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Coupling of gate-induced superconducting polythiophene layers through an insulating part

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We report on the enhancement of the critical temperature T_c for gate-induced superconductivity in polythiophene thin films using superconductor-insulator-superconductor stacks. The increase of T_c is ascribed to the coupling of the superconducting layers through an insulating barrier. This might be explained by an effective screening of the Coulomb interaction of charge carriers within different layers. Using a combination of superconducting polythiophene and lead layers evidence is found for coupling of charges of the two different layers via phonons.

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Recently, it has been demonstrated that the electrical properties of various materials can be tuned from insulating to superconducting using the field effect.[1–8] In the case of organic materials[5–8] the superconductivity is confined to a thin two-dimensional layer at the interface to an insulating layer. Several studies have suggested an influence of the insulator/superconductor-interface on the superconducting properties of the material.[9–12] Moreover, a significant dependence of the transition temperature T_c on film thickness has been observed in superconductor-insulator multilayers.[13–17] Several models have been proposed in order to explain such phenomena.[9–12] Here, we investigate the superconducting properties of polythiophene thin films, especially, the coupling of a superconducting film to a second superconductor (polythiophene or lead) via an insulating barrier, i.e., undoped polythiophene.

Field-effect devices were prepared on glass substrates using a sputtered Mo film as gate electrode and Al_2O_3 as gate insulator.[8] The electrical properties of the accumulation layer at the interface of the polythiophene [PT/regioregular poly(3-hexylthiophene)] film and the insulator were investigated by four-probe measurements using gold contacts[8] in the temperature range from 1.7 to 20 K. Thin (50–350 Å) PT films were solution cast from a dilute solution.[18,19] The thickness of the films was determined from the optical interference pattern. Due to regioregular head-to-tail coupling of the polymeric chains a nanocrystalline lamella structure is obtained giving rise to reasonably high charge carrier mobilities.[18–20] It has been demonstrated that gate-induced superconductivity can be observed in PT below ~2.4 K at high negative gate voltages (~−130 V).[8] The degree of self-organization has been found to be of extreme importance in this polymeric material, since disorder suppresses the occurrence of superconductivity in this material.[8] Without applied gate-bias the PT film is insulating ($\rho > 10^5\ \Omega$ cm). In addition, a second field-effect transistor (FET) was prepared on top of the thin film (see Fig. 1). For some devices a layer (~1500 Å) of Pb was evaporated onto the PT film instead of the second FET. The superconducting transition temperature of such Pb films was approximately 7.3 K, in accordance with values reported for bulk Pb. The coupling between the two superconducting layers (PT-PT or PT-Pb, respectively) through the undoped, insulating PT barrier is investigated. We would like to mention that the occurrence of superconductivity in PT was suppressed at too high gate voltages (~−150 V), which might be a result of correlation effects. However, in the gate voltage range, where superconductivity can be achieved, no variation of T_c was observed. The same gate bias (−135 V) was used throughout all experiments in order to achieve superconductivity.

Moreover, PT Josephson junctions have been prepared by patterning the gate-electrode, giving rise to a spatial modulation of the charge carrier density leading to in-plane superconductor-insulator-superconductor (SIS) structures.[21] Such devices can be used as tool to study the electron-phonon coupling[21–23] in the PT layers via tunneling spectroscopy. In addition, tunneling curves of the vertical SIS structure have been investigated. No signs of Josephson coupling have been observed in such structures.

Figure 2 shows the channel resistance of the top and bottom PT transistor for a 50 Å thick film. A T_c of 2.5 K can be estimated for either individual device. However, if superconductivity is induced in both, the top and bottom layer, the transition temperature is increased up to 4.4 K. Using PT films of different thickness an increase of T_c below 150 Å is observed (inset of Fig. 2). A similar effect can be noticed for

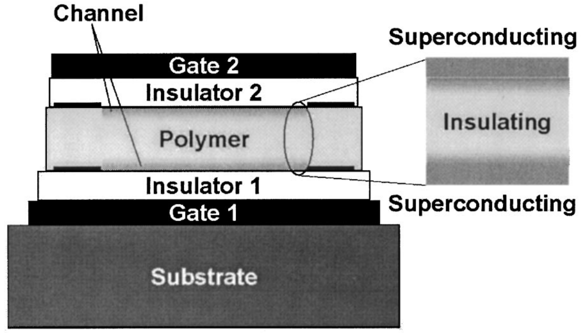

FIG. 1. Schematic device structure. A PT field-effect transistor is prepared on a glass substrate using a Mo-gate electrode (gate 1) and an Al_2O_3 gate insulator (insulator 1). In addition, a second transistor is prepared on top of the polymer film using a second Al_2O_3 layer (insulator 2) and an Au electrode (gate 2). By applying a high gate voltage superconductivity can be induced in a thin layer close to the PT/Al_2O_3 interfaces. The coupling of the superconducting layers through the insulating (PT) layer is studied.

