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## Constraints on the Origin of Coherence in Far-from-equilibrium Chemical Systems<sup>1</sup>

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Consideration of how dynamic coherences arise from less organized antecedents ('concrecence') is one of the central concerns of Whitehead's thought. Other chapters in this volume consider how his concepts may apply in various branches of physics, including quantum mechanics. But many of the coherences that are of human interest are macroscopic. Earlier interpreters generally regarded Whitehead's 'actual entities' as exclusively sub-microscopic. There is now a growing consensus that Whitehead's conceptual scheme ought also to be applicable to larger items. The origin of dynamic coherence in far-from-equilibrium chemical systems has been widely studied experimentally and theoretically, and is now rather well understood. The chemical level illustrates many features that are also characteristic of more complex aggregations (biological and social, for instance) but chemical systems are more amenable to experimental and theoretical investigation. This chapter considers some aspects of the origin of chemical coherence.

Logicians and analytical philosophers tend to be more comfortable with mathematics and mathematical physics than with chemistry. This seems apparent in current treatment of wholes and parts. The term 'mereology' (coined by Leśniewski) 'is used generally for any formal theory of part-whole and associated concepts'.<sup>2</sup> A 'mereological whole' is an individual that is comprised of parts that are themselves individuals. Any two (or more) individuals can comprise a mereological whole  $\frac{3}{4}$  the star Sirius and your left shoe, for instance. Concerning such aggregates, D. M Armstrong's 'doctrine of the ontological free lunch'<sup>3</sup> seems valid:

... whatever supervenes, or.. is entailed or necessitated, ... is not something ontologically additional to the supervenient, or necessitating, entities. What supervenes is no addition of being.... Mereological wholes are not ontologically additional to all their parts, nor are the parts ontologically additional to the whole that they compose. This has the consequence that mereological wholes are identical with all their parts taken together. Symmetrical supervenience yields identity.

This kind of mereology may well be quite appropriate to deal with some questions of mathematics,<sup>4</sup> but it seems quite inadequate for dealing with situations that chemists encounter. Chemical combination is not well understood in terms of mere addition of properties of components. Chemical combination generates properties and relations that are not simply related to the properties and relations of the components. Entities that are important in other branches of science, and in other parts of culture, have all the complexity of chemical combination  $\frac{3}{4}$  and more. It does not seem that the standard approach to mereological questions has wide applicability, much less *metaphysical* generality.

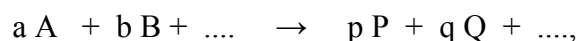
It may well be that every system of reckoning must have a unit  $\frac{3}{4}$  some thing, or class of things, that may *properly be taken* as simple (not composite), at least for the sake of the reckoning. For instance, Armstrong observes:

.... the existence of *atoms*, whether particulars or universals, is held to be a question for science rather than metaphysics, and one that we should at present remain agnostic about. The world divides, as Wittgenstein wrote, but it may divide *ad infinitum*, and there be no terminus, even at infinity.<sup>5</sup>

If, strictly speaking, there are no 'simples' (atoms, in the classical sense), but yet we need to deal with entities that are 'relatively atomic',<sup>6</sup> then we require what Armstrong calls 'unit-determining properties'  $\frac{3}{4}$  qualities that give rise to 'unithood'<sup>7</sup>.

Before getting into what sort of things unit-determining properties might be, we have to decide which level (spatial, temporal, or other) is appropriate to use in this discussion. Whitehead is usually interpreted<sup>8</sup> as holding that there is a fundamental level of description, perhaps at some sub-microscopic level of spatial size. But if we hold the alternative position  $\frac{3}{4}$  that there are no classical 'atoms', merely entities that are 'relatively atomic,' then it is not at all obvious that any spatial, temporal, (or other) level has priority  $\frac{3}{4}$  it seems that there may well be no 'fundamental' level. In this alternative view, we ought to be able to consider working at any spatial or temporal level whatsoever. We might even hope that we could identify *unit-determining* features that were operative at many levels  $\frac{3}{4}$  that might provide warrant for a consistent, adequate, and applicable metaphysics.

