This value, in conjunction with $S_a^{(2)}$ in Eq. (11), indicates that the photoreception concerned was performed with reliability 64%.

In Section 5, we could determine the minimum value $N_2^{(\min)}$ of N_2 to be about 1.618, derive the relation between N and α using $N_2^{(\min)}$ (see Eq. (38)), and calculate the time-dependent changes in N_2 and S (Tables 1 and 2; Figs. 1 and 2). We also described the physical meanings of the results of our calculations.

6. Concluding remarks

6.1. Conclusions of this paper

We first showed a general theory that reception of information from the outside in a receptor system is accompanied both by an inflow of entropy and by a generation of entropy depending on the reliability of the actual reception mechanism. Then, considering a case for the absorption of light by the visual-pigment system of visual cell,

we calculated the time-dependent change in the number of excited visual-pigments to obtain the entropy increase. Thus, we concluded the following four points: (1) One or two photons are detected in the visual-pigment system with a reliability of at least 54%; (2) In compensation for this detection, it is indispensable to generate the entropy of $1.12 \times \log 2$; (3) A photon of frequency ν which is incident into the visual-pigment system from the light source of temperature T_s yields an entropy of $h\nu / T_s$; and (4) Another entropy is also generated in proportion to $N_2(t)$, depending on the characteristics of the visual-pigment system as a photoreception device.

6.2. Variety of the visual-pigment systems

The results of our calculations in this paper can reasonably explain the entropy generation in the rod cells which exist in the surround of the retina in human eye. On the other hand, when we study the cone cells existing in the vicinity of the blind spot of the retina in human eye with regard to daylight and color visions, we should note that the energy intensity of incident light, the number of cone cells absorbing the light, the number of the pigments in a cone cell, the structure of their chromophore and opsin all differ substantially from those of the rod cells. Therefore, it is necessary for us to parameterize the respective values of $(N, \alpha, N_2^{(\min)})$ for cone cells which are different from those in Eq. (39c), Eq. (40a) and Eq. (37b). It would seem that the variety of the visual-pigment systems in various visual cells can reasonably be explained by the parameterizations mentioned above.

6.3. Maximum energy intensity I_{max} of incident light

When the visible light radiated from a surface A of luminance B images on a surface A', the illumination intensity J' is given by

$$J' = \pi B (n'/n)^2 \sin^2 \phi' \tag{44}$$

Here n and n' represent the respective indices of refraction of the thing space and the image space,

and ϕ' does the vertical angle of the cone of incident luminous flux in the image space [11].

When the values of n, n' and ϕ' in Eq. (44) are taken equal to

$$n = 1.000, \; n' = 1.333, \; \phi' = 0.1808 \tag{45a}$$

according to Donders' reduced eye [12], the illumination intensity J_B' of the image of the luminescence body of $B = 1 \; \text{cd}/\text{m}^2$ is calculated from Eq. (44) to be

$$J_B' = 0.1805 [\text{lm}/\text{m}^2] \tag{45b}$$

Because the spectral luminosity factor of human eyes to the visible light of $\lambda = 500$ nm is given by

$$L = 220.6 [\text{lm}/\text{watt}] \tag{46a}$$

the energy intensity E_B actually sensed by our eyes can be calculated as

$$E_B = 0.8182 \; [\text{erg}/(\text{cm}^2 \cdot \text{s})] \tag{46b}$$

And the value γ_{500} for $\lambda = 500$ nm is calculated to be

$$\gamma_{500} = 0.5899 \times 10^8 [\text{s}^{-1}] \tag{47a}$$

using Eq. (28b) and in the same way as Eq. (30b). Therefore, by using E_B of Eq. (46b), the intensity I_B of energy for the unit frequency can be obtained in the same way as Eq. (31a):

$$I_B = 0.8828 \times 10^{-8} [\text{erg}/\text{cm}^2] \tag{47b}$$

The maximum intensity $I_{\max}$ of the radiant energy which can be incident into human eyes has been estimated to be about 10 000 times the minimum intensity $I_{\min}$. Therefore, assuming

$$\begin{aligned} I_{\max} &= 1.200 \times 10^4 \times I_{\min} \\ &= 5.245 \times 10^{-8} [\text{erg}/\text{cm}^2] \end{aligned} \tag{48a}$$

the relation between this $I_{\max}$ and I_B of Eq. (47b) is found to be

$$I_{max} = 5.941 \times I_B \qquad (48b)$$

This fact can well explain the following experimental results: When human eyes were radiated by visible light of $\lambda = 500$ nm at the various kinds of light-source luminance, the luminance of the order of 10 cd/m^2 inactivated the function of the rod cells [13,14].

As described in the *Introduction*, the biological system has an excellent mechanism for entropy transmission. On the basis of the present findings for entropy generation, we will prove in the succeeding paper that the necessary-sufficient condition for realizing a 'negentropy state' of a biological system is the existence of an 'irreversible cycle' in the system, and that a model for a 'biological clock' can be expressed by linkage of a number of 'irreversible cycles' of cellular signal-transduction systems.

Appendix: Derivation of Eq. (23)

We first simplify $\log (N!/N_1!)$ using Stirling's formula Eq. (3):

$$\log(N!/N_1!) \doteqdot N(\log N - 1) - N_1(\log N_1 - 1)$$
$$= N \log N - N_2 - (N - N_2)$$
$$\times \log(N - N_2).$$

Here let $N_2/N = x \ll 1$, and hence we can obtain

$$\log(N!/N_1!) \doteqdot N \log N - Nx$$
$$- N(1-x)\log[N(1-x)]$$
$$= Nx(\log N - 1) - N(1-x)\log(1-x)$$
$$\doteqdot Nx(\log N - 1) + N(1-x)(x + x^2/2)$$
$$= Nx(\log N - x/2)$$
$$\doteqdot N_2 \log N$$

Thus, it is expressed that

$$\log[N!/(N_1!N_2!)] \doteqdot N_2 \log N - \log(N_2!)$$

On the other hand, the first term of the right side in Eq. (22), which is here divided by k for simplicity, can be rewritten as

$$N_2 \log(N_1/N_2) = Nx \log[(1-x)/x]$$
$$\doteqdot Nx[-x(1 + x/2) - \log x]$$
$$\doteqdot -Nx \log x$$
$$= N_2 \log N - N_2 \log N_2$$

Therefore, Eq. (23) can be expressed as

$$S(t)/k \doteqdot N_2 \log N - \log(N_2!)$$
$$\doteqdot N_2 \log(N_1/N_2) + [N_2 \log N_2 - \log(N_2!)]$$

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