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Physics aspects of ferromagnetic/superconducting superlattices

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Ferromagnetic/superconducting superlattices represent a new class of materials with the simultaneous occurrence of superconductivity and ferromagnetism. The mutual interaction of these antagonistic ordering phenomena is of vital fundamental interest and opens novel possibilities for spin-injection devices. We have prepared YBCO based superlattices with either $La_{2/3}Ca_{1/3}MnO_3$ or $SrRuO_3$ as ferromagnetic part by pulsed laser deposition with individual layer thickness ranging from 4 to 200 unit cells for YBCO and 10 to 500 unit cells for the magnetic layers, respectively. We studied superlattices of different modulation lengths especially with respect to the reduced phase transition temperatures to ferromagnetism and superconductivity. Conventional models to explain the reduction of T_C and T_{Curie} fail and novel concepts giving rise to a long-range proximity effect have to be introduced. Furthermore, in the case of ferromagnetic $La_{2/3}Ca_{1/3}MnO_3$ layers with high degree of spin polarization it is suggested that the pseudogap opening of the $YBa_2Cu_3O_7$ weakens the interlayer ferromagnetic coupling of the $La_{2/3}Ca_{1/3}MnO_3$ layers, thus contributing to the reduction of T_{Curie} and the reduced evolution of the ferromagnetic state.
Key words: superlattices, proximity effect, ferromagnetic superconductors, pseudogap.

1. INTRODUCTION

In a pioneering paper Ginzburg [1] addressed the question of coexistence of two different antagonistic long-range ordering principles such as superconductivity [SC] and ferromagnetism [FM]. He concluded that they can not coexist in a homogeneous system because SC requires the formation of pairs of electrons with antiparallel spins, whereas the exchange field in a ferromagnet forces the spins to align. This situation can be completely different in inhomogeneous systems where the local environment can cause parallel spin alignment and SC in different regions of the unit cell. Natural or artificial layered structures such as super-lattices [SL's] or heterostructures are examples for this. Ferromagnetic SC's such as $ErRh_4B_4$ and $HoMo_6S_8$ [2,3] with a Curietemperature T_{Curie} smaller than the transition temperature to SC, T_c, represent the class of layered structures. The competing ordering mechanisms lead to SC first and upon further cooling ferromagnetic order is energetically more favorable and the associated pair breaking effects kill SC. Recently, the family of $RuSr_2RECu_2O_8$ and $RuSr_2RE_{2-y}Ce_yCu_2O_{10}$ [4,5] com-pounds with RE = Gd, Eu has been discovered and $T_{Curie}>T_C$ has been postulated, representing the so called superconducting ferromagnets. The ruthenocuprates can be treated as a prototype of an intrinsic ferromagnetic/ superconducting superlattice.

In the past, SL's based on metal SC's and FM's have been fabricated and studied, both experimentally and theoretically [6-9]. All these experiments have been carried out in metal systems which have in common that the SC's are s-wave and their coherence length is always much larger as compared to the FM metal thickness. They showed a monotonic – in some cases even oscillatory – dependence of T_C with increasing FM layer thickness. Intermixing and hybridization of the conduction electron states in the FM with those of the SC host are accounted for the reduction of T_C.

