Nanorobots, NEMS, and Nanoassembly

ARISTIDES A. G. REQUICHA, FELLOW, IEEE

Invited Paper

Nanorobotics encompasses the design, fabrication, and programming of robots with overall dimensions below a few micrometers, and the programmable assembly of nanoscale objects. Nanorobots are quintessential nanoelectromechanical systems (NEMS) and raise all the important issues that must be addressed in NEMS design: sensing, actuation, control, communications, power, and interfacing across spatial scales and between the organic/inorganic and biotic/abiotic realms. Nanorobots are expected to have revolutionary applications in such areas as environmental monitoring and health care.

This paper begins by discussing nanorobot construction, which is still at an embryonic stage. The emphasis is on nanomachines, an area which has seen a spate of rapid progress over the last few years. Nanoactuators will be essential components of future NEMS.

The paper's focus then changes to nanoassembly by manipulation with scanning probe microscopes (SPMs), which is a relatively well established process for prototyping nanosystems. Prototyping of nanodevices and systems is important for design validation, parameter optimization and sensitivity studies. Nanomanipulation also has applications in repair and modification of nanostructures built by other means. High-throughput SPM manipulation may be achieved by using multitip arrays.

Experimental results are presented which show that interactive SPM manipulation can be used to accurately and reliably position molecular-sized components. These can then be linked by chemical or physical means to form subassemblies, which in turn can be further manipulated. Applications in building wires, single-electron transistors, and nanowaveguides are presented.

Keywords*—*Atomic force microscopes (AFMs), molecular machines, nanomachines, nanomanipulation, nanorobotics, scanning probe microscopes (SPMs).

I. INTRODUCTION

Nanorobotics is concerned with: 1) design and fabrication of nanorobots with overall dimensions at or below the micrometer range and made of nanoscopic components; 2) programming and coordination of large numbers (swarms) of such nanorobots; and 3) programmable assembly of nanometer-scale components either by manipulation with macro or micro devices, or by self-assembly on programmed templates or scaffolds.

Interest in nanorobotics is growing rapidly, e.g., within the IEEE, as evidenced by the papers and tutorials presented at the first IEEE international conferences on nanotechnology. A nanorobotics community is beginning to emerge. This growth of interest reflects the enormous potential of the technology, and also recent technical advances (for instance, in nanomachine synthesis) that suggest that nanorobots will not remain in the realm of science fiction much longer.

Nanorobots have overall dimensions comparable to those of biological cells and organelles. This opens a vast array of potential applications in environmental monitoring for microorganisms and in health care. For example, imagine *artificial cells* (nanorobots) that patrol the circulatory system, detect small concentrations of pathogens, and destroy them. This would amount to a programmable immune system, and might have far-reaching implications in medicine, causing a paradigm shift from treatment to prevention. Other applications such as *cell repair* might be possible if nanorobots were small enough to penetrate the cells. In addition, miniscule sensors and actuators are needed if the emerging vision of a physically coupled scalable information infrastructure is to come about. This is believed by many researchers to be the natural successor to the World Wide Web of today. It is a network of thousands or millions of nodes that can sense, process information, and act, and, therefore, are robots, albeit possibly simple ones. For this to be practical, very small devices are required and, therefore, this vision depends on progress in micro and nanorobotics.

Nanorobots are but one example of nanoelectromechanical systems (NEMS), which represent a new frontier in miniaturization, looming beyond the microelectromechanical systems (MEMS) that today constitute a multibillion-dollar industry.

A major obstacle facing nanotechnology today is the lack of effective processes for building the nanoscale

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The author is with the Laboratory for Molecular Robotics, University of Southern California, Los Angeles, CA 90089-0781 USA (e-mail: requicha@usc.edu).

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structures needed by the envisaged applications. Research at the University of Southern California, Los Angeles, Laboratory for Molecular Robotics and elsewhere shows that nanomanipulation with scanning probe microscopes (SPMs) provides an effective approach for constructing nanostructures from the bottom up, by assembling building blocks that result from chemical synthesis (e.g., molecules or colloidal nanoparticles). The primary shortcoming of this approach is its sequential nature and the associated low throughput. High throughput may be achieved, however, by massively parallel assembly operations using SPM multitip arrays, which are built by MEMS techniques. For example, an IBM group is building multitips for digital storage applications that are expected to achieve densities on the order of a few Tb/in^2 [69].

Single-tip SPM manipulation will be very useful for the foreseeable future as a device *prototyping* technique. Regardless of how a nanodevice will eventually be mass produced, prototyping is needed to ensure that the device will work as intended, and to optimize its parameters. The characteristics of a device, for example, its geometry, often can be altered easily by nanomanipulation, to study the sensitivity of the device to parameter variations. This is usually difficult to do by using self-assembly or other construction processes. Single-tip nanomanipulation may also be used to repair or systematically modify structures built by other means. Therefore, SPM nanomanipulation is here to stay.

The remainder of this paper is divided into two major sections. The first deals with nanorobot and NEMS construction, with an emphasis on nanoactuators, an area that has seen rapid development in the last few years and is of primary importance for future NEMS. The second section focuses on nanomanipulation with SPMs. Programming of robot swarms and sensor/actuator networks are complex topics that deserve separate treatment and are not covered in depth in this paper.

II. Nanorobots and NEMS

A. Background

Nanorobots, nanomachines, and other nanosystems discussed in this paper are objects with overall sizes on the order of a few micrometers or less *in all three spatial directions*, and which are assemblies of nanoscopic components with individual dimensions ~1–100 nm. Medical nanodevices traveling in the human body for therapeutic purposes have captured the public's imagination at least since the times of the movie *Fantastic Voyage* (Twentieth Century Fox, winner of the 1966 Oscar for best visual effects). Order-of-magnitude feasibility calculations [19], [25] indicate that nanorobots are not physically impossible. They would be extremely useful not only in the medical field but also in applications such as: 1) monitoring and interacting with harmful microorganisms in the air or in water and 2) building intelligent surfaces with a controllable (programmable) structure, for example, with variable roughness and friction. However, artificial nanorobots do not exist today, primarily because of the difficulties in building the necessary nanostructures. The only extant nanorobotic systems are biological, and provide an existence proof that such systems are indeed feasible.

