

Solid State Communications 113 (2000) 549-552

solid state communications

www.elsevier.com/locate/ssc

Random access of nanodevices

M.L. Cohen^{a,b}, S.G. Louie^{a,b}, A. Zettl^{a,b,*}

^aDepartment of Physics, University of California, Berkeley, CA 94720, USA ^bMaterials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

Received 2 November 1999; accepted 24 November 1999 by M. Cardona

Abstract

Systems containing ultrahigh densities of patterned nanometer scale devices are anticipated to be extremely difficult, if not impossible, to fabricate and access in an error-free fashion using existing technologies. In this work, we show that extremely high densities of on-tube and intertube nonlinear electronic devices are realizable in samples of randomly grown carbon nanotubes. Further we propose and demonstrate a strategy for accessing and exploiting the random collection of nanodevices in such systems. © 2000 Published by Elsevier Science Ltd. All rights reserved.

Keywords: A. Heterojunctions; A. Nanostructures; B. Nanofabrication; D. Electronic transport

With the continuing miniaturization of electronic device elements, the fabrication, access, and operation of highly packed submicron devices become increasingly difficult. Projecting present technologies to nanoscale dimensions reveals major technical and conceptual problems regarding issues such as manufacturing reliability, interconnect accessibility, power dissipation, etc. An alternative to the integrated microelectronics route in addressing these issues may be one of employing self-assembled nanoscale devices, synthesized in various physical and chemical processes. Although for certain applications it may be advantageous to pre-ordain the type and connectivity of the nanoscale devices, we here consider the situation where the assembly is completely random. While the manufacturing advantages of a random nanodevice network are obvious, less clear is the practical ability to access and make such a complex system functionally useful.

In this work we consider in a general way the random access of nanodevices, where the composite nanodevice system comprises a high-density, highly interconnected matrix of nanodevices with unknown internal structure. As an example, we explore a random collection of high-device density objects such as defected carbon nanotubes [1-10]. We find that in principle random connections made to such a matrix will access useful devices. Experimental demon-

stration of this principle is accomplished using an actual random network of carbon nanotubes rigidly bound in a solid matrix with 20 input/output terminals. Random access of the nanodevice structure yields, among other things, transistor action.

To explore the feasibility of random access of nanodevices (RAND), we estimate the accessibility of active elements in a nanodevice-rich interconnected matrix. The number of active elements which can be accessed is basically limited by two physical lengths—the diameter d of the wires used as contacts and the intrinsic connectivity length ξ among the internal device-containing elements (as we suggest later, these elements might be defected nanotubes). To illustrate this, let us consider a spherical matrix of diameter D with multiple leads inserted into it as shown schematically in Fig. 1. Theoretically, the number of wires which may be inserted at different depths near the surface into the matrix will be of the order of $N_{\rm w} \approx (D/d)^2$. However, in order for two wires to form a terminal pair, they have to be within a connectivity length between nanodevice elements. The number of effective wires within a connectivity area will be roughly given by $N_{\rm eff} \approx$ $N_{\rm w}(\xi/D)^2 \approx (\xi/d)^2$. The number of possible two terminal pairs would be $N_{\text{pair}} \approx N_{\text{w}} \times N_{\text{eff}} \approx (D/d)^2 (\xi/d)^2$. For illustration purpose, we consider the number of possible three-terminal active elements which may be sampled, for instance accessing the nonlinear current response of a pair of terminals by modulating the voltage on a third terminal. The number of such possibilities N_3 is of order

 ^{*} Corresponding author. Tel.: +1-510-642-4939; fax: +1-510-643-8497.
E-mail address: azettl@physics.berkeley.edu (A. Zettl).

^{0038-1098/00/\$ -} see front matter © 2000 Published by Elsevier Science Ltd. All rights reserved. PII: S0038-1098(99)00543-8



Fig. 1. Schematic diagram of a spherically shaped matrix of nanodevice-containing elements with multiple access wires inserted. Drawing is not to scale.

