Superconductor-Insulator transition in sputtered amorphous MoRu and MoRuN thin films

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Abstract. This work shows the experimental results of the superconductor-insulator (S-I) transition for amorphous molybdenum ruthenium (MoRu) and molybdenum ruthenium nitride (MoRuN) films. These amorphous films onto c-plane sapphire substrates have been interpreted to be homogeneous by XRD and AFM measurements. Electrical and superconducting properties measurements were carried out on MoRu and MoRuN thin films deposited by reactive sputtering technique. We have analysed the data on $R_{sq}(T)$ based on excess conductivity of superconducting films by the AL and MT term and weak localization and electron-electron interaction for the conductance. MoRu films which offer the most homogeneous film morphology, showed a critical sheet resistance of transition, $R_c$, of $\sim 2$ kΩ. This values is smaller than those previously our reported for quench-condensed MoRu films on SiO underlayer held at liquid He temperature.

1. Introduction

In field of condensed matter physics, the origin of two dimensional superconducting-insulator transitions (2D-SIT) is still unresolved [1]. For example, is there a universal critical sheet resistance?, that is quantum resistance, $R_Q = \frac{\hbar}{4e^2}$. If system is completely self-dual, the critical sheet resistance is a universal value, i. e. $R_Q$, independent of the kind of material and disorder. However, despite the amount of experimental work on SIT based on several 2D system over the past forty years, such a universal results had not found. Crauste et al. showed the relationship between effect of annealing and a changing of film thickness for SIT using amorphous NbSi films [2]. And they also reported the chemical composition is not changed and the film structure is still amorphous by annealing. However, by the influence of annealing, superconductivity of films is weaken due to the rearrangement of atoms. The effect appears though the sheet resistance, $R_{sq}$ by the variation of mean free path (mfp) $\ell$ of electron. Because $R_{sq}$ is defined by $R_{sq} = \rho/d$ and $\ell$ is obtain by $\frac{\hbar}{e^2\rho} \times (3\pi^2n^2)^{1/2}$. Here $\rho$, $d$ and $n$ are resistivity, films thickness and carrier density, respectively. In fermionic picture of SIT, the theoretical prediction
implicitly depend on mfp [3]. On the other hand, in bosonic picture, it is independent of kind of disorder for homogeneous 2D superconductor [4]. In this work, we compare results from two techniques on ex-situ method, using sputtering deposition, and in situ method, using an evaporator-cryostat composite, a series of amorphous MoRu films with various thickness.

2. Experimental detail

MoRu and MoRuN films were deposited at room temperature on c-plane sapphire substrates by RF reactive magnetron sputtering method. Collver and Hammond reported the relation between amorphous transition-metal alloys and transition temperature [5]. According to the results, when the electron-to-atom ratio (e/a) is close to 6.4, the $T_C$ has the maximum value of 9.4 K [6]. Therefore, we chosen the sputtering target of Mo75-Ru25 wt% alloys. The background pressure of the chamber was maintained at $5 \times 10^{-5}$ Pa. The relative amounts of argon and nitrogen introduced during sputtering were controlled by mass flow controllers. The total pressure was maintained at 0.6 Pa and the substrate was not heated intentionally during deposition. For MoRuN films, the flow rate of Ar gas was fixed at 50 sccm, while that of N$_2$ gas was of 25 sccm. The microscopic structure was investigated by X-ray diffraction measurement and atomic force microscopy (AFM). The XRD patterns show a broad peak which is the characteristic of amorphous films. And any granular structures are not observed with the surface roughness below 1 nm from the AFM measurements. The results indicate amorphous structure and smooth surface. Resistance and Hall effect measurements were carried out by four probe and Hall bar configuration. Samples were patterned using photolithography techniques and CF$_4$ reactive ion etching. Quench-condensed MoRu films was prepared using an evaporator-cryostat composite system. The results of details are described elsewhere [7,8].

