Magnetism, entropy, and the first nano-machines

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Abstract

The efficiency of bio-molecular motors stems from reversible interactions $\sim k_B T$; weak bonds stabilizing intermediate states (enabling direct conversion of chemical into mechanical energy). For their (unknown) origins, we suggest that a magnetically structured phase (MSP) formed via accretion of super-paramagnetic particles (S-PPs) during serpentinization (including magnetite formation) of igneous rocks comprising the Hadean Ocean floor, had hosted motor-like diffusion of ligand-bound S-PPs through its template-layers. Ramifications range from optical activity to quantum coherence. A gentle flux gradient offers both detailed-balance breaking non-equilibrium and asymmetry to a magnetic dipole, undergoing infinitesimal spin-alignment changes. Periodic perturbation of this background by local H-fields of template-partners can lead to periodic high and low-template affinity states, due to the dipole’s magnetic degree of freedom. An accompanying magnetocaloric effect allows interchange between system-entropy and bath temperature. We speculate on a magnetic reproducer in a setting close to the submarine hydrothermal mound-scenario of Russell and coworkers that could evolve bio-ratchets.

Key words: magnetic-reproduction; Brownian noise; H-field-controlled assembly; symmetry-breaking; magnetocaloric effect

Abbreviations: MSP - magnetically structured phase; S-PP - super-paramagnetic particle; MCE - magnetocaloric effect; ATP - Adenosine tri-phosphate; $k_B$ Boltzmann-constant; T-temperature.

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1.1 Introduction: Bio-molecular dynamics

Increasingly it is becoming apparent that the dynamics in biology at the nanoscale, such as in molecular motors, is part of the generic phenomena governed by the (linear) fluctuation-dissipation theorem [1,2]. The diffusive movement of the system traversing between two energy states, aided by random Brownian forces, gets rectified by coupling to a non-equilibrium force, e.g. ATP hydrolysis [3]. And such systems with low Reynold number, access to internal degrees of freedom, plus asymmetric interactions, can thrive on the best of both worlds—an equilibrium local state that can harness thermal fluctuations in a diffusive step as well as the directionality governed by a non-equilibrium reaction—leading to net movement in an asymmetric yet periodic energy landscape [4]. This brings to light two essential requirements for executing such dynamics: Firstly, weak bonds (hydrophobic, hydrophilic, van der Waals, H-bonds, etc) help to pin-up the motor temporarily in different equilibrium states. Secondly, the continuous nature of the energy landscape connecting different states shows a system that can absorb energy in an essentially continuous and reversible manner (adiabatic) by utilizing energy from random Brownian hits ($\sim k_B T$) which is of the same scale as rotational energy states in a molecule. This feature similarly enables a physical nanosystem to undergo periodic cycles a la Berry’s phase, between the two states [5].

1.2 Origins of weak bonds and reversible interactions

So the question arises: How could such reversible interactions and intermediate states, underlying the efficiency of these machines, have been physically realized by matter present at the dawn of Life? Note that these are almost impossible to achieve using chemical bonds that are the very basis of proposals for template-based processes using mineral crystal surfaces. Traditionally, origin-of-life theories concentrate either on its replication or metabolism aspects. On the other hand, the origin of the ubiquitous molecular motors, in and across all living systems, is seen as a later addition. According to Vale [6] two inventions were important in the development of motors: one-dimensional electrostatic sliding along polymers, and a conformational-change mechanism in the active site of a nucleotidase enzyme. How this happened however has been left unaddressed and is largely unknown. Here we suggest that Life’s origin was strongly linked to the emergence of nano-systems utilizing the thermal energy of the surroundings, just as in today’s biological nano-machines. A physical phase captured before the onset of the mineral crystallization could have also hosted template processes proposed by Cairns-Smith [7]. This is line with Dyson’s [8] proposal for ‘physical reproduction’ (not ‘chemical replication’) as having initiated Life, in a metabolically enriched environment. In
fact, physical forces seem to be well equipped to deal with some of the logical difficulties cropping up with ‘chemistry-only’ origin-of-life approaches. Indeed, the power of magnetic($H$) fields for external control on super-paramagnetic matter, seems to have gone unnoticed hitherto, despite their omnipresence in space and time. To begin with, H-field controlled super-paramagnetic particles (S-PPs) could have provided a ready basis at the origins of life for generating the various energy-transduction systems coupling the formation/use of energy-rich molecules with temperature or charge (redox, pH) transferring gradients [9]. Only later could the non-equilibrium and symmetry-breaking aspects of the field have been replaced by energy-rich molecules and asymmetric interactions. Besides, the long appreciated spin-system mimicking features across myriad phenomena displayed by biological soft-matter (landscape processes, orientational order in fluid state, etc.), would be easier to understand logically if we had begun with a magnetic Ancestor in the first place. After all, many bacteria play host to magnetosomes of of greigite ($Fe_3S_4$) [10, 11]. (And, biomineralization using magnetite ($Fe_3O_4$) – a close relative – is considered the most ancient matrix-mediated system that could even have served as an ancestral template for exaptation [12]). Sure enough, magnetic alignment of a particle to its partner in a template (an array of aligned particles) could have provided the beginnings for embodying weak bonds, typical in biology. Here, local reversibility at each infinitesimal step is achieved via effectively continuous spin alignment changes, where maximum/minimum interactions lead to association/dissociation, respectively. These could be driven by simple thermal fluctuations, just as in today’s bio-molecular motors.

