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ON THE ROLE OF COMPLEMENTARITY IN BIOGENESIS: A CRITICAL PHENOMENON APPROACH

by

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ABSTRACT

In the series of processes leading to the origin of life, the growth of information-containing self-replicating polymers from a mixture of oligomers probably was one of the very last equilibrium steps, before the onset of non-equilibrium phenomena which characterize living systems. We describe that crucial step as a critical phenomenon, treated within the renormalization group framework. We show that the diversity-stability duality of Darwinian evolution is achieved at this stage if we start from four different monomers capable of forming two complementary pairs.

Key-words: Biogenesis; Complementarity; Critical phenomenon; Renormalization group.

I INTRODUCTION

One of the earliest crucial stages in the origin of living systems is the growth of self-replicating information-containing polymers, starting from essentially uniquely characterized simple molecules (see, for example, Ref. [1]). Recently [2], we have discussed this question thought as a critical phenomenon and, as such, treated within the renormalization group (RG) framework [3,4]. We started with a system containing *two* complementary monomers (residues), A and B, complementarity being understood in the sense of the Crick and Watson base-pairs. We then showed that the effective fugacity (denoted by K, and corresponding to the complementary interaction) associated with the growing system, admits a *critical* value (denoted K* and corresponding to an *unstable* fixed point of the RG recursive relation), above which growth (i.e., polymerization) goes on indefinitely; it is clear that indefinite growth is just a first approximation of the problem (in fact complex chemical mechanisms, beyond the compass of the present theory but unimportant within the present context, will stop the growth when a certain large size is achieved). In principle, this growth mechanism (which will be recalled in detail later on, but which essentially consists in condensation reactions catalysed by fragments complementary to the original ones) provides, with equal chance, *any* sequence of monomers A and B (e.g., ABABAB..., AABBAABB..., ABBABB..., etc). In other terms, the preliminary version [2] of our model suffers from the restriction that, instead of producing potentially meaningful

(from a biological standpoint) sequences, we would end-up with a chaotic mass of unrelated strings. This limitation is common to other models of polymeric growth, such as the earlier one of Anderson and Stein [5], in which *diversity* is not restricted by *stability* [6]. On the other hand, models based on the minimization of the chemical potential [7], leading to a single stable sequence of oligomers are unsatisfactory, since diversity is a necessary ingredient in Evolution.

Anderson [6] has proposed a model based on a spin-glass-like Hamiltonian, which recovers both diversity and stability. We have now found that our model [2] can be modified to incorporate the diversity-stability duality, provided we start with *four* monomers (denoted A,T,C and G) capable of forming *two* (instead of *one*) complementary pairs (namely A-T and C-G). In spite of our notation the four residues A,T,C and G, could be distinct from the well known nucleotides: they could be their precursors, for example. In Section II we introduce the model and the RG formalism; in Section III we present the RG results; in Section IV we analyze the effects of intrachain interactions; finally, we discuss the main conclusions in Section V.

II MODEL AND FORMALISM

The monomers A,T,C and G can form strings by *intrachain* bonding (through condensation reactions leading to covalent bonds), but they also can form *interchain* bonds (through hydrogen bridges, for example). One such polymer could be

$$\begin{array}{c} -A-G-T-A-C-G- \\ \vdots\ \vdots\ \vdots\ \vdots\ \vdots\ \vdots \\ -T-C-A-T-G-C- \end{array} \qquad (1)$$

Let K_{AT} ($K_{AT} > 0$) be the fugacity (or bonding constant) of the A---T pair, and K_{CG} the fugacity of the C---G pair. Both K_{AT} and K_{CG} depend, in a complex unknown way, on a great number of thermodynamical equilibrium external parameters such as temperature, pressure, water concentration, various salts concentrations, etc.

We assume that the chain fragments will grow by the mechanism adopted before [2], and which can be illustrated as follows:

$$(A\text{-}G\text{-}T\text{-}A) + (C\text{-}G) \xrightarrow[\longleftarrow]{(A\text{-}T\text{-}G\text{-}C)}$$

$$(A\text{-}G\text{-}T\text{-}A)(C\text{-}G) \rightleftharpoons (A\text{-}G\text{-}T\text{-}A\text{-}C\text{-}G)$$
$$\quad\ \vdots\ \vdots\ \vdots\ \vdots \qquad\qquad\quad \vdots\ \vdots\ \vdots\ \vdots$$
$$(A\text{-}T\text{-}G\text{-}C) \qquad\qquad (A\text{-}T\text{-}G\text{-}C)$$

$$\rightleftharpoons (A\text{-}G\text{-}T\text{-}A\text{-}C\text{-}G) + (A\text{-}T\text{-}G\text{-}C) \qquad (2)$$

Notice that we have obtained the top sequence of (1). 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 a *catalysing fragment*.