In previous centuries, chemists used to say that certain chemicals had high 'affinity' for certain materials (those with which they reacted) and low affinity for yet others (with which they did not react). Zinc was said to have high affinity for acids, copper much less affinity, and gold hardly any affinity for those acids. Tables of chemical affinities were the stock in trade of many pre-twentieth century chemists. In Brussels in the 1920s, Theophile de Donder (1872-1957) put chemical affinity on a firm quantitative footing.<sup>9</sup> Consider a reaction:



the *law of mass action* asserts that there exists a constant,  $K$ , such that:

$$K = [P]_e^p [Q]_e^q \dots / [A]_e^a [B]_e^b \dots$$

Where brackets denote concentrations (or activities), exponents are stoichiometric coefficients (from the balanced chemical reaction equation) and, importantly, the subscript  $e$  indicates that the equation is valid only when the reaction has reached *chemical equilibrium*. The *chemical affinity of a reaction mixture* is defined:

$$A = RT \ln K / Q.$$

where  $Q$  is a quotient of the same form as the equilibrium constant ( $K$ ), but involving *actual concentrations* (activities) of the reaction mixture, whatever they may be, **not** equilibrium concentrations (activities). Chemical affinity ( $A$ , a 'state function') is the measure of the *driving force* of a chemical reaction. A reaction system with high affinity is said to be 'far from equilibrium'. If  $A$  is small, the system is considered to be 'close to equilibrium.' At equilibrium, the affinity is zero.

The concept of affinity was not used in Lewis and Randall,<sup>10</sup> the thermodynamics text regarded as definitive in the United States for the middle third of the twentieth century. Many chemists are uncomfortable with the concept. Probably logicians would find it odd as well. Affinity is a property  $\frac{3}{4}$  but a property of what? Not of an individual molecule, nor of a collection of like molecules, but of a *macroscopic reaction mixture*  $\frac{3}{4}$  a *collection* of molecules of diverse sorts. This is also true of such relatively un-problematic properties as pressure and volume. But, in contrast to pressure and volume, affinity *involves a relation*. Affinity is defined with *respect to a specific reaction*. One and the same reaction mixture might have various values of affinity with regard to the several possible reactions that could occur between the chemicals that constitute the reaction mixture.

It is a matter of discussion among logicians as to whether several distinct mereological wholes can have exactly the same constituents.<sup>11</sup> There is no doubt among chemists that one and the same chemical reaction mixture can produce a wide variety of different results, depending on the presence of diverse catalysts, for instance. A pot full of assorted amino acids can readily generate a myriad of different proteins (or a mixture of them), depending on what RNAs are present. The single pot would have an array of different values of chemical affinity, one value for each conceivable product.<sup>12</sup>

In a sense, chemical affinity is like 'distance'. What is the distance from Claremont?  $\frac{3}{4}$  Distance to where? Claremont is about forty miles from Los Angeles, but several thousand miles from Cambridge, England. The quantity 'distance from Claremont' is meaningful only if a specific destination is designated. Chemical affinity is a proper 'state function' (like volume or pressure) of a chemical sample, but *only if a particular reaction is specified*. It would not be surprising if some philosophers refused to accept such a notion.<sup>13</sup> Chemical affinity is an odd sort of property  $\frac{3}{4}$  but one that is rigorously defined and useful in both theory and practice

Ordinarily, chemical reaction mixtures change in such a way as to smoothly approach an equilibrium state  $\frac{3}{4}$  than is, concentrations change (either increase or decrease) while chemical affinity (with respect to the reactions that are occurring) steadily decreases. But certain far-from-equilibrium reaction mixtures behave quite differently. Rather than monotonously changing, concentrations of particular components of these mixtures *oscillate* regularly and repeatedly around an *unstable* steady state.<sup>14</sup> Often, these mixtures spontaneously become structured in space, as well as in the time dimension. If the reaction mixture is a 'closed' system (one that does not exchange energy or chemicals with its surroundings) then oscillations of

components gradually decrease, and spatial structures fade, as the system get closer to the condition of equilibrium. Such a gradual decay of oscillation can be avoided by arranging matters so that reactants are continuously fed in to the system, and reaction products are removed as they are formed. Such 'open systems' are always far from equilibrium <sup>3</sup>/<sub>4</sub> spatial structures and/or oscillations can be maintained indefinitely while these systems convert high-energy-content materials into low-energy-content materials. These spontaneously organized chemical systems are called 'dissipative structures'.<sup>15</sup> Such oscillating chemical systems resemble structures of more familiar types, in as much as they have the ability to *withstand disturbance*. A dissipative structure, if caused to deviate from its regular oscillatory pattern, will tend to *return* to the same pattern.