Nanorobotics and, more generally, NEMS research involves design (which often is biologically inspired), prototyping, fabrication, programming, and applications such as biomedical nanotechnology.

Robotics at any scale involves sensing, control, actuation and propulsion, power, communications, interfacing, and programming and coordination. In the following sections we discuss some of these issues, with an emphasis on actuation, which is a fundamental requirement for robotics. (We use the terms "machine," "motor," and "actuator" as synonymous in this paper.) We will often look toward biology, for instance to microorganisms such as bacteria, to see how nature has solved some of the problems that nanorobots will encounter.

B. Sensors

Artificial sensors that are truly nanoscopic do not yet exist, as far as we know. Devices that exploit the change in conductivity of nanotubes or nanowires when they are exposed to specific substances are perhaps the closest to true nanosensors [17], [34]. Sensitivity to different chemical species can be achieved by suitably functionalizing (i.e., attaching chemical groups to) the sensing elements. Although the tubes and wires used in these sensors are several micrometers long, it should be possible to make them shorter and still keep their sensing capabilities.

Chemical sensors based on microscopic cantilevers are being investigated by several research groups, and often called nanosensors, but they are really microscale devices [26], [67]. These sensors use two primary mechanisms; they either: 1) detect the deflection of a cantilever caused by surface stresses that arise when a chemical species binds to one of the two opposing sides of the cantilever or 2) measure the shift in the resonance frequency of a vibrating cantilever when its mass increases because of the deposition of the molecules being detected.

Tactile (i.e., force) sensing using functionalized SPM cantilevers is being investigated at the Laboratory for Molecular Robotics for applications in identification of marine microorganisms. Chemical sensing using similar techniques has already been demonstrated by Hinterdorfer's group at the University of Linz [29]. It may be possible to miniaturize these approaches by using nanoscale cantilevers, but this has not been done yet, as far as we know. Cantilevers with resonance frequencies around 1 GHz have been developed recently by using lithographic processes [31], but are still too large to be considered nanoscale objects according to the definition used in this paper. In addition, submicrometer cantilevers fabricated by lithographic techniques appear to have inherent elastic instabilities [13].

Fluorescent probes of nanoscopic dimensions have been demonstrated—see, for example, [14], [15], [22]. These probes can be injected into cells, and report on concentrations of chemicals inside the cells. However, a probe *per se* is not a sensor; it requires a light source and a fluorescence detector. These are normally of macroscopic dimensions,

and it is unlikely that they can be miniaturized for use in autonomous or semiautonomous nanorobots. Perhaps more promising is an approach that exploits the change of conformation (i.e., shape) of a protein when a binding event takes place [6]. The binding of a specific chemical species recognized by the protein causes motion about a "hinge," which results in a change of distance between an electrically-active component and an electrode and, therefore, can be detected electrically.

Bacteria may use sensors for such stimuli as magnetic fields or light, but mostly they sense chemical concentrations by using molecular transduction mechanisms. More generally, much of the sensing at the nanoscale that is done in nature appears to be chemical. These chemical sensors require *contact* between the receptors and the sensed chemicals. Macrorobot sensing strategies for navigation and other applications normally use sensor modalities such as sonar, which do not require physical contact with the sensed objects. Robotic strategies that relie only on contact sensing have not been studied, as far as we know.

Sensor technologies, especially chemical and biochemical sensing, are evolving rapidly, with a new sense of urgency that arises from the events of 11 September 2001, and perceived threats of chemical or biological warfare. Micro and nanosensors are especially interesting because they are small and unobtrusive, should be cheap to fabricate and operate, and—importantly—they should be more sensitive than their macroscopic counterparts. For example, as the dimensions of a vibrating cantilever sensor decrease, the change of mass that corresponds to a detectable shift in its resonance frequency decreases as well and, therefore, the sensitivity of the sensor increases. Networks composed of a large number of very small sensors will be able to acquire and process data with unprecedented spatial resolution and in environments that have been inaccessible until now. Sensor network research is rapidly emerging as an important interdisciplinary area—for example, the Center for Networked Embedded Sensing, a National Science Foundation Science and Technology Center, has been recently established. The Center is led by the University of California, Los Angeles, and includes the University of Southern California, Los Angeles; the California Institute of Technology, Pasadena; the University of California, Riverside; the California State University, Los Angeles; and the Jet Propulsion Laboratory, Pasadena.

The results of this burgeoning research activity on sensors and sensor networks will impact nanorobotics significantly, and will be useful in applications beyond security and defense. The fields of environmental monitoring and health care are expected to benefit greatly from new sensor technologies.

C. Actuators

1) Artificial Molecular Machines: There has been significant progress in the design and chemical synthesis of *molecular* machines in the last few years—see, for example, the surveys in [1] and [2]. These machines are either single molecules or supramolecular systems of interlocked molecules. In either case, they are atomically precise; that is, each atom is in a known and precisely established location with respect to the others. Power is supplied to these machines electrically, optically, or chemically by feeding them with some given compound. Chemical power tends to be inconvenient because it cannot be easily switched on or off—a machine will move until it runs out of fuel—and normally produces waste products that must be eliminated. Two of the most interesting molecular machines synthesized to date are light-driven small organic molecules: a linear shuttle [11], and the rotary motor shown in Fig. 1 [23]. Under irradiation with a suitable wavelength in the visible range one part of Feringa's molecule (the rotor, at the top of the figure) rotates continuously with respect to a fixed part (the stator, at the bottom of the figure) around the carbon–carbon double bond shown at the center of the figure. The rotation proceeds in four steps. First the light causes a cis-trans isomerization. This is a change of conformation (shape) of the molecule from a state in which two groups (of atoms) are on the same side of a bond (cis) to another in which the groups are on opposite sides of the bond (trans). The resulting conformation is unstable and spontaneously changes to a more energetically-favorable conformation, continuing the rotation. This is step 2. Steps 3 and 4 are similar to 1 and 2. In step 3 the light produces another cis-trans isomerization with an unstable result that spontaneously decays to the initial conformation, thus closing the cycle. Recent work by Feringa's group at the University of Groningen, The Netherlands, has yielded other motors whose rotors are substantially different from their stators and, therefore, should facilitate selective attachment to surfaces and other nanoobjects [24].