1.3 Need for a dynamical lattice: Power of magnetism

Now, field-induced structure formation as seen in non-ideal magnetic fluids (Sect.2.1), could bring about orientational long range order in the aqueous dispersed particles. Such a dynamic array as the ‘fountainhead of Life’ represents a major departure from conventional template approaches based on either crystal-surfaces, or where evolved chemicals had sufficient complexity for spatially asymmetric interactions for self-assembly (e.g. liquid crystals). However, no convincing explanation seems forthcoming as to how a reproducing life-like assembly from complex molecules had evolved from an immense medley of compounds. Consider instead, the role of magnetic dipolar interactions in giving rise to a dynamic assembly, without having to wait for the evolution of complex molecules, whose feasibility is crucial for the emergence in the Hadean of molecular motors – key players across kingdoms in biology. While a similar passage would be impossible with rigid lattices of mineral crystals, the simpler possibility of physically reproducing [8] magnetic particles exists for extending the horizons of traditional approaches by combining
chemistry with myriad physical effects. In this scenario, chemistry continues to play a role in the ligand shell reactions of colloidal S-PPs as in the simulations of Milner-White and Russell [13], but magnetic accretion provides the confining force for herding them together. Cutting through dipole-dipole interactions holding together layers of a magnetically ordered phase would require energy, orders of magnitude less than those cementing crystal layers. At the same time an orientation-based magnetically herded 'array' would retain the information transmission feature of ordered crystal surfaces [7]. Again, not only crystal surfaces, but individual layers of a dynamic array could be imagined as the very templates a la Cairns-Smith that hosted transfer reactions in the origins of Life. The tremendous increase in surface area vis-a-vis mineral crystal surfaces would also similarly stretch the prospects of catalytic activity, crucial for metabolism, and in line with today's spotlight on the nanoscale. Indeed, packing in physically accreted finite systems comes ready with built-in aperiodicity, as an effective substitute for the superimposed aperiodic distribution of metal ions on infinite periodic crystal lattices [14]. This very feature underlies the efficient packaging of information in nucleic acids, where the lack of correlations across sequences (random nature) satisfies Claude Shannon's maximum entropy requirement [cf. 15].

1.4 Outline of paper

With this background, we shall first briefly review magnetically structured phases (MSPs) of dispersed magnetic colloids (Sect.2.1), for trying to identify the ingredients required for extending this scenario to a possible Early Earth (Hadean) Ocean Floor setting (Sect.2.2). Next in Sects.3.1-3.5, we present a detailed correspondence (mapping) of the features of bio-molecular motors with those of super-paramagnetic particles diffusing through a magnetically structured phase. Finally, in Sects 4.1-4.2, we ask how bio-ratchets could have originated and suggest a greigite-based scenario; Sect.4.3 concludes with a discussion on the potential of magnetism.

2.1 Field-induced aggregates in non-ideal ferrofluids

The above brings us to the well known area of ferrofluids: colloidal single-domain magnetic nanoparticles (∼10nm) in non-magnetic liquids that can be controlled by moderate H-fields (∼tens of milliTesla) [16]. Coatings stabilize these dilute dispersions displaying ideal single-phase behaviour due to prohibited (chemical) inter-particle contacts. In contrast, the present application concerns the interactions within the magnetic subsystem, while the carrier re-
mains in the liquid state. The deviation from ideal magnetization behaviour shows up on increasing particle concentrations that can be understood in terms of H-field-induced inter-particle interactions leading to internal structure formation and manifesting in dense phases—a milder phase transition than to the solid-crystalline one. The structure of hydrated, heterogenous aggregates (e.g. chain-like, drop-like, worm-like micelles) would depend on factors like the strength of the applied field, the nature of the ferrofluid (molecular shape, susceptibility, etc.) [16, 17, 18]. An increase of chain size beyond a critical length, compactification due to interparticle magnetic interactions, formation of globules as nuclei for new dense phase, are all seen in the scheme of phase transitions leading to formation of bulk drop-like aggregates. As to the role of polydispersity, Wang and Holm [19] found that the fraction of large particles, with larger relative dipole moments in proportion to their volume, would overcome thermal forces more easily and respond to weaker fields and therefore dictate magnetization properties (e.g. initial susceptibility), even in case of dilute fluids. Furthermore, the solvent could also affect the aggregation, for Taketomi et al [20] observed field-induced macrocluster formation in water and paraffin-based ferrofluids but not in an alkyl-napthalene based one (even at 0.2 Tesla). In contrast, macroclusters formed in the water-based fluid at very low fields and remained even after removing the field. Li et al [21] have pointed out the dissipative nature of the field-induced aggregates that break up in response to thermal effects upon removal of field. They propose a gas-like compression model - a phase transition in which a particle concentrated phase separates from a dilute one, by following the orientation of the particle moments in the direction of the field. And, the higher the field intensity the more compact the aggregates; so that the aggregate space containing particles would decrease, just as in a compressed gas. In this model, the total magnetic energy of ferrofluids obtained from an applied field: \( W_T = W_M + W_S \); where \( W_M = \mu_0 MHV \) and \( W_S = -T \Delta S \) are the magnetized and the structurized energies, respectively, \( V \) is the volume of the ferrofluid sample and \( \Delta S \) is the entropic change due to the microstructure transition of the ferrofluid. An assumed equivalence of \( W_T \) (zero interparticle interactions), with the Langevin magnetized energy \( W_L = \mu_0 MHV \) necessitates to a correction in the magnetization, in terms of the entropy change. Evidently, these systems are well equipped to analyze the interplay between competing factors - dipolar interactions, thermal motion, screening effects, etc. leading to the emergence of MSPs [22]. Their colloidal state and magnetic entropy property can provide a ready basis for mapping with complex biological soft-matter. We now take a closer look at bio-molecular motors, as these systems capture many of the complexities of biosystems.
2.2 Magnetic assembly on the Ocean floor

Analogous to non-ideal ferrofluids with interparticle interactions, three ingredients are required for a dynamic lattice: (1) the presence of a moderate local H-field on the Hadean Ocean Floor; (2) a newly forming super-paramagnetic suspension turning into tiny magnets due to no. (1); and (3) charge on particles. Serpentinized and magnetized igneous rocks [23, 24] could offer a local field (see Sect.4.2), with geochemistry providing the rest (Sect.4.2). Note that in contrast to homogeneous synthetic ferrofluids, a suspension forming in situ in presence of rocks, would have likely been polydisperse, with larger particles dictating magnetization behaviour (Sect.2.1).

