



NANO-TO-MICRO INTEGRATED SINGLE-ELECTRON BIOMACROMOLECULAR ELECTRONICS FOR MINIATURIZED ROBOTIC "UNTETHERED FLYING OBSERVERS"

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ABSTRACT

A miniaturized "untethered flying observer" as a robotic free-flying vehicle supplied with MEMS/nano-to-micro payload and hosted on a much larger spaceship from which it would detach for short exploration missions is discussed. Downsizing of integrated electronic devices and nano-to-micro integration through *nanoelectronics* would allow high density MEMS-electronics packaging, very low power consumption and light weight to be realized. The nano-to-micro integration concept through single-electron molecular electronic devices leads to problems in the development of what could be called a "meta-system" as to electron physics involved in merging solid state (ss) and molecular devices. A single-electronics chip featuring biomacromolecular circuitry-to-ss Si integration for merging sensing, computation, communication and actuator functions would contemplate electron transport chain protein components and/or suitably functionalized DNAs. Single-electron controlled tunneling would work in such biomacromolecules even at room temperature due to the atomic size of the "well" and consequent less delocalization of the electron. Network design asks for quantum orbital calculations, for minimal use of molecular wires/connectors, for integrated nano-to-micro blocks and for nano-to-macro-world connections. Recent laboratory demos as to conductive molecular wires (fullerene nanotubes and chain molecules) must overcome difficulties involved in making their results into a practical technology. Langmuir-Blodgett ultra-thin organic films or lipid bilayers for construction and support of the integrated single-electron molecular component blocks would make scalars into vectors. Si for the biochip-hosting "platform" of the robotic nano-observer can act as a radiation shielding.

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that has been dubbed MicroElectroMechanical Systems (MEMS) technology, as well as for nano-to-micro integration of the emerging nanotechnologies both with MEMSs and microelectronics for sensors, transducers, actuators and control.

Miniaturized "untethered flying observers" (1g - 1kg)¹ have been envisaged as robotic free-flying vehicles supplied with MEMSs payload and hosted on a much larger spaceship from which they would detach for short exploration missions. After exploration of the celestial body, the *untethered flying observer* would be caused to deorbit by its microthrusters and to dock to its mother-spaceship.

As electronics generally is about 30 - 40% of the payload designed for such exploration systems, the downsizing of the present integrated electronic devices and nano-to-micro integration through the emerging field of *nanoelectronics* would allow high density MEMS - electronics packaging, very low power consumption and light weight to be realized.

As shown in the following, problems would arise in connection with the realization of the nano-to-micro integration concept through a molecular monoelectronics/solid state (ss) Si hybrid chip that, according to the MEMS philosophy, would merge sensing, computation, communication and actuator functions. Recent experimental achievements in the field of molecular electronics and in the construction of conductive molecular wires and connectors look promising^{2,3}. Further experimental steps would lead from lab demos to practical technologies.

The nano-to-micro integrated system whose physics is discussed in the following has been dubbed here "the *metasystem*", the prefix *meta* meaning "change", as it is a system in which a discrete transfer of charge coexists with the quasi-continuous charge transfer along a metallic conductor. Single-junction, two-junction and multi-junction molecular *nanodevices* are suggested, made up of monoelectronic molecular components (the *nanocomponents*), for single electron correlated tunneling (SECT) to ensure a circuitry allowing transition from the *quasi-continuous current* to the *discrete monoelectronic process* to occur, and at the same time logic and memory circuits based on digital devices to be realized. Anyway, as opposed to such application of

1. INTRODUCTION

Space research strives for integration of microelectromechanics with micromechanics into a technology

the so-called "universal computer conception paradigm"⁴ down to "one bit is one electron", the possibilities involved in other kinds of information processing techniques at the molecular level⁵ are briefly discussed.

The conception developed here departs both from the earliest concepts of complicated molecular systems embodying soliton logic functions⁶, and from Aviram's concept⁷ of electron tunneling through the chain system

metal — molecular electron acceptor fragment — nonconductive barrier — molecular electron donor fragment — metal

in which the energy of the highest occupied molecular orbital of the nonconductive form is less than the Fermi level of metal, so that no electron tunneling takes place until an electric potential is applied to two *microelectrodes* on the central axis of the molecule employed. Both such conceptions would imply very complicated technologies for the actual implementation and operation of the whole system.