 $N_3 \approx N_{\text{pair}} \times N_{\text{eff}} \approx (D/d)^2 (\xi/d)^4$. Hence, the RAND can be very large. For example, if we assume that D = 0.01 m, $d = 10^{-6} \text{ m}$, and $\xi = 10^{-4} \text{ m}$, then N_3 is near 10^{16} . For the general case of *m*-terminal active elements, the number of possibilities is given by $N_m \approx (D/d)^2 (\xi/d)^{2m-2}$. Given such a large number of possibilities, only a very small fraction of the available active elements needs to be of a desirable type. One possible strategy is to probe the nanodevice matrix and categorize a suitable number of active elements which might be designated for a specific use. For example, if a matrix of this kind containing an ultrahigh density of active elements is to be used in part for analog or digital computation, an *m*-terminal "functionality map" could be constructed, with each map tailored for a specific application and nanodevice matrix combination.

We now examine how the RAND method can be applied to a concrete physical nanodevice system: carbon nanotubes. Because of their nanometer dimensions, there are many interesting and often unexpected properties associated with nanotubes. For example, a local topological defect can change the structural and electronic properties of a nanotube even at infinite distance away from the defect. It has been shown that different carbon half-tubes may be joined with 5-member ring/7-member ring (pentagon-heptagon) pair defects to form junctions [4-10]. (A pentagon-heptagon pair is the smallest topological defect with minimal local curvature and zero net curvature which can be introduced into the hexagonal carbon network.) Such a carbon nanotube junction allows the formation of nanodevices [4]. Under appropriate conditions the electronic structure of these junctions can be very similar to that of standard devices, and



Fig. 2. Electron microscopy image of a random tangle of as-grown carbon nanotubes. Numerous random tube interconnects can be seen. (a) Corresponds to a pristine sample, while (b) corresponds to a sample that has undergone post-growth oxygen processing which alters the tube connectivity.

hence, they are atomic or molecular level devices composed of the single element carbon. Recently, experimental observations of nonlinear on-tube carbon nanotube devices, including rectifiers [11] and transistors [12], have been made.

The fact that some carbon nanotube junctions and other structures can exhibit highly nonlinear responses gives them great potential for nanoscale device applications. However, even though in principle one can make many device elements with "defected" nanotubes, it is not possible yet to pattern them in a controlled manner in synthesis. This leads to the proposal here of directly using the primordial structures (and hence the active device elements) in an as-grown material. That is, an as-grown sample of entangled tubes (see Fig. 2) will inevitably contain many randomly arranged device elements, perhaps with a "device density" orders of magnitude higher than that obtained from state-of-the-art Si technology. Because of the nanometer dimensions of the elements, such a system is expected to have an exceptionally high speed. Equally important, the high thermal conductivity [13] of the carbon





Fig. 3. Experimentally obtained *I*–*V* characteristics of carbon nanotube matrix system, measured between pins 16 and 18 (see inset for schematic representation of lead configuration to the cube-like matrix; the nanotubes themselves are not shown in this drawing). The solid line is the "pure" behavior, while the long-dashed-line was obtained by "gating" the device by applying a control voltage to pin 5 (for the data shown the gating current applied between pins 5 and 18 was 25 μ A, although substantially smaller gating currents still had a noticeable effect on the *I*–*V* characteristics between pins 16 and 18). The short-dashed line is an extrapolation of the low-field behavior of the ungated device and serves to quantify the deviation from "linear response".

network will give such a system outstanding thermal dissipation characteristics.

To demonstrate the feasibility of physically realizing a nanotube-based RAND system with potentially useful characteristics, a simple randomly connected carbon nanotube matrix device was constructed and tested electronically. Carbon nanotubes were synthesized using both arcplasma discharge [14,15] and laser vaporization methods [16]. For the matrix device to be described later, "ropes" of single wall carbon nanotubes were utilized. The tubes were first purified by heating to 1000°C in vacuum for 30 min to remove amorphous carbon contaminants. Randomly oriented tangles of the tube ropes were then dispersed in a non-conducting epoxy resin. While the epoxy matrix was still soft, 20 electrical leads, each consisting of 0.001 in. diameter gold wire, were inserted into the matrix in a somewhat haphazard way (one constraint obviously being that the leads not directly touch each other within the matrix). After the epoxy had cured the leads were wired to a 20-pin test fixture and the nanotube matrix was evaluated electrically at room temperature.

