By Robert Stadler

MOLECULAR, CHEMICAL, AND ORGANIC COMPUTING

Evaluating computation at the level of individual molecules.

In 1974, Aviram and Ratner introduced a concept for a molecular rectifier based on the use of a single organic molecule [1]. Approximately 30 years later, molecular electronics has become a very active field of research [5]. When discussing the application of organic molecules or materials for information processing or storage, three completely different fields must be distinguished, where each has its own paradigms and boundary conditions: chemical computing (CC), single organic molecular computing (SOMC), and organic electronics (OE). In CC, the photoabsorption or emission spectra in (bio)chemical reactions are reinterpreted as logic gates, which is interesting on a basic research level but unlikely to result in competitive electronic devices. SOMC is nanoscale computing with single organic molecules, which is also at the stage of basic research (in surface physics and chemistry) but envisioned to have a significant

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practical impact when it is fully developed. OE is based on bulk materials such as polymers and already commercialized for light-emitting diodes and display technology. Recent efforts have also been made in OE to develop polymer-based transistors [4]. This article briefly summarizes the progress in CC but primarily focuses on the current stage of development of SOMC.

CC: Chemical computing. An intermediate step for exploiting the variety of organic molecules and the conceptual analogy between chemical and biochemical reactions with information processing is realized in the growing field of chemical computing [2, 7]. In CC, both input and output can be defined to be either optical, electrical, or chemical and especially fluoerescence phenomena have been found to be suitable for being interpreted as rather complex algebraic operations, when the input/output pattern is translated into the truth table of a logic circuit [6]. The experimental implementations, however, have so far all been carried out on a macroscopic scale and in many cases the question needs to be further addressed whether these systems can be scaled down to the level of single molecules rather than an ensemble of molecules in a liquid.

SOMC: Electron transport through single organic molecules. The characterization of electron transport through single molecules is demanding, both theoretically and experimentally. For the theoretical description quantum mechanical equations must be solved numerically for a non-equlibrium system with open boundaries provided by long electrodes that maintain different chemical potentials due to an external bias. Within the framework of scattering theory, several approximations to this problem have been developed, where increased accuracy comes at the cost of increased computational expense. For experimental measurements the problem lies in the controlled and reproducible fabrication of nanojunctions, where two (or ideally three) metallic electrodes are positioned so close to each other that a molecule can be placed in the gap between them and connected to both. For this task scanning probe techniques are used, where a simultaneous imaging of details of the molecule-electrode contacts and measurement of the current/voltage (I/V) curves can rarely be achieved.

SOMC: Possible applications of single organic molecules as various components in electrical circuits. There have been several proposals for wires, diodes, transistors, memory cells, or logic gates based on the conductance properties of small single organic molecules. None of them has reached a stage of development where small circuits could be designed in theory and reproducibly demonstrated in experiment. In some of the theoretical work it is not clear whether the predicted device behavior is an artifact of the level of approximation chosen for the calculations. On the other hand, there is some controversy about theoretical explanations for device effects found in experiments, where negative differential resistance found in molecular wires has been explained by such diverse concepts as charging of the molecule and thermally activated rotation of functional groups. On the molecular scale any device effect is not a property of a particular molecule but must be attributed to the electrode/molecule/electrode nanojunction, where details of the interface between the molecules and the electrodes can qualitatively change the characteristics of the system. Since atomistic details of this contact can rarely be observed and much less so controlled in conductance experiments, the big variety of theoretical explanations for the same measurement is hardly surprising.

It is rarely addressed in the literature that for molecular versions of circuit elements to be useful, there must be the possibility to connect them together in a way in which their electrical characteristics—measured individually between electrodes—would be preserved in the assembled circuit. However, it has been shown that such a downscaling of electrical circuits within classical network theory cannot be realized due to quantum effects, which introduce additional terms into Kirchhoff's laws and let the classical concept of circuit design collapse [5].

Recently, schemes for computational architectures have been proposed that make use of the quantum properties on the molecular scale rather than

try to avoid them. Two examples for this are controlling the interference pattern of electron transport through aromatic molecules by modifying their chemical side groups [8], and cascading carbon monoxide (CO) molecules [3]. In the latter, CO molecules are arranged two-dimensionally on a copper surface with the tip of a scanning tunnelling microscope (STM), where the motion of one molecule causes the subsequent motion of another, and so on in a cascade of motion similar to a row of toppling dominoes. This movement is, however, dominated by quantum effects such as tunnelling from excited vibrational states. The arrival or absence of molecules at certain positions on the surface can represent a binary 1 or 0, respectively. The inputs for such a computation are provided by triggering the molecular movement at other positions through pushing them with a STM tip. Cascading logic gates such as AND and OR and their connectors have been experimentally realized based on this arrangement. C

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