In the case of SL's composed of cuprates and manganites this situation is changed fundamentally from the physics as well as materials point of view. According to dedicated X-ray analysis [10] as well as cross-sectional TEM investigations [11] structural intermixing is confined to roughly one unit cell at the interface. Furthermore, the order parameter in cuprate SC's has d-wave symmetry and the materials have an anti-ferromagnetic ground state. The coherence length is highly anisotropic with ~0.1nm along the c-direction and ~ 1-2 nm along the a, b plane. In the case of the manganites, e.g. $La_{2/3}Ca_{1/3}MnO_3$ [LCMO], the FM part has a nearly 100% spin polarization due to the combination of Jahn-Teller distortion of the MnO_6 octahedra surrounding the Mn^{3+} ions in combination with the Hund's rule. FM is caused by Zener double exchange rather than itinerant band correlations. Replacing the manganites by a bad metallic itinerant band ferromagnetic material such as $SrRuO_3$ [SRO] offers the possibility to probe the effect of spin polarization on the properties of the SL's. Due to the comparable deposition conditions for the cuprates and the ferromagnetic oxides high quality SL's can be fabricated by conventional techniques such as sputtering and pulsed laser deposition. Several papers have been published [12-19], demonstrating the coexistence of both ordering phenomena, SC and FM and a suppression of T_C as well as T_{Curie} has been observed. In these papers mainly the T_C depression has been studied and a characteristic length scale for the depression has been claimed much larger than predicted by the existing theories of the FM/SC proximity effect. Whereas in previous papers some fundamental aspects of the T_C reduction and free carrier response have been addressed, in this paper more attention is given how the superconductor affects T_{Curie} in the superlattices. Furthermore, some comparison is made how the different ferromagnetic materials in the SL's influence the transport and magnetic properties of the superlattices.

2.EXPERIMENTAL DETAILS AND RESULTS

Using $SrTiO_3$ single crystal substrates, superlattices consisting of m unit cells of $YBa_2Cu_3O_{7-x}$ and n unit cells of LCMO or SRO repeated N times have been deposited at 730^{0}C at an oxygen pressure of $5x10^{-3}$ Pa

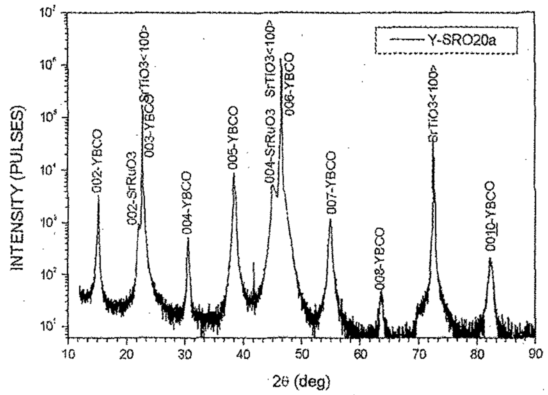

Fig.1. X- ray diffraction pattern of a $(80nm/20nm)_5$ YBCO/SRO superlattice.

by pulsed laser deposition technique [20,21]. To ensure complete oxygenation the SL's were in-situ annealed in 1atm oxygen for 1 hour at 530^{0}C. The deposition system (high vacuum chamber in conjunction with a KrF Excimer laser) is equipped with a FIR pyrometer to control the radiatively heated substrate surface and film temperature. A special computer program has been developed to account for the different emission coefficients of the substrates and the films, respectively. A computer controlled target exchange system accommodating up to 5 different targets facilitates the fabrication of the desired SL formation. Different modulation lengths, Λ, can easily be realized. Thickness control of the individual layers is done by pulse counting after some calibration runs to ensure the stability of the ratio film thickness per pulse. The X-ray diffraction analysis confirmed the phase purity of the c- axis oriented films (see Fig. 1). The X-ray diffraction pattern shows peaks with a rather large FWHM of $2^0 - 4^0$. Superlattice peaks as expected due to the additional scattering planes in a superlattice are barely to be identified. A determination of the SL peak positions according to the standard formula $\Lambda = \lambda/2(\sin\Theta_n - \sin\Theta_{n+1})^{-1}$ shows that for the first and second order peaks for our SL with the smallest modulation length Λ=13nm the peak positions to be expected are buried in the main peak. Cross-sectional high resolution TEM analysis (see Fig. 2) reveals the SL formation and shows atomically flat interfaces. The epitaxial relation is clearly "cube-on-cube" (Fig. 2b) [11]. In low magnification (Fig. 2a), however, the TEM figures show a certain waviness of the interfaces. This can arise from the different mechanisms for stress relaxation and a peculiarity generated by the Stranski-Krastanov growth mode for the YBCO film where growth islands at the surface affects the nucleation of the subsequent layers. However, it cannot be ruled out that the PLD technique applied is an additional source for the waviness due to