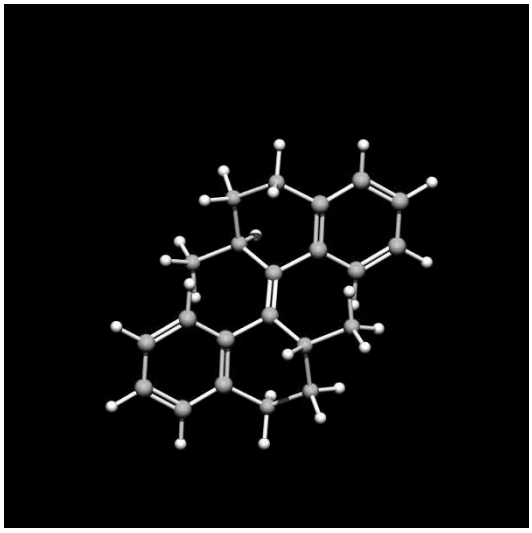

Fig. 1. The central part of a rotary nanomachine. (Figure courtesy of Prof. B. L. Feringa's group.)

The work on molecular machines is very interesting but in its current form has some drawbacks from the point of view of applications in nanosystems.

- The machines are synthesized and exist in solution. To be able to address each of them individually, it seems

necessary to attach them to a surface or perhaps to a three-dimensional (3-D) structure.
- Moving back and forth or rotating continuously without being attached to a load is not very useful. In general, the moving elements must be connected, or coupled, to other structures.
- The yield of an operation is usually much less than 100%. Thus, for example, if we apply radiation of the appropriate wavelength to a solution containing some of the light-driven molecular motors described in the literature, less than 50% may actually move. Design of mechanical systems with such a high tolerance for failure is very uncommon.
- Many of the molecules used in these machines are not rigid, whereas most of the design techniques for mechanisms at the macroscopic scale ignore flexibility.
- Chemical fueling is inconvenient and produces waste that must be removed.
- Light control may affect many machines because the wavelength of light is much larger than an individual machine. Electrical control typically requires wire connections. Approaches to building nanowaveguides [37] and nanowires [40], discussed later in this paper, may help here.
- The force/torque and energy characteristics of these machines have not been investigated in detail.

2) Biomotors: Another approach to mechanical nanosystem design involves harvesting (modified) biological motors. Biomotors tend to be on the range of 10 s of nanometers, and are typically larger than the synthetic molecular machines discussed above, which have overall sizes of only a few nanometers. Noji and coworkers were the first to directly image the motion of a biomotor [43]. They attached the F1-ATPase biomotor to a surface and also to a large actin filament that was visible in an optical microscope. Several laboratories are conducting interesting research on the applications of harvested biomotors to nanosystems—see, e.g., [18], [41].

Artificial motors built with biological materials have also been demonstrated. Typically they exploit certain properties of DNA. Seeman's group at New York University has reported an actuator that exploits a transition between two types of DNA [72], and a Bell Labs group has reported another actuator that exploits the tendency of short DNA segments to assume a rigid, linear conformation [62]. The Bell Labs machine is similar to a tweezer, which opens and closes when certain DNA strands are introduced in the solution.

Because biomotors have been successfully attached to surfaces and to loads, they are closer to applications than the synthetic molecular machines. But they are not without problems.

- They run on chemical fuel [usually adenosine tri-phosphate (ATP)], which has the drawbacks mentioned earlier.
- They are made of soft materials of limited durability.
- They operate in a narrow range of environmental conditions (e.g., temperature and pH).
- They are hard to control.
- They are very complex, and much is still unknown about their structure and operation.

3) Other Nanomachines: Larger, not atomically precise machines have also been demonstrated. Here the most interesting is perhaps a very recent nanotweezer development in Scandinavia [8]. This nanotweezer is based on a MEMS electrostatic motor with two cantilevers that bend under an applied voltage. Two very thin probes are grown on the tips of the cantilevers by deposition of carbonaceous material in a scanning electron microscope (SEM). Gaps between tips as low as 20 nm have been demonstrated. The nanotweezer could, in principle, be used to grasp nanoobjects and manipulate them in 3-D. This, however, has not yet been reported in the literature. An earlier nanotweezer built by gluing two carbon nanotubes to a probe was reported in [33] and demonstrated picking a 500-nm object. Strictly speaking, neither of these nanotweezers is a nanodevice, since they are microscopic devices with nanoscopic tips and auxiliary macroscopic components (much like an SPM).

D. Propulsion

Swimming or flying in fluids seems more attractive than walking or crawling on a surface, since most objects likely to be encountered on a surface are large and difficult to superate by a nanoscale walking or crawling machine. Bacteria are good models for nanorobots because they have sizes on the order of a few micrometers, which are likely to be comparable to those of future nanorobots, and move in fluids.

The characteristics of fluid motion are controlled by the Reynolds number, defined as $\mathrm{Re} = \rho.V.L/\eta$, where ρ is the specific mass, V a characteristic velocity, L a characteristic length, and η the viscosity. Plugging in typical values for a fish ($V = 1$ m/s, $L = 10$ cm) and for a bacterium ($V = 10$ μm/s, $L = 1$ μm) we find that the Reynolds number for a fish is on the order of 10^5, while for a bacterium it is 10^{-5}. This is a ten-order-of-magnitude difference, and has major consequences. Bacteria and nanorobots move in the Stokes (or low-Reynolds number) regime, which can be counterintuitive [7], [44]. For example, inertia is negligible and motion is controlled entirely by friction; coasting is impossible; propulsion cannot be achieved by symmetric motions; and jet propulsion does not work. Bacteria move in this regime typically by using cilia or rotating flagella.

