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Measurement of Electrical Conduction of C_{60} Derivatives Monolayer on Au Ultrathin Film

Fumihiko Matsuyama, Kazuo Nishitani, Teppei Maeda, Tomoyasu Oda, Nobuyuki Iwata and Hiroshi Yamamoto

College of Sci. & Technol. Nihon University, 7-24-1 Narashinodai, Funabashi-shi, Chiba 274-8501
Fax: 047-469-5457, e-mail: hyama@ecs.cst.nihon-u.ac.jp

Little and/or Ginzburg proposed low dimensional transport systems as an excitonic model to obtain a room temperature superconductivity. We have studied electrical conducting properties observed in self-assembly monolayers of C_{60}-O-C_8SH derivatives (C_{60}-SAM) (as a dielectric layer) on Au ultrathin films (as a conductive layer) to realize the Ginzburg's excitonic superconducting model. Electrode Au films were prepared on flat MgO(100) substrates trough the metal mask by sputtering, and then sputtered Au atoms passed around behind the holes of the mask. Au ultrathin films were successively deposited without the mask by sputtering. The Au-deposited specimens were soaked in a 0.01mM benzene solution of C_{60} derivatives for 20 h at room temperature to form C_{60}-SAM. Several specimens revealed the deviation from linear temperature dependence of resistance accompanying with steep decrease of resistivity in the temperature region less than 100K - 130 K. Such the resistivity anomaly was observed only in the specimens which had ultrathin Au film area with the dimension of less than about 10 μm.

Key words: conducting anomaly, exciton, C_{60}, ultrathin film, self-assembly monolayer

1. INTRODUCTION

As well known Little[1] or Ginzburg[2] proposed theoretical models of an excitonic mechanism to realize a high temperature superconductor with the 1-dimensional or the 2-dimensional transport system. Then expected T_c 's are above room temperature.

In the models, the candidate of conductive layers (the main chain) for the 2-dimensional system is a metal. Since the shielding length of the exciton in metals may be in the order of nanometers, the metal layer must be ultrathin and continuous. The dielectric layers (the side chain) must reveal strong interactions with free electrons through enough high energy elementary excitation.

Shi et al. [3] reported a novel way to prepare monolayers, which are C_{60} derivatives self-assembly absorbed on Au substrates with a highly closed packed density. The C_{60} derivatives formed a stable monolayer by the interaction between Au-S and/or by van der Waals's force among alkyl chains. As the side plane π-electronic molecules layers may be available because of the high excitonic energy in the order of a few electron volts. In C_{60} films optical absorption charge transfer (CT) excitons were experimentally identified between 2.5 eV and 3.1 eV by Holden et al. [4]. We have noticed the self-assembly monolayer of C_{60} derivatives (C_{60}-SAM) as the side plane because of its physical and chemical stability or easier handling.

In this study, the used C_{60} derivative was C_{60}-O-C_8SH. Au ultrathin films were prepared on MgO (100) substrates which had atomically flat surfaces. When the C_{60}-SAM was prepared on Au ultrathin films, we expect that this substance system becomes the Ginzburg model. Figure 1 shows the schematic excitonic superconducting model proposed here.

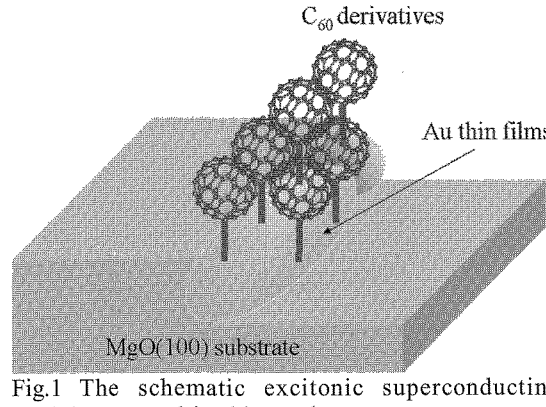


Fig.1 The schematic excitonic superconducting model proposed in this work.

The detailed experimental procedure will be explained. The morphology and structure of the surfaces are evaluated, and the electrode films configuration is studied. The interface structure of the specimen prepared is also evaluated and the observed temperature dependences of resistance are discussed in the view point of superconductivity.

2. EXPERMENTAL

The two MgO (100) substrates were faced to each other surface and were annealed at 1100℃ for 12 hours in the air .