We shall now construct the RG recursive relations through the configurational analysis of the aggregation of small parts of the large chain we want to produce. In Ref. [2] we performed the analysis of the configurations of the catalysing

fragments associated with the growth of a dimer and of a trimer (growing fragments). The consideration of larger oligomers will in principle improve the result (the exact result being hopefully attained when infinitely large growing fragments are considered). In the present paper, in order to illustrate the influence, on the critical phenomenon we are interested in, of the *sequence* of monomers in the final polymer, we shall consider growing fragments whose size goes up to nine monomers (*nonamers*). An additional benefit will be the numerical improvement on quantities such as critical points and critical exponents.

To make the RG procedure clear, let us first focuse a simple situation, namely the growth of the $K_{AT}K_{AT}K_{AT}K_{AT}\ldots$ type sequence. This corresponds to an infinite number of polymeric sequences (e.g., AAAA..., AAAT..., AATT..., ATAT ..., etc.); all these polymers will grow *simultaneously* within the present approximation (this unrealistic degeneracy will disappear with the better approximation discussed in Section IV). Let us arbitrarily choose the AAAA... sequence as a prototype, and first consider the AAA trimer as a growing fragment.

The configurational analysis of the catalysing fragments is indicated in Table I. To construct this Table, the following rules have been adopted: (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 1/2 when it can equally well be associated with the other growing fragment, and equals 0 (and is therefore absent from the Table) 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 disregarded (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 it is believed that any other "reasonable" set of rules would lead to results not essentially different from those we shall obtain.

Table I yields, through the sum of (weight) x (growing efficiency) x (fugacity), the following effective fugacity:

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

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

The subscript ^{3} stands for trimer. Eq. (3) coincides with Eq. (3) of Ref. [2].

We can treat the pentamer as a growing fragment (of the same sequence $K_{AT}K_{AT}K_{AT}K_{AT}...$) in the same way we have treated the trimer: the corresponding configurational analysis is indicated in Table II (where, for the present needs, *all* single fugacities are to be taken equal to K_{AT}). The corresponding effective fugacities are given by:

$$R_5^I(K_{AT}) = K_{AT} + 3K_{AT}^2 + 5K_{AT}^3 + 7K_{AT}^4 + 19K_{AT}^5 \quad \text{(procedure I)} \qquad (5)$$

$$R_5^{II}(K_{AT}) = K_{AT} + 3K_{AT}^2 + 5K_{AT}^3 + 7K_{AT}^4 + 29K_{AT}^5 \quad \text{(procedure II)} \qquad (6)$$

The RG recursive relation renormalizing pentamer into trimer is given by:

$$R_3^I(K'_{AT}) = R_5^I(K_{AT}) \qquad \text{(procedure I)} \qquad (7)$$

$$R_3^{II}(K'_{AT}) = R_5^{II}(K_{AT}) \qquad \text{(procedure II)} \qquad (8)$$

Both recurrences admit the trivial (stable) fixed points $K_{AT} = 0$ (corresponding to lack of growth) and $K_{AT} = \infty$ (corresponding to infinite growth). They also admit a critical (unstable) fixed point, namely $K^* = (\sqrt{277}-7)/38 \simeq 0.254$ for procedure I, and $K^* = (\sqrt{696}-7)/58 \simeq 0.334$ for procedure II..

The present calculation provides a further information: while approaching the critical value K^*, the *mean length* ξ of the growing fragment diverges as $\xi \propto (K^* - K_{AT})^{-\nu}$, where the

critical exponent ν is given (within the present RG approximation) by

$$\nu = \frac{\ell n(b/b')}{\ell n \left. \frac{dK'_{AT}}{dK_{AT}} \right|_{K^*}} = \frac{\ell n(b/b')}{\ell n \left[\frac{dR_b(K_{AT})/dK_{AT}}{dR_{b'}(K_{AT})/dK_{AT}} \right]_{K^*}} \qquad (9)$$

b(b') is the size of the original (renormalized) oligomer under analysis (in our present calculation, b = 5 for the pentamer and b' = 3 for the trimer), and $R_b(K_{AT})$ ($R_{b'}(K_{AT})$) the corresponding effective fugacity. For (b,b') = (5,3) we obtain $\nu \simeq 7.9$ for procedure I, and $\nu \simeq 4.3$ for procedure II. The smaller and more satisfactory (because more consistent with related calculations for standard growth models) value of ν obtained through procedure II, is to be attributed to the realism of the theoretical improvement introduced by the growth efficiency. To discuss the $K_{AT}K_{AT}K_{AT}K_{AT}\ldots$ sequence, we have illustrated the calculation by using (b,b') = (5,3), but no problem exists in principle in using b' = 2,3,4,... and b' = b + 1, b + 2,... (see Ref. [2] for (b,b') = (3,2) within procedure I). To conclude the one RG-parameter (K_{AT} in our case) case, it is worthwhile to mention the obvious fact that whatever has been said for the $K_{AT}K_{AT}K_{AT}K_{AT}\ldots$ type of sequence, holds as it stands for the $K_{CG}K_{CG}K_{CG}K_{CG}\ldots$ one.

