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Carrier Transport in Artificial Lattice of Self-Organized Gold Nano-Particles

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1. Introduction

Strongly correlated electron systems [1] are attractive candidates for future nano-electronic devices, since they have a rich variety of states: Mott insulating, metallic, (anti-)ferromagnetic, and superconducting states. In these systems, the metal-insulator transition takes place by changing the number of electrons per lattice sites (Fig. 1). Therefore, the high "ON"-"OFF" ratio of currents is expected, if a field-effect-transistor with a Mott insulator as an active channel material is achieved [2, 3].

The lattice site of theses systems is not restricted to a single atom or a molecule, but a quantum dot is also a candidate [4, 5]. With these *artificial lattice* systems composed of quantum dots, we can control both transfer (t) and interaction (U) energies of the Hubbard model at our disposal by designing the structure [4]. For example, the patterned Kagome lattice is expected to exhibit such an exotic behavior like flat-band ferromagnetism [4, 5].

Arrays of self-organized nano-particles also belong to this class of systems [6-9]. The size of these nano-particles (<5 nm) is enough small to expect the single electron charging effects. This system would be promising for flexible devices, since the fabrication process is based on the wet process at the room temperature. However, the basic transport properties of the mono-layer film are not completely understood, yet. R. Parthasarathy *et al.* reported nonlinear current-voltage (I-V) characteristics, reminiscent of the Coulomb blockade [8]. In contrast, K. C. Beverly *et al.* reported Ohmic conduction, and observed large dependence of resistivity on temperature (T) [9]. It is necessary to solve these discrepancies and to understand the transport mechanism of the film for a possible transistor application. In this paper, we discuss measured transport characteristics based on a theory of the dissipative quantum tunneling [10, 11].

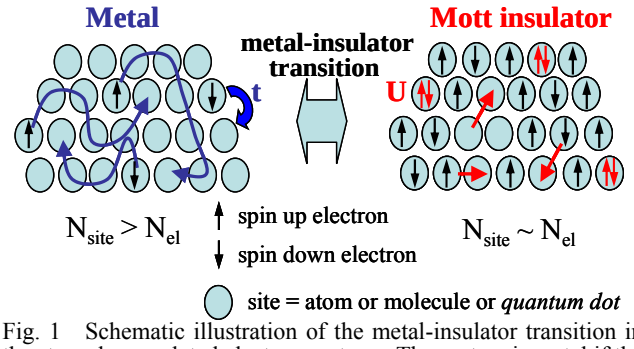

Fig. 1 Schematic illustration of the metal-insulator transition in the strongly correlated electron systems. The system is metal if the number of electrons N_{el} is small compared with the number of sites N_{site}, whereas the system becomes insulator if $N_{site} \sim N_{el}$.

2. Experiments

We chemically synthesized Au nano-partices [6-9] with an average diameter of 2.5 nm, whose charging energy is estimated to be 0.3 eV (~10kT at the room temperature). The surface of Au nano-particles is coated with mono-layer of C_4H_9SH molecules; organic ligands. The average distance between Au nano-particles is limited by the size of C_4H_9SH and is estimated to be approximately 2.0 nm.

The Si substrate with 200 nm SiO_2 was prepared and Au electrodes were made by conventional lithography and lift-off. The films of self-organized Au nano-particles were formed on the substrate (Fig. 2) by Langmuir-Blodgett (LB) techniques [6-9]. We prepared both mono-layer and sub-mono-layer LB films to examine impacts of structural defects in the arrays of nano-particles.

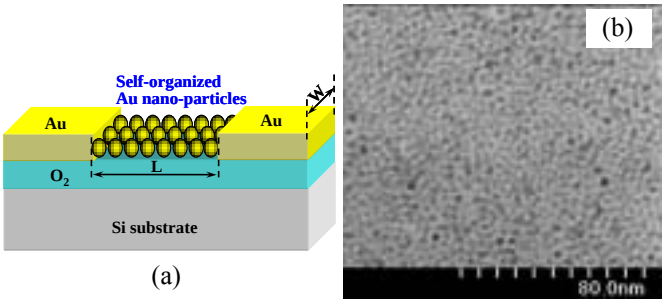

Fig. 2 (a) The device structure to measure I-V characteristics for the Langmuir-Blodgett film of the self-organized Au nano-particles. Nano-particles on Au electrodes are omitted for clarity. (b) The plain-view of TEM image for the Langmuir-Blodgett film of self-organized Au nano-particles with C_4H_9SH organic ligands.

3. Results and Discussions

Coherent Tunneling

Figure 3 shows I-V characteristics of the mono-layer film, where the linear I-V characteristics are maintained at least for 4 orders of magnitude down to the low temperature. Four terminal measurements confirmed that the contact resistance between the array and electrodes is sufficiently small compared to the sheet resistance. To achieve this linear behavior, we believe it is important to reduce structural defects inside the LB film, as confirmed by non-linear behaviors for films with defects, shown below. The linear behavior implies the coherent tunneling of carriers between neighboring nano-particles, and the formation of sub-bands due to the *artificial lattice* symmetry. As a result, the whole system exhibited a linear response similar to the conventional material.