Au/Mo electrodes were prepared on MgO (100) by on-axis RF magnetron sputtering method. The metal mask was used to prepare electrode films. Mo films were deposited with RF power of 100 W for 3 min at

150 ℃ substrate temperature, and Au films were deposited with 100 W for 9 min at 300℃. The distance between the target and the substrate was 50 cm. Base pressure of the chamber was 1.0×10^{-3} Pa. The pressure during sputtering was 1.5 Pa. Sputtered Au atoms passed around behind the holes of the mask. By adjusting the distance between the mask and the substrates, the diameter of deposited Au film electrodes was changed.

After the mask was removed, Au ultrathin films were prepared. The Au film was deposited with the RF power of 20W for 45 sec at the substrate temperature of 300 ℃. The distance between the target and the substrate was 50 cm. Base pressure of the chamber was 1.0×10^{-3} Pa. The pressure during sputtering was 1.5 Pa.

The detailed SAM process appeared in elsewhere[5]. The specimens were soaked in benzene solution of 0.01 mM C_{60} derivatives for 20 hour at room temperature. After the soaking non-adsorbed C_{60} derivatives were washed out by benzene and the specimens were dried by dry N_2 gas.

Au wire bonding was made on Au/Mo electrodes in order to measure the electric transport property of C_{60}-SAM/Au//MgO (100). The schematic cross section of the prepared specimen is shown in Fig. 2.

The surface morphology of Au and C_{60}-SAM/Au thin films was observed by Dynamic Force Mode (DFM). The images of the metal mask and Au/Mo electrodes were observed by the optical microscope. Temperature dependences of resistance were measured by a method of resistance measurement used two electrodes in the temperature range 77-300 K.

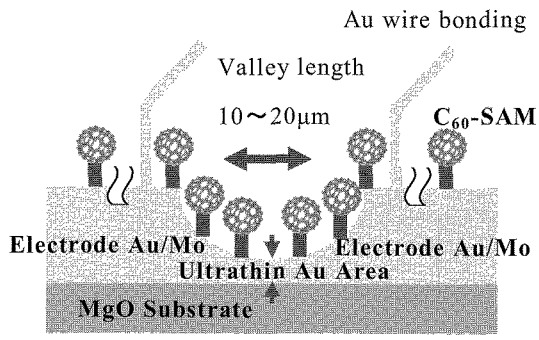
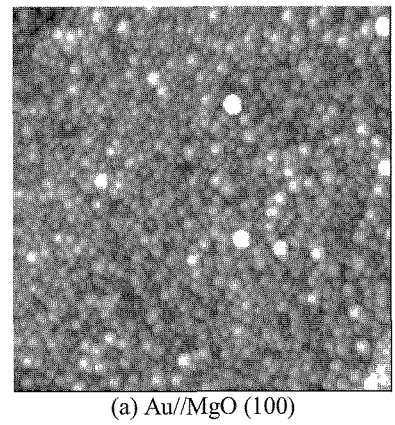


Fig.2 The schematic cross section of the specimen. The valley structure of ultrathin Au films was formed by deposited Au atoms passing around behind the metal mask holes.

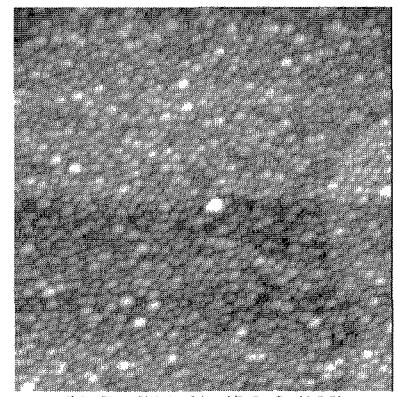
3. RESULTS AND DISCUSSION

3.1 Surface morphology

Figure 3 (a) and (b) shows DFM images of Au and C_{60}-SAM/Au thin films, respectively. A lot of Au particles with diameter of about 28nm were observed in the Fig. 3 (a) whole image. An average surface roughness (Ra) was 0.7283 nm. Particles with diameter of about 28nm were also found in the Fig. 3 (b) whole image. The Ra was 0.8877 nm.



(a) Au//MgO (100)

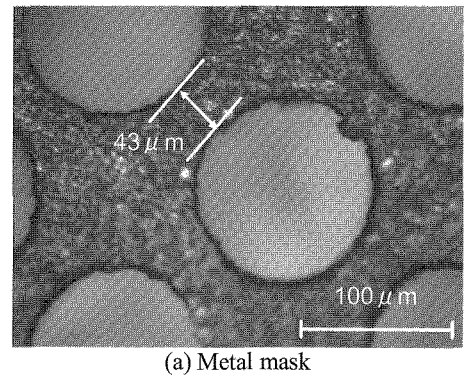


(b) C_{60}-SAM/Au//MgO (100)