Let us now treat the two RG-parameter case, namely, the (K_{AT}, K_{CG}) problem. Our purpose is to construct a RG recurrence in the (K_{AT}, K_{CG}) space, in order to show the influence of the sequence on criticality. We consider the sequences $K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ (which corresponds to the prototype ACAC...,

as well as to ACTC..., ACGC..., ACTG..., etc.) and $K_{AT}K_{AT}K_{CG}K_{CG}$... (which corresponds to the prototype AACC..., as well as to ATCC..., ATGG..., AACG..., etc.). They can be treated all together through the sequence $K_{AT}K_{XX'}K_{YY'}K_{CG}K_{AT}K_{XX'}K_{YY'}K_{CG}$..., by considering later on either $(K_{XX'}, K_{YY'}) = (K_{CG}, K_{AT})$ or $(K_{XX'}, K_{YY'}) = (K_{AT}, K_{CG})$. The simplest meaningfull RG choice consists in considering the nonamer $K_{AT}K_{XX'}K_{YY'}K_{CG}K_{AT}K_{XX'}K_{YY'}K_{CG}K_{AT}$ as original oligomer, and the pentamer $K_{AT}K_{XX'}K_{YY'}K_{CG}K_{AT}$ at the renormalized one. Were we to calculate even oligomers (e.g., the tetramer $K_{AT}K_{XX'}K_{YY'}K_{CG}$), we should have problems later on, namely *one* independent recursive (scalar) relation, instead of the *two* we need, the RG space being two-dimensional; this problem is due to the fact that the oligomer $K_{AT}K_{CG}K_{AT}K_{CG}$ (and the same holds for $K_{AT}K_{AT}K_{CG}K_{CG}$) is *invariant* under $K_{AT} \rightleftarrows K_{CG}$, and therefore preserves the effective fugacity.

We shall perform the calculations only within our best proposal, namely procedure II. The configurational analysis associated with the prototype pentamer AXYCA (corresponding to $K_{AT}K_{XX'}K_{YY'}K_{CG}K_{AT}$) is indicated in Table II. We obtain the following effective fugacity

$$
\begin{aligned}
R_5^{II}(K_{AT}, K_{XX'}, K_{YY'}, K_{CG}) &= K_{AT} + \frac{3}{2}\,K_{AT}K_{XX'} \\
&+ \frac{3}{2}\,K_{AT}K_{CG} + \frac{5}{2}\,K_{AT}K_{XX'}K_{YY'} \\
&+ \frac{5}{2}\,K_{AT}K_{YY'}K_{CG} + 7K_{AT}K_{XX'}K_{YY'}K_{CG} \\
&+ 29K_{AT}^2K_{XX'}K_{YY'}K_{CG} \qquad (10)
\end{aligned}
$$

hence

$$R_5^{II}(K_{AT}, K_{CG}, K_{AT}, K_{CG}) = K_{AT} + 3K_{AT}K_{CG} + 5K_{AT}^2K_{CG}$$

$$+ 7K_{AT}^2K_{CG}^2 + 29K_{AT}^3K_{CG}^2 \qquad (11)$$

and

$$R_2^{II}(K_{AT}, K_{AT}, K_{CG}, K_{CG}) = K_{AT} + \frac{3}{2}K_{AT}^2 + \frac{3}{2}K_{AT}K_{CG}$$

$$+ \frac{5}{2}K_{AT}^2K_{CG} + \frac{5}{2}K_{AT}K_{CG}^2 + 7K_{AT}^2K_{CG}^2 + 29K_{AT}^3K_{CG} \qquad (12)$$

The Table associated with the prototype nonamer AXYCAXYCA (corresponding to $K_{AT}K_{XX'}K_{YY'}K_{CG}K_{AT}K_{XX'}K_{YY'}K_{CG}K_{AT}$) is too lengthy to be reproduced here. It yields

$$R_9^{II}(K_{AT}, K_{XX'}, K_{YY'}, K_{CG}) = K_{AT} + \frac{3}{2}K_{AT}K_{XX'} + \frac{3}{2}K_{AT}K_{CG}$$

$$+ \frac{5}{2}K_{AT}K_{XX'}K_{YY'} + \frac{5}{2}K_{AT}K_{YY'}K_{CG} + 7K_{AT}K_{XX'}K_{YY'}K_{CG}$$

$$+ 9K_{AT}^2K_{XX'}K_{YY'}K_{CG} + \frac{11}{2}K_{AT}^2K_{XX'}^2K_{YY'}K_{CG}$$

$$+ \frac{11}{2}K_{AT}^2K_{XX'}K_{YY'}K_{CG}^2 + \frac{13}{2}K_{AT}^2K_{XX'}^2K_{YY'}^2K_{CG}$$

$$+ \frac{13}{2}K_{AT}^2K_{XX'}K_{YY'}^2K_{CG}^2 + 15K_{AT}^2K_{XX'}^2K_{YY'}^2K_{CG}^2$$

$$+ 89K_{AT}^3K_{XX'}^2K_{YY'}^2K_{CG}^2 \qquad (13)$$

hence

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

$$+ 7K_{AT}^2K_{CG}^2 + 9K_{AT}^3K_{CG}^2 + 11K_{AT}^3K_{CG}^3 + 13K_{AT}^4K_{CG}^3$$

$$+ 15K_{AT}^4K_{CG}^4 + 89K_{AT}^5K_{CG}^4 \qquad (14)$$

and

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

$$+ \frac{5}{2}K_{AT}K_{CG}^2 + 7K_{AT}^2K_{CG}^2 + 9K_{AT}^3K_{CG}^2 + \frac{11}{2}K_{AT}^4K_{CG}^2$$

