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# Suppression of the Berezinskii-Kosterlitz-Thouless Transition in 2D Superconductors by Macroscopic Quantum Tunneling

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## Abstract

The evolution with thickness of the properties of quench-deposited homogeneous amorphous bismuth (*a*-Bi) thin films with a 14.67Å amorphous antimony (*a*-Sb) underlayer has been studied. In contrast with the results of previous investigations on similar systems the transition between the insulating and superconducting regimes is not direct, but involves an intervening metallic regime over a range of thicknesses. For these metallic films the temperature dependencies of the resistances at temperatures above the metallic regime can be described by the Halperin-Nelson form suggesting the occurrence of a Berezinskii-Kosterlitz-Thouless (BKT) transition at lower temperatures. However, this transition never occurs as curves of  $R(T)$  flatten out as temperature is reduced. We suggest that this phenomenon is evidence of a crossover between a classical regime of thermal vortex unbinding at high temperatures and a regime of macroscopic quantum tunneling (MQT) at low temperatures. The latter prevents the BKT transition from occurring.

Over the last two decades, the electrical transport properties of disordered thin films near the onset of superconductivity have been studied extensively, especially in the presence of magnetic fields.[1–5] Of particular interest are the many reports that the resistance at the lowest temperatures becomes independent of temperature near the SI transition. This has been found with many different tuning parameters[2, 6–8]. These results have been interpreted in many different ways, which include the formation of a metallic state, the occurrence of macroscopic quantum tunneling, or just the failure to cool the carriers. The physics in this regime is still unclear. At the same time the physics of the Berezinskii-Kosterlitz-Thouless (BKT) transition has reappeared in the explanation of fluctuation phenomena in thin films [9], in systems exhibiting interfacial superconductivity [10], and in layered high temperature superconductors [11].

The behavior of the electrical transport properties near the superconductor-insulator (SI) transition should be affected by the degree of homogeneity of the order parameter near the transition. Some theoretical works have demonstrated the possibility that superconducting thin films undergoing a transition to the insulating regime as a function of disorder or magnetic field, may break into superconducting islands[12, 13]. Experimentally, scanning tunneling spectroscopy investigations of amorphous  $InO_x$  and polycrystalline  $TiN$  films near the transition temperature have revealed the presence of small superconducting patches embedded in an insulating background[14]. It is still unclear whether this is due to spacial chemical composition variations in these binary compound films or is an intrinsic property of highly disordered films in which the carriers are strongly localized. However, in either case, it is reasonable to treat such systems in a manner analogous to random Josephson junction arrays (JJAs). The two-dimensional ordered JJA has been well studied and has a fully developed theoretical description. The similarity between

quasi-2D thin films and JJAs is well known and been used to explain the conductivity on the insulating side of the SI transition[15]. There are also some theoretical works on the SI transition that are based on the JJA framework[16, 17]. Quite recently there has been work on the influence of quantum fluctuations on the BKT transition in films modeled as disordered JJAs [18].

In this letter, we present electrical transport measurements on a sequence of *in situ* deposited amorphous bismuth (*a*-Bi) films with a 14.67Å amorphous antimony (*a*-Sb) underlayer. An *ex situ* atomic force microscope (AFM) scan revealed that these films do not consist of isolated grains but are continuous, but with a significant thickness variation. Electrical transport measurements in the absence of magnetic field show a high temperature behavior indicative of a BKT transition. However this transition is not realized at low temperatures. This suggests that the vortex-antivortex binding phenomenon associated with the BKT transition is prevented from occurring by macroscopic quantum tunneling (MQT) processes. In the absence of a detailed theory it is found that aspects of models of ordered JJAs can be used to interpret the data.