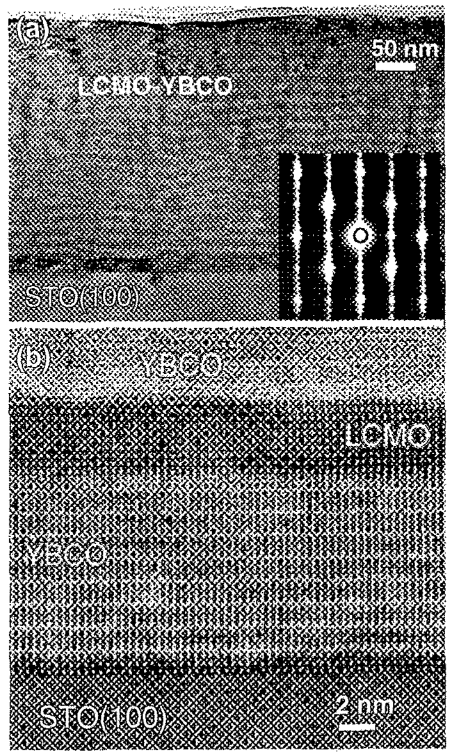

Fig.2 (a) low resolution and (b) high resolution transmission electron microscope and electron diffraction (shown on inset) images of a $(8nm/6nm)_{20}$ YBCO/LCMO SL.

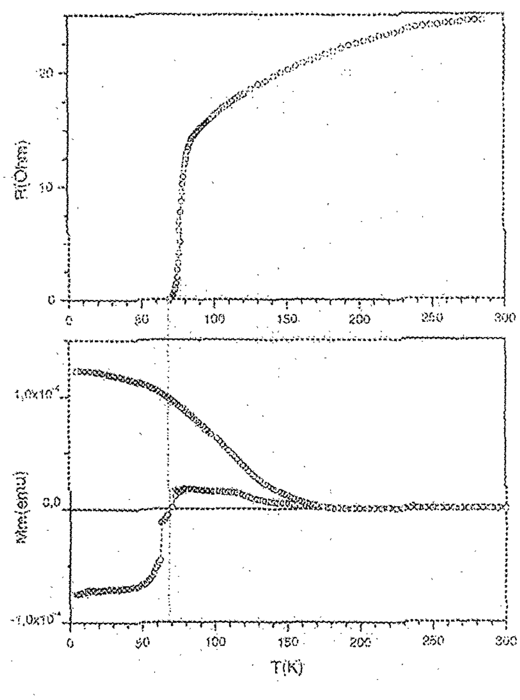

Fig.3. Resistance (upper panel) and magnetic moment (lower panel) of a $[40\ nm\ YBCO\ /20\ nm\ LCMO]_5$ superlattice as a function of temperature. The ZFC measurement shows the diamagnetic signal.

the pulsed growth and the high kinetic energy of the particles impinging the growing film. The magnetic moments of the films are determined using SQUID magnetometry; transport properties are measured by standard 4-point probe techniques using evaporated gold contacts and a probing current of 0.1 mA. Fig. 3 represents the R (T) and M (T) plots of a $[YBCO40nm/LCMO20nm]_5$ SL showing T_{Curie} = 150K and T_C = 70K. The transport normal state properties are expected to be a superposition of the linear dependence R ~ T of the YBCO and the features of the metal-insulator transition of the LCMO. The metal-insulator transition appears as a change in the slope of the R (T) around T_{Curie}. The main features in Fig. 3 are the reduction of the ordering temperatures to FM and SC, respectively, and a substantially higher resistivity as compared to the corresponding single layers. Optical measurements using spectroscopic ellipsometry as a bulk probe confirm this and rule out significant contributions by additional scattering at internal surfaces as the reason for the enhancement of resistivity [19]. For a systematic study of these effects we prepared $[t_{YBCO}/t_{FM}]_N$ - type SL's of different compositions, keeping either t_{YBCO} constant and change t_{FM} or vice versa. In Fig. 4 the increase of T_C for constant magnetic layer thickness [t = 20 nm] is given for both materials, LCMO and SRO, respectively.

