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The role of mesoscopic disorder in determining the character of the field-induced insulating regime of amorphous ultrathin films

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ABSTRACT

This work documents the conditions under which the field-tuned behavior of quench condensed films grown on relatively thick layers of a-Sb is essentially identical to that found for amorphous InO_x and polycrystalline TiN_x films. The electrical transport properties of a series of amorphous Bi (a-Bi) films of different thicknesses, grown with a 14.67 Å thick underlayer of amorphous Sb (a-Sb), were studied in perpendicular and parallel magnetic fields. A magnetoresistance (MR) peak was found in insulating films in both perpendicular and parallel magnetic fields. In all insulating films, Arrhenius type conduction was also found over all ranges of magnetic field. Neither behavior is found for a-Bi films grown on top of thinner a-Sb underlayers, which were substantially smoother. These observations, together with a quantitative analysis of film roughness profiles, highlight the role of mesoscopic scale thickness fluctuations in nucleating superconducting clusters or islands that apparently lead to the MR peak and Arrhenius conduction. This implies that the nature of the disorder plays a major role in determining the character of the insulating regime.

1. Introduction

The superconductivity of two-dimensional (2D) and quasi-2D films has been of interest because of its quantum critical behavior and the similarity of several of its properties to those of oxide superconductors. Recently magnetic fields applied perpendicular to the film plane have been the most extensively used tuning parameters in the study of superconductor–insulator (SI) transitions. There are many reports of large magnetoresistance (MR) peaks in the insulating regimes of such films [1–6]. Similar MR peaks have also been found by applying perpendicular magnetic fields to insulating films close to their disorder-tuned SI transitions [7]. In addition to the appearance of giant MR peaks, curves of resistance vs. temperature, $R(T)$, follow an Arrhenius form,

$$R = R_0 exp(T_0/T) \tag{1}$$

in magnetic fields near those associated with the magnetic-field-tuned SI transitions and the MR peaks [2,4,8]. This behavior is different from the variable range hopping usually found in disordered electron systems. The current–voltage (I–V) characteristics are also found to be non-linear in this regime [9,10]. It has been suggested that these unusual phenomena are associated with a special insulating state containing Cooper pairs. However, the detailed physics of the SI transition and of these phenomena is not clear.

The measurements exhibiting the above described phenomena were carried out on nominally homogeneous films of amorphous InO_x and polycrystalline TiN_x ranging in thicknesses from tens to hundreds of Angstroms, and on nano-honeycomb patterned amorphous Bi (a-Bi) films grown on alumina substrates. In work on much thinner a-Bi films prepared by quench-condensation onto very thin amorphous Sb (a-Sb) underlayers, a MR peak is never observed over a substantial range of parameter space. Such films are homogeneously disordered, smoothly covering the substrate because of the underlayer [11], and they exhibit a direct perpendicular field driven SI transition [12]. Also, the conductivity on the insulating side of the SI transition is found to be describable by variable range hopping [13,12]. However, in recent work on quench-condensed a-Bi films grown on top of relatively thick amorphous Sb (a-Sb) underlayers, giant magnetoresistance peaks and Arrhenius type conduction are found in the insulating regime [14].

The present work documents the conditions under which the field-tuned behavior of the insulating regimes of quench condensed films grown on relatively thick layers of a-Sb is essentially identical to that found for amorphous InO_x and polycrystalline TiN_x films. It addresses how the thicknesses and morphologies of a-Sb underlayers govern this behavior. To understand the role of the thickness of the underlayer, the morphologies of a-Sb films of different thicknesses were studied by atomic force microscopy (AFM). Thicker a-Sb films exhibit a granular-like structure with thickness fluctuations that increase with thickness. Substantially rougher a-Bi films are formed when they are grown on these relatively

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thick a-Sb underlayers. Such films exhibit MR peaks and Arrhenius conduction. This correlation suggests that roughness is essential to the observation of effects hitherto associated with amorphous InO_x and polycrystalline TiN_x films. Thickness fluctuations apparently aid in the nucleation of the superconducting clusters or islands that may be essential for the observation of magnetoresistance peaks and Arrhenius conduction.

2. Experimental methods

Ultrathin a-Sb and a-Bi films in this work were grown by the technique of quench-condensation. This involves the deposition of films onto substrates cooled to liquid helium temperatures, a procedure introduced by Shal'nikov [15]. In order to obtain reliable data one must perform depositions with precision, carefully tracking film history, and maintaining a consistently clean vacuum environment. A system was built to achieve this goal [16]. Its three main parts are an Oxford Kelvinox 400 dilution refrigerator, an ultrahigh vacuum (UHV) thermal molecular beam epitaxy (MBE) deposition system, and a liquid helium flow-through cryostat. This combination allows for the electrical transport properties of films to be studied *in situ*, without warming or removing them from vacuum. The dilution refrigerator has a cooling power of 400 μW at 100 mK and a base temperature of approximately 0.004 K. The vacuum condition of the MBE system during deposition is maintained in a pressure range of low 10^{-9} to high 10^{-10} torr. The distance between the Knudsen cell sources and substrate is 61 cm, which ensures that there is uniformity in the vapor flux density at the sample. Film thicknesses were monitored by a calibrated quartz crystal microbalance (QCM) during the deposition. The liquid helium flow-though cryostat can keep the sample temperature from exceeding 8 K during film growth.

