

## A Remark on the Energy Transfer in Biological Systems

Avery, Bay & Szent-Györgyi (1961) considered in their paper two mechanisms of energy transfer in biological systems: an individual and a collective one. The individual picture concerned an aggregate of molecules in which an energy quantum is deposited; this quantum excites a molecule and the excitation is transported from one molecule to another by random jumps until it reaches a sink in the aggregate. The authors stated that the individual picture would only be valid if  $\frac{\Gamma t_1}{n^2} > 1$ , where  $\hbar\Gamma$  is the width

of the excited level,  $t_1$  is the time needed for the excitation to jump to an adjacent molecule and  $n$  the number of paths an excitation can use to jump to a next nearest neighbour molecule.† The authors assumed that  $\Gamma$  would be of the order of  $10^{13} \text{ sec}^{-1}$  and calculated  $t_1$  using the number of random jumps which the excitation would need to reach the sink. They concluded that the individual picture cannot be valid in a system of 200 chlorophyll molecules. We have carried out the calculation in a slightly different way using a more accurate estimate of the number of random jumps and have arrived at the conclusion that it is not possible to give a definite answer as to the validity of the individual picture.

Let us consider, as Avery *et al.* (1961) did, a monomolecular layer. Let us, to simplify our calculation, assume that it is a square layer of  $N^2$  molecules and that the sink is in the centre of the square. To a good approximation this picture is equivalent to a square with a sink in each corner. Let us now take a coordinate system as illustrated (Fig. 1). We call  $D_{xy}$  the number of jumps to be taken at random to reach the sink from  $(x, y)$ . Only single jumps, parallel to one of the axes and to the nearest neighbour, are allowed. The boundary conditions are now:

$$D_{0,y} = D_{N,y} \text{ for all } y \quad (1)$$

and

$$D_{x,0} = D_{x,N} \text{ for all } x \quad (2)$$

It is clear that

$$D_{0,0} = 0 \quad (3)$$

Further it is clear that the number of jumps from  $(x, y)$  equals the average

† Our definition for  $n$  is equivalent to the definition used by Avery *et al.*:  $n$  is the number of intermediate neighbour molecules, which can be involved in a jump to a definite next nearest neighbour molecule.

of the numbers for the nearest neighbours of  $(x, y)$  plus one jump. This gives the recurrence relation:

$$D_{x,y} = \frac{1}{4}(D_{x+1,y} + D_{x-1,y} + D_{x,y+1} + D_{x,y-1}) + 1 \quad (4)$$

for all  $D_{x,y}$  except  $D_{0,0}$ .

We now consider the average  $D$  of all  $D_{x,y}$  over the aggregate:

$$D \equiv \frac{1}{N^2} \sum D_{x,y} = \frac{1}{N^2} \left( \sum_{\substack{\text{except} \\ (x,y)=(0,0)}} D_{x,y} + \frac{1}{4} \sum_{\substack{\text{nearest} \\ \text{neighbours} \\ \text{of } (0,0)}} D_{x,y} - \frac{1}{4} \sum_{\substack{\text{nearest} \\ \text{neighbours} \\ \text{of } (0,0)}} D_{x,y} \right)$$

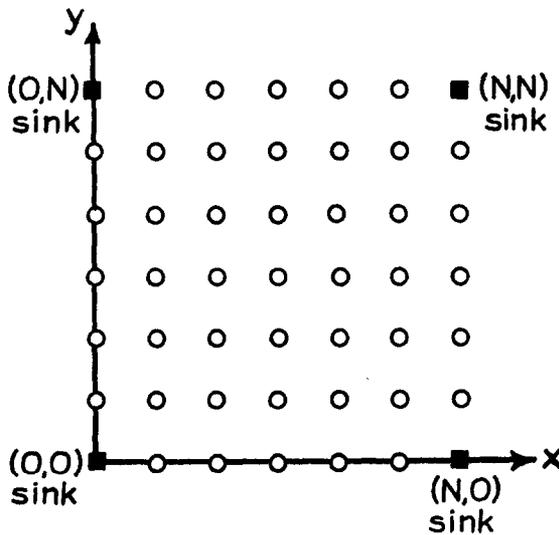


FIG. 1. Coordinate system for a square layer of  $N^2$  molecules with a sink in each corner.

Substituting equation (4) in the first term we find for the sum of the first and second term  $\frac{N^2 D + N^2 - 1}{N^2}$ . As the third term equals  $\frac{D_{1,0}}{N^2}$  we find:

$$D = D + \frac{N^2 - 1}{N^2} - \frac{D_{1,0}}{N^2}$$

So

$$D_{1,0} = N^2 - 1 \quad (5)$$

The general solution of  $D_{x,y}$  is then:

$$D_{x,y} = -(x^2 + y^2) + a(x + y) + bxy + c \quad (6)$$

Combining equations (1), (2), (5) and (6) we find

$$D_{x,y} = -(x^2 + y^2) + N(x + y) + N^2 - N \quad (7)$$

for all  $(x, y)$  except  $(0, 0)$  and

$$D_{0,0} = 0$$

This gives

$$D \equiv \frac{1}{N^2} \sum_{x,y} D_{x,y} \approx \frac{1}{N^2} \int_0^N \int_0^N dx dy D_{x,y} = \frac{4}{3} N^2 - N \quad (8)$$

The number of jumps to be taken is thus of the order of the number of molecules in the aggregate. Avery *et al.* stated that the number of jumps should be of the order of (number of molecules)<sup>2</sup>.

Let us now follow their calculation of  $t_1$  in a system of 200 chlorophyll molecules. More recent values than those employed by Avery *et al.* (1961) for the fluorescence yield of chlorophyll *in vivo* given by Latimer, Bannister & Rabinowitch (1957) indicate a yield of about  $2 \times 10^{-2}$ . The number of jumps is according to our formula about 250, so  $t_1$  should be about  $8 \times 10^{-13}$  sec and  $\frac{\Gamma t_1}{n^2} \approx 2$ .

This would lead to the conclusion that the individual picture can be valid, in contradiction with the conclusion of Avery *et al.* (1961). However,

the condition  $\frac{\Gamma t_1}{n^2} > 1$  for the validity of the individual picture is a neces-

sary, but not a sufficient one. Therefore it is not possible to give a definite answer about the validity of the individual picture. Both the mechanism which causes the excitation to jump and the probability of jumps longer than a single one should be studied before a sufficient condition can be formulated.

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