When Nature involves what seems to be a constant concentration of some chemical, close examination often shows that the concentration is maintained near an average value by some sort of oscillatory network of processes. Many technological devices work in a similar way. A home thermostat does not maintain a pre-set temperature steadily, but rather achieves the purpose for which it was designed by small oscillations of temperature above and below the target value. We now recognize that many — perhaps all — of the entities formerly thought to be substantial and perduring are, in fact, resultants of *networks of processes*. The question we need to consider is: what sorts of relationships between components must exist in order that a collection of processes would *behave as one unit*? Ivor Leclerc<sup>16</sup> discussed this question, but did not come to a specific conclusion of general relevance. We now consider chemical dissipative structures (oscillating reactions) as examples of how 'unit-determination' arises. Similar unit-determining properties (better, unit-determining closures of *relations*) can be seen to occur elsewhere <sup>3</sup>/<sub>4</sub> from the quantum level to the cosmological level, and at many intermediate ones.

In order for a chemical dissipative structure (oscillating reaction) to exist, the system must be *far from equilibrium* (have high affinity) and there must be no stable equilibrium state accessible to the system. When parameters change, it sometimes happens that a previously stable stationary state (or equilibrium) becomes unstable. For instance, at ordinary air temperatures, heat that is generated by fermentation inside a haystack diffuses out of the pile of dried grass, and a stable, quasi-equilibrium, steady state is eventually reached. However, if the ambient air temperature comes to be much higher than usual, the rate of exit of chemically generated heat would be reduced, and the haystack would heat up. Since fermentation occurs more rapidly at higher temperatures than at lower temperatures, retention of heat in the haystack would cause still further heat to be generated, and generated faster and faster. The non-equilibrium steady state would become unstable. The pile of hay would quickly become quite hot, and might burst into flame.<sup>17</sup>

The circumstance that increase of temperature causes increase of the release of heat, and yet further rise in temperature, is a particular example of *autocatalysis*. A chemical reaction produces a product (heat in this case) that *increases* the rate of production of that product. Autocatalytic processes abound in nature. Many nuclear reactions produce neutrons that can initiate yet further nuclear reactions. Genes are biological catalysts that make copies of themselves, through quite complicated mechanisms. Human groups socialize their members to engage in behavior that tend to increase the number of members of the group. Resources

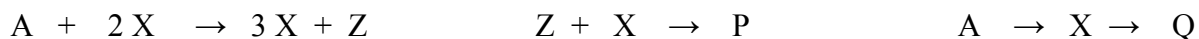
invested in bull markets produce yet further resources, which enable additional investment. Each such process can de-stabilize prior steady states  $\frac{3}{4}$  equilibria or quasi-equilibria.

Every dissipative structure must involve at least one autocatalytic reaction. The simplest kinds of autocatalysis (to which others can be reduced) can be represented:

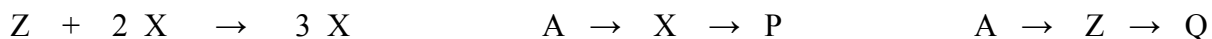


Any autocatalytic process can be the basis of a 'clock reaction'  $\frac{3}{4}$  a reaction that appears quiescent for a long period of time, and then bursts into activity  $\frac{3}{4}$  a solution changes color rapidly after a time without apparent change, or, a haystack suddenly bursts into flame. A virus (an autocatalytic agent) may lurk undetected for years, then replicate explosively. The length of the delay (the 'induction period') depends on the initial concentration of autocatalyst. The rate of production of autocatalyst will initially be low if the starting concentration of the autocatalyst is small. But no matter how low that rate is (so long as it is not exactly zero), the concentration of the autocatalyst continually increases, and the autocatalytic reaction steadily gets faster. This gives rise to the long induction period followed by a sudden rapid rise of the autocatalyst concentration. All dissipative structures are based on autocatalysis of one kind or another.

But something else is required to turn an autocatalytic process into a dissipative structure: there has to be some way to 'reset the clock'  $\frac{3}{4}$  to return the autocatalyst concentration to a low value. This requires the existence of at least one additional chemical, the 'exit species'. The autocatalyst is often called X, and the exit species designated Z. Here are representations of two types of chemical oscillators; both based on cubic autocatalysis, but each using a different strategy to reset the clock. (Both of these are *open* systems, with chemicals entering and leaving the system.)