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

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

The problem can now be considered as formally solved, as the RG recurrence (in the (K_{AT}, K_{CG}) space) we were looking for, is given by

$$R_5^{II}(K_{AT}', K_{CG}', K_{AT}', K_{CG}') = R_9^{II}(K_{AT}, K_{CG}, K_{AT}, K_{CG}) \qquad (16)$$

and (using the $K_{AT} \rightleftarrows K_{CG}$ symmetry)

$$R_5^{II}(K_{CG}', K_{AT}', K_{CG}', K_{AT}') = R_9^{II}(K_{CG}, K_{AT}, K_{CG}, K_{AT}) \qquad (17)$$

for the $K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ sequence, and is given by

$$R_5^{II}(K_{AT}', K_{AT}', K_{CG}', K_{CG}') = R_9^{II}(K_{AT}, K_{AT}, K_{CG}, K_{CG}) \qquad (18)$$

and (using the $K_{AT} \rightleftarrows K_{CG}$ symmetry)

$$R_5^{II}(K_{CG}', K_{CG}', K_{AT}', K_{AT}') = R_9^{II}(K_{CG}, K_{CG}, K_{AT}, K_{AT}) \qquad (19)$$

for the $K_{AT}K_{AT}K_{CG}K_{CG}\ldots$ sequence. The results are presented and discussed in the next Section.

III RESULTS

Eqs. (16) and (17) completely determine the RG flow for the $K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ sequence; the results are indicated in Fig. 1. Note that the $K_{AT} = K_{CG}$ axis constitutes a flow-invariant subspace, which recovers, in the present formalism, the situation depicted in Ref. [2], namely that where only *one* pair of complementary monomers exists. The trivial (fully stable; noted ■ in Fig. 1) fixed points $(K_{AT}, K_{CG}) = (0,0)$ and (∞,∞) respectively correspond to the *finite growth* (FG) and *infinite growth* (IG) regions. The critical (semi-stable, denoted ● in Fig. 1) fixed point $(K_{AT}, K_{CG}) = (K^*,K^*)$ determines the main universality class of the problem; in other words, the critical exponent ν occuring in the FG-IG critical line (separatrix of the RG flow in Fig. 1) is that of the *one* pair (of complementary monomers) problem, i.e. that given by Eq. (9). For the present (b,b') = (9,5) approximation we have obtained $K^* \simeq 0.544$ and $\nu \simeq 3.6$.

The critical line associated with the $K_{AT}K_{AT}K_{CG}K_{CG}\ldots$ type sequence is determined by Eqs. (18) and (19), and the results

are absolutely similar to those obtained for the $K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ type sequence (in particular K* and ν are precisely the same). An overall view is presented in Fig. 2. Above a given critical line, the corresponding sequence macroscopically(up to hundreds of monomers, in practice) grows. All the symmetric lines(with respect to the $K_{AT} = K_{CG}$ axis), such as that of the $K_{AT}K_{CG}K_{AT}K_{CG}\ldots$ and $K_{AT}K_{AT}K_{CG}K_{CG}\ldots$ sequences, satisfy the nucleotide ratio $(A+T)/(C+G) = 1$. In fact, this ratio can also be satisfied by other sequences (e.g., the sequence whose period is $K_{AT}K_{AT}K_{AT}K_{CG}K_{CG}K_{AT}K_{CG}K_{CG}$) whose crtical lines are not symmetrical. The pure sequences $K_{AT}K_{AT}K_{AT}K_{AT}\ldots$ and $K_{CG}K_{CG}K_{CG}K_{CG}\ldots$ either vanish or diverge the above mentionned ratio, and the sequence $K_{AT}K_{AT}K_{CG}K_{AT}K_{AT}K_{CG}\ldots$ yields $(A+T)/(C+G) = 2$. The ratios actually existing (at least in the biosphere) roughly belong[8] to the interval $[1/2, 2]$. Consequently, the present model, in spite of the non trivial fact that *it succeeds in differentiating classes of sequences*, is not free from a serious limitation. This is that any reasonable time evolution in the (K_{AT}, K_{CG}) space (see Fig. 2), i.e. a time evolution which starts (on Earth, several billions of years ago) in the neighbourhood of $(K_{AT}, K_{CG}) = (0,0)$ and arrives (on Earth, nowadays) in the IG region, *will unduly privilege the* (non existing in living organisms) *sequences close to the pure ones.*

A second important limitation of our model in its present stage is the already mentioned degeneracy which makes the growth of sequences such as AAAA...,AAAT...,AATT...,ATAT..., etc (i.e., families of monomer sequences which share the same sequence of K_{AT}'s and K_{CG}'s) *indistinguishable*. We argue, in

next Section, that *both* defects are simultaneously overcome if we take into account the effects of the intrachain (covalent) bonds.