The sequence of *a*-Bi films reported here was grown by quench-condensation *in situ* at liquid helium temperatures on a (100)  $SrTiO_3$  (STO) single-crystal substrate. First, platinum electrodes, 100Å thick, were deposited *ex situ* onto the substrate's epi-polished front surface to form a Hall bar configuration. A 14.67Å thick underlayer of *a*-Sb was used to precoat the substrates *in situ*. This underlayer exhibited zero conductance within instrumental resolution. The experiments involved repeated cycles of deposition and measurement carried out in a dilution refrigerator system designed to study the evolution of electronic properties with film thickness [19]. The sample measurement lines were heavily filtered so as to minimize the electromagnetic noise environment of the film. The approach was to use RC filters at 300 K (in series

with  $10k\Omega$  resistors) to attenuate 60 Hz noise, Spectrum Control #1216-001 pi-section filters at 300 K to attenuate radio frequency noise, and 2 m long thermocoax cables at the mixing chamber stage of the refrigerator to attenuate GHz Johnson noise from warmer parts of the refrigerator. All the transport measurements involved the use of a four-terminal configuration employing a DC current source with currents always in the linear regime of the current-voltage (I-V) characteristic. Examples of I-V characteristic and differential resistance ( $dV/dI$ ) are presented in the accompanying supplementary material in which the possible role of failure to cool the electrons is also discussed [20].

For quench-deposited *a*-Bi films, an important feature of the morphology is the thickness variation, which can be characterized by AFM. Unfortunately, such AFM studies cannot be carried out *in situ*. Thus, it is not possible to characterize every film during the process of sequential deposition. However, it is possible to characterize the thickest film, after the transport measurements are completed. In fact, the nominal thickness of the first film which exhibited measurable conductance below 1K was 19.74Å. The thickness at the onset of superconductivity was 22.24Å, and the last film was 23.42Å in thickness. The total thickness increment was 3.68Å over the entire thickness-tuned SI transition and was 1.18Å over the conductive branch of the SI transition. Therefore, apart from some dramatic structural change over this limited range of nominal thickness variation, the characterization of the surface of the last film should represent the thickness variation of the films on the conductive branch of the SI transition.

To carry out these measurements, the 23.42Å thick film was warmed slowly back up to room temperature. Figure 1(a) shows a  $300nm \times 300nm$  scan of the film. The average root mean square roughness,  $R_{rms}$  is 3Å relative to a total nominal thickness of 38.09Å, with 23.42Å of amorphous bismuth on top of a 14.67Å thick layer of amorphous antimony. This suggests that the film is well connected but with a thickness variation around 13% of the bismuth thickness, which is smaller than the total thickness. The cross-sectional analysis is shown in Fig. 1(b). It is reasonable to suggest that films with this structure near the SI transition have patches of nonzero order parameter as envisioned by Ghosal, Randeria and Trivedi [12], and might be expected to behave as random JJAs near the SI transition. Significant roughness is not found for film sequences exhibiting direct SI transitions tuned by either thickness, magnetic field or charge[21].

The I-V characteristics of the films were non-hysteretic and were linear at low currents, and non-linear when the applied currents were larger than  $1\mu A$ . However, at such large currents, Joule heating is significant, leading us to conclude that this non-linearity is due to heating. Therefore, the I-V characteristic cannot provide information on the critical currents of the films. The resistances vs.

temperature of *a*-Bi films with thicknesses ranging from 22.24Å to 23.42Å, and measured in linear I-V characteristic regimes, are plotted in Fig. 2. Instead of developing global superconductivity, the resistances of these films flatten out at the lowest temperatures.

To analyze the high-temperature part, we first define the resistance at 9K as the normal resistance  $R_N$ . Then we find the trend of resistance decreasing can be well described by the Halperin-Nelson form:

$$R(T) = R_0 \exp \left[ -b / (T - T_{BKT})^{1/2} \right] \quad (1)$$

where  $T_{BKT}$  is the BKT transition temperature [22]. This is the temperature above which vortex-antivortex pairs unbind thermally. Here  $R_0$  and  $b$  are constants. In the fits shown in Fig. 2, values of  $R_0$  are in the range from  $1.0 \times 10^5$  to  $1.7 \times 10^5$  while the values of  $b$  are from 2.5 to 3.2. The solid line in Fig. 2 represents this form for the resistance above the BKT transition. We find that in the regime between  $90\%R_N \sim 10\%R_N$ , the measured resistance values deviate by less than 3% from the fit. From the fit, the putative transition temperature,  $T_{BKT}$ , can be obtained for films of different thicknesses, as shown in Fig. 3(a).

