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A prospect for single molecule information processing devices*

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Abstract: Due to the foreseen limitations of the present information technologies, such as semiconductor technology as well as magnetic disk technology, next generation technology is desperately needed. ‘Single molecule devices’ have been proposed as candidates to supersede the present devices for more than quarter of a century, but have not been made practical yet. The main reasons are that it is very difficult to make an access to a single molecule, and that the electron states of the molecule are very complicated when connected to the electrode. The progress in scanning probe microscope technologies and simulation technologies have made it almost possible to handle a single molecule and to foresee characteristics. The aim of this paper is to review a prospect for single molecule devices for the future information technologies, placing emphasis on the possible advanced switching devices. Four major milestones towards the final goal are proposed, and the current status of the first milestone is summarized, which includes theoretical treatment, molecular synthesis and measuring technology developments. New architectures might also be necessary which are suitable for molecular information processing systems.

INTRODUCTION

Human beings have made an enormous progress in information technology since the early civilization, which began several thousand years ago. However, information processing, transmission and storage were almost all made on paper, since its invention about 2000 years ago until about 50 years ago, with the only exception of wireless communication using ‘noroshi’ or smoke, and calculation that use ‘soroban’ or abacus. It was only in the latter half of the 20th century, when electronic information processing technology, including computation, transmission and storage, started to emerge and to dominate the world, and finally completely changed the human life.

With regard to information processing devices, dozens of ‘semiconductor devices’ have been proposed since the middle of the 20th century [1], however, only metal-oxide-semiconductor (MOS) transistors survived the harsh competition with very few exception of bipolar transistors. Now, even the MOS transistors are facing very fundamental physical and chemical limitations [2,3], and again, dozens of ‘new concept devices’ have been proposed as candidates to supersede MOS transistors. However, the total performances of those devices are not necessarily superior to those of MOS transistors. Therefore MOS transistors are believed to survive until their ultimate limitations of several tens of nonometers in gate length, which might only be another 10 more years through the blind lane [4].

‘Single molecule devices’ have been proposed for more than 25 years [5,6], and they have been claimed to make it possible to realize very advanced devices and system characteristics [7,8]. However, the interest faded away very rapidly and they have not been made practical yet. The main reasons for this

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are, (i) there had been no means to have an access to single molecule to ascertain the superior characteristics, (ii) the electronic states of the molecule are very complicated to foresee when connected to the electrode and (iii) the possible switching speed of those 'molecular devices' is no faster than that of the existing MOS devices. Recent progress of scanning probe technology [9] as well as computer simulation technology [10] have made accessing technologies to molecules almost mature and nearly conquering the obstacles against the 'single molecule device' concept. A possible change in the computer architecture is also anticipated, and probable substitute for the present von Neumann architecture is proposed, such as quantum cellular automaton and quantum computing [11]. Therefore, advanced single molecule devices with very attractive characteristics would accelerate the research towards the realization of molecular information processing system.

The aim of this paper is to review the present status of the research towards the use of single molecule for advanced information processing systems, and to present the possible candidates of the information processing single molecule devices, which would supersede MOS devices beyond their limitations [12,13]. The paradigm-shifting device ideas, indispensable technologies for proving the possibility and unforeseen obstacles against the practical application of single molecule devices will also be discussed.

THE LIMITATIONS OF THE PRESENT DEVICES

The limitations of MOS transistors are discussed here, including scaling principle, physical/chemical limitation, and necessary characteristics of the superseding information processing devices.

Scaling principle and limitations of MOS transistors

Since the very rapid innovation of ULSI technology started in 1970, the integration was quadrupled, while the minimum dimensions of the transistors were reduced by about 70% every three years, respectively, as shown in Fig. 1. The basic principle behind this innovation is called, 'scaling principle' [14], in which the lateral as well as vertical dimensions of MOS transistors are shrunk by a factor of k, whereas the impurity concentration is increased by a factor of k, resulting in an improvement of switching speed by a factor of k and switching energy by a factor of k^3. Here, the principle demands 'the smaller the better'. The results shown in Fig. 1 also indicate that the performances of ULSIs, such as the clock frequency, are also quadruped every three years. In other words, it takes only 5 years to improve the performances of ULSIs by one order of magnitude.

