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A CRITICAL PHENOMENON APPROACH OF BIOGENESIS

by

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Abstract

A fundamental prebiotic stage of the origin of life is the formation, from a random assembly of oligomers, of information-containing self-replicating polymers. By adopting a growth mechanism (already used by Anderson and others) essentially based in the complementarity of the Crick and Watson base-pairs, we show that this stage may have occurred as a critical phenomenon. Within a simple real-space renormalization group framework we calculate the relevant critical lines, which are different for different sequences of base-pairs. The picture incorporates in a natural way Darwinian-like evolution, is consistent with reasonable nucleotide ratios (A+T)/(C+G), and suggests that polymers like ADN double-chains are more primitive than proteins.

Key-words: Biogenesis; Critical Phenomena; Renormalization group; Biopolymers.

1. Introduction

Life seems to have started on Earth about 3.8×10^9 years ago. The Earth itself was formed about 5×10^9 years ago. Consequently, if we assume that life in our planet started spontaneously, there was a period of approximately 1.2×10^9 years during which a complex set of steps were overcome in order to arrive up to the primitive biological systems. If life did not start spontaneously on Earth, but rather it came from elsewhere, then in that other place (or those other places) of the Universe a similar set of steps has presumably occured. Among the various prebiotic stages which probably occured during the transition from inanimate to living matter, one of great importance no doubt is the growth of codified self-replicating polymers starting from a random assembly of oligomers (dimers, trimers, etc). This is an important link of the chain which joins Organic Chemistry to Biology, and its study constitutes the central scope of the present work. This step remains, in spite of its obvious importance, insufficiently understood. This is due in part to the fact that the amount of related experimental work has not yet achieved the point where an enlightening and comprehensive view would be possible. This situation is, in some sense, in contrast with the present knowledge[1,2] of a more primitive step, namely the formation of nucleotides, animoacids, etc, starting from H_2O, methane, etc (the transition from Inorganic to Organic Chemistry, generally speaking); indeed, this step is now considered to be based on scientifically reliable grounds through the picture of violent non-equilibrium phenomena (electrical discharges, light and heat flashes, etc) occuring in relatively simple atmosphere.

The growth of codified self-replicating macromolecules (DNA- or RNA-like) has recently attracted quite intensive theoretical attention[3-10] within thermodynamic and/or statistical mechanics frameworks. The basic growth mechanism is assumed to be autocatalysis relying on a Crick and Watson-like complementarity[11], A and T as well as C and G thus constituting base-pairs. This is a convenient place for making clear that, although we shall use along the present work the notation A-T and C-G, *we do not necessarily refer to the well known nucleotides* (adenine, thymine, cytosine and guanine); the notation might as well refer to their *precursors;* even more, if the basic macromolecule is to be a RNA-like one[2,12-16], rather than a DNA-like one, the notation A-T would then refer to the A-U pair.

Other growth mechanisms, using clay[17], protein[18] and animoacid pairing[19] as basic ingredients, have been proposed.

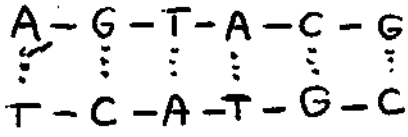
However, autocatalysis based on A-T, C-G pairings is very appealing[2,6,8-10,12-16], and this is the viewpoint we adopt in the present work.

We have developed since 1983 a thermal equilibrium critical phenomenon picture for understanding the growth of codified self-replicating polymers. Our first approach[20] assumed a *single* base-pair; it provided polymeric growth consistent with *diversity*, but with *no selection* in it. Our second approach[21] generalized the first one in the sense that the fugacity K_{AT} of the A-T hydrogen bridge might be *different* from the fugacity K_{CG} of the C-G hydrogen bridge; it showed that if *two* different base-pairs are assumed in the autocatalysis, *both diversity and selection* become possible, thus satisfying in a natural way the basic requirements for Darwinian evolution. The importance of having *four*, and *not two*, different monomers (capable consequently of forming *two*, and *not one*, complementary pairs) has also been emphasized by Anderson[10].

Our critical phenomenon theoretical picture is developed within the renormalization group (RG) framework[22,23], more specifically within a real space version of it, similar to those available in the literature[24-26] for polymer problems. A comprehensive and pedagogical review of our approach[20,21] of the above discussed prebiotic stage constitutes the central purpose of the present talk.

2. Model, formalism and results

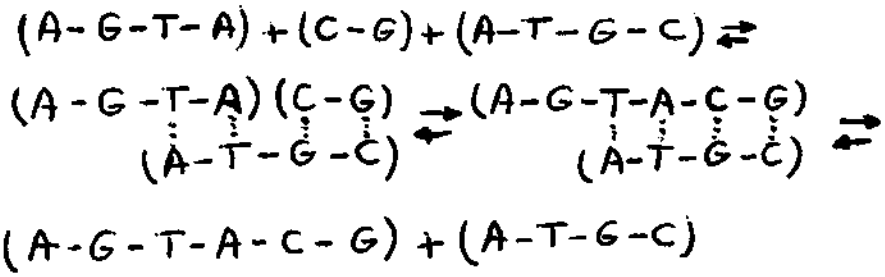
The monomers A, T, C and G can form double strings through *intrachain* covalent bonds (noted—in the illustration which follows) and *interchain* (hydrogen-like) bridges (noted...) as illustrated below:



(1)

Let K_{AT} and K_{CG} be the *fugacities* (or bonding constants) respectively associated with the *interchain* A-T and C-G bridges (K_{AT}, $K_{CG} > 0$). K_{AT} and K_{CG} depend, in a complex unknown manner, on all the thermal equilibrium (or almost equilibrium) external parameters (temperature, pressure, humidity, various salts concentrations, etc) which

characterize the *primordial soup*, assumed to contain arbitrary amounts of randomly codified oligomers (dimers, trimers, etc) like that of scheme (1). We further assume that oligomers can grow through the autocatalytic process illustrated below:



(2)

Notice that we have obtained, as a final product, the *hexamer* of scheme (1), whereas at the initial stage, we had nothing longer than *tetramers*. In the present illustration, (A-G-T-A) and (C-G) play the role of *growing fragments*, and (A-T-G-C) plays the role of *catalysing fragment*. We are assuming that the intrachain condensation (characterized in our illustration by the fugacity J_{AC} of the A-C covalent bond) between the two growing fragments is greatly favoured ($J_{AC} \gg 1$) in the presence of the catalysing fragment bonded, to *both* growing fragments, through the interchain bridges.