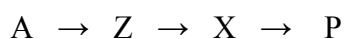
In this first set of reactions, Z is a byproduct of the autocatalytic process: Z also has the ability to remove the autocatalyst. As the autocatalytic reaction proceeds, the exit species Z builds up through the reaction shown on the left. In reaction-set shown in the center, the Z combines with X to remove the autocatalyst. In the rightmost set of reactions, the autocatalyst is fed into the system, and also departs from the system (e.g., decomposes). A is the (constant) supply of reagents; P and Q are products that play no other part in the reactions.



In the second case (illustrated just above), Z is not a product, but is a reactant in the autocatalytic reaction. Z is used up as the autocatalytic reaction (shown on the left) proceeds. Once Z is depleted, autocatalysis stops. Both the autocatalyst X and the exit species Z are fed into the system and both leave the system by decomposition (center and right reactions).

In these two sample cases, autocatalysis is controlled by an exit species. In the first case the autocatalytic reaction is choked off by buildup of a toxic byproduct. In the second case, the autocatalysis is starved by shortage of a necessary reactant. Any chemical oscillation that is based on cubic autocatalysis can be shown to correspond to one or the other of these two reaction types.<sup>18</sup> Oscillations based on quadratic autocatalysis must involve three (or more) variables, usually including a 'feedback species,' generally called Y. Like reactions based on cubic autocatalysis, oscillations based on quadratic autocatalysis are reducible to only a few basic patterns.

For simplicity, we confine our attention to a version of the second cubic autocatalytic system shown above, with spontaneous linear (not autocatalytic) formation of X from Z and spontaneous decompositions of X to produce an inactive product.



There are only two variables, X and Z. Important parameters in this system include proportionality (rate) constants for each of the three chemical reactions shown, and for the input of Z. We may examine the behavior of the system as two of these parameters are varied <sup>3</sup>/<sub>4</sub> while values of other parameters are held fixed. For parameter values that correspond to one region of this two-dimensional parameter space, there is a stable steady state with high autocatalyst concentration (the *thermodynamic* branch). Let's say this state is *blue*. For parameter values corresponding to another region of the parameter-space diagram, there is a stable (self-restoring after disturbance) steady state with low values of the autocatalyst concentration (the *flow* branch) <sup>3</sup>/<sub>4</sub> let's say that this non-equilibrium steady state is *red*. For parameter values corresponding to a third region of the two-dimensional parameter space, *both* of these non-equilibrium steady states are stable. In this region of parameter space, the system is said to be *bistable*. For parameters corresponding to points in this region of parameter space, the color of the system may be either blue or red, depending on its past history. If this region of parameter space is entered from the blue side, blue will prevail: if it is entered from the red side, red will obtain. (There is a third non-equilibrium steady state in the bistable system, but it is unstable <sup>3</sup>/<sub>4</sub> any fluctuation will cause the system to move to one or the other of the two stable non-equilibrium steady states, the red one or the blue).

A fourth region of the two-dimensional parameter space often exists. If the two variable parameters happen to correspond to a point in this fourth region, sustained oscillations of concentrations occur. In this region of parameter space, both blue and red non-equilibrium steady states are unstable. The system moves repeatedly from states approximating the blue non-equilibrium steady state to something approximating the red non-equilibrium steady state. Concentrations of all chemicals continuously oscillate in time. In such cases, a plot showing the state of the system at various times, in a two dimensional concentration space (one dimension for each of two concentrations), yields a *closed* trajectory that goes around a point corresponding to the unstable (third) non-equilibrium steady state. In contrast, parameter values that correspond to points in the other three regions of the two-dimensional parameter space yield a trajectory (in concentration space) that approaches either the red or the blue non-equilibrium steady state.

A proper dissipative structure corresponds to such a 'limit cycle' in some appropriate space. That is, there is a single, closed, curve that describes the sequence of states that the system follows over time. This same single, unique, trajectory is eventually attained no matter what the starting conditions might be. In the case that we are discussing, the appropriate space is the  $[X] - [Z]$  plane. If we wish, we could divide the curve in two-dimensional concentration space that describes this limit-cycle trajectory into four segments, using as dividers the points at which each of the two variables changes direction of motion (reaches a maximum or minimum in concentration).