Small objects in a fluid at room temperature are subject to thermal agitation and collisions. The result is a random walk, or *diffusion*. The distance L travelled by a set of diffusing objects in time t is given approximately by $L = (2\,D\,t)^{1/2}$, where D is the diffusion coefficient, which is approximately constant for a given type of objects in a given fluid and at a fixed temperature [7]. Distance is not proportional to time, but rather to its square root. For a small molecule in water at room temperature $L = 1$ μm is reached after a time $t = 0.5$ ms, whereas a distance of 1 cm corresponds to a $t = 14$ h. This shows that diffusion is fast for small distances and very slow for larger distances. In nature, objects with dimensions on the order of a few nanometers, such as the molecules used

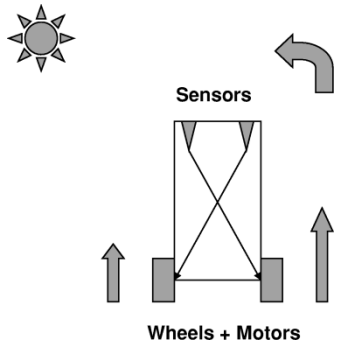

Fig. 2. Schematic of Braitenberg's Vehicle 2b.

for chemical signaling, are not self-propelled and rely on diffusion. In fact, it appears that there are no self-propelled organisms with sizes below 600 nm [21]. Attempting to propel and steer a smaller organism is ineffective because of the numerous collisions that will change its course unpredictably. Diffusion is then a better strategy. It follows that self-propelled nanorobots moving in a fluid should have dimensions on the order of a few micrometers. Luckily, this is precisely the size one would expect to achieve by assembling a relatively complex set of nanoscale components.

E. Control

Controllers for macroscopic robots are typically full-fledged computers. It is unlikely that the nanorobots of the near future will be able to carry inside of them the equivalent of a PC. But interesting behaviors are achievable with rather primitive control systems, which could probably be implemented at the nanoscale using emerging nanoelectronic technology. For example, Braitenberg's Vehicle 2b [10] is capable of steering toward a light source. It does this by using two sensors and two motors, which control the vehicle's wheels—see Fig. 2. The left sensor is connected to the right-wheel motor, and the right sensor to the left-wheel motor. When the left sensor sees a higher intensity of light, it tells the right motor to move faster, thus causing the vehicle to turn toward the light. The right sensor operates in a similar manner.

Bacteria provide another example of what can be done with a very simple control system. For example, *E. coli* move in a series of "runs" and "tumbles" [7]. A run is a motion in an approximate straight line. A tumble is a reorientation of the bacterium. An *E. coli* bacterium runs for a certain amount of time, then stops and tumbles, changing orientation to a *random* direction; it then runs again, and so on. *E. coli* manage to move toward higher concentrations of nutrients by using the following control scheme. The bacterium has chemical sensors for the nutrient, and takes several readings during a run. By comparing the sensed values it can determine whether the concentration is increasing or decreasing. If it is increasing, the bacterium will run a little longer than usual; if the concentration is decreasing, the bacterium will shorten the run and tumble sooner. Note that the tumble is always random, and the bacterium has no notion of where the nutrient is, or of which direction is best. All that it does is to bias its random walk, and this suffices to reach regions of high nutrient concentration. Randomness actually helps the bacterium move away from regions that become depleted, or from local minima of the concentration. The microorganism, in essence, executes a form of random search using only local information.

F. Communication

Communication among nanorobots by means of waves, be they acoustic, electrical, or optical, is likely to be difficult because of the small antenna sizes. If we look at what nature does, we find that bees communicate directly by dancing; ants communicate by releasing chemicals (pheromones) that change the environment (this is called *stigmergy* in the robotics field); and bacteria also release chemicals, for example, to assess the number of similar bacteria near them. This bacterial behavior is called *quorum sensing* and uses a very simple strategy. If each bacterium releases a fixed amount of a given chemical, it suffices to measure the concentration of the chemical to find how many bacteria are in a neighborhood. The vast majority of the communications between small objects such as cells and subcellular structures is done chemically, by using molecular recognition. As we noted above, in Section II-B, chemical signaling requires contact and poses interesting challenges for the design of robotic strategies.

G. Programming and Coordination

Each nanorobot by itself will have limited capabilities, but the coordinated effort of a multitude will produce the desired system-level results. Coordination is needed across the board—for communication, sensing, and acting—and poses a major research challenge. The scale and dynamics of nanorobotic systems precludes centralized coordination and global sharing of state. Therefore, we need coordination schemes that are inherently distributed and based on localized inputs, algorithms, and outputs.

In nature we find a range of approaches to the coordination of large numbers of cells or organisms. For example, bacteria show very limited coordination behavior; ants use elaborate algorithms [9]; and the human immune system has an extremely complex coordination and (chemical) signaling scheme, which is still far from being completely understood [16], [61]. The remarkable capabilities of the immune system appear to be linked to characteristics that are not normally found in human-designed systems.

- immune receptor degeneracy (any receptor binds more than one ligand and conversely);
- sensor degeneracy (a sensor responds to several stimuli, with different strengths and, therefore, several sensors respond to the same stimulus);
- pleiotropism (an agent causes multiple effects);
- effector redundancy (different agents have the same effect);
- context-dependent decisions/actions;
- random generation of new sensors/receptors.

Nature has produced biological systems that adapt and self-organize. How such concepts can be exploited in artificial systems is not yet clear. Programming nanorobotic systems is a research area with strong connections with several emerging fields of computer science and engineering: sensor/actuator networks (or physically coupled, scalable, information infrastructures), distributed robotics, and swarm intelligence.