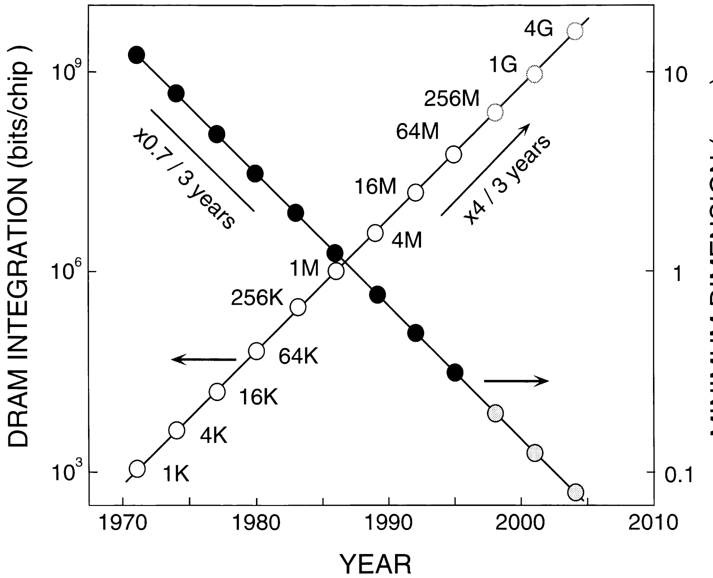

Fig. 1 The trend curve of ULSI technology innovation started in 1970. The integration quadrupled every three years, while the minimum dimension is decreased by ×0.7.

However, the progress is foreseen to cease sometime in the beginning of the centennial 2000s, when device, technology, material and physical limitations might be reached. There are various controversies regarding the limitation of the innovation, some are optimistic [15] and others are pessimistic [2,3]. The reasons for the obstacles against further scaling of MOS devices are the non-scaleable factors such as (i) built-in voltage of a p-n junction, (ii) band gap energy of an insulator, (iii) current capacity of a metal, (iv) statistical error of $\Delta N/N$ and (v) carrier mobility saturation [12]. All these limitations would prevent MOS transistors to scale down below around 100 m, which might be reached within 10 years from now. Therefore, superseding devices are definitely required for advanced information processing.

Necessary characteristics for the superseding devices

Here, the most essential characteristics for the information processing devices will be considered, because some of the 'new concept devices' only focus on the specified characteristics, such as switching speed. The characteristics are, (i) I/O signal balance, (ii) I/O isolation, (iii) fast operation speed, (iv) dense integration and (v) fabricability [12]. The reason why MOS transistors have been used so far and will be the sole device for information processing up to their physical/chemical limitations, is that they almost ideally fulfill those five factors in a very balanced manner. No other existing devices would be able to surpass MOS transistors, because only a limited performances are superior to those of MOS transistors. Of course, other factors such as noise immunity, reliability and power consumption have to be taken into consideration in integrated circuits. Though the scaling limitations would be reached sooner or later, the quest for a much higher performance information processing system would be accelerated even more. Therefore, information processing devices with orders of magnitude higher performances are inevitably necessary which supersede the silicon ULSI devices within 10 years from now.

SINGLE MOLECULE INFORMATION PROCESSING DEVICES

Why atom/molecule device for the next generation?

As has been discussed in the previous section, present device technologies are facing fundamental limitations arising from physical/chemical/materials nature. Therefore, innovation is definitely needed for the next paradigm shift, which makes it possible to realize orders of magnitude improvement in the performances. This will only be made possible if the innovation is achieved from the fundamental principles, such as physics, chemistry and materials. Which direction should we proceed? There is a proverb in Japan, that is pronounced as 'On-ko-chi-shin', which means that you have to look back the history in order to know what will come in the future. Figure 2 shows the switching speed and integration density of historical information processing devices. The figure indicates that human beings first started their electronic information processing using relays [1]. Then they were replaced by vacuum tubes and transistors, now finally integrated circuits dominate the information processing world. The figure clearly depicts that at least one order of magnitude improvement in such characteristics as switching speed and integration density is essential to bring the information devices to the paradigm shift. Therefore, the next generation devices would be switching at more than 1 THz and integrated at more than 10^9 devices/mm^2, which indicates that the superseding devices should be nm in size [12]. If all the information processing devices can be fabricated by single molecule based devices, not only would the switching speed and integration density of the devices be improved by orders of magnitude, but also the total energy necessary for the integrated system would be reduced by orders of magnitude. Therefore, single molecule based information system would satisfy the requirements of the high performance information processing devices and 'environment-friendly' operation characteristics.