- In one segment, X increases rapidly, while Z decreases. This segment is dominated by the autocatalytic reaction.
- In a second segment, the autocatalyst X begins to decrease while Z continues to decline. In this segment, the autocatalytic reaction competes with decomposition of the autocatalyst.
- In the third segment, X decreases to a low concentration and Z increases a great deal. The autocatalytic reaction is now largely shut off, but decomposition of the autocatalyst is proceeding, and the feed of Z is significant.
- In the fourth segment, both Z and X increase somewhat. The concentration of X has fallen so low that decomposition of the autocatalyst is no longer important. As the feed increases the Z concentration, non-autocatalytic production of X from Z becomes significant.
- When the X concentration passes a certain critical value, the autocatalytic reaction reaches a high rate and loss of Z exceeds the rate at which Z is being fed in, so a rapid increase of X and concomitant decrease of Z begins. This is the phase noticed first above. The cycle is complete. As James Joyce would have it, '*Finagin.*'

As with cycles of other sorts, there is no point at which this cycle may properly be said to begin. Also, division of this cycle into just four phases is quite arbitrary. *Closure* of the network of processes to yield the set of states of affairs described by the limit cycle is a real and significant feature of this collection of processes, but dividing the cycle exemplifies 'the fallacy of misplaced concreteness.'

In order for the oscillation to have long-term stability, its trajectory must pass through to the same set of conditions during each oscillation, a stringent condition.<sup>19</sup> Compared to the total range of parameter space, the region in which this condition is met may be quite small. But still, this region is often large enough that considerable tolerance (more or less, depending on the other parameters) exists for variation in parameters. Frequently, setting up such experiments using aqueous solutions requires only 'bartender's precision'. Variation of parameters, within the oscillatory region, gives rise to changes in the frequency and the amplitude of the oscillations.

\* \* \* \*

Here, then, are the constraints on origin of a dissipative structure in a chemical dynamic system:

- Affinity must be high. (The system must be far from equilibrium.)
- There must be an autocatalytic process.
- A process that reduces the concentration of the autocatalyst must exist.
- The relevant parameters (rate constants, etc.) must lie in a range corresponding to a limit cycle trajectory.<sup>20</sup> That is, there must be a *closure* of the network of reaction such that a state sufficiently close to the prior condition is achieved at the corresponding part of each oscillation.

If all these constraints continue to be met, the dissipative structure continues to exist, and may serve as a center of agency. Interactions of the system with the rest of the world are quite different in the presence of the dissipative structure than they would be in the absence of that self-organized coherence. In this sense, the closure<sup>21</sup> of a network of relationships that gives rise to a dissipative structure is the 'unit-determining' feature required to secure 'unithood', in Armstrong's terminology. The effects of the structure as whole are the resultant of the effects of the components, but the concentrations of the components that exist at any instant are the effects of the *closure* of the limit cycle. This is a definite example of a kind of 'downward causation,'<sup>3/4</sup> an influence on the components arising from the thing those components constitute.

Similar situations, where *closure* of a network of processes has important effects, occur in many fields. The postulate of de Broglie, central to the development of quantum mechanics, is strikingly similar in some respects to the positions being advocated here. In systems involving electrons and atomic nuclei, there are stringent conditions on the *closure* of sets of relationships. Once that closure is attained, a system maintains its coherence indefinitely, and can function as a unit in yet higher-level coherences. Networks of interaction also abound in biochemistry, molecular biology, organismic biology, and ecology. In favorable cases, systems in all these areas display 'unit making' closure of relationships quite similar to those displayed by dissipative structures.

The 'toy model' used to illustrate the discussion of chemical coherence given above is basically the same as the model generally used to understand the generation of calcium oscillations in biological cells.<sup>22</sup> These oscillations are known to function in the control of complex biological organs, such as the human brain. Remarkably, it is the frequency, not the amplitude of the calcium oscillations that is decoded as the controlling signal.<sup>23</sup> The signal is generated by the unit-determining *closure* of the regulatory network that defines a dissipative structure composed of processes in the brain, and the information that is transmitted results from subtle alteration in the parameters that control that oscillation. There seems to be no way to deal with effects of this sort using standard mereology. This difficulty is of central significance in questions of philosophy of mind and of human action<sup>24</sup>, but its outline is clear even in the simpler chemical cases discussed in this paper.