III. Nanoassembly With the SPM

A. Background

The SPM was invented in the early 1980s by Binnig and Rohrer, of the IBM Zürich Laboratory, and earned them a Nobel Prize. SPMs opened a new window into the nanoworld and have been a major force driving the current development of nanoscience and engineering. Although SPMs are normally used for imaging, it was recognized soon after their invention that they can also modify the samples. Eigler's group at the IBM Almadén Laboratory demonstrated that the scanning tunneling microscope can be used to manipulate atoms [64]; a well-known example of their work is the IBM logo written with xenon atoms. Other pioneering research on atomic manipulation was done by Avouris' and Aono's groups [36], [68]. Atom manipulation is typically performed in ultrahigh vacuum and at low temperature ($\sim$4 K). More recently, interesting work on atomic manipulation has been done in Rieder's group at the University of Berlin—see, e.g., [5]. They have shown that it is possible to determine if atoms are being pushed or pulled on a surface by examining the signals acquired by the scanning tunneling microscope during the manipulation operation.

Building nanoobjects atom by atom in ultrahigh vacuum at 4 K is not very practical. An alternative approach, initiated by Samuelson's group at the University of Lund [32], starts with larger, molecular-sized building blocks and assembles them with an atomic force microscope (AFM) in ambient conditions. Our group at the University of Southern California's Laboratory for Molecular Robotics has been investigating this approach for several years. Work on AFM-based manipulation has also been reported by other groups [39], [60], [63], [65], [66].

An AFM is both a sensor and a manipulator, and we do not have an independent measurement of "ground truth" when we navigate the tip over the sample. Operating the AFM in the chamber of an SEM provides a separate sensing capability. Visual feedback from the SEM can be used for the manipulation, much like one normally does with optical microscopes at a larger scale [70]. Manipulation inside an SEM was pioneered by Sato's group [42], [59] for microscale objects, and has been used at the nanoscale by the Ruoff/Zyvex group [73] and Fukuda's [20]. SEM sensing is not appropriate for all samples, because it normally requires a vacuum environment and involves bombarding the sample with high energy electrons. SEMs also tend to have lower resolution and be more expensive than SPMs. In this paper we focus on AFM manipulation without SEM imaging.

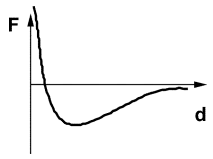

Fig. 3. Force between tip and sample as a function of their relative distance.

B. The AFM as a Robot

The AFM is a conceptually simple apparatus [48], [58]. A micrometer-scale cantilever with a sharp tip (diameter $\sim$10–50 nm) is scanned over a sample at distances on the order of a few nanometers. Interatomic forces between the tip and the sample are sensed by the cantilever, whose deflection is measured (usually) by a laser and a photodetector. (Piezoresistive cantilevers can also be used, and may be more amenable to on-board sensing for autonomous or semi-autonomous micro or nanorobots.) The force experienced by the tip varies nonlinearly with the tip–sample separation, as shown in Fig. 3. (In the figure positive forces are repulsive.)

In contact mode operation, the tip is in the repulsive region of the curve, and the force is kept constant during the scan by a feedback circuit that monitors the photodetector signal. A tip in contact mode exerts a relatively large normal force on the sample, and also a substantial lateral force. As a result, fragile samples are damaged, and tips tend to wear out rapidly. In addition, the deflection signal is low-pass and the process is subject to low-frequency noise.

The preferred mode of operation often is dynamic force microscopy, which uses a vibrating cantilever and avoids the force and noise problems of contact mode. There are two versions of dynamic force microscopy. In noncontact mode, the tip oscillates above the sample in the attractive force regime, whereas in intermittent contact mode, the tip contacts the sample for a short time interval ("taps") during each cycle of the oscillation.

The standard use of the AFM is as an imaging instrument. Constant force is maintained by using feedback and the tip is scanned in the x, y plane by using piezoelectric actuators. The vertical, or z, motion required to keep a constant force is the output signal, which approximates the topography of the sample $z(x, y)$. (This is a very simplified description; for more details, see, e.g., [52] and [58].)

Because of the many causes of error discussed below, the only truly reliable way of measuring x, y, z is by using feedback. This is the approach taken in machine tools and robots in the macroscopic world. Position feedback is used in some AFMs for large scans and features. For example, some commercial instruments offer scanners with a range of $\sim 100\ \mu$m and with feedback-controlled x, y positioning. Note, however, that a typical 256×256 pixel image with a scan size of 100 μm has a resolution or pixel size of $\sim$400 nm, which is quite large. For the work done in our lab, scan sizes are usually $< 1\ \mu$m and accuracies < 1 nm are required. Sensors and feedback circuits cannot normally offer such accuracies; for instance, a 2-nm root-mean-square noise level is typical in

commercial instruments. Hence, these instruments are operated open loop for small scan sizes and high resolution. The z axis is feedback controlled using the cantilever (plus the photodiode and associated optics) as a sensor and, therefore, the accuracy in z is much higher than in x, y. (New instruments just becoming available claim feedback circuitry with accuracies and noise levels < 1 nm; these SPMs will greatly facilitate nanomanipulation.)

There are many sources of spatial uncertainty in AFM measurements.

- *Tip Effects*—When the tip moves in contact with a sample, it traverses a contact manifold in what is called in robotics a *configuration space* [35]. Therefore, we obtain the image of the configuration space obstacle that corresponds to the sample rather than the image of the sample itself. This is sometimes called a "convolution" of the sample and tip and has an effect akin to low-pass filtering with an associated broadening of sample features. For a discussion of tip effects and their compensation, see, for example, [71].
- *Drift*—The major cause of spatial uncertainty in our lab is thermal drift between the tip and the sample. We work at room temperature, in ambient air, and without careful temperature and humidity control. A typical value for drift velocity is 0.05 nm/s. This implies that for an image with 256×256 pixels obtained in a 1-Hz scan, an object will drift by $\sim$12.5 nm per scan, which is approximately the size of the particles we usually image.
- *Creep*—A large voltage step will produce a rapid displacement of the tip followed by a slow creeping motion, which can last several minutes. Typically, creep values can reach 50 nm over a 1-min interval for a 1000-nm offset.
- *Hysteresis*—The extension of a piezo depends on the history of the voltages applied to it. For example, scanning right-to-left or left-to-right produces different results. The differences can be large. For example, for a 500-nm scan one can find a displacement of $\sim$15 nm.
- *Other Nonlinearities*—Even ignoring hysteresis, the piezo's response is not linear with the voltage. In addition, the tube scanners used in most AFMs move approximately in a circle and not in a straight line.