Initial ideas of single molecule devices

Theoretical simulation of a single molecule diode idea was first proposed by Aviram & Rattner in 1974 [5], followed by an experimental demonstration of molecular photodiode by Fujihira *et al*., in 1976 [6]. The 'molecular device' idea, proposed by Carter in 1980 [7], gathered so much attentions from the many scientists that numerous 'single molecule devices' have been drawn on papers. However, they have not been made practical for almost a quarter of a century. The main reason is that there has not been any

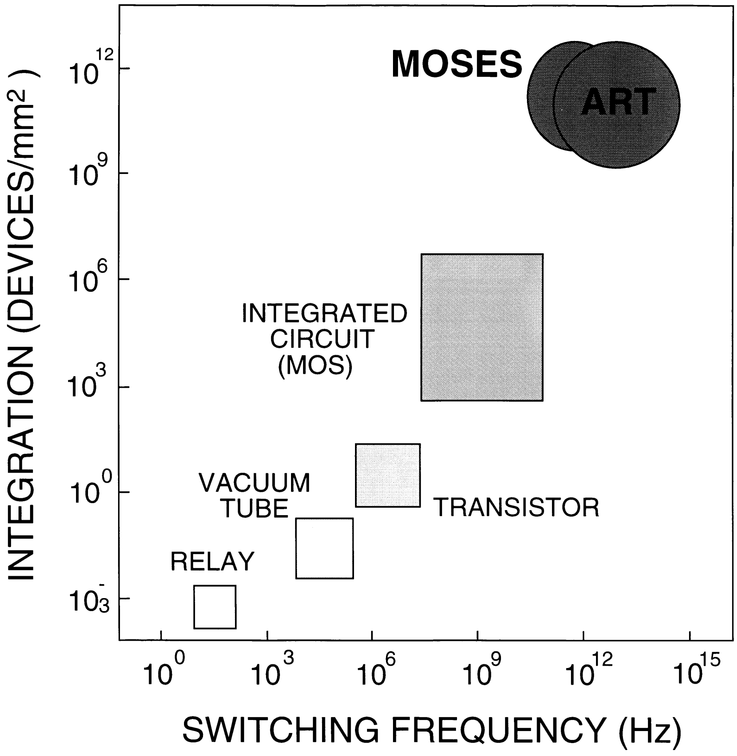

Fig. 2 Relationship between switching speed and integration density of historical information devices: two–three orders of magnitude improvement is essential to bring the information devices to the paradigm shift.

method that enables to have an access to one single molecule before the invention of the scanning tunneling microscope [9]. In addition, most of the 'devices' were proposed because they demonstrated very interesting characteristics as molecules, but not necessarily from the information processing point of view. Because of the strong competition among the semiconductor devices, the 'molecular device' ideas were only discussed within academia and this wave never reached the industry [8].

Single molecule device ideas revisited

Because the performances of the scanning tunnelling microscope have improved [9], it was almost made realistic to access to a single molecule. In addition, the 'limitations' of MOS devices are beginning to be discussed much more seriously than before. Therefore, the 'single molecule device' ideas were being considered as a possible substitutes for MOS devices, and many workshops have been held very actively since the middle of the 1990s. Several examples are listed in refs [16–18]. The following section summarizes some of the switching device ideas, classified by the switching mechanisms.

Switching devices based on the conformable change of a molecule

This classical device idea was based on the conformable change of the molecule, because chemists played a central role in the first step towards the construction of molecular device ideas. Some of the typical examples are described in refs [16–18], where the molecular structure change, such as *cis-trans* transition, was used as the major switching principle. These devices would not switch at more than several kHz, because as usually the case, the reverse conformational transition is much slower in the molecular system. These non-symmetrical switching characteristics might be due to the energy differences between the two molecular structure states. Present information processing requires symmetrical switching characteristics, because it relies on switching between '1' and '0', and no predetermined or preferential switching direction exists. Information processing speed is dominated by the slowest process in the existing architecture, and any slow switching step would lower the total performances of the system.

Switching by a single electron transfer principles

A high performance device based on single electron transistor (SET) principles was proposed [19]. The switching speed of the SET was simulated, and it was predicted that the operation speed should be more than THz, if the quantum dot size is reduced to a nm regime, as shown in Fig. 3 [20]. Conventional nano lithography technologies [21–23] would not be able to fabricate a quantum dot in the nm regime at the designated accuracy, and the only solution for the reproducible device fabrication would be to use molecules. Figure 4 schematically shows the Molecular Single Electron Transistor (MOSES) idea [24,25]. The figure suggests that by using conducting molecules for quantum dot, source, drain and gate electrodes, as well as insulating molecules for tunnel junctions and gate insulator, it would be possible to achieve more than 1 THz operation of MOSES.