The inset to Fig. 3 shows a schematic drawing of the RAND nanotube matrix device with protruding access wires. The electrical response of this 20-terminal device was found to be very complex. For initial characterization, the dc current (I)-voltage (V) characteristics between various terminal pairs were measured. As expected for a random matrix with high intrinsic device density, a rich response spectrum was observed. In general, no two

terminal pairs showed identical response. The main body of Fig. 3 shows one example of the I-V response curve, in this case between terminals 16 and 18 (solid line). The response is clearly nonlinear, even for relatively low applied bias voltages (the short-dashed line shows a linear extrapolation of the low-bias behavior). Apparently, with terminals 16 and 18 we are accessing some specific (but possibly quite complicated) nanotube nonlinearity within the matrix.

The nanotube matrix of Fig. 3 was found to be "interactive". That is, in addition to interesting and potentially useful two-terminal response characteristics, three-terminal and higher-order terminal devices are found. For example, the long-dashed line in Fig. 3 again shows the I-V characteristics of terminals 16 and 18, but with a "control" signal (gate voltage) applied to terminal 5 (referenced to terminal 18). The I-V characteristics of terminals 16 and 18 are markedly altered with the control signal on; in fact, the high-bias resistance between terminals 16 and 18 changes (decreases) by 16%. In analogy with more conventional three-terminal active semiconductor devices, terminals 5, 16, and 18 behave functionally something like a transistor. The current gain (β) of the "transistor" of terminals 5, 16, and 18 is $\beta = 0.014$. Although this gain figure is significantly less than 1, it does not imply that local nanotube "devices" cannot or do not have true gain figures that are much larger. Here the control current injected into terminal 5 explores large portions of the matrix and thus serves as the control signal for numerous multi-terminal devices, accessed via other terminals. The matrix system is highly interconnected; therein lies both its complexity and potentially far-reaching functionality. Interestingly, applying a control voltage to terminal 6 had little effect on the terminal 16 and 18 I-V characteristics, as when applying a control voltage to terminal 17. In this RAND system with a highly complex set of randomly interconnected nanotubes, close physical proximity of device terminals does not guarantee useful electrical interaction.

By its very nature a RAND system, composed of highdensity nanotubes or otherwise, does not have a predictable nor exactly reproducible architecture. The characteristics differ greatly from one RAND device to another. Obviously, it would take an impractically long time to map out all interesting and potentially useful terminal connection combinations of the nanotube RAND system using manual methods as were used to obtain the data of Fig. 3. Each independently constructed nanotube RAND matrix is expected to have a completely different internal "wiring diagram". For many foreseeable applications each matrix would presumably require some initial characterization. One characterization possibility is an electronic *n*-terminal switcher with conventional hardware but utilizing appropriate optimized characterization software. To be useful, RAND necessitates novel device characterization and screening algorithms, and novel implementation hardware and software matched to the extreme device variability.

Although the system examined here experimentally is a

matrix of carbon nanotubes, other material systems also have excellent potential for RAND application. Examples include other conducting wires or molecules in contact with nanoswitches or nanocrystals, BN, BC₂N, and BC₃ nanotubes [17–27] (which in principle can be doped to achieve the desired properties, or combinations of $B_xC_yN_z$ tubes with carbon nanotubes to achieve desirable connectivity). The matrix systems can also be tuned (even after final assembly) through electrical or optical pulses to change their characteristics.

In summary, RAND sidesteps many of the usual difficulties in using nanodevices and changes the emphasis from hardware to software development. Once a complex system such as a RAND computer is operating, a natural "spare time" task may be for it to continually map its own patterns of devices and hence with time become increasingly efficient.

Acknowledgements

We thank S. Chase and P. Collins for technical assistance in the construction and testing of the RAND nanotube matrix device, and U. Varadarajan for synthesizing the nanotube materials used in this study. R. Smalley kindly provided additional nanotube samples. This research was supported by National Science Foundation Grant nos. DMR-9520554, DMR-9501156, and DMR-9801738, and by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the US Department of Energy under Contract no. DE-AC03-76SF00098.