The electron transport chain (ETC) properties of many proteins, together with the present progress in protein engineering and in functionalizing DNAs, offer a good opportunity for constructing nanoscale single-electron devices. Electron localization in molecules is given by molecular orbitals, while electron transport between localized states can be controlled through the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) separation (i.e. through the electronic coupling tunneling matrix), the Franck-Condon factor (nuclear modes coupling to the electronic states in question) and the electronic-nuclear motion coupling.

A host of results can be found in the literature as to charge transfer in small molecules and in macromolecules and as to ETC in proteins; the latter phenomenon, arising from chains of *redox centers* in many cellular proteins, is a basis of many life processes. Knowledge of *intramolecular* and *intermolecular* electron transfer is of the essence for translating a molecular electronics theoretical design into a real molecular construction. So from the science of ETC proteins a practical molecular electronics technology could be obtained. As shown in the following, molecular nanoelectronics, and its integration with ss microelectronics, is a crossdisciplinary problem involving quantum chemistry and quantum electron dynamics (outside the "universal computing paradigm", e.g. outside the conventional

computer *rigid* architectures, it will also be a problem in far-from-equilibrium quantum electron kinetics).

Accordingly, components consisting of ETC proteins and/or of suitably functionalized DNA molecules will be considered here as the basic components for the *granular, controlled tunneling electronics* contemplated for integration with ss Si microelectronics into a biochip for a robotic nanosatellite (1g - 1kg) free-flying observer. The main features of an envisageable structure of the latter will also be discussed.

2. DESIGNING THE METASYSTEM

The monoelectronic character of really *molecular single-electronics* requires the introduction of the notion of "current" as the number of electrons through the conductive molecule in a time interval, one electron only being transferred in every separate event along a given orbital.

The *Coulomb blockade*⁸ of tunneling will give the SECT effect. The driving force of electron transfer is determined by the redox centers potential difference. The system design must take into account the fact that such electron transfer is a real reaction: it affects the immediate surroundings, as for instance the Langmuir-Blodgett monolayer employed for support and/or chain-like or layer-like connection of the active biomacromolecular units.

The structure of the proposed nanoelectronic circuit will thus consist of a number of *redox centers*, whose properties are to be described through quantum chemistry principles, which are the *building blocks* or the "*nodes*" of the *network*, i.e. according to standard quantum chemical terminology the *donor* (D) and *acceptor* (A) biomacromolecules or biomacromolecule parts separated by "*spacers*" (S) acting as the tunneling barrier. The principles of single electron tunneling (SET)⁸ will be applied to the determination of the structure of biomacromolecular nanodevices and their electron dynamics for realizing logic circuitries: separate electrons, captured through *Coulomb blockade*⁹, will be used to represent digital bits. First the nature of such nodes (the *nanocomponents* consisting of biomacromolecules) will be discussed, not from the standpoint of their chemical constitution, but as to their nanoelectronics; second, the *nanodevices* realizing SECT; third, their *connections* (the problem of wiring, nano-to-micro and nano-to-macro-world connection, i.e. connectors and addressing for communication between the macro- and the mesoscopic worlds, and dissipation); fourth, their network interaction (the *circuit*) will be discussed.

2.1 THE NANOCOMPONENTS

ETC proteins and/or suitably functionalized DNA molecules are suggested for the metasystem in question. Electron transfer in proteins occurs from liquid helium temperature up to the temperatures of survival of some bacteria, e.g. 120°C. There are reasons to think that electron transfer in proteins can be realized even at higher temperatures. Recent research on DNA¹⁰ would suggest that the



arrangement for realizing a molecular scale size tunnel junction and then SECT molecular monoelectronic circuitry, suitable for matching the (quasi)-continuous current bulk metal or semiconductor system, can be made up of both such kinds of biomacromolecules.

ETC research in living systems would supply the molecular electronic research with a host of data and processes to be exploited for this emerging technology. Continuous progress in Protein Engineering would allow the "customizing" of natural protein properties to the desired values.