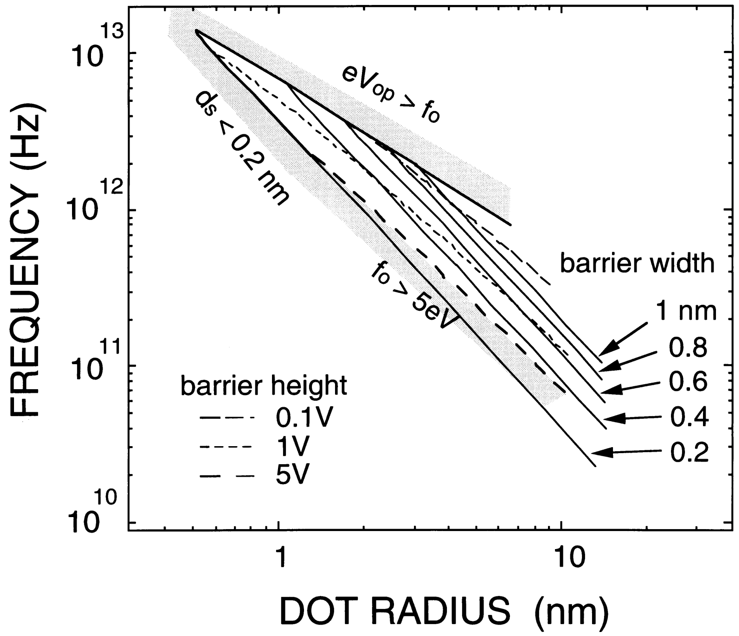

Fig. 3 Simulation results of the switching speed of single electron transistor (SET). The quantum dot size should be smaller than several nm in order to operate at more than THz regime.

The detailed analysis of the MOSES operation characteristics would permit the precise design of the quantum dot size, tunnelling barrier height and width, gate insulator specification and the electrode characteristics. Chemical synthesis of the molecule should be a very interesting challenge. Theoretical prediction of the conductance of a molecule [26] suggests that molecules, with several eV wide band gap such as those proposed in ref. [27], would have very high transmission probability by choosing the appropriate connection and carrier injection conditions. However, due to the fluctuation of potential in the molecule, practical use of those molecules would not allow such precise injection conditions, and the conductance would decrease logarithmically with the increase of molecule length [28]. In addition, a theoretical prediction of the tunnel barrier characteristics would contribute to the thorough understanding of the molecule-electrode complex system, when the electrodes are connected to the external system. Solving these problems would enhance the development the single molecule device.

Switching by mechanical movement of an atom in the molecule

An atom size device, Atom Relay Transistor (ART), was proposed in which the mechanical motion of an atom causes conductance change or switching of an atom wire, as schematically shown in Fig. 5 [12]. The switching speed of ART was simulated by the first principles method, and the switching atom can move at the frequency of more than several 10 THz [29], to allow ART to operate at a very high speed. The switching characteristics were also predicted by simulation, and it was shown that a displacement of the

