anamnestic responses. Thus, with both antigens, hemolysin titer was, in general, reciprocally related to per cent transfer during the initial immunization, but transfer sometimes remained high at peak titer during the second and especially during the third immunization (Figs. 1–4). This result, with respect to transfer in the rabbits immunized with SStr, was associated with a progressive increase in the γ_2 hemolysin with a high transfer rate and a decrease in the γ_1 hemolysin with a low transfer rate (Figs. 5 and 6). Inasmuch as little or no γ_2 hemolysin has been found by various investigators in response to AStr, the high rates of transfer in the anamnestic responses to AStr were possibly associated with a relative increase in normal hemolysins as production of immune hemolysins decreased.

Transfer was ascertained in terms of the number of 50 per cent units of hemolysin needed to be adsorbed on unlabeled red cells to give the net transfer of one 50 per cent unit of hemolysin to unsensitized Cr^{51}-labeled red cells. The slope of the von Krogh graphs in the tests was usually 0.4 for the γ_1 hemolysin and 0.2 for the γ_2 hemolysin.

† A preliminary note by the authors has appeared in *Science*, 132, 1500 (1960).

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NEGATIVE ENTROPY AND PHOTOSYNTHESIS

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As was suggested by E. Schrödinger,[1] the maintenance of the high organization of living beings is due to a continuous influx of negative entropy. Animals get this negative entropy by eating plants (as well as one another) while plants get their negative entropy, along with energy, from the sun's rays. The point is that solar radiation arriving on the earth is not in a state of thermodynamical equilibrium. Indeed, using Wien's formula λ max T = const. for the maximum of the energy in the spectrum, we obtain T = 6,000°K, i.e., the surface temperature of the sun.

On the other hand, since solar radiation is highly diluted while expanding into the surrounding space, its energy density is much smaller when it arrives at the earth, and using the Stefan-Boltzmann formula $e = aT^4$ we get $T = 300°K$, i.e., the surface temperature of the earth.

When this diluted solar radiation falls on the material surface which permits the exchange of energy between different frequencies, an irreversible process is taking place in which the original high-frequency quanta are transformed into a much larger number of the quanta with much lower frequencies. This process results in the increase of the radiation entropy which can be calculated from the quantum statistical theory of radiation. If the surface performing this transformation is made of simple inorganic material, like an iron roof of a house, the total entropy of the system increases and the incident visible light is re-emitted in the form of heat rays.

In the case of the chloroplasts in plant leaves, however, the situation is different, and the increase of the entropy during the irreversible transition of the incident radiation is used to compensate for the decrease of entropy in building organic molecules from the molecules of H_2O and CO_2. Although the mechanics of the photosynthetic process is not yet well understood, purely thermodynamical considerations require that the decrease of entropy per organic molecule formed must not be larger than the increase of entropy of the light quanta participating in the process in their transformation from a high energy inequilibrium state into a low energy state of equilibrium. The purpose of this article is to calculate entropy changes of the incident diluted solar radiation, and to compare them with the measured decrease of entropy in the building process of organic molecules.

Figure 1 shows energy distribution in the spectra of equilibrium (black body) radiation at two temperatures, T_1 and T_2. Suppose that the high temperature radiation, T_1, is diluted to such extent that its integrated energy denisty (the area under the curve A') is equal to the integrated energy density of equilibrium radiation corresponding to the lower temperature T_2 (the area under the curve B). The process taking place when the energy exchange between different frequencies is permitted can be compared to a flow of heat from a reservoir at the temperature T_1 to a cooler reservoir at the temperature T_2. If E is the total
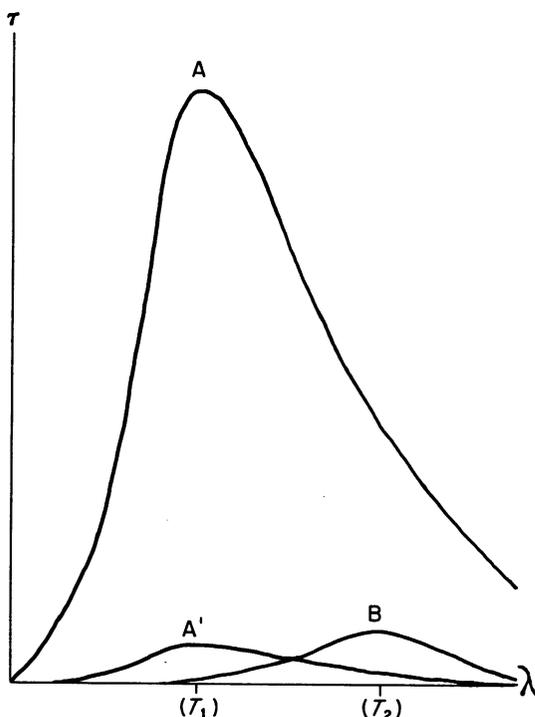
FIG. 1.

amount of energy involved, we may write for the change of entropy:

$$\Delta S \cong E \left(\frac{1}{T_2} - \frac{1}{T_1} \right) \tag{1}$$

The above-given plausibility derivation of this formula can be justified by strict quantum statistical considerations given in the Appendix.