IV INTRACHAIN INTERACTIONS

All our effective fugacities (such as Eqs. (3) - (6), (10) and (13)), on which the RG recursive relations are constructed, were formulated supposing that *any* appropriately binded complementary fragment would *inevitably* increase the size of the growing fragment. This is clearly an oversimplification, since the chain growth also depends on the different *intrachain* binding constants (these will be denoted J_{AA}, J_{AT}, J_{AC}, J_{AG}, J_{TT}, J_{TC}, J_{TG}, J_{CC}, J_{CG} and J_{GG}; they are 10 in number, within a nearest-neighbour picture). In fact, our approach thus far corresponds to assign to these 10 constants the value infinity. It is intuitive that *finite* values for these constants will make it *more difficult* to attain the point of indefinite growth of the polymers. We have indicated in Fig. 3 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 Fig. 2) to shift towards *higher* values of K_{AT} and K_{CG}. This shift is *different for differing sequences of monomers, even if they preserve the same sequence of* K_{AT}*'s and* K_{CG}*'s*. The result is indicated in

Fig. 4, with the (desired) disappearance of the privileged point existing in Fig. 2 (denoted ●). The particular situation depicted in Fig. 4 is consistent with the choice

$$H_{CC} \simeq H_{CG} \simeq H_{GG} < H_{AA} \simeq H_{AT} \simeq H_{TT} < H_{AG} \simeq H_{TC}$$

$$< H_{AC} \simeq H_{TG} < \infty .$$

We have not carried on actual RG calculations corresponding to finite J's, but they are in principle tractable (although somewhat burdensome, because of the increasingly large number of RG parameters). It is clear, in any case, that this is a realistic path for overcoming the two difficulties mentioned in the previous Section.

V CONCLUSIONS

One of the important steps of the prebiotic stage of the origin of life is the transition from a random assembly of oligomers to an information-containing self-replicating polymer. We have recently described this phenomenon as a critical one[2]; in the present paper we introduce a more realistic model, involving *two pairs* of complementary monomers, and making allowance for the monomer interactions along the chain.

Our polymer is essentially single-stranded; it is self-replicating since a given sequence along the chain say, ...ATCT... can act as a catalysing fragment, provided growing fragments,

or oligomers for that matter, are available in the surroundings.

The model has not a built-in feature to assure equal (or at least finite) size for the polymer molecules. For most proteins, of course, size homogeniety is essential, but the strict requirement was probably relaxed at an early equilibrium stage, such as the one we describe. Anyhow, on critical phenomena grounds, the present theory yields, for the picture with *two* pairs of complementary monomers, the same universality class obtained within the picture with only *one* of such pairs.

The *diversity-stability* duality, typical of Darwinian evolution, is incorporated in the model in a natural way. This vital aspect is introduced by the finite, differing values of the intrachain binding constants J's. Figure 4 shows that, as the system evolves in time, one sequence is bound to reach the critical point before the others, and will therefore grow and dominate its environment. Which is the favoured sequence depends on the values of the K's and J's, which are, in their turn, functions of temperature, pressure, dielectric constant, ionic strength and other physico-chemical properties of the surroundings. If these conditions, as well as the concentrations of particular oligomers, change, some other sequence might become the dominant one. Anyhow it is important to stress that the present proposal is consistent with reasonable nucleotide ratios $(A+T)/(C+G)$. With respect to the type of sequence, no "a priori" restriction exists, and it is only because of computational simplicity that we have mainly focused periodic sequences.

Our model, giving the overhelming importance of complementary

interactions, clearly favours a RNA (or DNA) precurser over a protein one. It is possible, however, that some residual complementarity existed between pairs of early amino-acids (for a recent discussion on amino-acid pairing, see ref. [9]). In fact, pairwise complementarity is not infalible, and consequently it is a source of errors in the sequence of oligomers during growth and replication.

In the last years, other theoretical schemes have been proposed to describe the prebiotic evolution of self-replicating macromolecules. Besides the extensive phenomenological model of Eigen and Schuster [7], Anderson [6], Dyson [10] and Demetrius [11] have tackled this problem within Statistical Mechanics frameworks. It would be most interesting to make a comparative analysis of these models, but we leave this for a forthcoming opportunity.

Let us conclude by saying that, through the present proposal, a picture emerges of what might have been the *last* important thermodynamical equilibrium prebiotic step, before the beginning of essentially non-equilibrium phenomena.

CAPTION FOR TABLES AND FIGURES

Table 1 - Configurational analysis of the catalysing fragments corresponding to the growth of a *trimer* (sequence type: $K_{AT}\,K_{AT}\,K_{AT}\ldots$)

Table 2 - Configurational analysis of the catalysing fragments corresponding to the growth of a *pentamer* (sequence type: $K_{AT}\,K_{XX}, K_{YY}, K_{CG}\,K_{AT}\,K_{XX}, K_{YY}, K_{CG}\ldots$)

Fig. 1 - 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 type $K_{AT}\,K_{CG}\,K_{AT}\,K_{CG}\ldots$. Arrows and dashed lines indicate RG flow; the central dot indicates the "isotropic" critical fixed point (responsible for the universality class of the whole critical line).