However, the data points of  $R(T)$  flatten out at the lowest temperatures. As shown in the inset of Fig. 2, this flattening happens at higher temperatures in thicker films, whose resistances are smaller than those of thicker films and therefore should produce less Joule heating at the same applied current. This suggests that the flattening is not due to either heating or the failure to cool the carriers, and can be taken as evidence of MQT below  $T_{BKT}$ .

We then use the value of resistance at 50mK as the resistance associated with MQT, and substitute it into the Halperin-Nelson form, *i.e.*, Eq. (1) to obtain a crossover temperature  $T_{cr}$ , between thermal and quantum processes, as shown in the inset of Fig 2. In Fig. 3(b), we plot  $T_{cr}$  versus  $T_{BKT}$  and find that the two temperatures are linearly proportional to each other.

In the absence of a detailed theory, we turn to modeling these films as JJAs. The non-hysteretic I-V characteristics suggest that the films are in the overdamped limit. Although they would be random arrays we will use the framework developed for ordered arrays.

The BKT transition temperature of an ordered array can be expressed as:

$$k_B T_{BKT} = (\pi/2) E_J \quad (2)$$

where  $E_J$  as the Josephson coupling energy of a single junction.[23] If the films behave as JJAs, the Josephson coupling energy  $E_J$  should increase with increasing thickness because the localized superconducting order becomes more robust. From Eq. (2),  $T_{BKT}$  would then be

expected to increase with increasing thickness, which is consistent with the data in Fig. 3(a). In the JJA model, a vortex moving in real space is analogous to the motion of the phase of a single junction in a tilted washboard potential[24]. We hypothesize that the crossover temperature  $T_{cr}$ , remarking the boundary between the thermal and quantum vortex motion regimes, calculated for a single junction applies to an array. This temperature increases with the plasma frequency, which is linear proportional to the coupling energy  $E_J$ . In the small current and overdamped limits, it has been estimated to be:

$$k_B T_{cr} = (R/R_Q) E_J \quad (3)$$

where  $R_Q = h/4e^2 \approx 6450\Omega$  is the quantum resistance of electron pairs and  $R$  is the shunt resistance of a single junction [23]. Substituting Eq. (2) into Eq. (3), we obtain  $T_{cr} = (2R/\pi R_Q) T_{BKT}$ , assuming that system is overdamped and that the effective shunt resistance remains nearly constant for films of different thicknesses. This is qualitatively consistent with the data plotted in Fig 3(b). However there is a non-zero value of  $T_{cr}$  even when  $T_{BKT} \rightarrow 0$ . This indicates that a crossover between the thermal and quantum regimes happens even when the  $T_{BKT} \rightarrow 0$ . This is not included in the JJA model but may emerge in a theory which directly treats the affect of quantum fluctuations on vortex unbinding and quantum motion of vortices.

The curves of  $R(T)$  in low perpendicular magnetic field also flatten out at the lowest temperatures. In Fig. 4, we show an Arrhenius plot of resistance vs. inverse temperature at fields ranging from 0.01 to 3 T in the temperature range from 100mK to 500mK. Over a significant temperature range, the resistance at field higher than 0.4T can be fit by an Arrhenius form.

Now, we focus on the branch of the data at low temperatures for which  $dR/dT > 0$ . The resistance also deviates from the Arrhenius form for films in low magnetic fields as in the zero field case. Moreover, the deviation happens at higher temperature for lower fields. This observation can be used to argue that the charge carriers are not failing to cool, because the films have a lower resistance and therefore lower dissipation in lower magnetic fields. As in the zero-field case, these temperature-independent resistances at low temperature may be due to MQT of vortices in the tilted washboard potential. Previous experimental results on ordered JJAs in perpendicular magnetic fields also similar to the present findings with *a*-Bi thin films [25].