C. Manipulation Phenomena and Protocols

Nanoscale objects such as nanoparticles can be pushed mechanically by the tip of an AFM. There are several protocols for manipulation by pushing, all of which share the following aspects. First, image the sample to determine where the desired particle is. Then move against the particle, but change the operating parameters so that a force higher than that used for imaging is applied. In our lab we usually push by imaging in dynamic force mode and then moving with the feedback off along a straight line that goes through the center of the particle. Sometimes we also decrease the tip–sample separation by moving in z when we turn off the feedback.

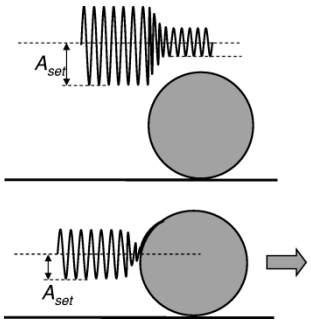

Fig. 4. Mechanically pushing a nanoparticle.

This pushing protocol is almost 100% successful when the tip is sharp and we hit the particle very close to the center. We use relatively stiff cantilevers (spring constants on the order of 10 N/m) and sharp tips (radii on the order of 10–20 nm), and operate in ambient air or in liquids, at room temperature, and without strict environmental controls.

We have studied carefully the phenomena involved in pushing in a series of papers [3], [4], [12], [45], [46], [53], [56]. We observe that when the tip is oscillating relatively far from the surface the amplitude decreases as the tip approaches the particle but the particle does not move (top part of Fig. 4). When the tip is sufficiently close to the surface, the vibration amplitude goes to zero as the particle is approached, the average (or dc) cantilever deflection becomes nonzero, and the particle moves, if the deflection is above a certain threshold dependent on the cantilever and various other characteristics of the setup (bottom part of the figure). The changes in vibration amplitude and cantilever dc deflection can be used to monitor the manipulation in real time, and verify with a high degree of confidence that it is successful, without further imaging.

Recently, we have discovered that it is also possible to push a particle in a purely lateral mode, without any vertical deflection of the cantilever. Qualitatively, this happens when the tip is very close to the surface when it approaches the particle, but we have no quantitative data for reliably predicting when manipulation will take place in this no-deflection mode.

Other interesting approaches to pushing are possible. For example, if we superimpose a tip vibration (dither) in the x, y plane to the trajectory of the tip as we approach a particle, the particle moves in the desired direction. The dither motion must be approximately normal to the undithered trajectory—see Figs. 5 and 6. More research is needed to determine under what conditions this approach is successful. Observe that the dithering motion simulates a straight-edge end effector, parallel to the x, y plane. This may decrease the trajectory accuracy required for successful pushing of particles, and may also facilitate the manipulation of more complex objects such as nanorods or nanotubes, which are difficult to move controllably with our usual protocols and tips [30]. Interestingly, it may open a new application area for the rich theory of object orientation with straight-line end effectors, which has been developed for the macrorobot world [27].

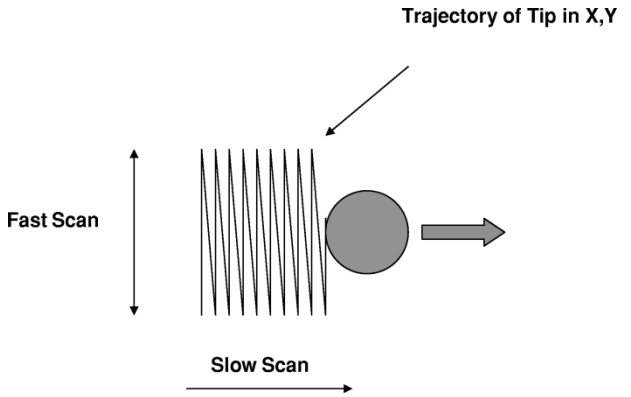

Fig. 5. Pushing with a simulated edge.

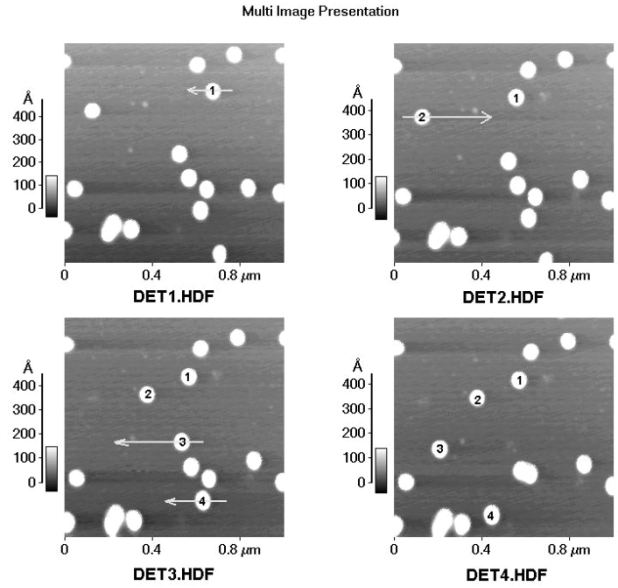

Fig. 6. Examples of successful nanomanipulation with dither.

D. Nanoparticle Patterns

Nanoparticle patterns are attractive nanostructures because 1) there are many known methods for synthesizing nanoparticles with a variety of characteristics (e.g., metallic, semiconducting, or magnetic) and the state of the art is steadily improving; 2) the particles have more uniform sizes (i.e., are more monodisperse) than structures of comparable sizes made by competing techniques such as electron-beam lithography; and 3) arbitrary planar patterns of nanoparticles can be built by nanomanipulation using the protocols discussed earlier.