Simulations of field-induced dissipative structures in non-ideal ferrofluids (see above) postulate the energy of a constituent particle to have contributions from the dipole-dipole interactions with neighbours; repulsive (charge/steric) effects; and its energy accruing from its orientation w.r.t. the H-field [25]. In the absence of a complete theory of dipolar fluids, and on the basis of available literature [25, 26], we envisage the emergence of an MSP upon gradual build-up of particles interacting via dipole-dipole interaction, just above the rocks. This, in turn would increase magneto-viscosity, impeding particles from rotating freely. Contributing factors for dipole ordering with energy-minimization include material properties, ligand-field effects, polydispersity, H-field strength, apart from ordering-variation [27] from parallel to anti-parallel. And, transient chains/arrays forming due to interacting dipoles forming layers of the MSP, could serve as magnetic templates for enabling bio-molecular motor-like transport. Figure 1 represents roughly parallel orientational correlations with resultant magnetization of MSP along the rock H-field. Finally, requirement no. (3) (charge on particle) is chosen in view of the key role of conflicting forces - here attractive magnetic, and repulsive electrostatic, – in bringing about a dynamic assembly [see 28]. Again, in the absence of ‘synthetic coatings’, the high ionic strength (screening effect) of the sea water [29] would have further encouraged dipole-dipole interactions.

It is interesting to compare a corresponding build-up of particles under ambient temperature, in the absence of a magnetic field. These would gradually form a colloidal network which would be expected to age by passing on to the crystalline phase. Thus it seems that a moderate local H-field pre-empts this process by capturing the build-up of magnetically tunable particles, thereby enabling Life-like dynamics in an otherwise inaccessible magnetically ordered fluid phase. Note that unlike a chemically bonded thermally formed gel, a magnetic gel would retain the potential of reverting back to its colloidal components just like colloid-gel transitions pointed out in living systems [30].


3.1 Directed motor movement: questions of origins

Thus far, we have considered the possibility of a magnetically ordered phase on the Hadean Ocean floor. But, how could directed diffusion as in bio-molecular motors migrating on templates like proteins and nucleic acids, have occurred for particles diffusing through these MSPs? In brief, motor proteins normally display unidirected transport, walking towards either the plus (e.g. kinesins) or the minus (e.g. dyneins) ends of the template (e.g. filaments, microtubules) that are polar polymers, arranged in a head-to-tail fashion. With cargo bound to their tail end, the motor diffuses back and forth till its capture by sites ahead in the progress direction; the greater likelihood of which follows asymmetry in binding affinity. The key change due to the ATP ligand is thus an altered energy landscape potential, leading to states with altered binding affinity. Although the free energy of this 'bound' conformation is larger than the minimum free energy of the ligand-free protein, thermal motion makes this conformation accessible [31]. Recent experiments by Taniguchi et al [32], led them to propose an entropic basis of rectification for the directed migration of kinesin; the backward step leads to a significantly lower entropic state than in the forward one.

Two outstanding clues are thus retrieved from motor dynamics: First is the capacity of the motor to combine with local anisotropy to bring about net movement which stems from its internal degrees of freedom allowing it to take on a different trajectory (different intermediate states on altered landscape potential) for the second half-cycle in each period. Second is its apparent capacity to undergo infinitesimal conformational changes by extracting energy $\sim k_B T$ from the thermal bath with help from close-to-equilibrium coupling to a non-equilibrium source for rectifying these fluctuations. Then in the Hadean, in the absence of complexity, we encounter the following question: How about particles bearing internal degrees of freedom having carried out similar entropy-reducing Maxwell Demon-like feats, as occurs for small systems [1], in the origins of Life? To address this question, we shall extrapolate this scenario to the directed diffusion of S-PPs through layers of an MSP, and check if the latter can supply both a topologically and energetically satisfying correspondence to the rectified diffusion of molecular motors on templates.

3.2 Directed diffusion of dipole through MSP

Recall that a magnetic particle moves in response to a field-gradient. A uniform field can orient a magnetic dipole but as the forces on its north and south poles would be balanced, there would be a zero net translational force acting on it. This situation would change in the presence of a field gradient.
And, magnetic field lines due to rocks would be cutting through the MSP. Their nature would be expected to be non-homogeneous, albeit changing in intensity in a very gradual manner. If the variations were so gentle as to appear small as compared to the radius of the diffusing S-PP, it would sense an effectively isotropic local environment [33,34]. The symmetry-breaking effect of the gradient would be felt by the particle at greater distances and bias the directional preference for diffusion. This diffusion of the nano-particle (negligible inertial effects) would further slow down to a net drift if it were to take place in a magneto-viscous medium formed as a result of magnetic dipolar forces between S-PPs. Next, two changes are expected upon ligand binding: lowering of both rotational freedom and coercivity [35] on ligand-bound end. Thus, while unconstrained rotation of ligand-free particles enables alignment and propagation of the ‘information’ in the magnetic dipole-ordered assembly (‘reproduction’), ligand-binding aids diffusive passage.