In aggregate these results support the idea of that strongly disordered superconducting films may break up into islands near the SI transition. Although theory predicts these islands may form intrinsically[12, 13], in the present instance the islands may be associated with structural inhomogeneity. The AFM scans reveal that the films are continuous, but with a 13% thickness variation

on mesoscopic length scales. This is in contrast with a direct SI transition, without an intervening metallic regime, found for smoother films. The data in all regimes appear to be qualitatively consistent with a model of overdamped JJAs. Curves of  $R(T)$  at higher temperatures in zero field are consistent with the form predicted for the BKT transition. At lower temperatures, the resistances become temperature-independent with and without presence of perpendicular magnetic field, which suggests that the electrical transport is dominated by MQT effects.

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## Figures

Figure 1: (a) Surface height AFM scan of the 23.42Å thick *a*-Bi film. The scan is taken *ex situ* after a slow warming process. (b) Cross sectional analysis of a horizontal cut in (a). It shows that the surface has thickness variations, but the film is well connected.

Figure 2: Sheet resistance vs. temperature of 22.24Å (top), 22.36Å, 22.63Å, 22.89Å, 23.15Å, and 23.42Å (bottom) thick *a*-Bi films in zero magnetic field. The solid lines are a fit by the Halperin-Nelson form. The same set of  $R(T)$  curves at low temperature is enlarged as shown in the inset to emphasize the deviation of the data from the Halperin-Nelson form at low temperatures. The horizontal dashed lines represent the resistances at 50mK and their intersections with solid lines are taken to be the crossover temperatures.

Figure 3: (a) Berezinskii-Kosterlitz-Thouless transition temperature versus thickness of sequentially-deposited *a*-Bi films in zero magnetic field. The transition temperature  $T_{BKT}$  is obtained by fitting the resistances with Eq. (1). (b) Crossover temperature plotted vs. the BKT temperature. The crossover temperature is obtained by inserting the value of the flattened resistance into Eq. (1). The straight line is a guide to the eye.

Figure 4: Arrhenius plot of the resistance of the 23.42 thick film in a perpendicular magnetic field. The magnetic fields applied are (bottom) 0.01, 0.05, 0.1, 0.15, 0.2, 0.4, 0.6, 0.8, 1, 1.4, 2T (top). The straight lines are guide of eye for Arrhenius fits. One can see that in the low fields, the resistance flattens out at low temperatures.