References

- [1] S. Iijima, Nature 354 (1991) 56.
- [2] P.M. Ajayan, T.W. Ebbesen, Rep. Prog. Phys. 60 (1997) 1025.
- [3] M.S. Dresselhaus, G. Dresselhaus, P.C. Eklund, Science of Fullerenes and Carbon Nanotubes, Academic Press, New York, 1996.
- [4] L. Chico, V.H. Crespi, L.X. Benedict, S.G. Louie, M.L. Cohen, Phys. Rev. Lett. 76 (1996) 971.
- [5] L. Chico, L.X. Benedict, S.G. Louie, M.L. Cohen, Phys. Rev. B 54 (1996) 2600.

- [6] B.I. Dunlap, Phys. Rev. B 49 (1994) 5643.
- [7] R. Saito, G. Dresselhaus, M.S. Dresselhaus, Phys. Rev. B 53 (1996) 2044.
- [8] J.C. Charlier, T.W. Ebbesen, Ph. Lambin, Phys. Rev. B 53 (1996) 11108.
- [9] T.W. Ebbesen, T. Takada, Carbon 33 (1995) 973.
- [10] P. Lambin, J.P. Vigneron, A. Fonseca, J.B. Nagy, A.A. Lucas, Synth. Met. 77 (1996) 249.
- [11] P.G. Collins, A. Zettl, H. Bando, A. Thess, R.E. Smalley, Science 278 (1997) 100.
- [12] S. Tans, A.R.M. Verschueren, C. Dekker, Nature 393 (1998) 49.
- [13] J. Hone, M. Whitney, C. Piskoti, A. Zettl, Phys. Rev. B 59 (1999) 2514.
- [14] D.S. Bethune, C.H. Kiang, M.S. de Vries, G. Gorman, R. Savoy, J. Vasquez, R. Beyers, Nature 363 (1993) 605.
- [15] C. Journet, W.K. Maser, P. Bernier, A. Loiseau, M. Lamy de la Chapelle, S. Lefrant, P. Deniard, R. Lee, J.E. Fischer, Nature 388 (1997) 756.
- [16] A. Thess, R. Lee, P. Nikolaev, H. Dai, P. Petit, J. Robert, X. Chungui, Y.H. Lee, S.F. Kim, A.G. Rinzler, D.T. Colbert, G.E. Scuseria, D. Tomenek, J.E. Fischer, R.E. Smalley, Science 273 (1996) 483.
- [17] A. Rubio, J.L. Corkill, M.L. Cohen, Phys. Rev. B 49 (1994) 5081.
- [18] X. Blase, A. Rubio, S.G. Louie, M.L. Cohen, Euro. Phys. Lett. 28 (1994) 335.
- [19] Y. Miyamoto, A. Rubio, S.G. Louie, M.L. Cohen, Phys. Rev. B 50 (1994) 18360.
- [20] Y. Miyamoto, A. Rubio, S.G. Louie, M.L. Cohen, Phys. Rev. B 50 (1994) 4976.
- [21] X. Blase, A. Rubio, S.G. Louie, M.L. Cohen, Phys. Rev. B 51 (1995) 6868.
- [22] Z. Weng-Sieh, K. Cherrey, N.G. Chopra, X. Blase, Y. Miyamoto, A. Rubio, M.L. Cohen, S.G. Louie, A. Zettl, R. Gronsky, Phys. Rev. B 51 (1995) 11229.
- [23] O. Stephan, P.M. Ajayan, C. Colliex, Ph. Redlich, J.M. Lambert, P. Berniew, P. Lefin, Science 266 (1994) 1683.
- [24] N.G. Chopra, J. Luyken, K. Cherrey, V.H. Crespi, M.L. Cohen, S.G. Louie, A. Zettl, Science 269 (1995) 966.
- [25] P. Gleize, S. Herreyre, P. Gadelle, M. Mermous, J. Mater. Sci. Lett. 13 (1994) 1413.
- [26] A. Loiseau, F. Williame, N. Demoncy, G. Hug, H. Pascard, Phys. Rev. Lett. 76 (1996) 4737.
- [27] K. Suenaga, C. Carbon, N. Demoncy, A. Loiseau, H. Pascard, F. Williame, Science 278 (1997) 653.