Tunnel junction areas obtainable through molecular monoelectronics would be in the 1nm² range. According to the Coulomb blockade theory this would lead to critical temperatures (i.e. where thermal fluctuations suppress the Coulomb blockade) much higher than room temperature. So the molecular SECT would work within a much larger temperature range than the ss-based tunnel junction. Moreover, the operational times of a molecular SECT would be in the ps range.

A general picture embodying the controlling physicochemical aspects involved in molecular nanocomponent operation in a SECT circuit is given in Fig. 1, which also stresses the role of the *system surrounding* through its "reorganization energy" λ which is to be taken into account in the design. Quantum mechanical calculations for the non-adiabatic and the (much less likely) adiabatic electron transfer can be carried out by well established methods^{11,12}. Here it is to be stressed that the *component connection problem* is intertwined with the *fabrication technology* adopted, e.g. the well known Langmuir-Blodgett monolayer technologies.

The surrounding of the active



molecule (the *donor - acceptor plus the spacer* system) is marked by the hatched area. The *ks* are the electron transfer rate constants. These and λ can be

calculated by quantum chemical methods^{11,12}. Excitation by light quanta has been assumed here.

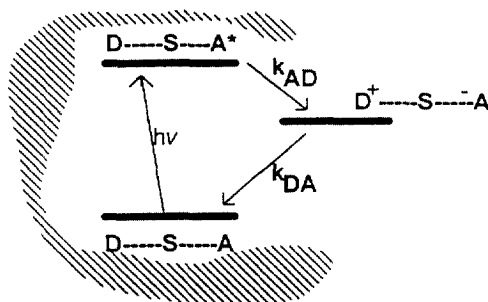


Figure 1

Illustrating the simplest electron transfer reaction; tunneling through the "spacer" S can be modulated by spacers of different lengths

The *nanoscale molecular minimal building block* for the SECT circuitry of Fig. 1 should be designed so that both λ and the surrounding fluctuations do not affect the *redox centers* through which the ETC would be realized.

2.2 THE NANODEVICES

Single-junction, or otherwise two-junction/multi-junction SECT would be realized by

- 1) ETCs made up of protein molecules according to the scheme

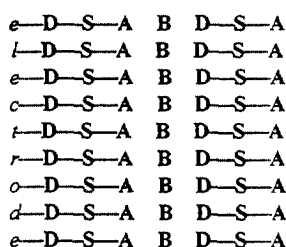


arranged in the shape of arrays of ETC chains between micro- or macroelectrodes:

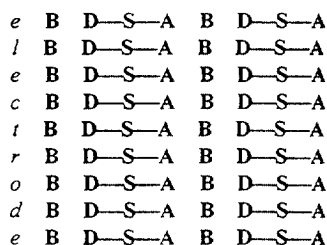
(metal or semiconductive electrode)—D—S—A— ...

or as ordered layers obtained through thin film technology:

a)



b)



where *electrode* stands for a metal or semiconducting electrode, and B is an electronic saturated (i.e. insulating) chemical structure, suitably calculated for the desired electron transfer rate, acting as a mere bridge to avoid electron-hole recombination and for charge localization, or, in case b), to obtain electron transfer with the electrode and no change in the molecular layer potential

or

2) by special arrangements of the biomacromolecular D—S—A units, e.g. into one of the possible forms of a SET (Fig. 2 and Fig. 3) and/or in the form of a single-electron pumping device or of the so-called "turnstile" device designed for ss electrode - junction devices¹³ (Fig. 4 and Fig. 5 respectively):