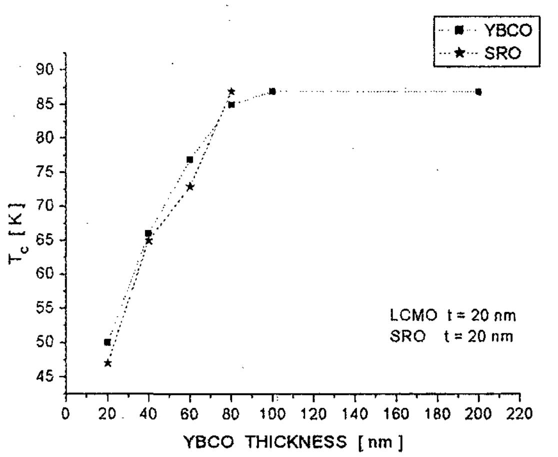

Fig.4 Evolution of T_C as a function of the YBCO thickness in YBCO/LCMO and YBCO/SRO super-lattices with $t_{LCMO/SRO}$ = 20 nm.

There is an approach to the T_C bulk value for layer thickness > 70nm; both materials, SRO and LCMO behave quite similar. The change of T_{Curie} for constant YBCO film thickness of t = 20nm [Fig. 5] reveals a smooth approach to the bulk value and again no significant differences for SRO and LCMO. The main results are summarized as follows: (i) Coexistence of ferromagnetism and superconductivity, (ii) composition dependent reduction of T_C and Tcurie, and (iii) reduction of conductivity.

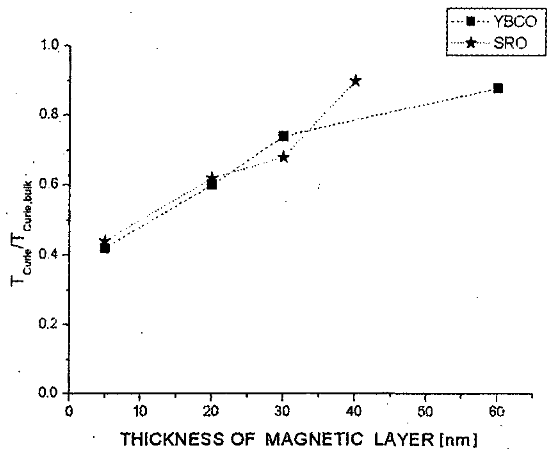

Fig. 5 Evolution of the Curietemperature for YBCO based superlattices with t_{YBCO} = 20nm.

3. DISCUSSION

The reduction of T_C in cuprate based superlattices with an insulating or normal spacer is not a new discovery; it has been investigated extensively in the past decade and a wealth of experimental data has been revealed and summarized in several review papers [22, 23, 24]. Amongst the mechanisms accounted for the T_C reduction, material related extrinsic effects have been discussed such as interfacial lattice strains, incomplete oxygenation, disorder, grain boundaries, and weak links due to the interface of the SC and non-SC layers [23]. Systematically ruling out these effects to be dominant in high quality SL's, intrinsic effects are treated as possible origin for the T_C reduction. Suggested explanations like interlayer coupling as a mandatory prerequisite to achieve high T_C, Kosterlitz-Thouless transition, long-range Josephson coupling and charge transfer mechanisms show that no conclusive quantitative interpretation of the T_C reduction in SL's exists. Jansen and Block [25] have achieved a quantitative agreement of the experimental data for YBCP/PrBCO SL's on the basis of a microscopic approach based on an indirect-exchange Cooper pairing between quasi-particles and oxygen anions.