![Figure 1. Temperature dependence of sheet resistance $R_{sq}$ with various film thickness. Numerals denote the film thickness.](image)

3. Results and discussion

Figure 1 shows temperature dependence of sheet resistance, $R_{sq}$ ($T$) with different thickness. We found that the $R_{sq}$ ($T$) shows a monotonic decrease with decreasing temperature down to the lowest temperature in our measurements. For in-situ films, a slight increase in film thickness changes the
transport properties from insulating to superconducting behavior. $R_{sq}(T)$ also did not have reentrant behavior as seen frequently for granular 2D films. We consider the relation between $T_C$ and $R_{sq}$. In Fig. 2, we plot the $T_C$ as a function of the $R_{sq}$ for in-situ [8] and ex-situ MoRu and MoRuN films. We used the $R$ at 10 K as $R_{sq}$ since $R$ was less dependent on temperature around 10K for ultrathin MoRu films with low $T_C$. This figure shows that $T_C$ decreases as $R_{sq}$ for all samples. For MoRu films both series, we fitted the experimental data using expression which applied to homogeneous two-dimensional disordered system with uniform thickness. According to Finkel'stein [3], lowering of $T_C$ from a bulk transition temperature $T_{C0}$ follows the expression:

$$\frac{T_C}{T_{C0}} = \exp\left(-\frac{1}{\gamma}\left[\frac{1+\left(\frac{r}{2}\right)^{1/2}}{\gamma - \frac{r}{4}} - \frac{1-\left(\frac{r}{2}\right)^{1/2}}{\gamma - \frac{r}{4}}\right]\right)^{1/\sqrt{2}},$$  \hspace{1cm} (1)

where $\gamma = \ln\left(k_B T_{C0} \tau_0 '\right)$ which is characterized by $T_{C0}$ and the transport relaxation time (elastic scattering time) $\tau_0'$. $\gamma$ is used as a fitting parameter. And the parameter $r$ is $\left(\frac{e^2}{2\pi\hbar}\right) R_{sq}$. We also used this theory on different system: amorphous Bi [7], Mo-alloy[8,9] and NbN [10]. The red and black theoretical curves for $\gamma = -0.17$ and $\gamma = -0.11$ are fitted to initial gradient of experimental data for in-situ and ex-situ MoRu films respectively. The mfp is obtain from $l = \frac{v}{\nu \tau_0}$, using the above value of $\tau_0'$, where the Fermi velocity $v\nu$ is estimated from the free electron model. From the value of $\gamma$ the transport relaxation time is evaluated to be the $1.1 - 2.2 \times 10^{-16}$ s which, using a typical Fermi velocity, gives a mfp of about few nano meter.

**Figure 2.** The $T_C$ as a function of the $R_{sq}$ for the ex-situ films (closed circle) and in-circle (empty circle) [8], and the red and black solid lines are enhanced Coulomb interaction model [3].
Comparing the reduction of $T_C$ for the two series, we found that change in ex-situ MoRu films are sharper than in-situ MoRu films. By theoretical curve of ref. 3, the value of $R_C$, which means $T_C(R_{sq}^N) = 0$ can be estimated to be nearly 2 kΩ for ex-situ films. On the other hand, it becomes about 5 kΩ for in-situ films. This difference may reflects film structure, that is a homogeneous films for in-situ case and granular film for ex-situ case. The R(T) of ultrathin granular films of conventional superconductors exhibited quasi-reentrant behaviour [8]. In ex-situ films, we can check the film morphology using AFM. On the other hand, it is difficult to observe the film surface of in-situ films using conventional AFM, because the amorphous structure of films change to poly-crystalline one around 180 K. To clarify the difference, we believe that it is necessary to observe the surface while keeping the film at low temperature and in vacuum.

Next, we have carried out the fluctuation analysis to evaluate the quantities. In our present results, the mechanism on suppression of superconductivity might be related to Coulomb interactions using Eq.(1) of Finkel’stein’s theory. Therefore, we need to know these localization and interactions effect for $R_{sq}(T)$ of the region above the $T_C$, i.e., para-conductivity regime, where there is enhanced conductivity due to presence of superconducting fluctuation. The superconducting fluctuation theory is denoted as the form of the excess sheet conductance $1/R_{sq}(T)$ - $1/R_{sq}^N(T)$ for a uniform superconducting film in terms of the following fluctuation conductance

$$\frac{1}{R_{sq}(T)} = \frac{1}{R_{sq}^N} \frac{e^2}{16\hbar} \left(\ln \left(\frac{T}{T_C}\right)\right) + \frac{e^2}{8h \ln \left(\frac{T}{T_C}\right) - \delta} \ln \left(\frac{T}{T_c}\right),$$

(2)

Figure 3. Reduced temperature $\ln(T/T_C)$ dependence fluctuation conductivity. The empty circle are experimental data. The dotted line means the contribution from only the AL theory. The Solid line is a fit to the data using Eq. (2).