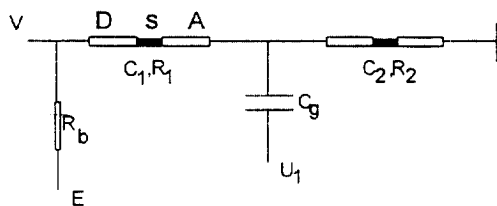


Fig. 2

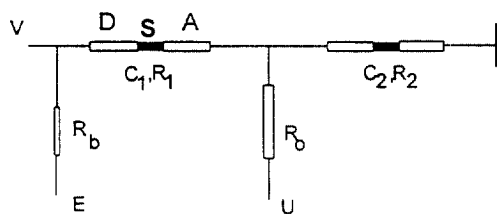


Fig. 3

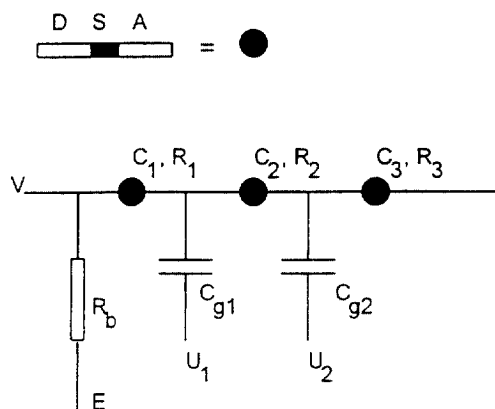


Fig. 4

The Single-Electron Pump

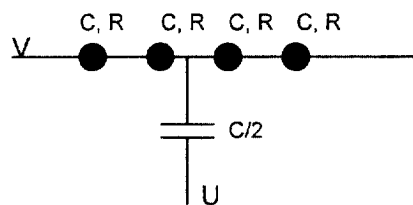


Fig. 5

The Single-Electron Turnstile

While the circuits of Fig. 2 and Fig. 3 can control only the average value of the current, as the times when electrons pass through the device are random, the circuit of Fig. 4, through the action of two control voltages allows electrons to be transferred one by one in the three-junction device, that has been nicknamed the "single-electron pump"¹⁴, as the charge can flow in a direction opposite to that imposed by the transport voltage V on reversal of the same. So this circuit gives the possibility of controlling charge flow at the single-electron level, just like that in Fig. 5, but the latter is irreversible.

In all figures showing the equivalent circuits of assemblies including macromolecular nanoelectronic components and microelectronic circuitry, i.e. macro-components, the standard macroscopic concepts employed to describe circuits (V as the transport voltage sources to deliver a static current, U's as the control gate voltage, C's as capacitances, R's as resistances, the subscript b for biasing) have been adopted; however, the *network dynamics* discussed in the following involves a description that takes into account both the ongoing at the macromolecular (i.e.

mesoscopic) level and their connection with the macroscopic level of the microelectronic circuitry in the *metasystem*. Thus, from the theory of Coulomb blockade of tunneling, e.g. in Fig. 1 the single-electron Coulomb energy E_c coming into play is

$$E_c = e^2/2(C_1 + C_2 + C_g)$$

and it is a basis to describe the electron flow just if the quantum levels and their transitions in the mesoscopic components making up the whole nano-to-micro integrated circuit are known. It is to be recalled here that the tunnel resistance, usually pointed out jointly with the tunnel capacitance, does not imply any dissipative process: it means just that at the junction barrier the wave function of the electron undergoes partial transmission and partial reflection. It is an entirely elastic process.

While the standard logic circuitry could be implemented at the molecular level through such devices, with integration of a monoelectronic device with a macroscopic microelectronic device into a *metasystem*, other kinds of information processing, e.g. those of special purpose kind, could be realized. It is worth recalling that the underlying operational principle (the single-electron tunneling) also works under *far-from-equilibrium* conditions, so lending itself to new concepts in robotics⁵.

Molecular quantum wells can be introduced in the *metasystem* by HOMO - LUMO calculations as to the structure and suitable size of the molecular or macromolecular S group, whose ionization potential and electron affinity must be well separated from those of D and A for correct charge transfer rates. They would work even at room temperature as to Coulomb blockade, due to the small size of the junction and consequently the high localization of the electron.

2.3 CONNECTIONS AND THE NETWORK DYNAMICS

The problem of "connection", meaning the choice of *connectors* in the *metasystem* and a means for *addressing* the mesoscopic level (i.e. communication between the macroworld and the nano-level, down to definite segments of a macromolecule) is closely intertwined with the problems of *fabrication* and of *network dynamics*.