Further, diffusion is expected to be faster for particles with increased magnetization. Although this would also depend on the nature of ordering in particle clusters, our choice of criterion no. (3) ensures that only small size particles would diffuse through the MSP. A charge on the S-PPs would have not only aided in self-organization of the structured magnetic phase, but also its layers of similarly charged particles, would have repelled the entry of large clusters of similar charge-carrying particles. This effect was likely accentuated due to the low effective shielding (low concentration of sea-water) inside the layers of the dense MSP. And while mono- or di-mers could be expected to diffuse through, higher molecular weight members with increased surface charge would face resistance to passage, due to greater repulsive effects. (The oriented diffusion of S-PPs (see Figure 1) is imagined in the direction parallel to that of the field lines, so $\sin \theta = 0$ and therefore no force is exerted by the $H$-field on a charge moving parallel to it). In figure 1, the S-PPs (in blue) have both motional and spin degrees of freedom, in contrast to their MSP-counterparts embedded in a magnetically bonded network with orientational correlations (in black).

### 3.3 Interactive cycles

The periodically changing landscape potential of complex bio-molecular motors seems to match the periodic perturbations on the background rock $H$-field due to superimposed local $H$-fields of particles, constituting the MSP layers (the ‘templates’), as ‘seen’ by the S-PPs drifting in the gentle gradient. This is caused by the variations in orientations of individual template-partners, even while the resultant MSP-magnetization remains along the rock $H$-field direction. (For, the local magnetic field acting on a particle is the sum of the external field and the dipolar fields of the other particles [26]). Thus, spin-ordering in a diffusing S-PP, oriented along the rock $H$-field - State 1 having lower
Fig. 1. Directed interactive diffusion of S-PP through MSP (with parallel correlations). MSP represented in black; State 1/State 2: lower/higher template-affinity states of the ligand (L)-bound S-PP, in blue; green lines signify alignment in State 2; T.E. or thermal energy from bath; rock H-field direction indicated on top of figure, see text.

template-affinity – would change for aligning to the local H-field of a template partner–State 2 having higher template-affinity. These changes would be similarly facilitated by thermal excitations from bath [c.f. 36, 37], with rectification by either the gentle H-field gradient or local template-partner H-fields. Indeed, this scenario of changing H-fields for modulating intrinsic dipole-dipole interactions closely resembles the simulations by the Korenivski group [38, 39] who propose a ferrofluid-based associative neural network for pattern storage where the respective transition probabilities satisfy detailed balance. These demonstrate how local variations of the external H-field (via Zeeman effect) can be used to influence the positions and spin orientations of individual particles that (in contrast to ferromagnets) do not retain a magnetization upon removing the applied field. This spin degree of freedom of a magnetic dipole has an obvious parallel with the internal degree of freedom of molecular motors. Thus are recovered the features of Maxwell demon-effects, as well as Complexity, that allows a different route for regeneration in the other half-cycle. Furthermore, while a monomeric particle (Sect.3.2) in the diffusive searching phase could even lose track of its initial template, a dimer of particles would remain associated with the starting template, if the diffusing phase of the first particle coincided with the template binding phase of
3.4 Other motor aspects; magnetocaloric-effect

Further, in bio-molecular motors no overall macroscopic potential gradients are present, even if ATP hydrolysis in each cycle has the effect of raising the local temperature [40]. This ‘heat-engine effect’ upon ATP-coupling ensues from vibrational energy drilled into the molecule. Again, in the entropy-feeding motor mechanism proposed by Matsuno and Paton [41], coupling to ATP-hydrolysis leads to release of energy in very tiny quanta (similar in energy to Brownian hits) and thus an effective temperature of almost zero Kelvin for the actinomyosin complex. For correspondence to the S-PP scenario, it is interesting that a direct non-biological mechanism enabling interchange between a system’s environmental temperature and its own entropy is provided by the (anistropic) magnetocaloric effect (MCE) [42], which is the property of some magnetic materials to heat up when placed in an H-field and cool down when they are removed (adiabatic). And, recent evidence shows that at the nanoscale too, the heat capacity turns out to be a few-fold higher than that of bulk systems, thanks to MCE [43]. This effect can also be seen in another related study, although the paradoxical phenomenon of cooling by isentropic magnetization in high fields, in this six-particle system [44], is about one order of magnitude higher than the conventional cooling mechanism by isentropic demagnetization and is related to the ring-chain transition. The cross-over of states emanating from a conflict between magnetic and structural order underlies this paradoxical effect. In contrast, our present context involves no such conflict, since it embodies a conventional magnetic system where diffusing ‘hard’ S-PPs interact with H-fields of template partners. Anyhow, this simulation does provide a concrete example of a nano-scale manifestation of the MCE, in contrast to ferrofluid systems with a large number of particles considered for use in magnetocaloric heat engines [44]. And, a periodic manifestation of MCE -due to two entropic degrees of freedom (magnetic ($S_M$) vs thermal ($S_T$)) – follows as a logical consequence of periodic change superimposed on a background potential, provided by H-fields of template-partners for S-PPs drifting along the rock field gradient.