In order to better understand the RG framework within which we shall perform calculations, let us first discuss the *single* base-pair particular case ($K_{AT} = K_{CG} \equiv K$; both A and C denoted by A; both T and G denoted by B). We perform the configurational analysis associated with the growth of a small oligomer (e.g., a *dimer* in Figure 1(a), where procedure I has been illustrated), according to the following rules: (i) we consider all the growth-active configurations of all the catalysing fragments whose size is not longer than twice the growing fragment under consideration (we want to retain only the most probable mechanisms, and the probability of occurrence of catalysing fragments *much longer* than the growing fragment is rather poor); (ii) the "weight" equals 1 when the catalysing fragment is unambiguously associated with the growing fragment under consideration, equals ½ when it can equally well be associated with the other growing fragment, and equals 0 (and is therefore absent from the figure) when it is unambiguously associated with the other fragment (to be more precise, when the number of non-connected residues at any given end of the catalysing fragment exceeds the number of its residues actually connected to the growing fragment under consideration); (iii) the number of growth-active ends (1 or 2) of the catalysing fragment can be disgarded (*procedure* I) or taken into account (*procedure* II) by introducing a "growth efficiency" which equals the number of growth-active ends; (iv) the interchain bonds are assumed independent

(hence the effective fugacity of a given set of simultaneous bonds is just the *product* of the corresponding fugacities); (v) multiple catalysing processes (involving more than one catalysing fragment) or similar complex processes are neglected because of a presumably low probability of occurrence. These set of rules obviously involve a certain degree of arbitrariness; however we believe that any other "reasonable" set of rules would lead to results not essentially different from those we shall present.

Figure 1(a) yields, through the sum of (weight) × (growing efficiency) × (fugacity), the following effective fugacity:

$$R_2^{I}(K) = K + 4K^2 \qquad \text{(procedure I)} \qquad (3)$$

Analogously we obtain, within procedure II,

$$R_2^{II}(K) = K + 5K^2 \qquad \text{(procedure II)} \qquad (3')$$

The subscripts 2 stand for dimer. We now repeat the configurational analysis for the growth of a longer oligomer (e.g., a *trimer* in Fig. 1(b), where procedure I has been illustrated). We obtain the following effective fugacity:

$$R_3^{I}(K) = K + 3K^2 + 8K^3 \qquad \text{(procedure I)} \qquad (4)$$

and also

$$R_3^{II}(K) = K + 3K^2 + 11K^3 \qquad \text{(procedure II)} \qquad (4')$$

We can now write down the RG recursive equation, namely

$$R_2^{\alpha}(K') = R_3^{\alpha}(K) \qquad (\alpha = \text{I or II}) \qquad (5)$$

Both recurrences admit the trivial (stable) fixed points $K = 0$ (corresponding to lack of infinite growth, and characterizing the *finite growth (FG) phase*) and $K = \infty$ (characterizing the *infinite growth (IG) phase*). They also admit a critical (unstable) fixed point, namely $K^* = 1/8 = 0.125$ for procedure I, and $K^* = 2/11 \simeq 0.18$ for procedure II. The present calculation provides further information: while approaching the critical value K^*, the *mean length* ξ of the growing fragment diverges as $\xi \propto \left(K^* - K\right)^{-\nu}$, where the critical exponent ν is given (within the present RG approximation) by

$$\nu = \frac{\ln(b/b')}{\ln\left(\frac{dK'}{dK}\right)_{K^*}} = \frac{\ln(b/b')}{\ln\left[\frac{dR_b(K)/dK}{dR_{b'}(K)/dK}\right]_{K^*}} \qquad (6)$$

where b(b') is the size of the original (renormalized) oligomer under analysis (in our present example, b = 3 for the trimer and b' = 2 for the dimer) and $R_b(K)$ ($R_{b'}(K')$) the corresponding effective fugacity. We obtain $\nu \simeq 7.0$ for procedure I, and $\nu \simeq 3.3$ for procedure II. The smaller and more satisfactory (because more consistent with related calculations in polymer physics) value of ν obtained through procedure II, is to be attributed to the higher realism introduced by the growth efficiency. Anyhow it is completely out of the scope of the present very crude approximations to obtain reliable numbers for K^* or ν: our arguments concern only the qualitative facts of the picture.

Summarizing, we have seen that the autocatalytic mechanism might lead, when approaching a critical value for the interchain fugacity, to the growth of codified self-replicating polymers. This already seems to us a very suggestive conclusion. However, if a *single* base-pair is assumed, all codes grow, and all do so at the same value of K: this is fine regarding diversity, *but*, from the biological standpoint, completely unsatisfactory in what concerns selection! We shall next see that the (realistic) assumptions of *two* (or more) different base-pairs, will lead to a remarkable improvement.