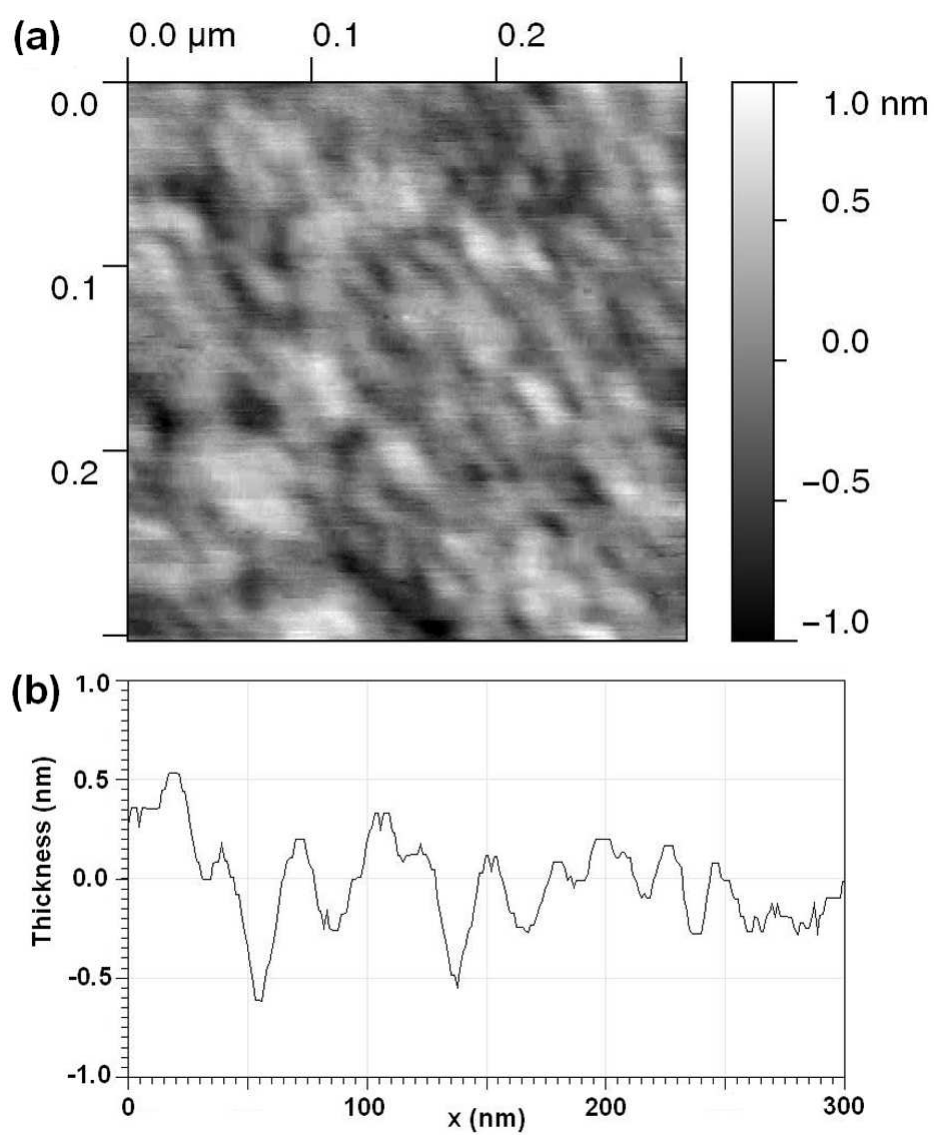


Figure 1 LL12739 15May2012

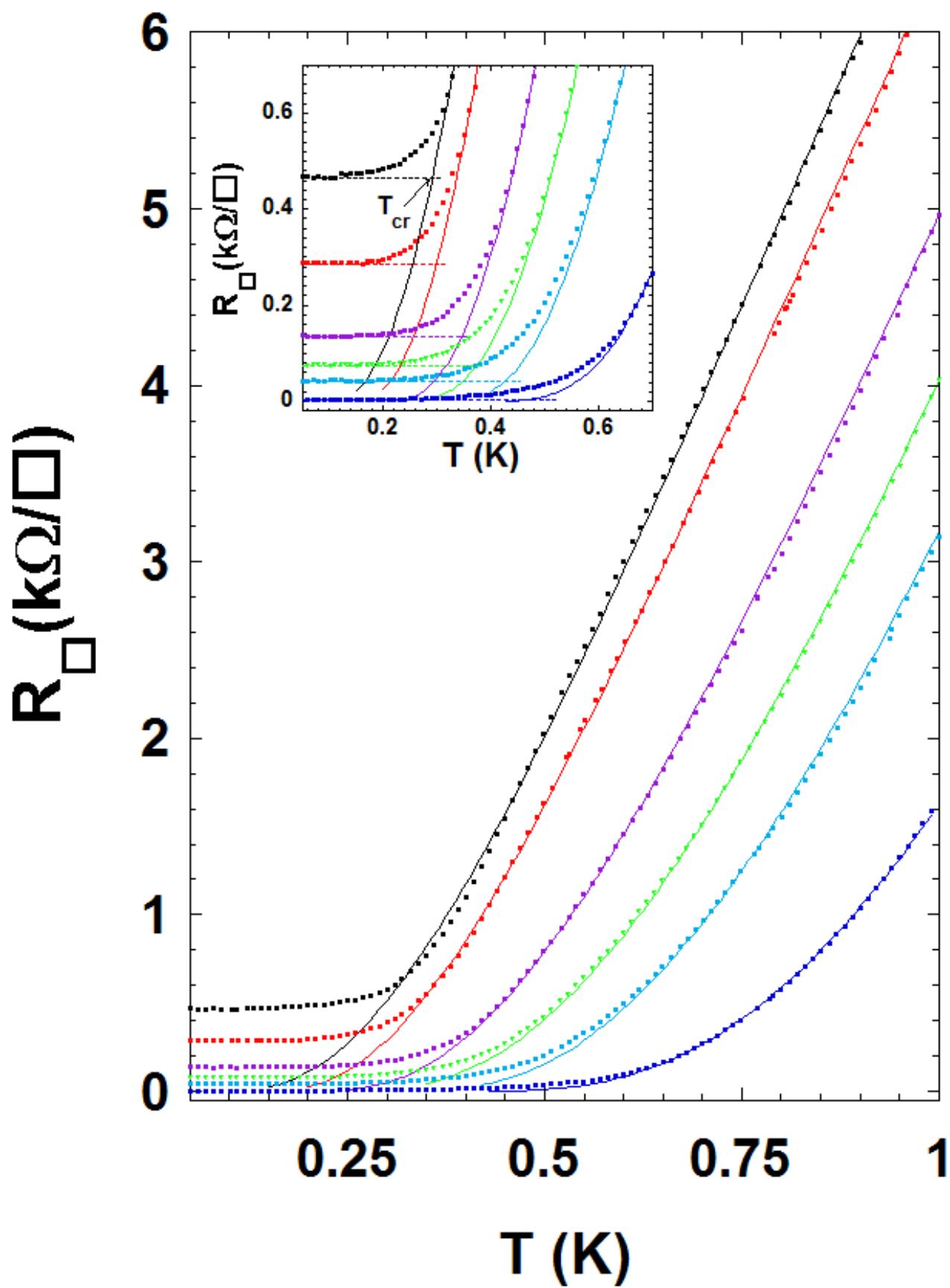