3.5 Motor vs magnetic-dipole

The Table offers some non-trivial parallels between motors moving on biopolymers and motion of S-PPs on layers of particles bound by magnetic dipolar forces. Their nano-size would give a negligible inertial term in the Langevin
| Correspondence in features | Bio-molecular motor | Particle diffusing through MSP (Hadean) |
|----------------------------|---------------------|----------------------------------------|
| Templates                  | Biopoly-mer like protein filaments and nucleic acids | Layers of magnetic particles with dipolar interactions |
| Low Reynolds number        | Nano-particle diffusing in intracellular viscous milieu | Nano-particle diffusing in magneto-viscous medium |
| Movement direction         | Amino end to Carboxyl end of template or vice versa | North to South pole or vice versa; gentle H-field gradient cuts through MSP. Cause vs effect |
| Conversion to mechanical energy directly: Symmetry breaking via infinitesimal decreased potential in progress direction | Gentle decrease in chemical potential upon ATP coupling for conformations having greater affinity for sites in preferred direction. | Gentle decrease in magnetic potential due to gradient, for diffusion in preferred direction till captured by H-field of template partner. |
| Switching between low and high template-affinity states by non-eqforce; energy flow from bath | Via different conformations due to altered landscape potential by ATP/ADP+Pi binding/release cycles, where thermal diffusion is rectified | Via different spin ordering due to altered magnetic potential by periodic presence/absence of template partner, helped by thermal excitation from bath |
| Small systems allowing for time-reversed trajectories | Nano-scale | Nano-scale |
| Time-reversible degree of freedom $\sim$ thermal hits ($k_BT$) | Can undergo infinitesimal conformational changes. | Can undergo infinitesimal spin alignment changes |
| Bond for stabilizing eq state? | Weak, e.g. H-bond, etc | Weak -alignment to partner |
| Increased apparent local temp of medium in each cycle and lowered temp. of that of motor/ magnetic particle? | Yes, ATP-coupling leads to enhanced vibrational motion (cannot account for transport via thermal ratchet); energy released in bits $\sim k_BT$, leading to nearly 0 K of motor. | Yes, MCE due to local partner could cause heat increase of S-PP consequently released to bath, with spin-relaxation away from partner, lowering S-PP temp. |
| Processivity vs attachment points | Enhanced for two heads vs one | Similar enhancement for dimers vs monomers |
| Self-propelled, template-interactive transport: non-eq energy and asymmetry | Non-eq, time-dependent repetition of asymmetry (‘seen by complex motor’) enables generation of drift velocity by averaging over thermal noise. | Non-eq, asymmetry from H-field grad for magnetic dipole. Cyclic template interactions due to H-fields of template partners in MSP. |
equation, which together with frictional forces, due to viscous medium gives a low Reynolds number. At each point velocity is the direct result of an external force, acting on the particle that achieves its terminal velocity instantaneously; thus thermal hits cause random diffusion. Both systems offer high efficiency mechanisms for direct conversion of a non-equilibrium source into mechanical energy, rather than via an intermediary state, e.g., heat for thermal engines [45].

The slight decrease in magnetic potential energy \( \sim -M(dH/dz)z \); assuming a constant gradient at small distances, where \( M \) is magnetization) of the diffusing particle has a parallel in the slight decrease in chemical potential of the motor believed to occur in the preferred direction of diffusion [5, 34]. Both are examples of small systems where time-reversed microscopic equations of motion allow for time-reversed trajectories. In both, thermal hits get rectified for the periodic recycling between higher and lower template-affinity states, by close-to-equilibrium coupling. In the motor, a slow modulation of chemical potential by thermodynamic energy from ATP hydrolysis biases conformational changes towards stickiness for forward binding sites, on the locally asymmetric but periodic template. ATP coupling breaks the microscopic reversibility and drives directed diffusion from N- to C-terminal or vice versa, with motors binding in similar orientations in either situation and not facing opposite directions. This asymmetric template-affinity is remarkably similar to how a gentle increase of field lines, to the front of or behind, a North to South oriented dipole can cause its drift in the forward or backward directions, respectively. Here detailed balance is broken by gentle changes in flux lines (non-eq) due to rocks, while interactive cycles with alterations in magnetic ordering are brought about thanks to alignment with local H-fields of consecutive template-partners in the MSP. For the ratcheting motor, the different trajectories in the two half-cycles enable net movement via asymmetric track-binding of intermediate states. Since trajectory in the first half-cycle is not retraced, neither is the motor velocity, as template binding capacity is changed [4, 46, 47]. Herein lies the difference: The non-equilibrium force does not push the motor directly but by rectified thermal diffusion via asymmetric motor template interactions. In contrast, for the diffusing magnetic dipole this symmetry-breaking drift would have been a direct consequence of the gentle flux changes, but superimposed local secondary H-fields would have generated periodic particle-template interactions, with altered magnetic ordering.

4.1 Ratchets replaced magnetic effects?

Clearly, bio-molecular motors are not driven by a macroscopic external force. But compare this to a slow directed diffusion of a magnetic dipole in a very gentle gradient due to a non-homogeneous magnetic field from rocks, a logical
scenario rooted in basic physical principles. To recapitulate, the combination for self-generated transport – non-equilibrium and asymmetry – are both provided by an H-field for a magnetic particle only; not ordinary matter. And, template interactions, with cycles between low and high affinity states, are seen as a consequence of local H-fields of consecutive partners in the MSP (itself another ramification of the rock H-field via magnetic-dipolar interactions between the particles). As to their origins, the first part of the puzzle seems to be one of searching for a driving force that could have enabled self-assembly, while simultaneously driving other responses, such as movement.

And, the second is to look for both an external driving force as well as the complexity of matter being driven. It does not seem to help if we only search an external force, e.g. a thermal gradient could have facilitated transport of ordinary matter, but quickly activated the crystal formation phase, thus limiting access to a soft self-assembled phase. The other possibility is to look for matter that could have been present in the Hadean with access to internal degrees of freedom, underlying the complexity of today’s biomolecules. Conceptually, these could have undergone self-assembly, and additionally used gradients, e.g. thermal, for eliciting a response. But how could the emergence of such matter be explained, in a limited time-frame? Now, in the origins of Life, S-PPs could have themselves turned into magnetic sources of energy in the presence of a moderate inducing H-field. It is this handshake between the magnetic features of a moderate field, S-PPs (with spin degrees of freedom), and the components of the MSP, that makes it all conceptually feasible in a Hadean Ocean Floor setting. It is possible that evolution of this magnetic system translated and merged the directed gradient-driven diffusion and (interactive) periodic alterations in magnetic alignment potential therein, into another with periodic alterations in landscape potential, as in bio-molecular motors. In the latter, the continuous interactive movement is via close coordination of chemical (hydrolysis) and mechanical (association-dissociation) cycles. Such evolving complexity (conformational changes connecting intermediate states via different trajectories) enabling close-to-equilibrium coupling to drive the macroscopic system uphill in its landscape potential—a bio-ratchet—could have helped disengage the local magnetic ladder.