The whole picture of the proposed technology to bridge the microscale and the nanoscale physics contemplates a kind of *modular chemistry* and the interpretation of molecular electronic structures as artificial *supramolecular structures*, i.e. the counterparts of natural supramolecular structures to be found

in living beings. *Elemental modules* carrying the basic electronic functions could be assembled through the already well developed molecular recognition techniques into complex-structure modules, keeping the size of such modules within the nanoscale level. Biomacromolecules would be especially suitable for such "self-assembly" technology, and the combination with Langmuir-Blodgett technologies¹⁵ for thin film and even for monomolecular layer production would lead to molecular monoelectronic *blocks* making up the supramolecular electronic structure to be connected with macroscopic microelectronics. The intervention of so-called *dry nanotechnology* through STM manipulation, capable of working on one molecule at a time, would be limited to a few blocks for final connection. Some wiring through the envisaged nanoscale conductive assemblies (conductive nanoscale fullerene wires have been realized recently by the Nobelist Richard Smalley at Rice University, Texas) might be employed.

From this and other laboratory demos² about the feasibility of molecular conductive wires it can be envisaged that practical technologies for their mass production will be available in a near future, especially if a market demand will develop. Indeed, e.g. polyene $(CH)_n$ unsaturated (conjugated) molecular chains, suitably doped for charge delocalization, would be endowed with extremely large conductivities; i.e., they could be employed as molecular wires through which one-dimensional electron transport would be possible over distances *very large on the molecular scale*. Anyway, conductive though such molecular connectors may be, minimization of their number and length in favor of an *integrated supramolecular structure* and of efforts towards very low resistance electrical contacts to macroscopic microelectronics are necessary requirements to overcome the main problem met with in realizing extremely large integrated electronic architectures, i.e. the problem of heat removal, which, though the power dissipated in a SECT nano-to-micro integrated system would be very low, should occur through nanoscale size surfaces.

A mesoscopic system is the frontier between the microscopic world that requires a quantum mechanical description and the macroscopic world which can be described by Newtonian mechanics. The *metasystem* considered here joins the two worlds; so an adequate way must be adopted to connect them. The envisaged supramolecular structure of two-level, D-S-A macromolecular electroactive elements, that make up the mesoscopic structure, is supplemented by macroscopic electrical connections for external control. Direct *addressing* of single macromolecules

or their segments (macro-to-nano connection) for communication with the mesoscopic world would also be desirable. So the *dynamics* of this *network of redox centers* as the *electrical nodes* in the ETC protein or any biomacromolecule can be pictured through the Hamiltonians of the single mesoscopic nodes modified by the node - node interactions which give rise to the *electronic eigenstates* of the network. In Fig. 6 the hatched area depicts the immediate environment of the mesoscopic structure, i.e. the portion of the environment capable of affecting both the quantum state of each node, mainly through the already mentioned *reorganization energy* λ , and the *capacitance of each node* in the Coulomb blockade process. The size of environment that would affect C will be discussed in the following. As to the value of such capacitance C of a macromolecular node (the molecular tunnel junction), it looks like reasonable to assume that the charging energy measured by $n^2 e^2 / 2C$ for a 10 nm junction in ss circuits can be substituted in a 1 nm meso-object described through microscopic terms with the total energy difference between the ionized and the neutral state. The "environment" also acts as a thermal bath.

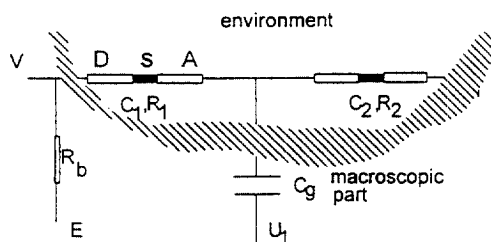


Fig. 6

This picture would be particularly illustrative in case the voltage V drives the system far from equilibrium, as it stresses that, in developing a quantum kinetic treatment of the system, the *macroscopic part* of the same would act as a set of boundary conditions.

As open systems, the integrated systems considered here including MEMS, digital logic circuitry controlling micromachines, sensors, etc. for use in an autonomous robotic untethered nanosatellite should be treated rigorously as nonequilibrium systems. However, the assumed closeness to the equilibrium state allows them to be considered at equilibrium.