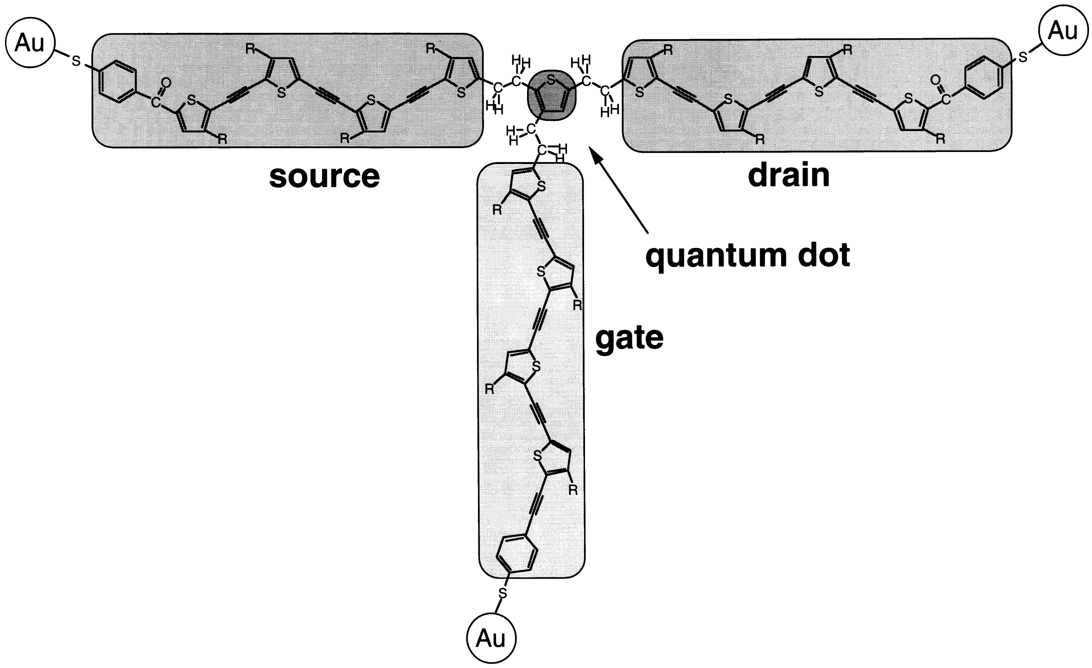

Fig. 4 Schematic figure of Molecular Single Electron Transistor, MOSES, with quantum dot, tunnel junctions and gate structures.

switching atom by only one atom diameter would change the conductance of the atom wire by orders of magnitude [30]. A molecular version of ART was also proposed [31], and the rotation of the molecule would allow the conductance of the atom wire to change according to the position of the molecule. The rotation speed should far exceed THz regime, and highly superior switching characteristics are also expected. Therefore, molecular scale 'mechanical electronics' (relay) might be reviving after 50 years of absence from the main current of information processing [12].

Switching devices based on other principles

One of the interesting switching principle relies on the potential modulation in the molecule, where the electronic structure, i.e. the impedance, of the molecule is expected to change according to the differences of the molecular structures [32]. In such an operation scheme, potential injection and detection would also be necessary as other devices with conventional carrier transport devices, and the low conduction problem in such high impedance molecules cannot necessarily be solved. This operation principle also poses the question whether the adsorption on the solid surface affects the operation principle. In addition, the potential propagation speed within a large molecule (switching speed) has to be predicted, in order to see whether this operation principle satisfies the above mentioned requirements of 'novel devices'.

TECHNOLOGY DEVELOPMENTS FOR PUTTING SINGLE MOLECULE DEVICE IDEAS INTO REALITY

The final destination of single molecule information processing devices is the 'molecular supercomputer' concept, as schematically shown in Fig. 6. The core part is made up of a THz molecular processor and GByte cache memory, whereas the peripheral part is occupied by more than a 10 GByte semiconductor main memory and interface circuits. Thus, the advantages of very high speed processing of single molecule devices are made the best use of, while the man/machine interface is taken charge of by the peripheral semiconductor circuits. However, this goal would not be able to be reached in one step, and the following four milestones might have to be considered: (i) measure the two terminal conductance of a single molecule, (ii) demonstrate the functions of two terminal molecules, such as light emission, (iii)