4.2 Search for field controlled assembly in the Hadean

The search for super-paramagnetic matter that could have been externally controlled by means of a magnetic-field developed through serpentinization of the igneous rocks comprising the ocean floor, led us to the mound scenario conceived by Russell and coworkers [9]. The substance could well have been greigite (a non-stoichiometric Ni-bearing iron sulphide phase, \( \sim \text{NiFe}_5\text{S}_8 \)) whose similarity to complexes in enzymes considered ancient, helped link Life’s
origins to the Hadean Ocean Floor.

### 4.2.1 The mound scenario

The mound builds up slowly as iron-nickel sulphides precipitate along with other components in an envisaged environment enriched with gradients (moderate temperature, redox, pH), leading logically to a host of metabolites concentrated in membranous compartments, thereby endowing this scenario with rich metabolic potential (see Figure 2). Namely, water percolating down through cracks in the hot ocean crust reacted exothermically with ferrous iron minerals, and returned in convective updrafts infused with H$_2$, NH$_3$, HCOO$^-$, HS$^-$, CH$_3$; this fluid (pH $\sim$ 10 $\leq$ 120$^\circ$ C), exhaled into CO$_2$, Fe$^{2+}$ bearing ocean waters (pH $\sim$ 5.5 $\leq$ 20$^\circ$ C) [48]. The interface evolved gradually from a colloidal FeS barrier to a single membrane and thence to more precipitating barriers of FeS gel membranes. Since fluids in alkaline hydrothermal environments contain very little hydrogen sulphide, the entry of bisulphide, likely to have been carried in alkaline solution on occasions where the solution met sulphides at depth [49], was controlled. This was perhaps important for the envisaged gel-environment, since colloids often form more readily in dilute solutions - suspension as a sol- than in concentrated ones where heavy precipitates are likely to form [28]. Further, theoretical studies by Russell and Hall [50] show the potential of the alkaline hydrothermal solution (expected to flow for at least 30,000 years) for dissolving sulphydryl ions from sulfides in the ocean crust. The reaction of these with ferrous iron in the acidulous Hadean ocean (derived from very hot springs [50]) is seen as having drawn a secondary ocean current with the Fe$^{2+}$ toward the alkaline spring as a result of entrainment [51]. Significantly enough, the super-paramagnetic property of greigite ($\leq$ 30-50 nm [52]), one of the components of the FeS colloidal barrier, brings to light its possible magnetically reproducing aspect. And, framboids observed in the chimneys [53, plate. 2], reveal the role of physical forces in producing these dynamically ordered forms under mound conditions [28].

### 4.2.2 Field estimate from W-B model

The associated H-field with rocks, needed for overcoming temperatures $\sim$ 50C in the mound, is estimated by extrapolating the Wilkin and Barnes (W-B) model [57] for formation of framboidal pyrite. This is based on the alignment of precursor greigite (taken as single domain crystals), under the influence of the weak geo-magnetic field that would help overcome the thermal energy of particles above a critical size. Ferrimagnetic greigite has a saturation magnetization value $M_{sat}$ at 298K ranging between 110 and 130 kA/m. Assuming
Fig. 2. The hydrothermal mound as an acetate and methane generator. Steep physicochemical gradients are focused at the margin of the mound (see text for details). The inset (cross section of the surface) illustrates the sites where anionic organic molecules are produced, constrained, react, and automatically organize to emerge as protolife (from Russell and Martin [54], and Russell and Hall [49], with permission). Compartmenal pore space may have been partially filled with rapidly precipitated dendrites. The walls to the pores comprised nanocrystals of iron compounds, chiefly of FeS [55] but including greigite, vivianite, and green rust occupying a silicate matrix. Tapping the ambient protonmotive force the pores and bubbles acted as catalytic culture chambers for organic synthesis, open to H$_2$, NH$_3$, CH$_3^-$ at their base, selectively permeable and semi-conducting at their upper surface. The font size of the chemical symbols gives a qualitative indication of the concentration of the reactants.

In a spherical geometry, the critical grain diameter of constituent crystallites comprising the framboid interior $d_c = 2a$, where $a > 1$, is given by

$$d_c = \left(\frac{6k_BT}{\mu_0\pi M_{sat}|H|}\right)^{1/3}$$

This result can be obtained from the inequality $W_{WB} > k_BT$ where we define $W_{WB} \equiv \mu_0 M_{sat} VH$. Here $k_B$ is the Boltzmann’s constant and $\mu_0$ the permeability of vacuum. When aligned parallel to weak geomagnetic field ($\sim 70 \mu T$), $d_c = 0.1 \mu m$. According to this formula, a rock H-field for accreting 10nm sized particles would have to be 1000 fold higher. This also is of the same order of magnitude $\sim 10 mT$, seen for magnetite-based ferrofluids [16].
Fig. 3. Iron and FeNi particles derived from the subhypervelocity flux of iron and of NiFe-metal-containing chondritic meteorites and micrometeorites are distributed on the ocean floor below, and sparsely scattered throughout, the mound. Some of the nickel may have been incorporated into catalytic mineral clusters such as greigite; clusters later co-opted into proto-metalloenzymes [50, 13]. Unpublished work of Ostro and Russell (2008), kindly provided by M.J.Russell.