The parameter space of our problem will now be a two-dimensional one, namely determined by K_{AT} and K_{CG} (all intrachain fugacities are assumed infinite at this level of approximation). The RG flow will now be determined by (explicit or implicit) recursive relations of the following type:

$$K'_{AT} = f_{bb',\sigma}(K_{AT}, K_{CG}) \tag{7.a}$$

$$K'_{CG} = g_{bb',\sigma}(K_{CG}, K_{AT}) \tag{7.b}$$

where $f_{bb',\sigma}$ and $g_{bb',\sigma}$ are functions which will in general depend on the respective sizes b and b' of the original and renormalized oligomers we have, chosen to work with, *as well as on the particular code which is growing* (and which is denoted by the index σ). Examples of such codes are the following: $\ldots K_{AT}K_{AT}K_{AT}K_{AT}\ldots$ ($\sigma = 1$), $\ldots K_{CG}K_{CG}K_{CG}K_{CG}\ldots$ ($\sigma = 2$), $\ldots K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ ($\sigma = 3$), $\ldots K_{AT}K_{AT}K_{CG}K_{CG}K_{AT}K_{AT}K_{CG}K_{CG}\ldots$ ($\sigma = 4$), $\ldots K_{AT}K_{AT}K_{CG}K_{AT}K_{AT}K_{CG}\ldots$ ($\sigma = 5$). The single base-pair particular case can be obtained through three different limits, namely: (i) $K_{AT} = K_{CG} \equiv K$ and arbitrary σ, therefore $f_{bb',\sigma}(K,K) = g_{bb',\sigma}(K,K) \equiv F_{bb'}(K)$; (ii) $K_{AT} \equiv K$, arbitrary K_{CG}, and $\sigma = 1$, therefore $f_{bb',1}(K,K_{CG}) \equiv F_{bb'}(K)$; (iii) $K_{CG} \equiv K$, arbitrary K_{AT}, and $\sigma = 2$, therefore $g_{bb',2}(K,K_{AT}) \equiv F_{bb'}(K)$. Furthermore, for codes

which are invariant through $K_{AT} \rightleftarrows K_{CG}$ permutation (e.g., $\sigma = 3,4$, but not $\sigma = 5$) the following property must be satisfied: $f_{bb',\sigma}(X,Y) = g_{bb',\sigma}(X,Y)$ for arbitrary (X,Y). Several of the above properties can be verified on the following examples.

1st example: growth of the $\ldots K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ sequence ($\sigma = 3$). The RG equations are given by

$$R_5^{II}(K'_{AT},K'_{CG},K'_{AT},K'_{CG}) = R_9^{II}(K_{AT},K_{CG},K_{AT},K_{CG}) \tag{8}$$

and

$$R_5^{II}(K'_{CG},K'_{AT},K'_{CG},K'_{AT}) = R_9^{II}(K_{CG},K_{AT},K_{CG},K_{AT}) \tag{9}$$

where

$$R_5^{II}(K_{AT},K_{CG},K_{AT},K_{CG}) = K_{AT} + 3K_{AT}K_{CG} + 5K_{AT}^2K_{CG} + 7K_{AT}^2K_{CG}^2 + 29K_{AT}^3K_{CG}^2 \tag{10}$$

and

$$R_9^{II}(K_{AT},K_{CG},K_{AT},K_{CG}) = K_{AT} + 3K_{AT}K_{CG} + 5K_{AT}^2K_{CG} + 7K_{AT}^2K_{CG}^2 + 9K_{AT}^3K_{CG}^2 + 11K_{AT}^3K_{CG}^3 + 13K_{AT}^4K_{CG}^3 + 15K_{AT}^4K_{CG}^4 + 89K_{AT}^5K_{CG}^4 \tag{11}$$

See in Figure 2 the associated RG flow, which determines the corresponding critical line, and also exhibits that the two base-pairs case belongs to the same university class as the single base-pair case.

2nd example: growth of the $\ldots K_{AT}K_{AT}K_{CG}K_{CG}K_{AT}K_{AT}K_{CG}K_{CG}\ldots$ sequence ($\sigma = 4$). The RG equations are given by

$$R_5^{II}(K'_{AT},K'_{AT},K'_{CG},K'_{CG}) = R_9^{II}(K_{AT},K_{AT},K_{CG},K_{CG}) \tag{12}$$

and

$$R_5^{II}(K'_{CG},K'_{CG},K'_{AT},K'_{AT}) = R_9^{II}(K_{CG},K_{CG},K_{AT},K_{AT}) \tag{13}$$

where

$$R_5^{II}(K_{AT},K_{AT},K_{CG},K_{CG}) = K_{AT} + \tfrac{3}{2}K_{AT}^2 + \tfrac{3}{2}K_{AT}K_{CG} + \tfrac{5}{2}K_{AT}^2K_{CG} + \tfrac{5}{2}K_{AT}K_{CG}^2 + 7K_{AT}^2K_{CG}^2 + 29K_{AT}^3K_{CG}^2 \tag{14}$$

and

$$R_9^{II}(K_{AT},K_{AT},K_{CG},K_{CG}) = K_{AT} + \tfrac{3}{2}K_{AT}^2 + \tfrac{3}{2}K_{AT}K_{CG} + \tfrac{5}{2}K_{AT}^2K_{CG} + \tfrac{5}{2}K_{AT}K_{CG}^2 + 7K_{AT}^2K_{CG}^2$$

$$+9K_{AT}^{3}K_{CG}^{2}+\frac{11}{2}K_{AT}^{4}K_{CG}^{2}+\frac{11}{2}K_{AT}^{3}K_{CG}^{3}+\frac{13}{2}K_{AT}^{4}K_{CG}^{3}$$

$$+\frac{13}{2}K_{AT}^{3}K_{CG}^{4}+15K_{AT}^{4}K_{CG}^{4}+89K_{AT}^{5}K_{CG}^{4} \qquad (15)$$

The corresponding RG flow is similar to that presented in Figure 2.