Fig. 2 - Critical lines (in the (K_{AT}, K_{CG}) fugacity space) corresponding to the growth of selected sequence types (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 Fig. 1; the dotted line is a symmetry axis of some of the sequence types (e.g., $K_{AT}\,K_{CG}\,K_{AT}\,K_{CG}\ldots$ and $K_{AT}\,K_{AT}\,K_{CG}\,K_{GC}\ldots$). The arrows indicated a plausible (slow) time evolution of K_{AT} and K_{CG}.

Fig. 3 - Indicative FG-IG critical line (one and the same for all 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 "isotropic" critical point of Fig. 1.

Fig. 4 - Indicative FG-IG critical lines corresponding to the growth of *different polymer sequences* (not only different sequence types; see the text). 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 A growing fragment	Weight	growth efficiency I	growth efficiency II	fugacity
T T catalysing fragments	$\frac{1}{2}$	1	1	K_{AT}
T T	$\frac{1}{2}$	1	1	K_{AT}
T T T	1	1	1	K_{AT}^2
T T T	1	1	1	K_{AT}^2
T T T T	$\frac{1}{2}$	1	1	K_{AT}^2
T T T T	1	1	1	K_{AT}^3
T T T T	1	1	1	K_{AT}^3
T T T T	$\frac{1}{2}$	1	1	K_{AT}^2
T T T T T	1	1	1	K_{AT}^3
T T T T T	1	1	2	K_{AT}^3
T T T T T	1	1	1	K_{AT}^3
T T T T T T	$\frac{1}{2}$	1	1	K_{AT}^3
T T T T T T	1	1	2	K_{AT}^3
T T T T T T	1	1	2	K_{AT}^3
T T T T T T	$\frac{1}{2}$	1	1	K_{AT}^3

TABLE 1

growing fragment	catalysing fragments	Weight	growth efficiency I	growth efficiency II	fugacity
A X Y C A	G T	$\frac{1}{2}$	1	1	K_{AT}
	T X'	$\frac{1}{2}$	1	1	K_{AT}
	G T X'	1	1	1	$K_{AT}\ K_{XX'}$
	G T X'	1	1	1	$K_{AT}\ K_{CG}$
	Y' G T X'	$\frac{1}{2}$	1	1	$K_{AT}\ K_{XX'}$
	G T X' Y'	1	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}$
	Y' G T X'	1	1	1	$K_{AT}\ K_{YY'}\ K_{CG}$
	G T X' Y'	$\frac{1}{2}$	1	1	$K_{AT}\ K_{CG}$
	Y' G T X' Y'	1	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}$
	G T X' Y' G	1	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}\ K_{CG}$
	X' Y' G T X'	1	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}\ K_{CG}$
	Y' G T X' Y'	1	1	1	$K_{AT}\ K_{YY'}\ K_{CG}$
	X' Y' G T X' Y'	$\frac{1}{2}$	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}$
	Y' G T X' Y' G	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	G T X' Y' G T	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	T X' Y' G T X'	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	X' Y' G T X' Y'	1	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}\ K_{CG}$
	Y' G T X' Y' G	$\frac{1}{2}$	1	1	$K_{AT}\ K_{YY'}\ K_{CG}$
	X' Y' G T X' Y' G	1	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}\ K_{CG}$
	Y' G T X' Y' G T	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	G T X' Y' G T X'	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	T X' Y' G T X' Y'	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	X' Y' G T X' Y' G	1	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}\ K_{CG}$

(continued)

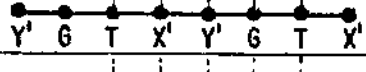
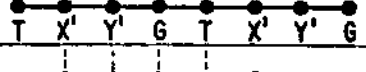
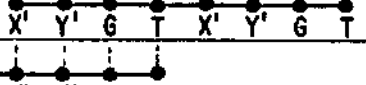
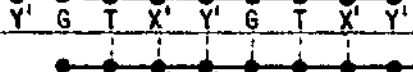
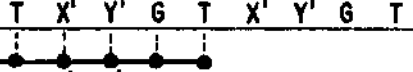
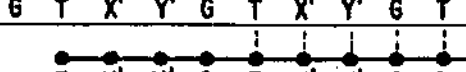
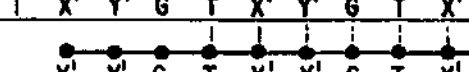
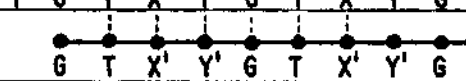
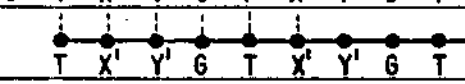
	$\frac{1}{2}$	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	$\frac{1}{2}$	1	1	$K_{AT}\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	$\frac{1}{2}$	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	1	1	2	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$
	$\frac{1}{2}$	1	1	$K_{AT}^2\ K_{XX'}\ K_{YY'}\ K_{CG}$