The saturation magnetization of magnetite ($M_s = 4.46 \times 10^5$ A/m) is about 3.5 times greater than that of greigite; from this one expects proportionate values for the fluid susceptibility of a corresponding greigite suspension, building up slowly in the ocean waters (see above). Further, the dipole-dipole interactions between negatively charged greigite particles, under mound conditions where pH is well above 3 [57] is likely to be aided by the screening effect due to ionic strength of natural waters [29]. This description of an aqueous suspension of super-paramagnetic greigite matches that of aqueous ferrofluids, although the particles would lack the protective (steric-stabilization) coating of their synthetic counter-parts. Hence such a dispersion should show non-ideal behaviour even under dilute conditions, where magnetic dipole forces would attract particles, as in non-ideal ferrofluids where dissipative internal structures are known to form in the presence of an external field. Of course, if energy due to dissipative structure formation were also included, the effective local field would be only higher; this can be checked by comparing $W_{WB}$ (see above) with the equation $W_T = W_M + W_S$ of Li et al [21] for a non-ideal ferrofluid (Sect 2.1), where the (positive) second term $W_S$ represents the effect of interaction, and effectively increases the value of the H-field present in the first term $W_M$. 

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Thus a moderate rock H-field $\sim 50$ -100mT would lead to magnetic accretion of nano-sized greigite particles – a soft magnetic assembly (a new phase) – not accessible without an H-field. Magnetic dipolar forces would provide the compression for ‘packing’ (in 3d-space) in this finite system, giving it access to aperiodic order. Indeed, this very logic underlies the ‘magnetic reproduction proposal’ of Breivik [58] where his model of templates formed from ferromagnetic ‘monomers’ offers a means to study the direct link between thermodynamic and the information-theoretic concept of entropy.

4.2.3 Local field due to magnetic rocks

So the issue is to look for how magnetic rocks could provide a moderate local H-field $\sim 50$-100mT. Now, low levels of magnetization in rocks leading to crustal magnetic anomalies on the present day Ocean floor are typically understood in terms of (apart from mechanisms like sedimentation) the classical mechanism of thermo-remnant magnetization (TRM) – acquired when newly formed minerals cool below their Curie temperature in the presence of the geo-magnetic global field. On the other hand, for achieving a local field we note that subsurface magnetic rocks are known to create sufficiently intense magnetic anomalies (w.r.t. geo-magnetic field) used to track their location. As an example consider the rich iron ore province in the Pilbara region of Western Australia with a background ambient magnetic field of about 55 $\mu$T, where a helicopter survey recorded high anomaly amplitudes of up to 120 $\mu$T, indicating the high percentage of iron ore composition [59]. Since field strength decreases rapidly with distance ($\sim r^{-3}$) from the magnetic medium, the corresponding value on the rock surface is expected to be higher by a few orders of magnitude. This overwhelms the contribution of the ambient geo-magnetic field, which was already about half as strong 3.2 billion years ago as it is today [60]. A stronger reason for the irrelevance of the geo-magnetic field vis-a-vis local (rock) H-fields comes from the fact that $\sim 4.1$-4.2 Ga, the time when Life is believed to have been already initiated ($\sim 4.2$-4.3 Ga [9, 50]), the geomagnetic field did not even exist (!) [61]. This leaves the local field due to magnetic rocks as a primary candidate governing the initial conditions leading to Life.

4.2.4 Magnetism from extraterrestrial sources

Now the present geomagnetic field strength is too weak to explain the high NRM (natural remanent magnetization) to SIRM (saturation isothermal remanent magnetization) ratios of lodestones, i.e., natural magnets with magnetic field strengths varying upto 0.1 Tesla [see 62], as the initial magnetization
depends on the strength of the inducing field. This eventually led to lightning remnant magnetism as a plausible mechanism [63]. Further, Tünyi et al. [64] have examined the possibility of nebular lightnings as a source of impulse magnetic fields (in the context of accretion of Earth and other planets, that is seen as rendering the gravitational accretion process more efficient) by magnetizing the ferromagnetic dust grains to their saturation levels. Quite possibly, the most important contribution to the crust was thanks to the presence of accreted highly magnetized meteoritic matter, with acquired isothermal remanent magnetism, such as seen in meteorite, lunar samples, etc. Indeed, the Wasilewski group [65] propose magnetization of chondrules that cooled while spinning and translating through a magnetic field, in view of their matching demagnetization profiles with that of melt slag droplets. They also employ the properties of metallic systems for explaining remanence in lunar and meteoritic samples containing iron and iron alloys in contrast to that of terrestrial ones comprising oxides. And, they describe specific structures and microstructures associated with magnetic remanence effects for the Fe-Ni system, produced by various transitions and transformations with or without diffusion [66].

4.2.5 Extra-terrestrial magnetic matter in the mound

Now, the presence of ferromagnetic matter due to vestiges of iron and iron-alloy-containing meteorite bodies in the primitive Hadean crust, seems relevant in view of conditions in the primitive crust that were highly reducing (in contrast to today’s picture) with the redox state depicted at Fe-FeO (Wustite) [67, 68]. While the oceans are believed to have been formed around 4.3Ga, life is thought to have emerged between 4.3 and 4.2 Ga [9, 50], when conditions in the newly formed crust still seem to have been extremely reducing [61]. Indeed, impact craters formed by asteroids and comets that offer a route for delivery of extraterrestrial iron from iron-containing meteorites, have been pointed out as hosting conditions important for the emergence of Life, e.g. catalytic reduction of CO$_2$ that is linked to the origins of metabolic pathways [69]. In an extension of this scenario, Ostro and Russell (2008; unpublished results kindly provided by MJ Russell) suggest that similarly reducing Ocean floor accumulations may also have resulted from the non-cratering (sub-hypervelocity) flux of NiFe-metal containing meteorites and micrometeorites onto the Earth’s surface. As shown in Figure 3, in addition to acetate production by reduction of dissolved CO$_2$ by precipitated Fe(II)-bearing minerals (Figure 2), the presence of Fe and FeNi particles accumulated around the base of the mound could have allowed CO$_2$ reduction all the way to CH$_4$. Their analysis is based on extrapolating available statistics on current flux of extraterrestrial matter vis-a-vis its metal fraction back over four billion years. Although exposure to water is expected to lead to corrosion, apart from the fact that external oxidized layers would hamper the weathering process, they argue that the presence
of nickel would have helped in enhancing the resistance of meteoritic metal to oxidation (as in stainless steel alloys). They have pointed out that owing to the then powerful tidal currents \[70\], dense meteoritic matter -from fine grained particles to larger ones- would tend to be trapped in local basins, e.g. collecting around protuberances like hydrothermal mounds. Thus, in such an environment with possibility of magnetic elements (magnetite, awaruite, and iron-nickel alloys), as observed in meteorites \[61\] the chances of producing a local magnetic environment seems highly plausible. Still another possibility is an internal mechanism like spontaneous magnetization.