Equation (10) (Eq. (14)) has been established by making the configurational analysis associated with the sequence $\ldots K_{AT}K_{XX},K_{YY},K_{CG}K_{AT}K_{XX},K_{YY},K_{CG}\ldots$ (see Figure 3), calculating the effective fugacity $R_5^{II}(K_{AT},K_{XX'},K_{YY'},K_{CG})$, and then taking XX' = CG and YY' = AT (XX' = AT and YY' = CG). We have proceeded analogously to obtain equations (11) and (15).

We have indicated in Figure 4 the critical lines corresponding to various typical sequences. We notice an important improvement with respect to the one base-pair model: the picture presents now *both* diversity and selection! In other words, a microscopic basis for Darwinian evolution is now achieved. However, and in spite of this interesting achievement, the model is not yet free from two important limitations: (i) if we assume a reasonable time evolution of K_{AT} and K_{CG} (see Figure 4), the most privileged codes are those presenting either very low or very high (A+T)/(C+G) ratios, a fact which is not easily consistent with the values ($\frac{1}{2} \lesssim (A+T)/(C+G) \lesssim 2$; see [11]) associated with modern living systems (at least in the biosphere); (ii) the critical line is one and the same for all sequences of nucleotides which correspond to a single sequence of bonds (e.g., ...ACACAC..., ...AGAGAG..., ...ACTGTCTG..., etc, correspond to the sequence $\ldots K_{AT}K_{CG}K_{AT}K_{CG}\ldots$), a fact which has no biochemical support. Both limitations disappear by considering the different intrachain fugacities (they are, within a nearest-neighbor picture, 10 in number, and will be denoted by $J_{AA},J_{AT},J_{AC},J_{AG},J_{TT},J_{TC},J_{TG},J_{CC},J_{CG}$ and J_{GG}). In fact, our approach thus far corresponds to assign to these 10 constants the value infinity. It is intuitive that finite values for these fugacities will make it *more difficult* to attain the point of infinite polymeric growth. We have indicated in Figure 5 the expected critical line assuming say that all the J's are equal among them (and equal to J), and that $K_{AT} = K_{CG} \equiv K$; note that K approaches K^* when J diverges. The fact that the actual J's are *finite and different from one another*, will make all the critical lines (of Figure 4) to shift towards *higher* values of K_{AT} and K_{CG}. This shift is in general *different for differing sequences of nucleotides, even if they preserve the same sequence of* K_{AT}*'s and* K_{CG}*'s*. The result is indicated in Figure 6, by arbitrarily choosing $J_{CC} \simeq J_{CG} \simeq J_{GG} < J_{AA} \simeq J_{AT} \simeq J_{TT} < J_{AG} \simeq J_{TC} < J_{AC} \simeq J_{TG} < \infty$.

We have not carried on actual RG calculations corresponding to finite J's . They are in principle tractable, though burdensome because of the large number of RG parameters. It is clear, in any case, that this is a realistic path for overcoming the two limitations mentioned previously.

Let us add that if we were to pursue calculations along this line, a further realistic element would have to be included in the model, namely a certain amount of cross-links which do exist between different points of the (folded) double-chain. The effect of these cross-links would presumably be (as observed, for instance, in hemoproteins) to make the (intrinsic) fractal dimensionality of the macromolecule higher than one. This would be necessary for having phase transitions at finite (not vanishing) temperatures in the presence of finite (not infinite) J's.

3. Conclusions

Within a critical phenomena approach we have studied the prebiotic stage concerning the growth of codified self-replicating polymers (DNA or RNA-like) starting from small oligomers (dimers, trimers, etc). The growth mechanism that has been adopted essentially is autocatalysis through Crick and Watson like pair complementarity. The picture which emerges can be synthetized as follows:

(i) A *single* base-pair ($K_{AT} = K_{CG}$) yields critical growth consistent with *diversity* but not with *selection;*

(ii) Two (or more) base-pairs ($K_{AT} \neq K_{CG}$) yield polymeric growth consistent with *both diversity and selection;* these fundamental ingredients naturally come into the theory, thus providing a microscopic basis for Darwinian evolution; life would have then appeared from a certain amount of self-replicating codes, and not from a single one (those different codes would have grown at different, though close, moments of the Earth evolution);

iii) The role played by *finite* values for the interchain covalent fugacities (J_{AA},J_{AC}, etc) is to make possible realistic values for the nucleotide ratio (A+T)/(C+G) (roughly between ½ and 2);

(iv) In the old querelle "which came first: nucleic acids or proteins?", our picture suggests a more primitive role for the nucleic acids;

(v) The polymeric growth we are concerned with *can* be thought as

essentially being a *thermal equilibrium* stage (no assumptions of processes far from equilibrium are needed).

As a final remark we might add that, if analogies with spin ½ magnetic systems are to be done for the prebiotic stage under study, the roughest "reasonable" model seems to be that in which *the binary code* ($S = \pm\frac{1}{2}$) *refers to the base-pairs* (A-T or C-G) *and not to the nucleotides* (purine or pirimidine).

One of us (C.T.) acknowledges hospitality received at the Service de Physique Théorique, CEN-Saclay, where the present review was written.

CAPTION FOR FIGURES

Figure 1: Configurational analysis, within procedure I, of the catalysing fragments corresponding to the growth of a *dimer* (a) and of a *trimer* (b).

Figure 2: Critical line (full line) in the (K_{AT},K_{CG}) fugacity space, separating the finite growth (FG) phase from the infinite growth (IG) one of the sequence $\ldots K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ Arrows and dashed lines indicate RG flow; the central dot indicates the single base-pair critical fixed point (responsible for the universality class of the whole critical line).

Figure 3: Configurational analysis, within both procedures I and II, of the catalysing fragments corresponding to the growth of a *pentamer* (sequence $\ldots K_{AT}K_{XX},K_{YY},K_{CG}K_{AT}K_{XX},K_{YY},K_{CG}\ldots$).