Figure 2 LL12739 15May2012

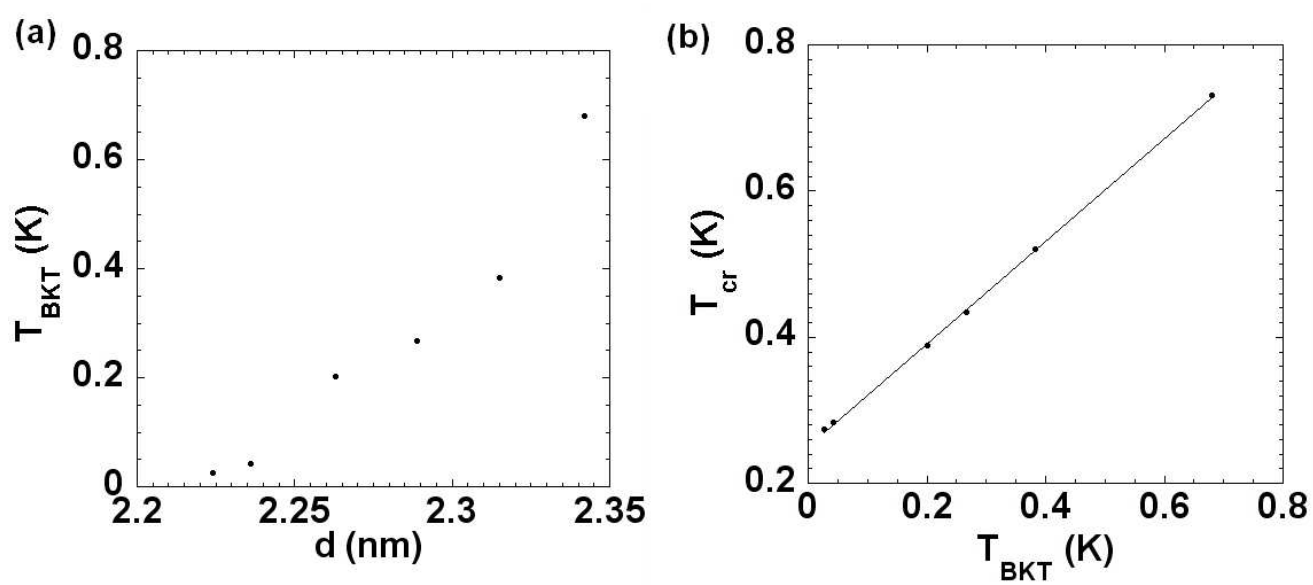