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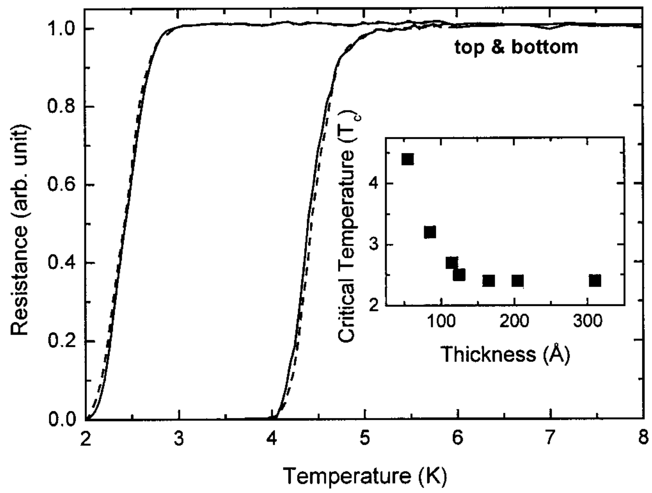

FIG. 2. Channel-resistance as a function of temperature for the top (dashed) an the bottom (solid) transistor. The increase of T_c from 2.4 to 4.4 K upon coupling of both superconducting layers is clearly observable. The inset shows the thickness dependence of the superconducting transition temperature for the coupling of two superconducting PT layers. An increase for films below 150 Å is clearly observable. A maximum T_c of 4.4 K is observed for 50 Å thick films.

PT and Pb multilayers (Fig. 3). A maximum T_c of 8.4 K is observed for coupling of superconducting Pb and PT (gate induced). It is particularly noteworthy, that this T_c is higher than that of bulk Pb. Tunneling spectra of PT films using in-plane SIS structures[8] reveal coupling to excitations of 8 and 10 meV (Fig. 4). These values are in good agreement with phonon energies reported for this material[24] suggesting phonon-mediated coupling in PT. Additional peaks in the tunneling spectrum of PT are observed in the presence of an adjacent Pb film (Fig. 4). These resonances can be explained by coupling of the charge carriers in the PT film to transverse (~6 meV) and longitudinal (~10 meV) Pb phonons.[23]

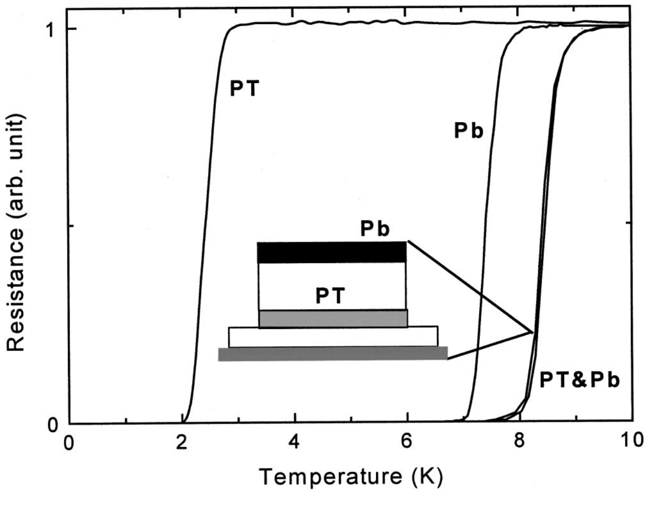

FIG. 3. Channel-resistance as a function of temperature for the Pb film (dashed) and the bottom PT transistor (solid). A T_c of 8.4 K is observed upon coupling of both superconducting layers through the thin insulating PT barrier (~50 Å). The inset shows the schematic device structure (not to scale).