Figure 4: Critical lines (in the (K_{AT},K_{CG}) fugacity space) corresponding to the growth of selected sequences (the dashed line is indicative); FG (IG) denotes the finite (infinite) growth phase. The point at $K_{AT} = K_{CG} = K^*$ reproduces the fixed point of Figure 2; the dotted line is a symmetry axis of some of the sequences (e.g., $\ldots K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ and $\ldots K_{AT}K_{AT}K_{CG}K_{CG}K_{AT}K_{AT}K_{CG}K_{CG}\ldots$). The arrows indicate a plausible (slow) time evolution of K_{AT} and K_{CG}.

Figure 5: Indicative FG-IG critical line (one and the same for al sequence types) corresponding to $K_{AT}\ K_{CG} \equiv K$ and all J's equal among them (and equal to J). K^* refers to the single base-pair critical point of Figure 2.

Figure 6: Indicative FG-IG critical lines corresponding to the growth of different *nucleotide sequences* (not only different sequences of interchain links). The dashed line is a symmetry axis of some sequences (e.g., ...ACAC..., ...AGAG...); the dotted lines indicate the value K^* of all previous figures. The arrows indicate a plausible (slow) time evolution of K_{AT} and K_{CG}.

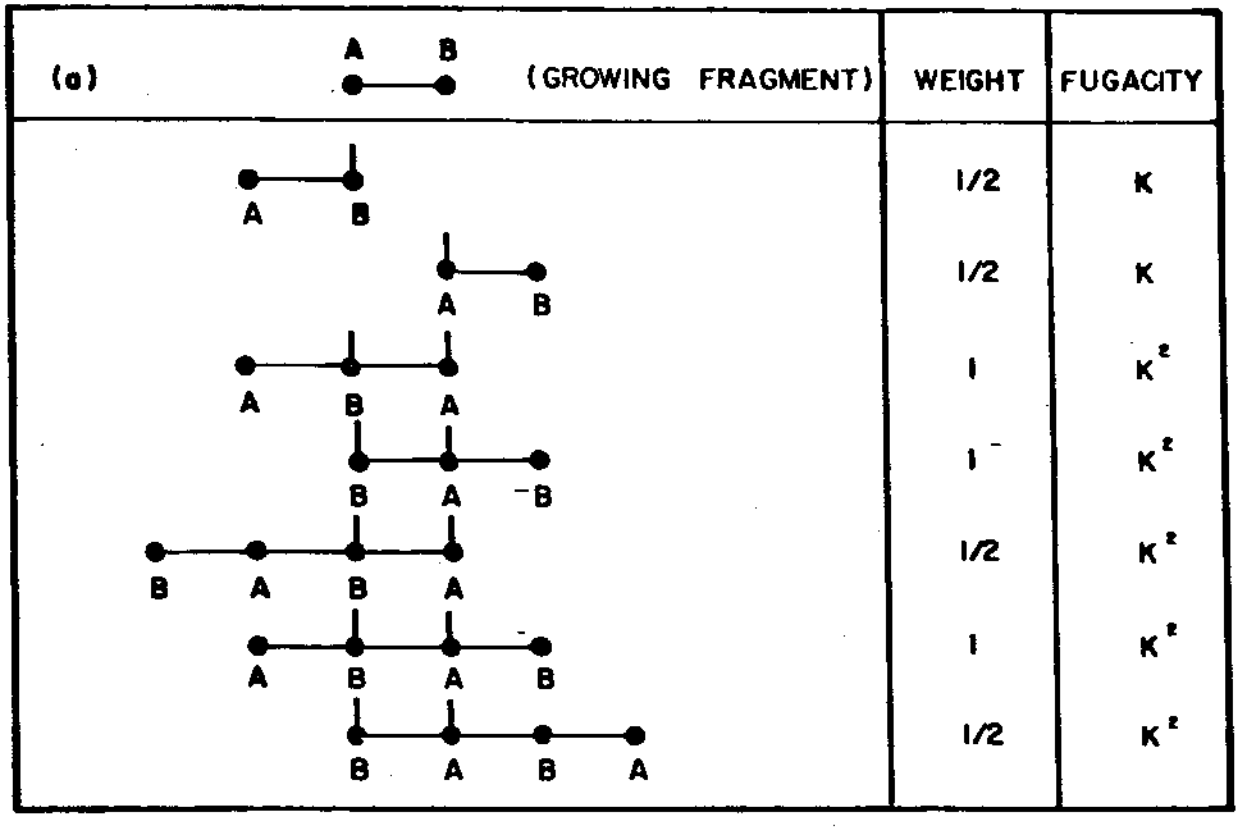

(a)
A B
(GROWING FRAGMENT)
WEIGHT
FUGACITY
A B
1/2
K
A B
1/2
K
A B A
1
K²
B A B
1
K²
B A B A
1/2
K²
A B A B
1
K²
B A B A
1/2
K²

