



Integration of molecular functions into Si device for nanoscale molecular devices



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ABSTRACT

Recent progress on nanoscale molecular devices is reviewed. The advantages of organic molecules are highlighted in terms of their unique features such as quantum effects based on their nanometer size, optical properties originating from photochromism and chemical syntheses for various structural designs. This review covers three topics: a scanning probe microscope-based evaluation of individual molecules, nano-gap electrodes for electrical current through single-molecules and the incorporation of molecular functions into Si-based devices. In particular, the importance of the incorporation of organic molecules into Si-device architecture is emphasized with a view to realizing the large-scale integration of nanoscale molecular devices.

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1. Introduction

Since the scaling of Si-based transistors is approaching its physical limit, intensive work is being undertaken on alternative materials and device architectures. A promising approach to overcoming the impending problems is to provide current complementary metal oxide semiconductor (CMOS) devices with functionalities. These challenges are referred to using the terms “beyond-CMOS” or “More than Moore”. Two-dimensional (2D) materials, such as graphene [1] and molybdenum sulfide (MoS_2) [2], are fascinating candidates because of their superior carrier mobilities and compatibility with existing device architectures. However, these 2D materials have a disadvantage in terms of device size reduction. On the other hand, organic molecules offer advantages as regards their small size (nanometer scale) and diverse functionalities, which are unobtainable with their inorganic counterparts [3–6]. These potentialities mean that organic molecules have been receiving increasing attention as components of nanoscale electronic devices. In this short review, we describe the background, recent progress and current status of molecular optoelectronic devices. The development of molecular electronics can be divided into three stages: scanning probe microscope (SPM)-based analyses of single molecules, nano-gap electrodes with molecules as preliminary devices and large-scale integration with Si-based device architectures. The first part of this review is devoted to SPM-based single-molecular devices, and concentrates on the merits of SPM techniques in view of their accessibility to individual molecules. Although at this stage most studies were related to fundamental evaluations of molecular properties, this work triggered an interest in single-molecular electronics. The second part discusses electrical measurement techniques related to nano-gap electrodes. In particular, it reviews the recent progress made possible through the use

of the mechanically controlled break junction (MCBJ) technique. The great contribution of this technique has been to form a bridge between basic science and application development. The last part of this review describes molecular devices in Si-based structure. This is the most promising way to realize the large-scale integration of molecular functions and to provide existing Si devices with functions.

2. Scanning probe microscopes for single molecular devices

A family of scanning probe microscopes (SPM) including the scanning tunneling microscope (STM) and the atomic force microscope (AFM) were invented to allow the structural observation of the surfaces of materials. Moreover, soon after these epoch-making inventions, SPM techniques were recognized as effective tools for characterizing electronic states on an atomic scale. For example, Joachim et al. showed that STM can be used to measure the electrical properties of an individual molecule. They determined the resistivity of a single C_{60} molecule by making a stable electrical contact using an STM tip [7] (Fig. 1(a)). Tour and his co-workers examined the conductivity of a conjugated molecular wire. For this purpose, a conjugated molecule with a rod-like structure was inserted into a non-conductive self-assembled monolayer on an Au(111) surface [8] (Fig. 1(b)). Porath and Millo analyzed the discrete energy levels of an isolated C_{60} molecule from its tunneling current–bias voltage (I – V) characteristics [9,10]. This is called scanning tunneling spectroscopy, where occupied and unoccupied electronic states are visualized from differential I – V curves (dI/dV curves). Since these pioneering studies undertaken in the late 1990s, SPMs have been widely used to study electronic states on a single molecular scale [11–16]. In addition to STM, AFM is also useful for measuring the conductance of single molecules [11]. Engelkes et al. used conductive cantilevers in an AFM setup as illustrated in Fig. 1(c). This technique allows us to observe non-conductive self-assembled monolayers, and to measure the charge transport through

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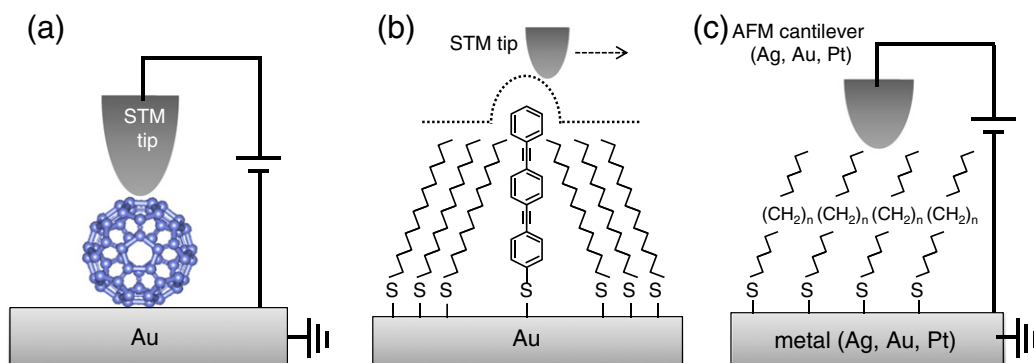


Fig. 1. Applications of SPM techniques for measuring electrical conductivities of individual molecules. (a) The direct contact of an STM tip on a C₆₀ molecule determined the single-molecule resistivity (ref. [7]). (b) A π-conjugated molecule in a non-conductive self-assemble monolayer gives high contrast in an STM image (ref. [8]). (c) The length and work function dependence of the charge transport was examined with conductive AFM cantilevers (ref. [11]).

the molecules. They discussed the tunneling mechanism between the cantilever and the substrates by systematically changing the length of the alkyl chains and the work function of the metals.