TABLE 2

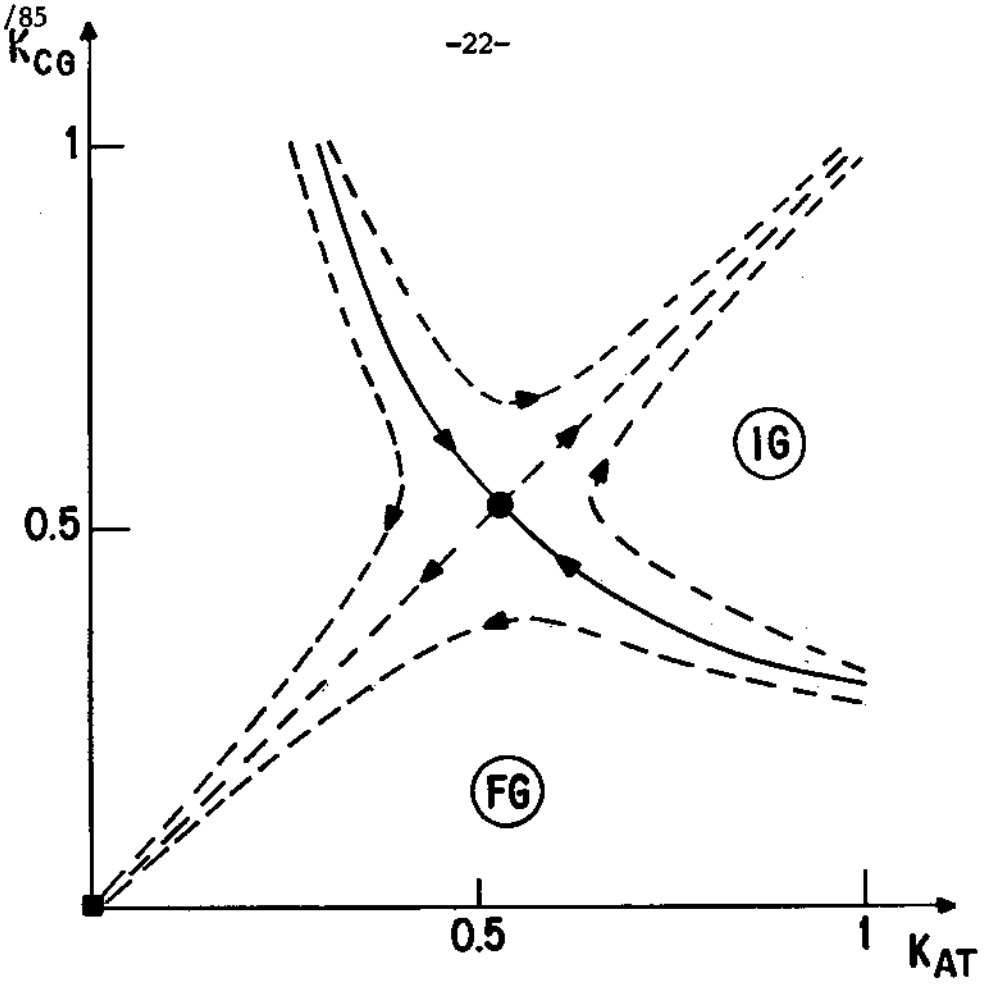

K_{CG}
1
0.5
IG
FG
0.5
1
K_{AT}

FIG.1

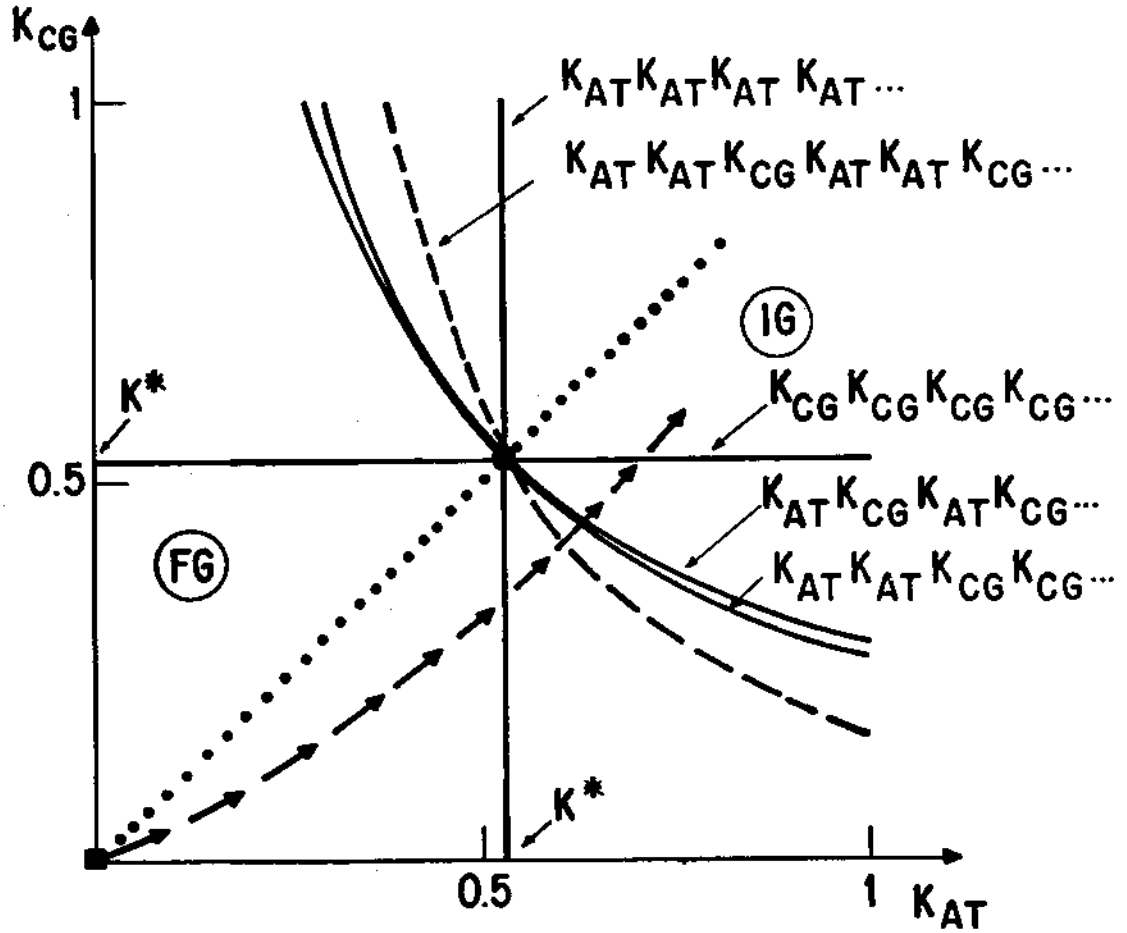

K_{CG}
1
$K_{AT}K_{AT}K_{AT}K_{AT}\cdots$
$K_{AT}K_{AT}K_{CG}K_{AT}K_{AT}K_{CG}\cdots$
IG
$K_{CG}K_{CG}K_{CG}K_{CG}\cdots$