FIG. 1

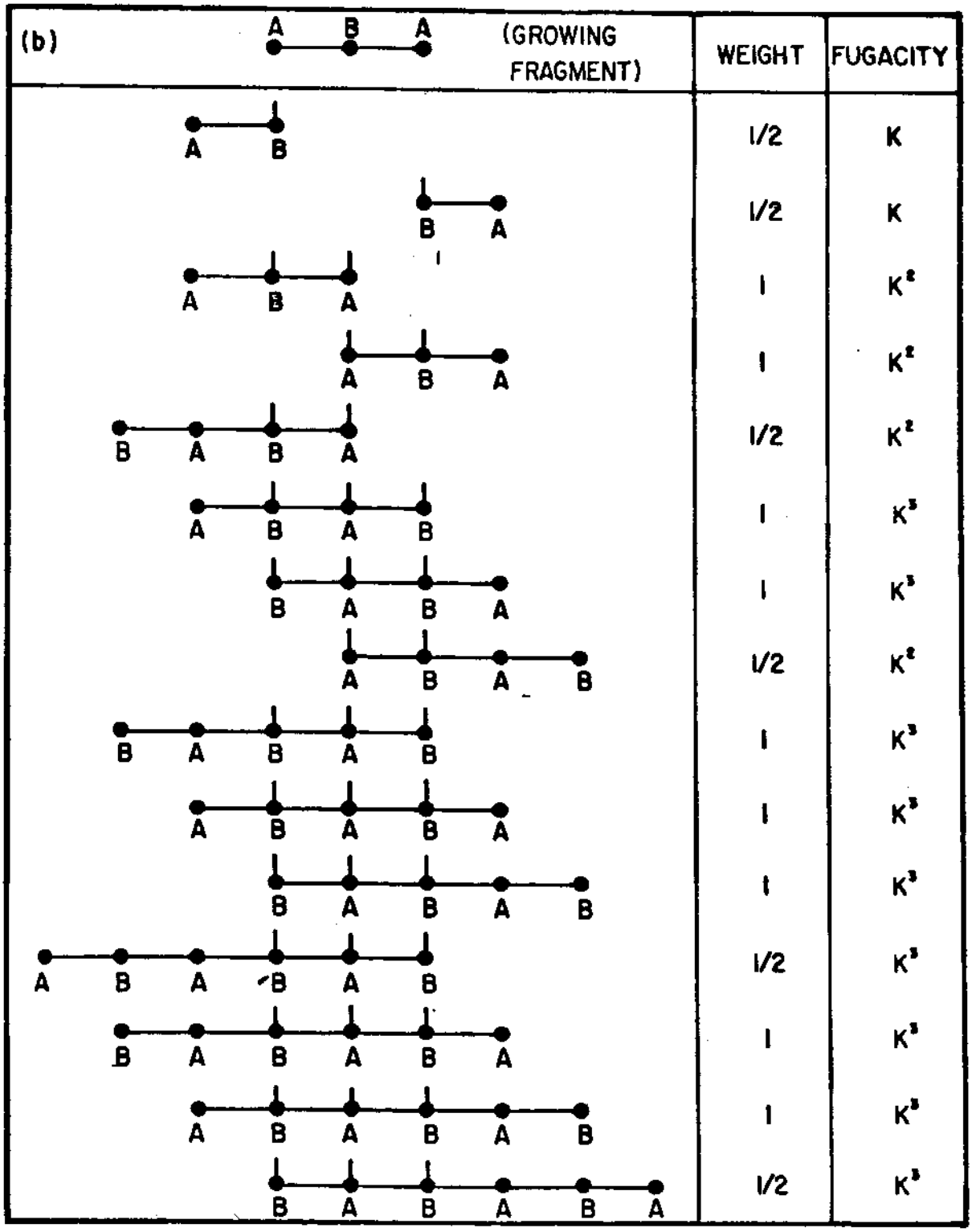

(b)
A B A
(GROWING FRAGMENT)
WEIGHT
FUGACITY
A B
1/2
K
B A
1/2
K
A B A
1
K²
A B A
1
K²
B A B A
1/2
K²
A B A B
1
K³
B A B A
1
K³
A B A B
1/2
K²
B A B A B
1
K³
A B A B A
1
K³
B A B A B
1
K³
A B A B A B
1/2
K³
B A B A B A
1
K³
A B A B A B
1
K³
B A B A B A
1/2
K³