Transport properties of the films were measured using a standard four-terminal technique. The sample measurement lines were heavily filtered so as to minimize electromagnetic noise. The starting point of the measurements involved first identifying the linear regime of the current–voltage (I–V) characteristic, and choosing two bias currents in this regime. The resistance was then determined by measuring the voltages at these two bias currents and computing the slope. The temperature of the dilution refrigerator was determined using a calibrated resistance thermometer measured using a self-balancing resistance bridge. The samples have been shown to be in thermal equilibrium with the dilution unit down to about 60 mK [17]. Measurements below 1 K were made by setting the temperature of the dilution refrigerator with a temperature controller. This could be done with a precision of 0.5 mK. Above 1 K, a temperature range where the dilution refrigerator temperature could not be easily stabilized, resistance measurements were obtained by slowly cooling at a rate of about 0.05 K per minute. The maximum applied magnetic field was 10 T. The rate of change of the external magnetic field was kept below 0.1 T/min to avoid Eddy current heating.

In the experiments reported here, samples could be positioned with their planes either parallel or perpendicular to the applied magnetic field. On the sample holder, a specially designed screw with a hollow ring at its end was attached to the sample platform through a joint pin. The sample holder was optimized to minimize errors in the alignment of the film both perpendicular and parallel to the applied magnetic field. The misalignment was estimated to be less than 3 degrees in these two positions. During tests, several calibrated resistance thermometers were attached to the sample holder at different positions. The sample was rotated while the sample holder was attached to the dilution refrigerator. We found that the cooling power from the refrigerator could keep the sample below 4 K during the entire process of rotation.

The transport data of the present work were obtained from studies of homogeneous a-Bi films that were sequentially grown by quench-condensation *in situ* at liquid helium temperatures on (100) STO substrates precoated, also *in situ*, with a 14.67 Å underlayer of a-Sb. The underlayer exhibited zero conductance within instrumental resolution. In a sequential thickness study, the sample is transferred to the dilution refrigerator for *in situ* measurement right after a deposition step. When the measurements are finished, another deposition is performed to increase the thickness of the sample. This process is repeated until the sample becomes superconducting.

After all the electrical measurements were completed, the thickest film of the set was also warmed slowly back up to room temperature and its thickness variation was determined by AFM. Although we can only characterize the surface variations of the thickest film of the whole series of films, the total thickness change was only about 3 Å over the entire thickness-tuned SI transition. Therefore, it is reasonable to assume that the thickness variation established at an early stage of film growth and the thickness variation of the thickest film are representative of the whole series of films. Furthermore it has been shown that mesoscopic scale features of ultrathin quench condensed films do not change during warming [18]. Thus, *ex situ* AFM studies would be expected to reveal mesoscopic scale features present *in situ*.

In order to compare the morphologies of a-Sb underlayers of different thicknesses, 10 Å, 15 Å, and 20 Å thick a-Sb films were grown separately by quench-condensation on (100) $SrTiO_3$ (STO) substrates using growth parameters identical to those employed for the growth of the films of the transport studies. These a-Sb films were warmed slowly up to room temperature to preserve their structures. Then their thickness variations were determined by AFM.

3. Results

In this section, after presenting details of the characterization of film roughness, we present detailed transport measurements to establish that films grown on thick a-Sb underlayers exhibit most of the full range of properties found for films of amorphous InO_x and polycrystalline TiN_x. Films are categorized as belonging to the superconducting regime if their resistances decrease with decreasing temperature and to the insulating regime if they increase. We compare measurements of the resistance of a sequence of films in the insulating branch in the presence of perpendicular and parallel magnetic fields with the results of work on amorphous InO_x and polycrystalline TiN_x. Finally, the magnetoresistance of films in the superconducting branch in perpendicular field is presented.

3.1. Characterization of the surfaces of amorphous antimony and amorphous bismuth films

Representative 500 nm × 500 nm AFM surface height scans of 10 Å, 15 Å, and 20 Å thick a-Sb films are shown in Fig. 1(a)–(c) respectively. Note that these figures share the same false-color scale. A well-defined terrace structure of the STO substrate can be easily observed in Fig. 1(a) but cannot be seen in Fig. 1(b) and (c). Furthermore, the 20 Å thick film exhibits granular-like features with a grain size of around 5–10 nm. This suggests that the growth mode depends upon thickness, which is consistent with previous *in situ* STM studies of ultrathin quench condensed Au films [18]. Although films grown by quench-condensation are amorphous, in the case of these Sb films the growth follows the Stranski–Krastanov mode [19]. It is layer-by-layer for the first 10 Å but columns which appear as grains or islands form when the films become thicker than 10 Å. This