K*
0.5
$K_{AT}K_{CG}K_{AT}K_{CG}\cdots$
FG
$K_{AT}K_{AT}K_{CG}K_{CG}\cdots$
K*
0.5
1
K_{AT}

FIG.2

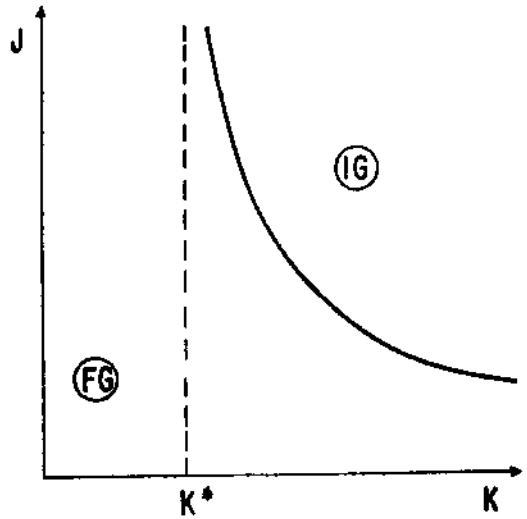


FIG.3

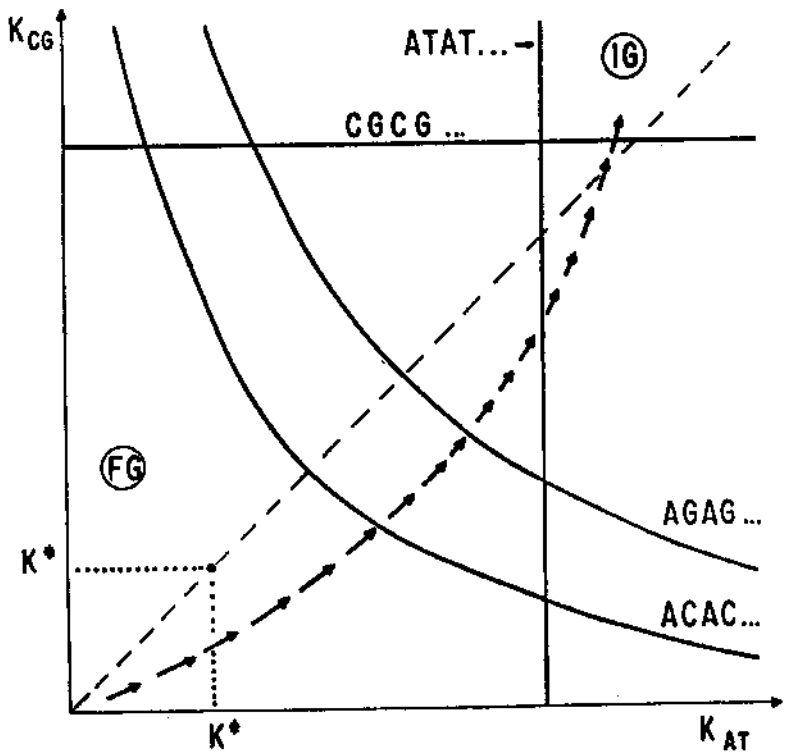


FIG. 4

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