4.2.6 Reinforcing H-field by serpentization

In an earlier version we had simply assumed the presence of magnetic rocks in the mound, say via mechanisms such as lightning remnant magnetism \[66\] and on the lines of Túnyi et al \[64\] that could have provided a local field up to \(\sim\) Tesla \[24(ii)\], while only moderate fields \(\sim\) tens of mTesla would suffice for accretion. Improving on this scenario, magnetic rocks are seen as situated immediately beneath the mound and to have been produced during the serpentinization of ocean floor peridotites in a process that generates magnetite \[71, 23\], and also awaruite \[72\].

Further, in the present paper, molecular motor-like diffusion (close-to-equilibrium) of greigite nano-particles through the MSP is envisaged as being propelled by a gentle flux gradient - a scenario which is rather naturally simulated by the non-homogeneous H-field generated by magnetic rocks. This is in contrast to an earlier approach \[24\] where we had considered the possibility of the temperature gradient in the hydrothermal system itself as having driven this molecular motor-like passage, and therefore the possible co-evolution of both reproducing and metabolic aspects of greigite simultaneously in the same location. But the problem of a single – location origin, in dealing with far-from-equilibrium gradients supporting metabolism with close-to-equilibrium driven diffusion in an identical location (the MSP being a delicate phase), has necessitated a change: a close but separately situated origins of the two wings of Life. And, not too far from the gradient, magnetic rocks in cooler waters, would coax a gentle build up of greigite particles into an MSP (aided by MCE due to the rock field that could give rise to a mild turbulence). The complexity of the MSP, in turn, would evolve thanks to a continuous supply of chemicals diffusing from the metabolic counterpart in the mound. It’s coupling to energy-rich ones may have led to bio-like ratchets, permitting exit from the confines of the magnetic rock field, with the two faces of greigite enabling complex energy transduction mechanisms (see Sect.4.3).
4.3 Conclusions: Double-origins revisited

The vicinity of a physical self-reproducer (via magnetic rock-controlled S-PPs) to its metabolic counterpart, as in the proposed double origins [8], could have allowed replacements via a chemical genie. And a driven system where a coherent energy source - the H-field - maintained phase correlations between constituents of the assembly would provide a natural selection basis for its chemical replacements with capacity for such anisotropic dynamics in the absence of the H-field. This is in contrast to conventional proposals of randomly evolving chemical reactions that by chance, led to the emergence of Life but in a non-specified time frame. The requirements of a starting magnetically controlled phase do offer a basis for explaining the emergence of coherently coupled systems comprising non-equilibrium sources like ATP on the one hand, and on the other of evolving soft matter with their internal degrees of freedom that can exist in different equilibrium states, inter-convertible by harnessing random Brownian motion. And note that the potential of magnetic particles for evolving transduction mechanisms lie in their capacity to interact with other sources of energy. For example, a magnetic Soret effect [73] can provide a means for rectified diffusion, on analogous lines to the thermophoretic Soret-effect [74], due to infinitesimal changes in: susceptibility vs solute-solvent interfacial tension, respectively.

Finally, a remarkable spin-off of directed movement of cargo-loaded magnetic particles, across a packed array, is a logical symmetry-breaking enrichment of one from a pair of ligated optical isomers, by the ‘grinding effect [see 75] due to space constraints on surface-transfer reactions. Note that magnetic effects can non-invasively resolve intractable mixtures [76] of magnetic and non-magnetic components; they can also show up invasively by controlling spin states in biological systems: from chemical reactivity (due to spin-selectivity of reactions; see [77]) to quantum coherence [78]. The Maxwell Demon-like potential of S-PPs diffusing through an MSP, due to a gentle H-field gradient gives access to coherent dynamics. Indeed, such a system seems to fit the requirements of Davies [79] quantum computing origins-of-life proposal, as also acknowledged by him in [80]. Again, the larger information storage-capacity of a DNA-motor system, than the usual 1 bit/base basis, results from several internal states of the motor itself [81]. And, such an information-processing network of DNA-motor-bath could have its natural origin in a magnetic Ancestor.

The common material constituents comprising all kingdoms of Life are certainly important clues for the origins of Life. But, the list of commonalities also feature non-material aspects like sense, induction, search capacity, sensitivity to fields, adaptation potential, feedback within a hierarchically assembled network where local units dictated collective properties emerging at the global level, to name some. The traditionally accepted picture is that these key fea-
tures of Life evolved at different space-times but merged together somehow to produce the replicating wing of Life. The other possibility considered here is a simple physical system having the above physical properties, including the capacity for computational searches. Armed with this potential and with help from the metabolic arm of the origins of life, it could have set about training chemistry, persuading it to behave like it does in biology, instead of as in non-living systems. Did we have a computing Ancestor directing its own evolution? Maybe condensed-matter physics could help in this search . . .

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