A characteristic feature of organic molecules is their diverse molecular functionalities, which can be obtained by chemical synthesis techniques. SPM-based techniques have shown that such molecular functions can be realized even on a single molecular level. Various SPM applications have been reported in relation to charge transport switching through a redox center [17], the switching of molecular orientation [18], a high density non-volatile memory in C₆₀ multilayers [19], giant magnetoresistance through a single molecule [20], a rectifying effect [21] and the optical switching of conductance through a photochromic molecule [22]. Typical examples are shown in Fig. 2. Yasutake et al. revealed that the orientation of an endohedral metallofullerene was reversibly switched (Fig. 2(a)) [18]. Here, the driving force is an applied bias voltage from an STM tip. Meanwhile, Nakaya et al. found that electrical current from an STM tip triggered a chemical reaction on a single molecular level [19]. They demonstrated that dimerization ([2 + 2] cycloadditive reaction) between neighboring C₆₀ molecules was induced by a tunneling current. Importantly, the opposite tunneling current returned such bonded molecules to their original unbonded state (Fig. 2(b)). They proposed applying this reversible reaction to a non-volatile memory. These studies have encouraged the hope that a variety of functions can be incorporated into individual molecules and ultra small optoelectronic

devices can be produced based on single molecules. However, the SPM-based techniques provide a two-terminal device configuration. The next step towards making this hope a reality is to arrange a single molecule in a three-terminal device structure, namely a molecule between a pair of electrodes on a substrate, for preliminary device operations. In the following section, we review such rudimentary molecular devices.

3. Break junctions for single molecular devices

Three-terminal device geometries such as a transistor structure are indispensable for the further development of molecular electronic devices. An important point is that a tiny gap with dimensions comparable to those of a single molecule should be formed between the source and drain electrodes. Such nano-gap electrodes, which are mostly produced by lithographic techniques or by breaking thin metal wires, have made it possible to reveal the potential of organic molecules as quantum dots. Song et al. reported that transport current through molecule was directly modulated by an external gate voltage, where electrons were tunneling resonantly via molecular orbital [23]. McEuen and his co-workers investigated the coupling between a single-electron transfer and the quantized mechanical oscillation of single C₆₀ molecules in the transistor geometry with nano-gap electrodes [24]. They observed a Coulomb blockade and the Kondo effect through cobalt-coordinated molecules by using the same techniques

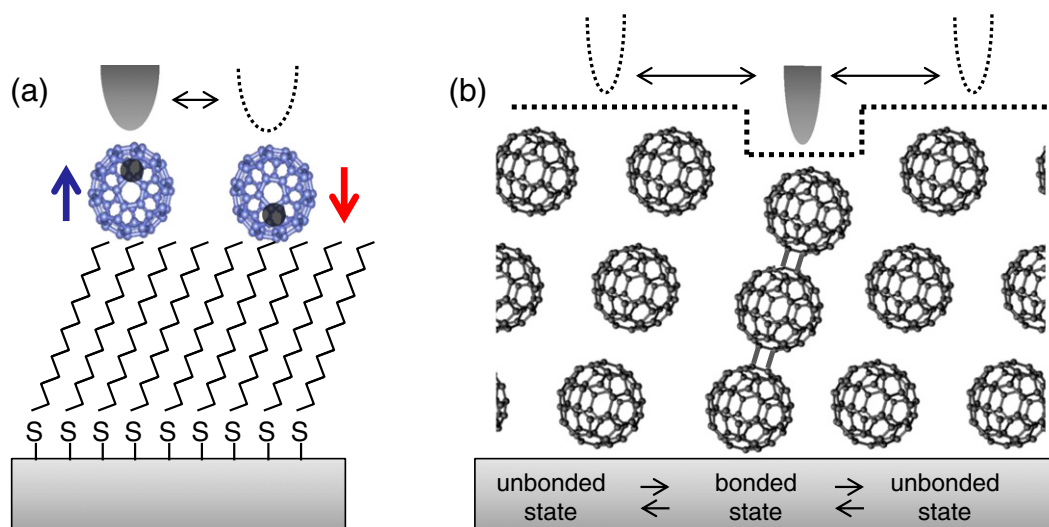


Fig. 2. Active uses of STM techniques for single-molecule switching and memory devices. (a) Applied bias voltages switched the orientation of an individual endohedral metallofullerene (ref. [18]). (b) Tunneling current induced the dimerization ([2 + 2] cycloadditive reaction) of C₆₀ and reverse bias voltage returned it to the original unbonded state (ref. [19]). Both showed reversible reactions, thus demonstrating potential as switching and/or memory devices.

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