If we understand the word 'mereology' in its general sense, as a theory of parts and wholes, (rather than as the specific system of Leśniewski), it seems clear that a new, more discriminating, mereology is needed.



An adequate logic of wholes and parts must be capable of dealing with coherences of the types considered here, in which closure of a network of relationships gives rise to significant effects (both external and internal) that would not exist, absent that closure.

### References and Notes

- 1) An earlier version of this paper was presented at the Second Summer Symposium on the Philosophy of Chemistry of the International Society for the Philosophy of Chemistry, Sidney Sussex College, Cambridge University, England, August 3-7, 1998 and (*in absentia*) at the Silver Anniversary Whitehead Conference, The Center for Process Studies, Claremont, CA. August 4-9, 1998.
- 2) Peter Simons. *Parts, A Study in Ontology*. Oxford: Clarendon Press, 1987, p. 5.
- 3) D. M Armstrong, *A World of States of Affairs*, Cambridge: Cambridge University Press, 1997, p.12.
- 4) David K. Lewis, *Parts of Classes*, Oxford: Blackwell's, 1990.
- 5) D. M. Armstrong, *op. cit.*, p. 263.
- 6) D. M. Armstrong, *op. cit.*.
- 7) D. M. Armstrong, *op. cit.*, pp. 189-191.
- 8) For an alternative interpretation, see J. E. Earley, Sr. "Towards a Reapprehension of Causal Efficacy," *Process Studies*, 24, 34-38, **1995**, and J. E. Earley, Sr. "Naturalism, Theism, and the Origin of Life," *Process Studies*, 27, 267-279, **1998**.
- 9) Dilip Kondepudi and Ilya Prigogine, *Modern Thermodynamics: From Heat Engines to Dissipative Structures.*, N.Y., Wiley, 1998.
- 10) Gilbert Lewis and Merle Randall, *Thermodynamics*, N.Y. McGraw-Hill, (2nd ed.) 1961.
- 11) D. M. Armstrong, *op. cit.*,
- 12) In such a case the reaction products would be kinetically determined not thermodynamically determined and the catalysts, not the affinities, would account for which product(s) were, in fact, produced.
- 13) It seems worth noting that Peter Van Inwagen asserts that distances do not exist! Seminar at Georgetown University, April 17, 1998.

- 14) Stephen Scott, *Oscillations, Waves, and Chaos in Chemical Kinetics*, Oxford: Oxford University Press, 1992.
- 15) Gregoire Nicolis and Ilya Prigogine, *Self-Organization in Far-from Equilibrium Systems*, New York, Wiley, 1977.
- 16) Ivor Leclerc, *The Nature of Physical Existence* (Washington: University Press of America, 1986) and *The Philosophy of Nature* (Washington: Catholic University of America Press, 1986).
- 17) Scott, *op. cit.*
- 18) Eiswirth, M.; Freund, A.; Ross, J. *Adv. Chem. Phys.*, **1991**, 190, 127; *J. Phys. Chem.* **1991**, 95, 1294.
- 19) In certain systems (that are more complex than the simple examples discussed here), this requirement is somewhat relaxed, and the oscillation becomes irregular, even chaotic. In these cases, the closed trajectory in two-dimensional space lies on an object of fractal dimension (a strange attractor) rather than on single curve.
- 20) Or to a strange attractor. (See preceding note.)
- 21) Joseph E. Earley, Sr., "Varieties of *Chemical Closure*" in *Closure; Emergent Organizations and Their Dynamics*, J. L. R. Chandler and G. Van de Vijver, editors, *Annals of the New York Academy of Sciences*, 901, (2000), pp. 122-131.
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- 23) James W. Putney, Jr. "Calcium Signaling: Up, Down, Up, Down...What's the Point?" *Science*, 279, 191-192 (9 January), **1998**. "Sensitivity of CaM Kinase II to the Frequency of Ca<sup>2+</sup> Oscillations," Paul De Koninck and Howard Schulman, *Science* 279, 227-230 (9 January) **1998**.
- 24) Alicia Juarrero. *Dynamics in Action; Intentional Behavior as a Complex System*. Cambridge, MA., MIT Press, 1999.