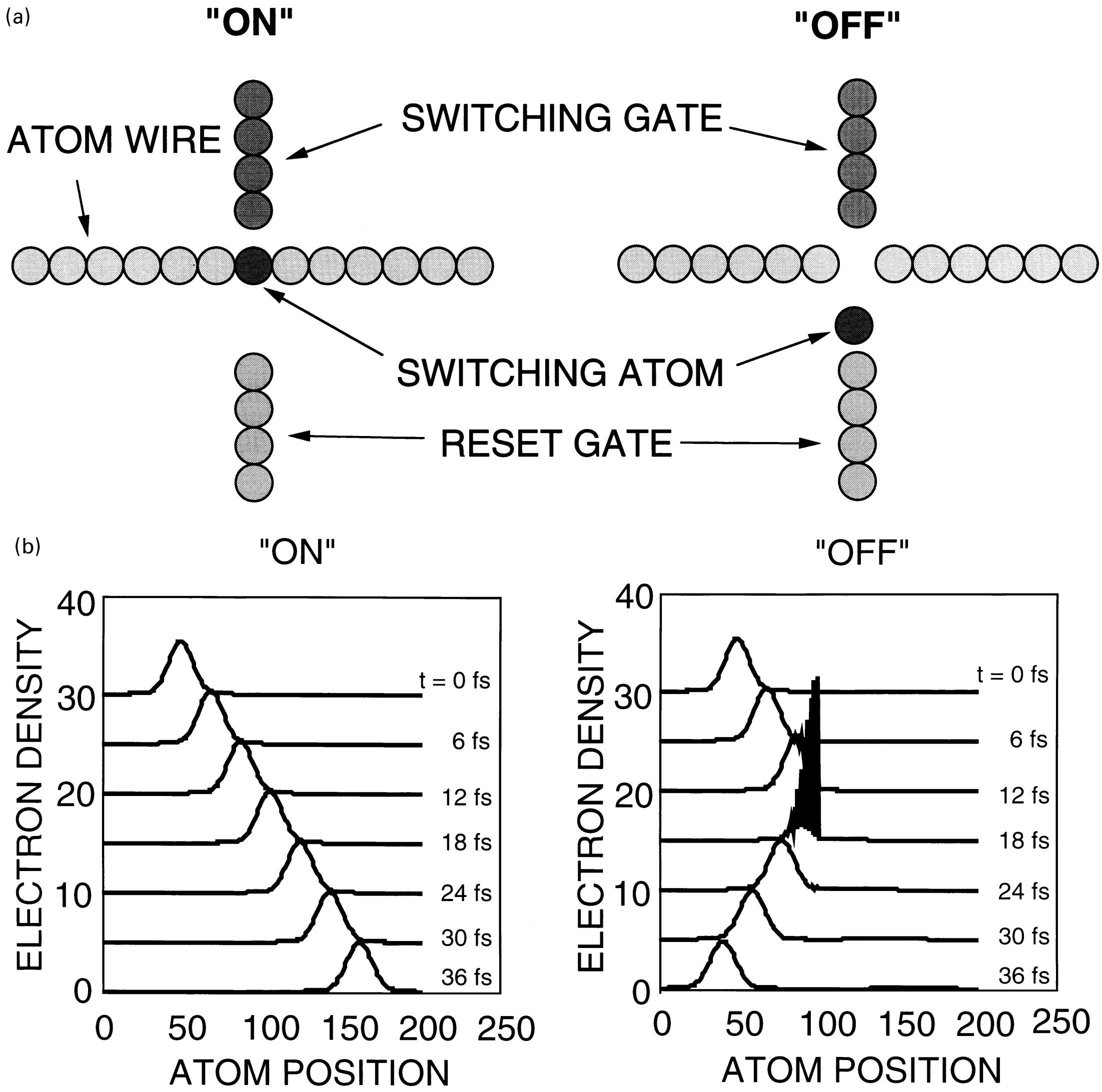

Fig. 5 Schematic figure of Atom Relay Transistor, ART, with atom wire, switching atom and switching gates.

verify the operation of three terminal molecules, such as transistor action and (iv) integrate the functions of the 'molecular supercomputer'.

Molecules for two terminal electrical measurements

The first milestone in the technological development towards the 'single molecule electronics' is to address the single molecule between two electrodes and to measure the electronic characteristics. However, despite of those very impressive and attractive 'single molecule device' proposals, electrical characterization of one single molecule has not been thoroughly demonstrated yet. Several access methods to a single molecule have been proposed, and some very impressive experiments were carried out. However, almost no results provided enough experimental data that persuaded one single molecule conductance. Here, the achievements are summarized to show the possible problems in the measurements.

Theory of electron transport through linear single molecule

Carrier transport within a molecule should pose very interesting theoretical problems, and many theoreticians are eager to demonstrate the possibility of the conductance in single molecule. A very plain

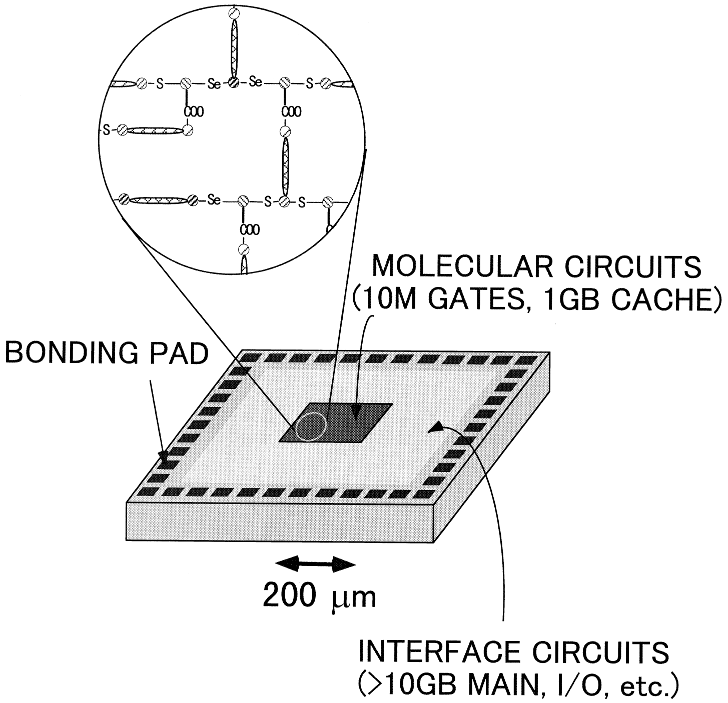

Fig. 6 Schematic figure of 'single molecule supercomputer', in which the core part is made up of 1 THz molecular processor and 1 GByte cache memory, whereas the peripheral part is occupied by more than 10 GByte semiconductor main memory and interface circuits.

plant explanation of the conductance simulation is reported in ref. [28], in which the conductance of a molecule shows a logarithmic decrease on the length of a molecule. Therefore, a molecule with about 10 nm in length should exhibit a transmission of about 10^{-16}. In order words, even a 5 nm long molecule is a very good 'insulator', which shows about 10^{-10} transmission. Therefore, conventional molecules would not provide any possibility of formulating an 'electronic circuit' in an existing sense.