Fig. 7 shows a sequence of manipulation steps in the construction of a pattern that encodes ASCII characters in horizontal rows of nanoparticles on a surface. The presence of a particle at a node of a regular two–dimensional (2-D) grid is interpreted as a one and its absence as a zero. The pattern, read from the top to the bottom encodes "LMR." The particles have diameters of 15 nm, and the grid nodes are spaced with a 100-nm pitch. The areal density is on the order of 60 Gb/in^2, and it should be possible to increase this density by over an order of magnitude by using smaller particles and tighter spacing. This would give densities approaching

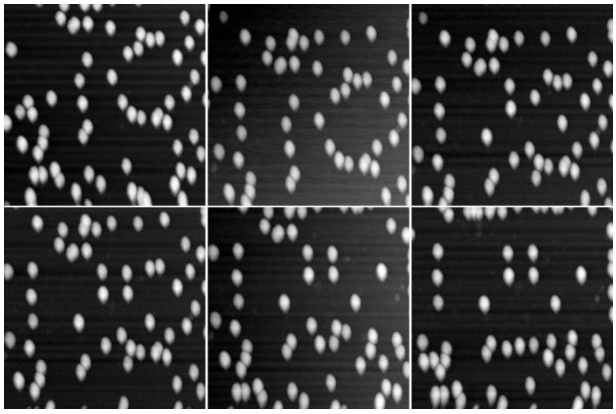

Fig. 7. Steps in the construction of the LMR pattern by nanomanipulation.

the Tb/in^2. This digital storage technique is a candidate for an editable nano compact disk. However, there are two main hurdles that must be overcome for it to be practical. The first is the need for high speed in reading and writing. Speed can be increased dramatically by using multitip arrays [47]. The second is more pernicious: for swift reading and writing, the particles must be positioned originally at the vertices of a grid. Changing a random configuration of particles (as deposited) into a regular grid configuration by nanomanipulation is a very time-consuming process. What is needed is a self-assembly technique that automatically places the particles at every grid node with a pitch sufficiently large to permit easy manipulation of the deposited particles. Thus far, no self-assembly technique with these properties has been discovered.

Manipulation of nanoparticles can also be used to build prototypes of electronic and optoelectronic devices. In fact, many of the nanoelectronic devices built until now have either relied on chance to place an element in the desired relationship with others, or have used SPM manipulation. For example, placing a nanoparticle at tunneling distances between two electrodes (the source and the drain) can be used to make a prototype for a single-electron transistor. Fig. 8 illustrates the process. The left panel of the figure shows three gold electrodes that correspond to the source, drain, and gate of an FET. The electrodes have widths $\sim$100 nm and were built by electron-beam lithography by Dr. P. Echternach at the Jet Propulsion Laboratory in a collaboration with our lab. The center panel of the figure is a zoomed-in view of the source-drain gap, and identifies two gold nanoparticles with diameters of $\sim$15 nm (near the bottom of the figure). The right panel shows the result of pushing the marked particles into the gap.

A prototype for an optoelectronic device—a "plasmonic" waveguide—was built in a collaboration between our lab and Prof. H. Atwater's group at Caltech. The initial idea was as follows. First, place (by nanomanipulation) colloidal gold nanoparticles with diameters on the order of 30 nm at equal distances from each other in a chain, plus a fluorescent latex particle at the end of the chain [37]. Then, inject energy at a wavelength in the visible range into the gold particle at one end of the chain, and demonstrate that the

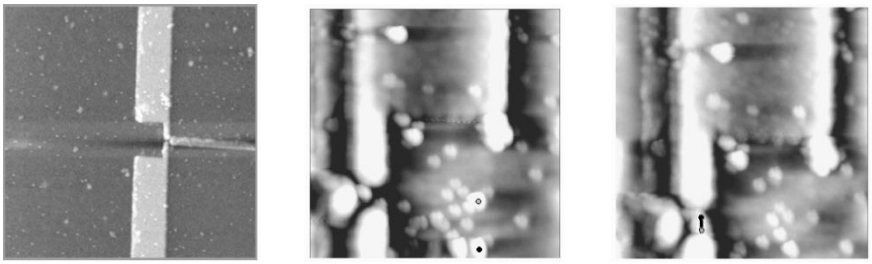

Fig. 8. Building a single-electron transistor by nanomanipulation.

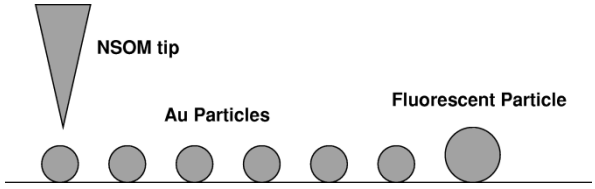

Fig. 9. Schematic of a plasmonic waveguide.

energy propagates through the chain by observing the fluorescence of the latex ball (Fig. 9). Theoretical computations and experimental studies at larger scales by the Caltech group showed that this device should function as a waveguide, exploiting near-field effects that involve the plasmon resonance of the particles. Very recently, the feasibility of the nanowaveguide concept was demonstrated experimentally, but using silver nanorods instead of gold nanoparticles [38]. This nanowaveguide is interesting because it has transverse dimensions much smaller than the diffraction limit for the wavelengths in the hundreds of nanometers that are being used. It may serve in the future to feed light to *individual* molecular machines without exciting other machines in the same neighborhood.

E. Linking and Embedding

Patterns of unlinked nanoparticles can be useful, as we just saw in the previous section. However, many applications require "solid" nanostructures of specific shapes. These can be approximated by groups of suitably positioned and *linked* nanoparticles [49], [50]. We have investigated several approaches to linking. The first uses covalent (i.e., chemical) bonding to a linker [54], [55]. For example, gold particles can be connected with di-thiols. (Di-thiols are organic molecules with sulfur end groups.) The di-thiols self-assemble to the gold and serve as chemical glue. We have demonstrated two variants of this approach: 1) first deposit the particles, position them, and then immerse the sample in the di-thiol solution to link them or 2) deposit the particles, apply the thiols, and then manipulate the particles into contact, thus linking them. We also have shown that it is possible to push a group of nanoparticles linked by di-thiols as a whole [54], [55]. These results demonstrate hierarchical assembly at the nanoscale, i.e., the construction of assemblies of components, which are themselves (sub-) assemblies of other components or of primitive building blocks.