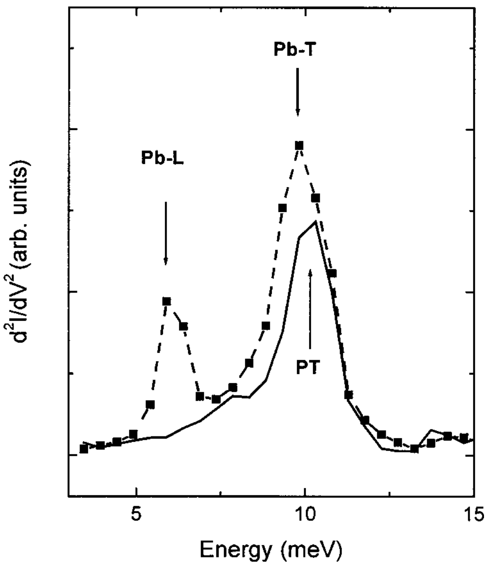

FIG. 4. Second derivative of the tunneling current in a PT superconductor-insulator-superconductor junction as a function of the applied bias. The peaks correspond to coupling to phonons. The solid line is observed for gate-induced superconductivity in a single PT film. The dashed line corresponds to coupling of a superconducting PT and a superconducting Pb film. The additional peaks are ascribed to coupling of charge carriers in the different layers via Pb phonons.

The increase of the superconducting transition temperature T_c resembles some similarities to the T_c enhancement in Sn-SiO or Al-Al_2O_3 superconductor-insulator stacks.[13–15] In addition analogous results of finite size effects have been observed in ultrathin $NbSe_2$ single crystals,[25] ultrathin quench-condensed Pb/Sb and Pb/Ge multilayers,[16] or cuprate superlattices.[26–28] Several models have been proposed to explain the modification of T_c in superconductor-insulator multilayers, especially with respect to high-T_c superconductivity in cuprates. Ginzburg[9] suggested that changes in the superconducting interaction at the film surface could lead to changes in the film T_c. However, for our devices it is not obvious how the superconducting interaction might change at the interface between the insulating and superconducting PT, since the superconductivity is turned on and off via gate doping without modification of the interfaces. Further models proposed exciton mechanisms at interfaces rather than phonon mediated pairing.[11] However, the tunneling spectra seem to be in contradiction with this assumption. Other models attributed a rise in T_c to disorder-effects,[15,29] which can also be ruled out in our experiments since the disorder does not change upon gate-induced superconductivity in the second film. In addition, a lowering of phonon frequencies compared to bulk material has been proposed for the T_c enhancement,[15] which can also be excluded as explanation since the PT superconductor-insulator multilayer consists of the same material. Hence, a lowering of the phonon modes upon gate-induced superconductivity in the second transistor seems to be unreasonable. Another possibility is Josephson coupling of the superconducting layers which has been suggested for high-T_c cuprates.[16,30–32] However, no signs of the

Josephson effect could be observed in vertical SIS tunneling structures in contrast to in-plane devices.[21] Hence, this explanation seems to be unlikely although it can not be ruled out completely. In other models electron transfer through localized states of the insulator[16] or the proximity effect account for a T_c variation in multilayer stacks, but it seems to be questionable that these effects can result in an increase of T_c above the bulk value, which is observed in our experiments.

Cohen and Douglass suggested the possibility of pairing of charge carriers separated by a barrier layer.[10] Since an insulating layer does not present a barrier to phonons, it is possible that a charge carrier in layer 1 can emit a phonon, which is then absorbed by a charge carrier in layer 2. This will result in an attractive, phonon-induced interaction V^{12} comparable in magnitude with the interaction within the layer V. Since the charge carriers on opposite sides of the barrier feel a lower, screened Coulomb interaction[10,33] the resulting superconducting transition temperature can be increased. Moreover, it has been suggested that interlayer interactions can mediate Heisenberg-type coupling between order parameters in adjacent superconducting layers.[12] The tunneling spectra of PT/Pb stacks indeed reveal the coupling of charge carriers in the PT film to Pb phonons. Hence, it might be reasonable to assume that the charge carriers in the two superconducting films can couple via the Cohen-Douglass mechanism giving rise to an increase of T_c up to 8.4 K for the combination of Pb and PT and up to 4.4 K for PT SIS structures. In the latter case of two superconducting PT layers the increase of T_c can be ascribed to the screening of the Coulomb interaction of the charges confined in the two layers due to the insulating barrier. Obviously, the explanation using the Cohen-Douglass mechanism is still speculative and further specific studies of the observed T_c enhancement will be necessary in order to understand the microscopic physics in more detail. Nevertheless, this model can account very well for the experimental observations. In addition, it would also be extremely interesting to study similar effects in other organic or gate-doped materials, especially, hole-doped C_{60}, where a T_c of 52 K has been observed in a single field-effect device.[7]

In summary, the T_c enhancement in polythiophene multilayer structures is tentatively ascribed to phonon-mediated coupling of charge carriers in adjacent superconducting layers through an insulating barrier. This leads to a strong reduction of the Coulomb interaction. Moreover, a T_c of 8.4 K is observed for coupling between polythiophene and lead. Evidence of coupling to lead-phonons is found in tunneling spectra of the polythiophene films.

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