The node Hamiltonian for the electronic eigenstates of an N nodes network with I states for each node is thus for single-electron tunneling

$$H = N \sum_{n=1} H_n + (1/2) \sum_{n \neq m} H_{nm}$$

where

$$H_n = I \sum_{i(n)=1} E_{i(n)} |i_n\rangle \langle i_n|$$

$$H_{nm} = \sum_{i(n), i(m)} U_{i(n)i(m)} |i_n\rangle \langle i_m|$$

and $|i_n\rangle$ as the eigenfunctions of H_n whose eigenvalues are $E_{i(n)}$.

This set of equations as a quantum mechanical description for instance of the circuit of Fig. 2 would become particularized into

$$H = \Delta G + H_{t1} + H_{t2} + H_{t3} + H_1 + H_2 + H_3 + H_4$$

where ΔG = Gibbs free energy; H_t 's are tunnel junction Hamiltonians, H_1 are the D and A Hamiltonians; moreover, with currents as shown in Fig. 7, and $C_s = C_1 + C_2 + C_g$,

$\Delta G = (Q/2C_s) - E[n_1(C_2 + C_g)/C_s + n_2 C_1/C_s]V$ + an integration constant from

$$Q_0 = \int I_0 dt + C_g U$$

$$Q = e(n_1 - n_2) + Q_0$$

with n_1, n_2 as the number of electrons already transferred through the junctions

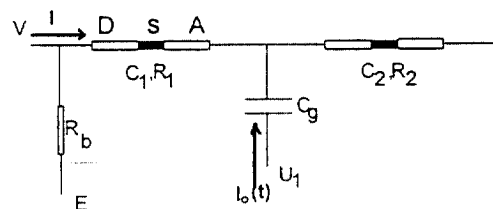


Fig. 7

As to the problem of nanowires or of macroscopic connectors affecting the value of the capacitance for the Coulomb blockade effect, the *limiting environmental size* of each quantum mechanical node could be determined according to the Büttiker - Landauer conjecture¹⁶ formulated in the early studies of charge tunneling processes. On application of such criterion to the network macromolecular nodes, according to Relativity principles the limiting size would be the radius r of the horizon of events of each node calculated from the tunneling process time and the time of restoration of the quantum state of the D and A parts of the nanosize junction. Such times can be estimated to amount to about 10^{-14} s, so that

$$r \approx c \times (10^{-14}) \text{ m} \approx 10^{-6} \text{ m}$$

and connector portions of the size up to 1 μm could affect the node capacitance. The capacitance of usual micro-connectors of that size would be of the order of the fF, and this low value might spoil the advantages of macromolecular electronic technologies with respect to ss nanolithography.

The *addressing* of a *single node* of the mesoscopic network would occur through STM techniques, which allows very small spot areas to be addressed, or through suitable developments of "*spectral hole burning*" techniques aiming to address molecules through their remarkable selectivity to light frequency and externally applied fields; or through *holographic spectral hole burning*, if some spatial delocalization of the information bit is allowed with respect to the "*one bit one molecule*" or "*one bit one specific area of a biomacromolecule*" philosophy is (just a little *bit*) relaxed.

3. BIOCHIP-BASED ROBOTIC FLYING OBSERVERS

The fundamentals of the envisaged nanosatellite technology show that the structure, shape and power sources of a nanosatellite, defined as a satellite whose mass is between 1g and 1kg, strongly depend on the mission duration and purposes¹⁷.

The general purpose autonomous robotic miniaturized "*Untethered Flying Observer*" proposed here would consist of a spherical shape, 2 cm radius all-silicon structure for suitable radiation shielding and for an acceptable ballistic coefficient even for exploration missions on planets with an atmosphere density quite similar to that of the Earth. The structure, whose weight would slightly exceeds 80 g (Si density is about 2.4 g/cm³) would be highly integrated, according to the conceptions discussed above, and so made up of nanoelectronic and microelectronic circuitry, and of microsystems such as physical, chemical¹⁸ and biochemical sensors¹⁹, pumps, valves, fluid channels, making up a MEMS. The subsystems, mounted on a silicon platform, would include monitoring microdevices (pumps, fluid flow sensors, mixers, reactors, and nano/microelectronics for a whole *processing* integrated subsystem) both for physical, chemical and biochemical experiments. Accordingly, the highly integrated structure would also be *modular for design flexibility and adaptation to special missions*.