where, $\delta$ is a pair-breaking parameter, and $R_{sq}^N$ is the normal state sheet resistance. The second term can be described by the Aslamazov-Larkin (AL) model of fluctuating Cooperpair[11]. The third term is due to Maki-Thompson contributions by a pair-breaking interaction[12,13]. To analyze present data within this work, we use three parameters to describe each curve: $R_{sq}^N$, $T_C$ and $\delta$. We determined $R_{sq}^N$ from the intercept of the straight line in the plots of $1/R_{sq}(T)$ vs $1/T$ around $T_C$. Figure 3 shows normalized inverse
fluctuation conductivity \( \sigma/\sigma^*(= R_{sq}(T)/[R_{sq}^N-R_{sq}(T)] \) as a function of normalized reduced temperature \( \ln(T/T_c) \) at zero magnetic field. The dotted and solid lines are determined from the only AL term and AL + MT terms, respectively. Experimental data cannot be fitted with only AL term, represented by the dotted line. Use of AL + MT term, the data can be explained well by the fluctuation theory. We think that the agreement means the homogeneity of films. According to weak localization theory, the \( \delta \) is represent by \( \delta = \pi \hbar/(8k_B\tau_m) \) using the inelastic scattering rate \( \tau_m \) [14]. In addition, the rate of \( 1/\tau_m \) can be expressed as

\[
\frac{1}{\tau_m} = \frac{1}{\tau_{\text{nc}}} + \frac{1}{\tau_{e-e}} + \frac{1}{\tau_{e-ph}} = \frac{e^2}{2\pi \hbar} R_{sq}k_B T \ln \left( \frac{2 \ln 2}{\eta + \beta} \right) + \frac{e^2}{2\pi \hbar} R_{sq}k_B T \ln \left( \frac{\pi \hbar}{e^2 R_w} \right) + \frac{14 \pi \zeta(2) k_B}{\hbar} T^3, \tag{3}
\]

where \( \eta = \ln(T/T_c) \), \( \zeta \) is the zeta function, \( \lambda \) is the electron-phonon interaction strength and \( \Theta_D \) is the Debye temperature[15,16]. The first, second and third terms on the right side can be expressed as the \( \delta_{\text{nc}} \) due to the superconducting fluctuation effect, \( \delta_{ee} \) due to the electron-electron interaction, and \( \delta_{eph} \) due to the electron-phonon interaction, respectively. The first and second term are given by \( R_{sq}^N \) at a fixed value of \( \eta \). The third term is the origin of \( \delta_{\text{ph}} \) in the above expression \( \delta = \delta_0 + a(R_{sq}^N)^b \). For details of the estimation procedure on of \( \delta \), see our previous work [9]. As a results, we obtain that the red solid line in Fig. 4 is calculated from eq. (3) at \( \eta = 0.1 \) under the assumptions \( \Theta_D = 750 \) K which is the same as that of NbN [17] and MoN [9] and \( \lambda = 1 \). It is similar that the \( R_{sq}^N \) dependence of \( \delta \) show the effect of weak localization and Coulomb interaction in spite of there is deference of \( R_C \) between in-situ films and ex-situ film. Though we do not know yet the \( R_{sq}^N \) dependence. To obtain the qualitative explanation, it is need to study the magneto-conductance for clarification of the \( R_{sq}^N \) dependence of \( \tau_m \). We will discuss an analysis of magneto resistance for \( \tau_m \) in the near future.

![Figure 4](image)

**Figure 4.** \( R_{sq} \) dependence of the \( \delta \) as a function of the \( R_{sq} \) for ex-situ and in-situ MoRu films.

4. **Summary**

We have demonstrated S-I transition using MoRu and MoRuN films with various thickness deposited by reactive DC magnetron sputtering method. In addition, we compared the experimental results
between in-situ and ex-situ films. As for $R_{\text{sq}}$ dependence of $T_C$, it seems that the data can be explained by enhanced Coulomb interaction theory. It is important to note that this result for the thickness-tuned SI transition has been obtained for MoRu films. Since there is no report, as far as we know, on the electron tunneling measurement of MoRu films near the thickness-tuned S–I transition, it would give us further information as to the effects due to the difference of films deposition method. Although we believe that this difference come from film structure, a full understanding of the thickness-tuned SI transition in a thin MoRu film would need further investigation from the point of view of the electrical transport properties combined film structure analysis.

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