Figure 3 LL12739 15May2012

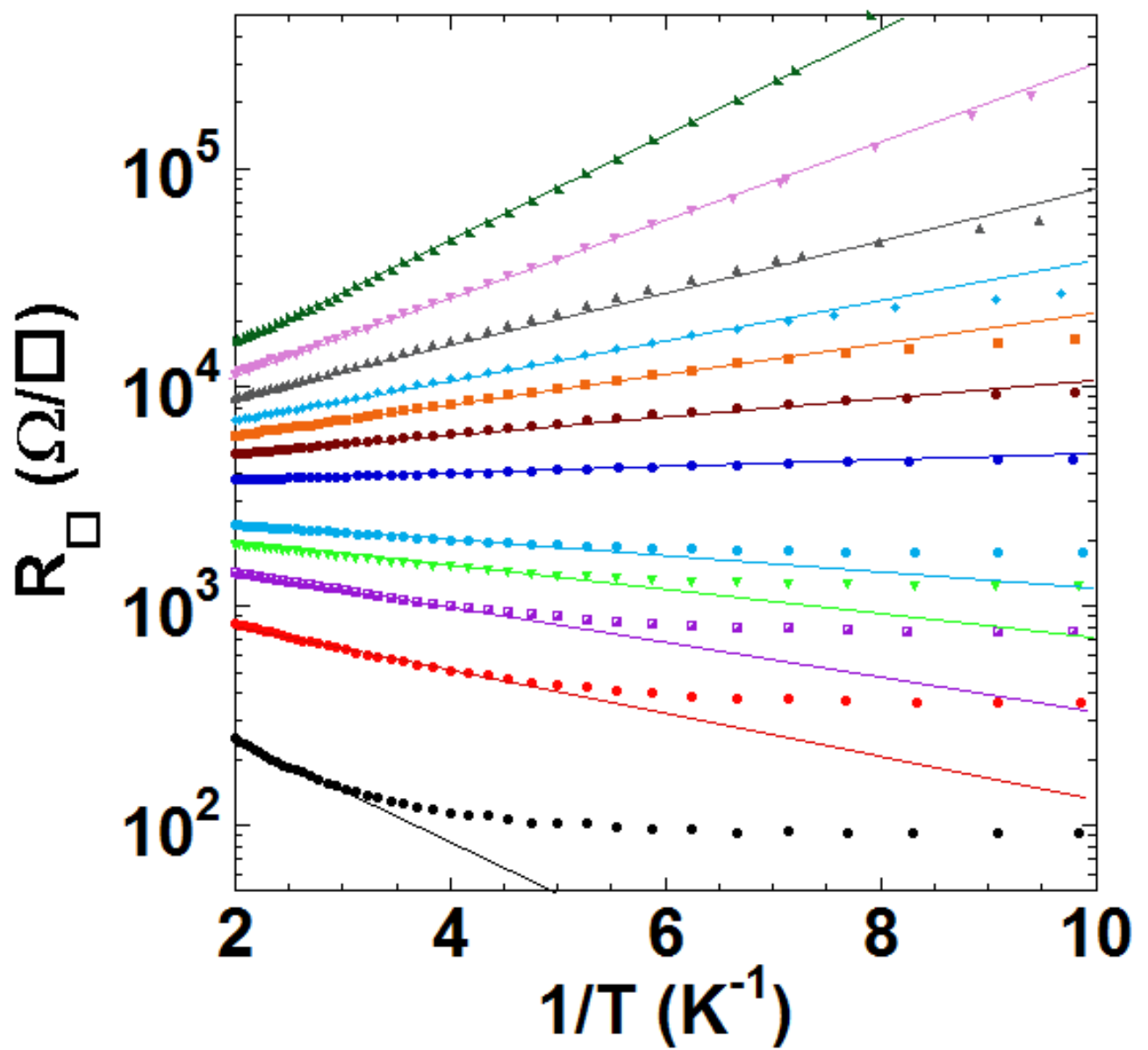


Figure 4 LL12739 15May2012