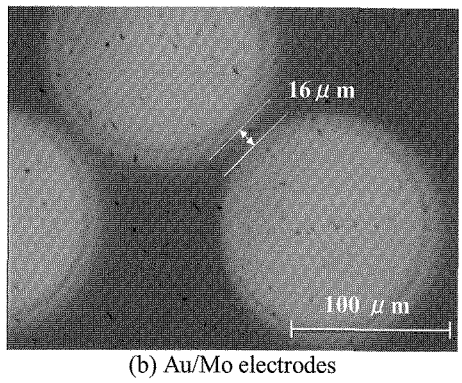
Fig.3 DFM images of Au//MgO (100) and C_{60}-SAM/Au //MgO (100). (a) 1 μm^2 DFM image of Au ultrathin film. (b) 1 μm^2 DFM image of C_{60}-SAM/Au thin film.

3.2 Image of metal mask and Au/Mo electrodes.

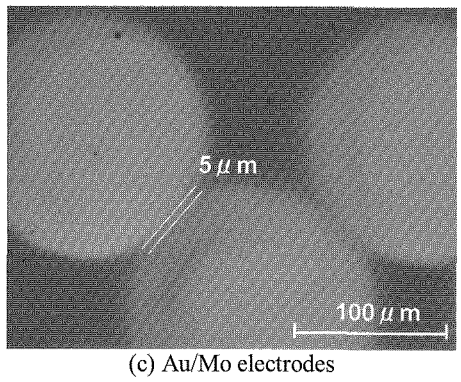
Figure 4 (a) –(c) shows the optical microscope images of metal mask and the typical Au/Mo film electrodes, respectively. As shown in Fig. 4 (a) the width between two holes of the metal mask was about 43 μm. The valley width of Au ultrathin area between two electrodes of the specimen was less than 20 μm as shown in Fig. 4 (b) and (c). Since sputtered Au atoms passed around behind the holes of the mask, the two electrodes distance became narrower.



(a) Metal mask



(b) Au/Mo electrodes



(c) Au/Mo electrodes

Fig.4 Optical microscope images: (a) the metal mask (b), (c) the typical two Au/Mo film electrodes deposited through the mask by changing the distance between the mask and the substrate.

3.3 Electric transport property

Figure 5 shows the typical temperature dependence of resistance in the Au thin films. The resistance decreased linearly from 1 Ω at room temperature to 0.4 Ω at 77 K, and the resistivity gradient was constant. This result was understood as the typical metallic conducting property

Figure 6 shows the three examples of temperature dependence of resistance in the C_{60}-SAM/Au thin films.

In the case of Fig. 6 (a) the valley width between two electrodes of the specimen was less than 16 μm. The resistance non-linearly increased from about 500 kΩ at room temperature to about 600 kΩ at 77 K.

In case of Fig. 6 (b), the valley width between two electrodes of the specimen was less than 11 μm. The resistance was comparatively high, and decreased linearly and slowly from about 35.2 Ω at room temperature to about 34.9 Ω at 130 K with almost the same resistivity gradient. The resistivity, however, more steeply decreased in the lower temperature than about 130 K. The value of the resistivity gradient in the lower temperature region was obviously larger than that in the higher temperature region.

In case of Fig. 6 (c), the valley width between two electrodes of the specimens was less than 5 μm. The resistance linearly decreased from about 600 mΩ at room temperature to about 200 mΩ at 100 K with almost the constant resistivity gradient. The resistivity gradient, however, became slightly large in the lower temperature than about 100 K.

The anomalous conduction accompanying with the change of the resistivity gradient was observed only in the specimen which had an electrode distance less than about 10 μm. On the other hand the resistance decreased linearly as decreasing temperature in normal metal films. It was expected that the observed resistivity anomaly was caused by the influence of excitonic interactions between free electrons in the metallic Au films and electric dipoles excited in the mono-molecular C_{60}-derivative layers. In this stage we cannot discuss, however, more because the measurement of resistivity was done only in the high temperature region, above 77 K. Furthermore the dependence of the resistivity on measurement current was not studied in this work.

We already reported and discussed the similar anomalous resistivity in the similar C_{60}-SAM/ultrathin Au thin films.[6] In that case the resistance decreased rapidly and deviated from the linear dependence around 140 K. Then it was noticed that the temperature dependence changed by the measurement current. When the measurement current was about 10 mA, the anomaly around 140 K disappeared, while the anomaly was observed in the case of comparatively small current, 0.1 mA. Those observed results might suggest a superconductivity break by exceeding critical current I_c. So we will do the same measurement for the specimen in this work. Also the resistivity measurement should be done under high magnetic field. Those intimate experimental investigations and analyses of resistance in the micro-scaled area of the specimens are now proceeded.