Putting $E = nh\nu$ where n is the number of light quanta participating in the photosynthesis of a single molecule, and neglecting $1/T_{sun}$ compared with $1/T_{earth}$, we obtain

$$\Delta S \cong \frac{nh\nu}{T_{earth}} \tag{2}$$

for the maximum entropy change per molecule formed. Numerically with $\nu \cong 4 \cdot 10^{14}$ sec^{-1} (for red light) and $T_{earth} \cong 300°$K, we obtain

$$\Delta S \cong 10^{-14} \cdot n \, \frac{erg}{1°} \, (\text{per molecule}) = 150 \cdot n \, \frac{cal}{1° \, mol} \tag{3}$$

The over-all photochemical reaction:

$$6CO_2 + 6H_2O \rightarrow C_6H_{12}O_6 + 6O_2 \tag{4}$$
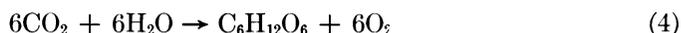
requires at least 3 light quanta and corresponds to the entropy change of about 40 cal/1° mol. Thus we conclude that, if the photosynthetic process has at least 10 per cent efficiency in the entropy conversion, the growing of a plant in the sunlight is consistent with the second law of thermodynamics.

Appendix.—The quantum statistical treatment of radiation in temperature equilibrium is well known.[2] In particular this theory leads to the expressions

$$e = aT^4 \tag{5}$$

and

$$s = \frac{4}{3} aT^3 \tag{6}$$

for the energy density and entropy density of black-body radiation, where a is the Stefan-Boltzmann constant,

$$a = \frac{8}{15} \frac{\pi^5 k^4}{(hc)^3}. \tag{7}$$

For the nonequilibrium situation, it is possible to define a temperature[2] for each frequency and also an entropy flux which can be expressed in terms of the energy flux.[3] However for the problem of a black-body sphere radiating into empty space, the problem can be handled directly in a simple manner.

A black-body sphere of radius R_1 will radiate energy E_1 per unit time and entropy S_1 per unit time in amounts given by:

$$E_1 = 4\pi R_1^2 \cdot \frac{1}{4} c \cdot aT_1^4 \tag{8}$$

and

$$S_1 = 4\pi R_1^2 \cdot \frac{1}{4} c \cdot \frac{4}{3} aT_1^3 \tag{9}$$

respectively. In the steady state these same amounts pass through a larger sphere of radius R_2:

$$E_1 = 4\pi R_2^2 \epsilon_2, \tag{10}$$

$$S_1 = 4\pi R_2^2 \sigma_2, \tag{11}$$

where ϵ_2 is the energy flux through the sphere and σ_2 is the corresponding entropy flux. Relation (10) is valid because of conservation of energy. That (11) is valid may be established by noting that there is no mechanism for the production of entropy in the pure radiation field. (Actually the scattering of light by light can increase the entropy, even in empty space, but this fourth-order effect is neglected here.) Another way of establishing (11) is by noting that the transport equation for radiation in space is just the Liouville equation, which is completely reversible with respect to time. The ratio of expressions (11) and (10) is:

$$\frac{\sigma_2}{\epsilon_2} = \frac{4}{3} \cdot \frac{1}{T_1} = \frac{s_1}{e_1}, \tag{12}$$

which shows that the entropy-energy ratio at any distance from the black body corresponds to the *temperature of the black body* emitting the radiation. If we now consider some of the entropy S_1 and energy E_1 initially in the radiation field to be captured in an enclosure and allowed to come to equilibrium, the temperature will become lower than T_1 because of the increase in entropy. In fact, if T_2 is the final temperature and S_2 the final entropy:

$$\frac{S_2}{E_2} = \frac{S_2}{E_1} = \frac{4}{3} \cdot \frac{1}{T_2} \tag{13}$$

and

$$\Delta S = S_2 - S_1 = \frac{4}{3} E_1 \left(\frac{1}{T_2} - \frac{1}{T_1} \right) \tag{14}$$

which, apart from the factor 4/3, is the first displayed equation in the text.

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