Silicon would thus be employed both for MEMS and for the structural parts. Indeed, its mechanical and thermal properties (high yield strength, 7,000 MPa as a single crystal; Young's

Modulus of 170 GPa; and 3.0 MN-m/kg strength-to-density ratio; high thermal conductivity, 150 W/m K; high melting temperature (1,700 K) and low thermal expansion, 2.5×10^{-6} cm/cm K) make them the material of choice with respect, e.g., to aluminum. reover, silicon wafers are now routinely produced with diameters up to 20 cm, and microfabrication techniques for integrated sensors and microsystems are continuously developing. So batch-fabricated, all-silicon nanosatellites and probes can be considered as a near term goal.

The 80 g nanosatellite as an untethered object, employed together with a number of its replicas into a swarm of probes for exploration missions or as a member of a cluster of probes linked to each other, might need no active thermal control system due to Si heat conduction properties. However, such vehicle, being conceived as a robotic system hosted on a much larger spaceship from which it would detach for an exploration mission and to which it would be docking after deorbiting when its task is over, would be endowed with a propulsion microsystem, be it an electric (microresistojet or micro ion engine) or a chemical propulsion (liquid or solid fuel micro-thruster) system. A chemical propulsion microsystem would be capable of providing very small *kicks*, e.g. a few mN, for attitude control. Such systems would add just a small weight to the satellite/probe, including the propellant microtank, pressure microsensors, microvalves etc. Anyway, the modular structure of the propulsion system would allow flexibility of choice according to mission requirements.

If the mission asks for transmission of data from a nanoprobe not intended for being recovered, an antenna and its bulky auxiliary devices would increase weight even in case of transmission over short distances and adopting the presently available smallest antennas, but the weight of the complete vehicle would keep within nanosatellite definition size limits.

While the whole surface area of the 2 cm radius all-silicon nanosatellite could be covered with solar cells as its power system, the employment of the nanosatellite as an exploration means on board an interstellar spaceship would require different conceptions as to the power system. The modular structure of the suggested general-purpose robotic nanovehicle would allow the most proper selection of the power system and of its architecture to be made by the designer.

Let us suppose that the nanosatellite in question is to be stocked for a long time on the mother spaceship before being released for an observation mission. Moreover, let the design be carried out on an envisageable near-medium term technological projec-

tion (i.e., more than 5 years). The modular structure would allow two choices for the power source and the corresponding architectures of the whole power system (solar arrays, primary bus, power converters etc.): the adoption of lithium primary batteries for short missions, with exclusion of solar arrays; or the use of the newly introduced technology of lithium ion cell chemistry, under development at the present time, which is based on intercalated lithium ions. They would store the energy generated by multi-junction photocells. Unfortunately, data about the cycle life of Li^+ ion cells are unavailable as yet. Should the foreseeable technology efforts fail to obtain satisfactory values of cycle life for space missions, batteries based on the well known NiCd chemistry could be introduced in the design²⁰.

4. CONCLUSION AND OUTLOOK

The smart autonomous, ultra-low power consumption nanosatellites discussed here ask for a considerable, yet stimulating and commercially invaluable technological jump. Translating the results in nanoscale science already obtained, and those envisageable in the near future, into practical, market-reaching technologies would be rewarding and would open the way to new concepts in robotics of particular interest for space exploration.

Laboratory results as to nanoscale construction of electronic components look promising and microfabrication techniques toward MEMS are advancing quite rapidly. Yet some crucial, but not insurmountable problems must be tackled, mainly in connection with mass production. The inquiry into the behavior of such integrated nanosystems under far-from-equilibrium conditions⁵ would disclose new ways to the synthesis of much more efficient, smart micro- and nanorobots for space exploration. The possibility of material embodiment of new, biosystem inspired information processing and computing concepts, and of construction of the related nanostructured actuator members as well is at stake.

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