The second approach to linking also uses selective self-assembly. Additional material is deposited on the particles until

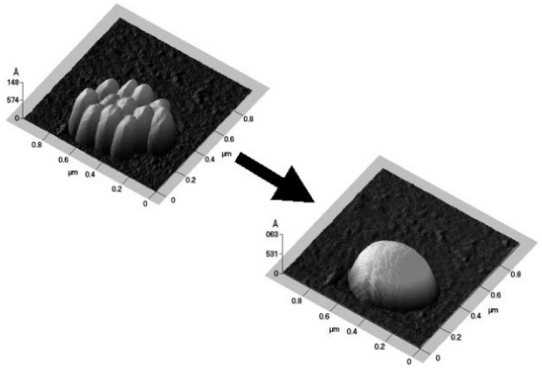

Fig. 10. Connecting latex nanoparticles by sintering.

they become connected. The material and experimental conditions must be selected to ensure that the material assembles to the particles but not to the remainder of the sample. For example, we have shown that a pattern of gold nanoparticles can be used as a template for the electroless deposition of additional gold. Gold wires of arbitrary geometry can be built by first manipulating the particles into the desired geometry and then linking them by immersion of the sample in the electroless solution with a specific set of parameters such as immersion time, concentration, and so on [40].

A third approach discovered very recently uses sintering to connect fluorescent latex nanoparticles. The particles are first manipulated to form a desired template. The template is then heated and the particles melt together into a single nanostructure [28]. Fig. 10 shows on the left a pattern of 12 latex nanoparticles with diameters $\sim$100 nm placed by nanomanipulation so as to form a disk. On the right is the 3-D nanoscale solid that results from sintering the nanoparticle pattern.

For certain applications, we may need to ensure that nanocomponents are fixed on the substrate. This can be done by selective self-assembly. We need a material that will assemble to the substrate, but not the particles, and, thus, will embed the particles in a thin layer. We have demonstrated particle embedding in a silicon oxide layer by first depositing particles and manipulating them, then depositing a monolayer of a silane (which attaches only to the substrate), and finally oxidizing the silane layer with ultraviolet light [57]. (Silanes are organic molecules containing silicon atoms.)

We have used embedding of particles in successive layers for a proposed new rapid prototyping technique at the nanoscale, called layered nanofabrication [51]. We build successive layers of a 3-D object by nanoparticle manipulation, and planarize each layer by adding a molecular sacrificial layer whose top surface serves as support for the next processing step. The sacrificial layers are removed in a final step. Thus far, we have demonstrated that it is possible to build sacrificial layers and to manipulate gold nanoparticles on top of them.

IV. Summary and Outlook

Nanorobotics is an emerging and highly interdisciplinary field that involves computer science, chemistry, physics, biology, and other disciplines. Very few people (if any) master all of these disciplines; therefore, teamwork and collaboration between experts in different fields are essential for progress in nanorobotics.

Construction of nanorobots and NEMS is still in its infancy. However, progress in exploiting biological motors and in developing artificial nanomachines has been rapid over the last few years, and the first (and fairly primitive) nanorobots are likely to emerge from research labs within the next five to ten years. Building and testing of nanodevices, and coupling of nanodevices to build integrated systems that can be interfaced with the micro/macro world, continue to be major challenges.

AFMs provide effective means for fabricating nanodevice and nanosystem prototypes and products in small quantities. They interface the nanometer world of the tip with the micrometer scale of the cantilever and the centimeter scale of the instrument. An AFM is both a sensor and an actuator. The tip is akin to a mobile robot, which must map the sample, navigate over it, and modify it.

AFM manipulation can be used to accurately and reliably position molecular-sized components. Unlike its macroscopic counterparts, which are primarily governed by classical mechanics, nanomanipulation phenomena fall mostly in the realm of chemistry. Linking and assembling of nanoscale objects can be done by chemical and physical means, by using techniques such as "gluing" with suitable compounds, chemical deposition, or simply heating.

Demonstrations that may lead to useful applications of nanoassembly are beginning to appear. However, increased levels of automation in nanomanipulation are needed to prototype more complex and useful devices and systems. Pick-and-place operations and the construction of 3-D nanostructures are still very primitive and need further development. Finally, mass production methods (which are likely to be based on "programmed" self-assembly rather than nanomanipulation) and NEMS applications are still at the embryonic stages.

Acknowledgment

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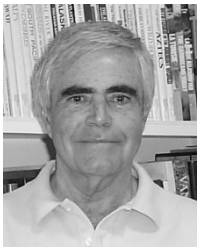

Aristides A. G. Requicha (Fellow, IEEE) received the Engenheiro Electrotécnico degree from the Instituto Superior Técnico, Lisbon, Portugal, in 1962 and the Ph.D. degree in electrical engineering from the University of Rochester, Rochester, NY, in 1970.

He is currently the Gordon Marshall Professor of Computer Science and Electrical Engineering at the University of Southern California, Los Angeles, where he also directs the Laboratory for Molecular Robotics. He has authored some 150 scientific papers, and has served on numerous journal editorial boards. His past research focused on geometric modeling of 3-D solid objects and spatial reasoning for intelligent engineering systems. Currently he is working on robotic manipulation of nanometer-scale objects using scanning probe microscopes, and on its applications in nanoelectronics, nanoelectromechanical systems, and biomedical nanotechnology. The long-term goals are to build, program, and deploy nanorobots and sensor–actuator networks for applications such as environmental monitoring and health care.

Dr. Requicha is currently a Member of the IEEE Nanotechnology Council AdCom, representing the Robotics and Automation Society and serving on the publications committee. He has served on numerous conference program committees. He was a college and high school Valedictorian.