FIG. 1

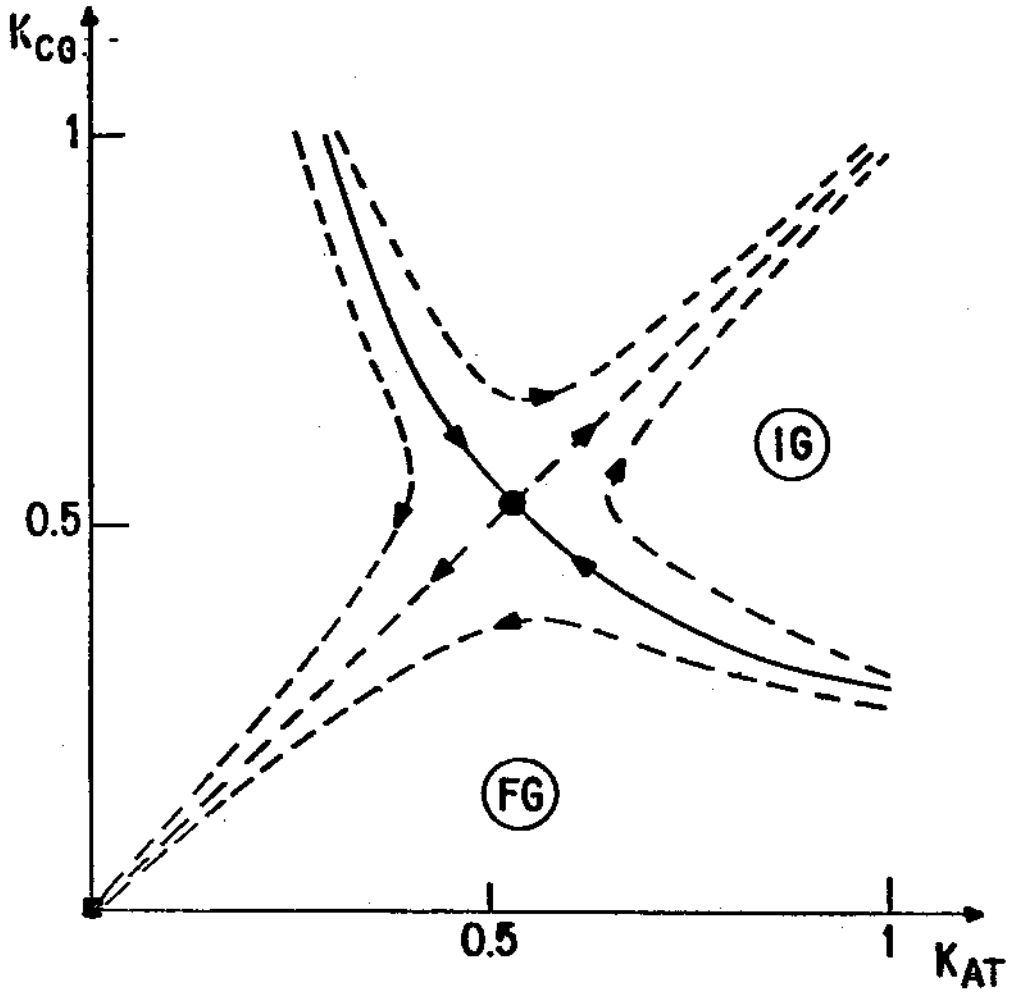


FIG. 2

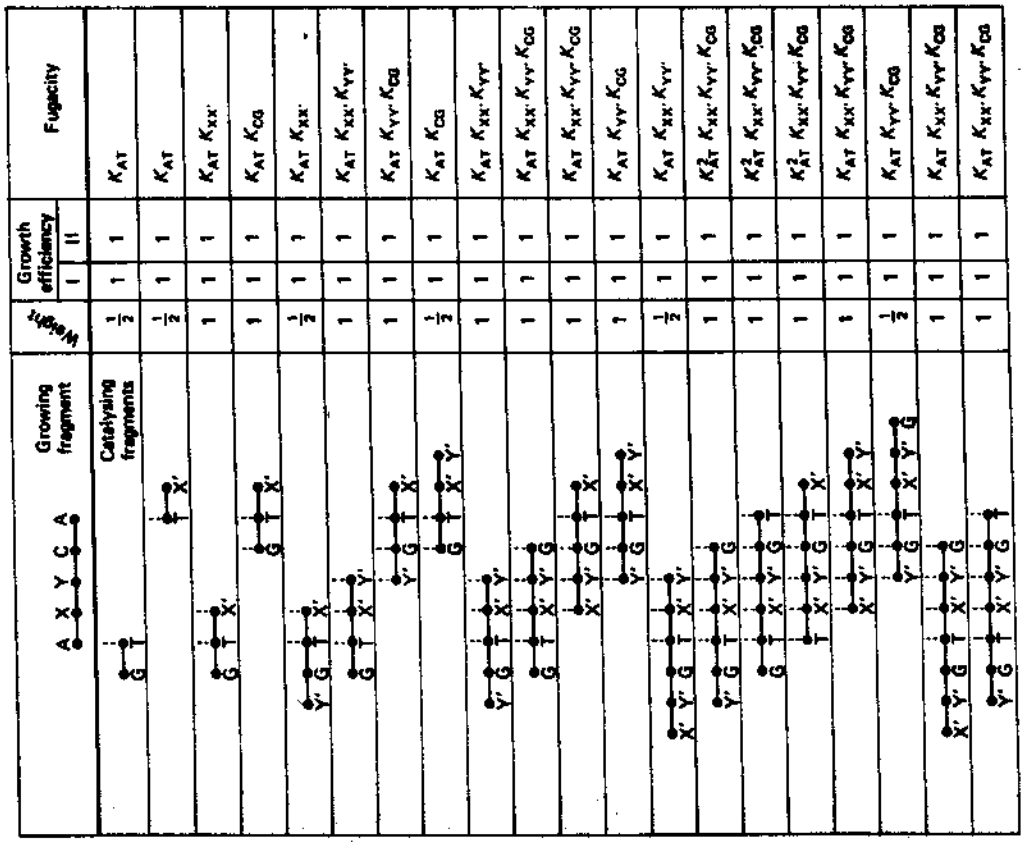

Growing fragment
Catalysing fragments
Weight
Growth efficiency
I
II
Fugacity

FIG. 3

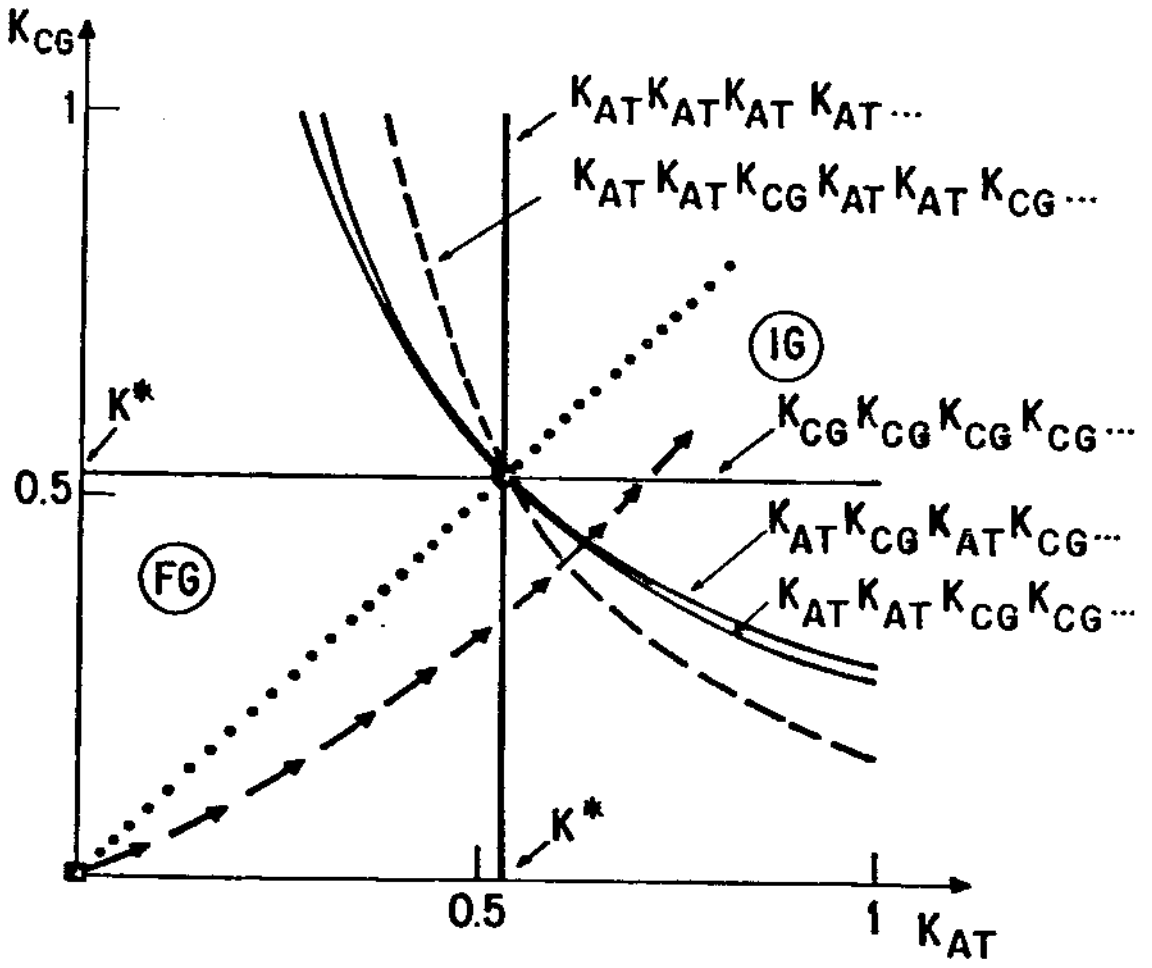

K_{CG}
1
0.5
K^*
FG
IG
$K_{AT}K_{AT}K_{AT}K_{AT}\cdots$