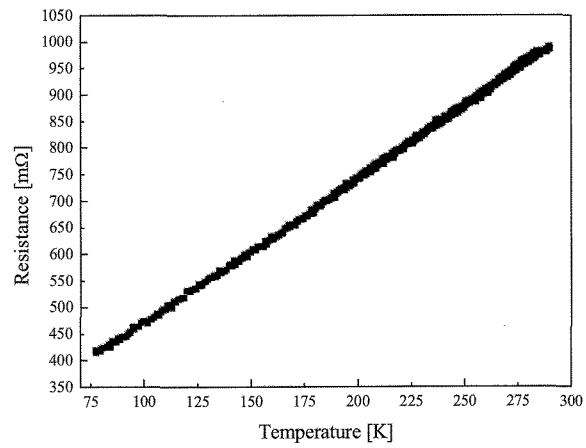
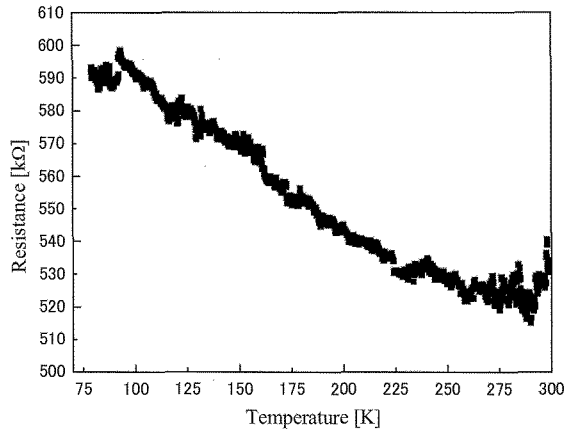
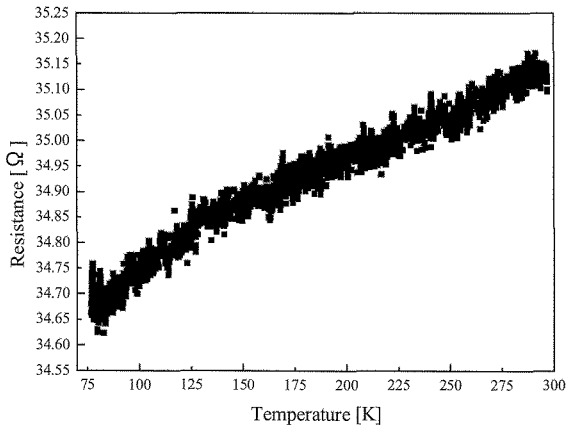


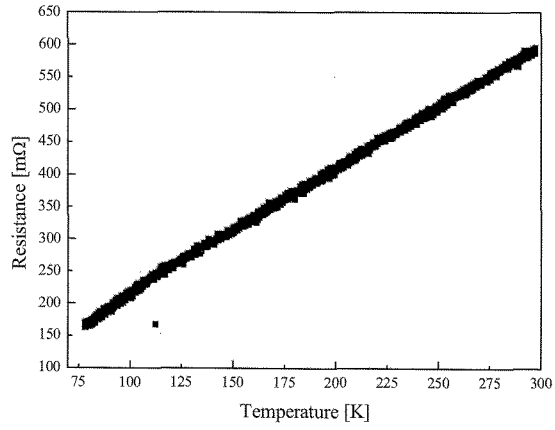
Fig.5 The typical temperature dependence of resistance of the Au//MgO(100) specimen. The resistance decreased linearly as decreasing temperature as expected in the typical metallic conduction.



(a) C_{60}-SAM/Au//MgO(100)



(b) C_{60}-SAM/Au//MgO(100)



(c) C_{60}-SAM/Au//MgO(100)

Fig. 6 Temperature dependence of resistance of C_{60}-SAM/Au//MgO(100). (a) The valley width between two electrodes was *ca.* 16 μm. (b) The valley width between two electrodes was *ca.* 11 μm. (c) The valley width between two electrodes was *ca.* 5μm.

4. Conclusion

The Au ultrathin films were deposited on very flat MgO (100) substrates with Ra less than 1 nm by on-axis RF magnetron sputtering. The C_{60}-SAM adsorbed on the Au films. The morphology of the specimen surfaces was observed by DFM. The some specimens showed the anomalous decreases of resistance which were observed in the temperature range of 100K-125K. The anomalous conduction was observed only in the specimen which had the valley width less than about 10 μm. The observed anomaly may be caused by the influence of excitonic interactions between free electrons in ultrathin Au films and electric dipoles excited in the C_{60}-SAM. However, experimental analyses are not enough to discuss the mechanism of the observed phenomena. The detailed studies are the future subject.

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