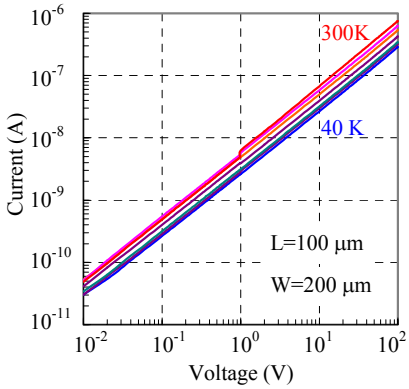

Fig. 3 The Ohmic conduction behavior down to low temperature for the LB film with reduced amount of structural defects.

Pinning and Depinning Behavior

Next, we examined the samples of the *sub-monolayer* film. In this case, we intentionally introduced voids in a controlled manner by reducing the coverage of the film. We observed the nonlinear *I-V* characteristics, as shown in Fig. 4. This behavior can qualitatively be explained by the pinning and depinning phenomena. In the low bias regime, the carriers are trapped in the local potential minima, which are generated by the structural defects. On the other hand, carriers would be depinned if the applied bias is sufficiently large. Such a behavior is described by the carrier motion in a washboard potential, famous for the dynamics of superconducting phases or charge density waves pinned by impurities. Our results suggest that a new system, self-organized nano-particles with structural defects, is added to this family.

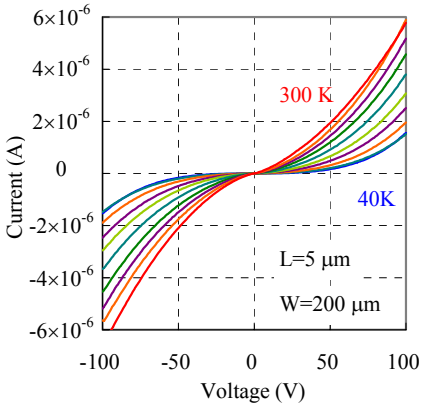

Fig. 4 Non-linear characteristic for the sub-mono-layer LB film with structural defects.

Scaling Properties of Dissipative Quantum Tunneling

In order to confirm above pictures, we applied the theory of dissipative quantum tunneling [10, 11]. In our system, we have many degrees of freedom, such as conduction electrons inside nano-particles and phonons of organic ligands, which behave as *environment*. The coupling of carriers with such environment is known to reduce the probability of the quantum tunneling. According to the theory of biased double well potential, the differential conductance behaves as

$$\frac{dI}{dV} \propto T^{\alpha} \coth\left(\frac{eV}{2kT}\right)\left|\Gamma\left(1+\frac{\alpha}{2}+i\frac{eV}{2\pi kT}\right)\right|^2 . \quad (1)$$

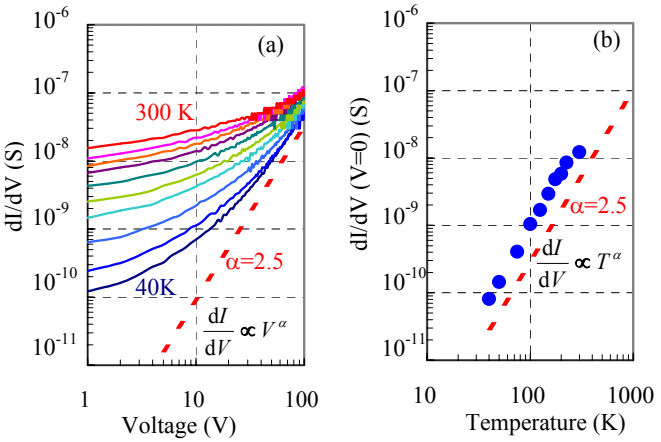

Fig. 5 Power law behaviors for the differential conductivity (a) as a function of voltages in the high field regime and (b) as a function of temperatures in the low field regime.

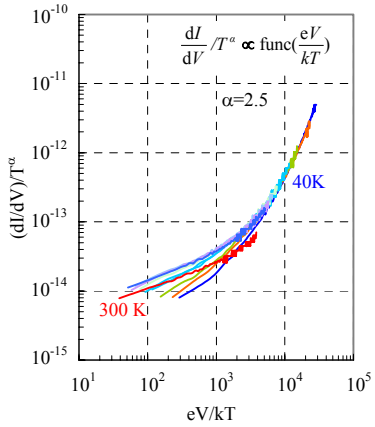

Fig. 6 Universal scaling law behaviors consistent with the theoretical predictions of dissipative quantum tunneling.

We compared our experimental data of Fig. 4 with Eq. (1). In Fig. 5, we confirmed that the differential conductance exhibits the power law behaviors as functions of V and T in high and low bias regimes. According to the exponent, we obtained the coupling constant $\alpha=2.5>1$, which means that the dissipative coupling is sufficiently strong to result in de-coherence of tunneling. In addition, the plot of Fig. 6 shows the universal scaling law independent on T, consistent with the theoretical predictions. This agreement suggests that the carrier transport in Au nano-particles with structural defects is limited by dissipative couplings with environment.

3. Conclusions

We examined the transport mechanism of *artificial lattice* of Au nano-particles. We observed the incoherent tunneling for films with structural defects, consistent with a theory of dissipative quantum tunneling. The precise control of ordering is important for achieving the coherent tunneling, which is indispensable for the formation of sub-bands in *artificial lattice*. The present work is the first step towards the realization of Mott transition of self-organized nano-particles by field-effect-transistors for the application of flexible devices.

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