$K_{AT}K_{AT}K_{CG}K_{AT}K_{AT}K_{CG}\cdots$
$K_{CG}K_{CG}K_{CG}K_{CG}\cdots$
$K_{AT}K_{CG}K_{AT}K_{CG}\cdots$
$K_{AT}K_{AT}K_{CG}K_{CG}\cdots$
K^*
0.5
1
K_{AT}

FIG. 4

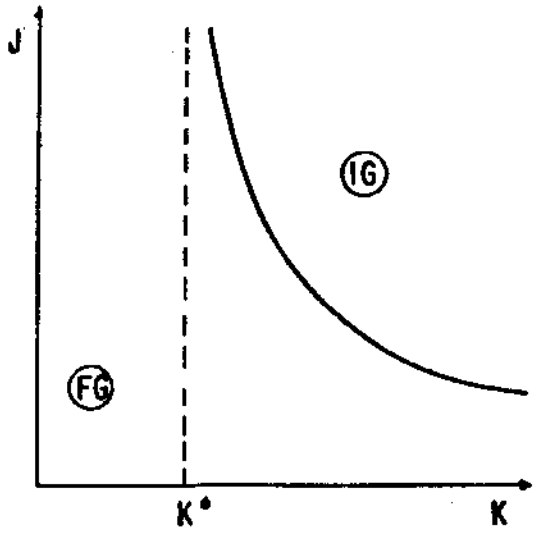


FIG. 5

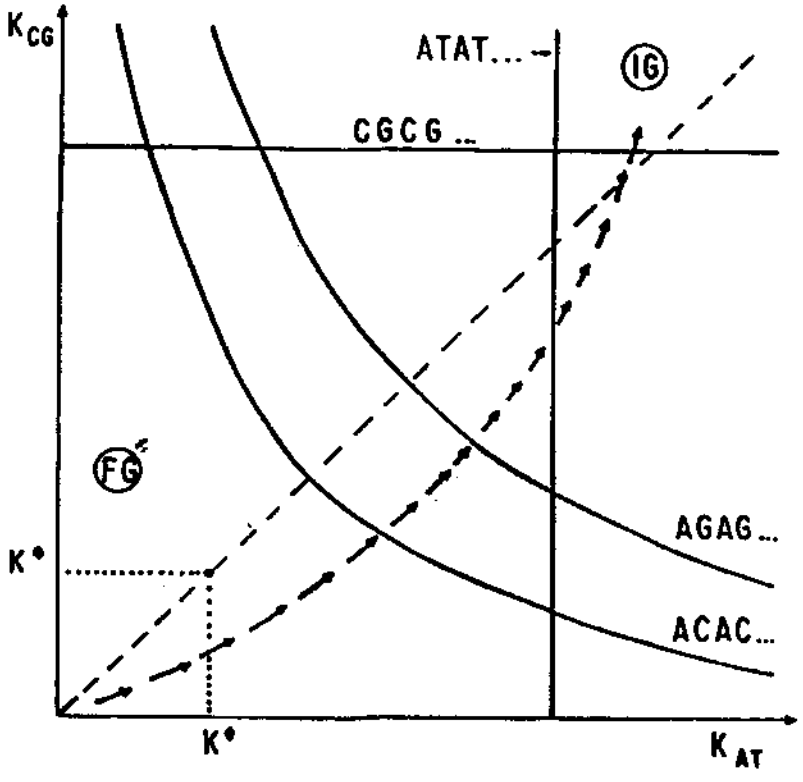


FIG. 6

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