Some of the very encouraging results are reported in ref. [26], in which it is claimed that if the center of a parallel molecule is connected, the resonant injection should make the molecule a very high transport material. Even a 10 nm long molecule should have a transmission almost equal to a single atom. However, since the potential within a circuit changes according to their logic functions, it would be almost impossible to formulate integrated systems based on this principle. Therefore, in order to synthesize a functional molecule as shown in Figs 4 and 5, it would be appropriate to use a molecule with 'metallic conduction characteristics' and/or doped semiconductor characteristics. One interesting prediction regarding the conductance of a single molecule connected between two electrodes is that the injected electron might transfer coherently within the molecule below a certain length [33]. If the first principles simulation result is correct, the molecule synthesis strategy described below would be greatly influenced.

Synthesis of linear molecules

The following molecular characteristics have to be fulfilled in order to accurately measure the conductance of single molecule. They are, (i) conductive, hopefully metallic, for low resistance measurement, (ii) solid and rigid for easy-to-connect between two electrodes, (iii) covalent connection sites on both ends of the molecule for low resistance contacts and (iv) isolated from the substrate like enamel wire for preventing short circuits. Many molecules have been proposed for the single molecule measurement [34–36], however, most of them do not necessarily satisfy those requirements, especially 'low resistance' characteristics, because of their 2–3 eV band gap. Therefore, their conductance characteristics would follow the predictions described in ref. [28]. There have been several efforts towards the 'zero band gap' molecules [37], and it is expected that this 'metallic' molecule would make the accurate conductance measurement possible.

'Conductance' measurement of linear single molecule

As described before, the first milestone toward the practical development of single molecule devices would be to demonstrate the 'conductance measurement' of a single molecule. This section reports several efforts of the current status towards the goal.

Use of solid electrodes to address a single molecule

Several ideas have been proposed so far and one of the most conventional methods to measure the conductance of a linear molecule is to address it between two solid electrodes with few nm wide gap [38]. Then, the molecules are attached to the electrode structures by either immersion in a solution or vacuum evaporation. Thus some molecules are expected to attach between the electrodes. The shortcomings of this method are, of course, (i) possibility of attaching large number of molecules and (ii) leakage current through the surface of the substrate. Therefore, very few experimental results are reported so far, although many experiments have been done on the fabrication of the nm gap structures.

Nano connection technology using piezo-driven devices

Another possible method for measuring the single molecule conductance is to use piezo-electrically driven devices. Two methods have been reported so far, one is called 'mechanically controlled breakable junction (MCB)' [39] and the other uses STM [40]. The former pushes very thin gold electrodes attached with molecules by piezo-electric devices to widen the gap between the electrodes, so that the gap width could be controlled down to a pm regime. The latter images the surfaces by STM, on which the molecules are arranged. It is planned that if the objective molecule protrudes from the other molecules, it can be accessed by the STM tip. Both experiments report that the resistance of the single molecule can be in the order of several mega-ohms. The reason for the very high resistance might be due to the insulating nature of the measured molecule. In addition, the very top portion of the electrode structure has not been investigated, so that there remains a possibility that the electrodes can be very flat to accommodate so many hundreds of molecules between them. 'Needle Formation and Tip Imaging' (NFTI) method [41] described below might make it possible to evaluate the electrode structures at an atomic scale. Further investigation should be required for concrete results. Another problem might be the covalent bond information between the molecule and the electrode.

Micromachine technology for addressing a single molecule

A micromachine STM was fabricated for single molecule addressing [42]. A scanning electron micrograph of the micromachine STM is shown in Fig. 7, in which the comb electrode moves the main body back and forth by applying voltages between the two electrodes. In order to investigate whether the area between the two electrodes, tip and sample, are small enough to accommodate a single molecule, transmission electron microscope (TEM) investigation of the tunnel gap was carried out. The results are shown in Fig. 8 [43], in which it is indicated that the area of the adjacent two electrodes is small enough so that only one molecule could connect the two electrodes. Another possible nano-electrode formation technology is called, 'Needle Formation and Tip Imaging (NFTI)'. Very small needle structures, as small as 2 nm in diameter, can be grown on the silicon surface [41] which should be small enough for addressing single molecule on the top. The remaining problem is the mechanical strength of the structures, so that neither the high electric field nor the mechanical force would break the small structures.

Three terminal connection ideas

The third milestone towards the 'single molecule electronics' would be to connect the three terminals to the molecules. The idea was stated in ref. [13], in which selective reaction between the specific part in the molecule and corresponding atom species is described. For example, the selective reaction takes place between the sulfur atom and gold atom as well as carbonic acid and silicon atom. Thus, by

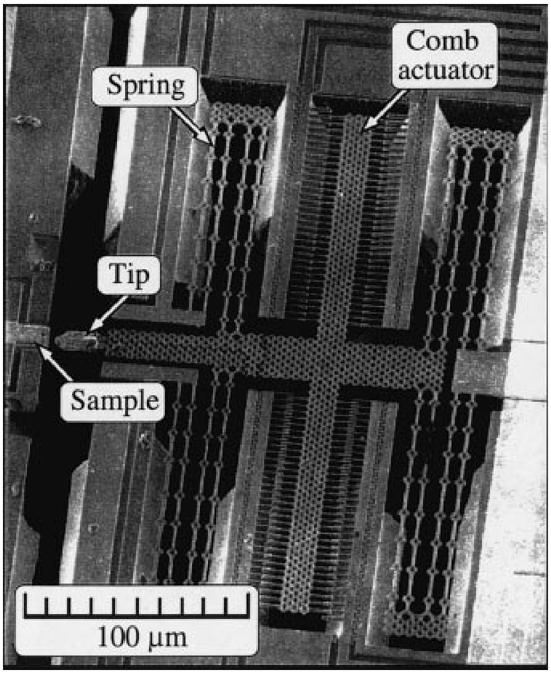

Fig. 7 Scanning electron micrograph of the fabricated one dimensional (1-D) μ-STM.

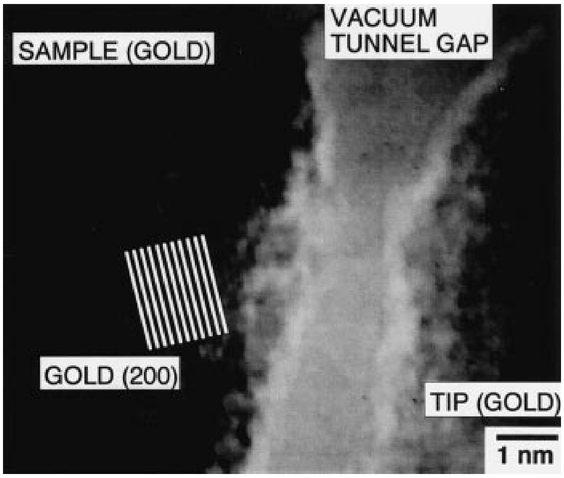

Fig. 8 TEM micrograph of the tunneling gap between the μ-STM tip and sample.

choosing appropriate reagents, the molecule would be able to be placed on a designated position of the surface.

REMAINING PROBLEMS

There are several remaining problems to be solved along with the achievement of the above mentioned four milestones. They are, (i) architecture, (ii) man/machine interface, (iii) clock distribution, (iv) wiring, including multilayered wiring technology and transmission delay, (v) power consumption, (vi) noise immunity, (vii) self-assembled arrangement of molecules on a chip, (viii) functional change of the molecules due to connection to other devices, including electrodes and (ix) very large functional molecular device synthesis. These problems have to be solved before the real integrated device chip, schematically shown in Fig. 6, would be fabricated. Other possible application of single molecule device would include light detection devices [44], storage devices [45], light emission devices and sensor devices. Those devices would also contribute to the high performance information processing and enrich the human life.

CONCLUSION

The present information processing devices should reach their fundamental limitations within a decade, therefore, superseding devices are definitely required. This paper summarized the single molecule device ideas for high performance information processing, and the necessary technology development towards their realization. Four major milestones would have to be cleared before the next paradigm of a single molecule device system would be developed. The first milestone would be to measure the conductance of a single molecule, and the current research status was summarized, including theoretical predictions, molecular synthesis and measurement technologies. Micromachine scanning probe technology was reported which would provide means for access a single molecule. Three terminal device ideas were also reviewed, and the possible need for a new information processing architecture, which is suitable for the molecular information processing, was pointed out.

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