Qualitatively, all these arguments can be used in the case of the FM/SC SL's; quantitatively the T_C suppression is much more pronounced as in the case of e.g. YBCO/PrBCO. In Fig. 6 the experimental data for YBCO/LSMO as well as YBCO/SRO SL's with t_{YBCO}=20nm and increasing thickness of the ferro-magnetic spacer layers are represented and compared with the data reported by Lowndes [26] for the YBCO/PrBCO system for 3 unit cell [3,5nm] and 6 unit cell [7nm] thick YBCO layers in the SL. The data points in parentheses are extrapolated from the plot given in [28]; this extrapolation seems to be justified according to the theory of Jansen and Block and the experimental results of our group and others. It is obvious that the reduction of T_C in the SL's containing ferromagnetic spacer layers is much stronger compared to the

YBCO/PrBCO case consequently additional interaction mechanisms must be effective. Furthermore, the reduction in T_C is stronger for the Zener double-exchange ferromagnetic material with a saturation magnetization of $3.5\mu_B$ such as LCMO as compared to the itinerant band ferromagnet SRO with $1.5\mu_B$. As already discussed in [19] magnetic correlations can play an important role in determining the temperature of the phase transitions, possibly due to a novel magnetic proximity effect where the charge carriers are coupled to different and competing kinds of magnetic correlations. Conventional proximity effect at a metallic superconductor/ ferromagnetic metal interface is governed by the ratio $hv_F/2\pi\ \Delta E_{ex}$ with v_F being the Fermi velocity and ΔE_{ex} the exchange splitting. Even for classical metals this quantity is around 1nm, in the case of oxides it should not exceed that value.

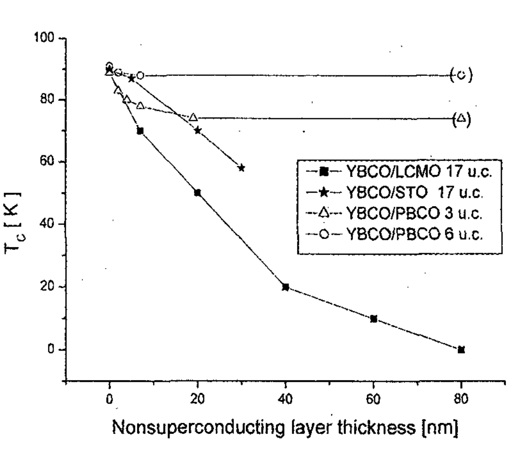

Fig.6 Comparison of T_C vs. thickness of non-superconducting layers in YBCO/LCMO, YBCO/SRO, and YBCO/PrBCO superlattices (explanations in the text).

The physical origin of the reduction of T_{Curie} is much less clear. Extrinsic effects such as incomplete oxygenation and epitaxial strain can be ruled out according to the arguments given in [11]. Massive charge transfer of holes into the ferromagnetic layer can qualitatively reduce T_{Curie} in the case of LCMO. According to the temperature/doping phase diagram of the La-Ca-Mn-O system, however, for bulk material – as well as relaxed thin films- at 33% Ca doping T_{Curie} is 270 K and an increase of the Mn^{4+} concentration – equi-valent to increased doping - leads to a minimum T_{Curie} of 195K at 50% doping. Higher doping will lead to an antiferromagnetic charge-ordered insulating state. The Curie temperatures measured in YBCO/LCMO SL's [c.f. Fig. 4] are much below this value consequently the argument of hole transfer to the LCMO cannot quantitatively explain the reduction of T_{Curie}. Additionally, such a massive charge transfer would drive the YBCO into the non-superconducting insulating state in contrast to the experimental observations. Charge transfer will not be completely ruled out due to the different chemical potential of YBCO and the ferromagnetic spacer layers. According to the simple depletion layer model for semiconductors, the Poisson equation is solved approximately with a quadratic potential difference on both sides of the interface giving a total depletion layer $d = [2\varepsilon\Delta\Phi/(\pi Ne)]^{1/2}$. Here, N is the volume density of free carries – assumed to be comparable for the two materials involved -, $\Delta\Phi$ is the potential difference between the bulk and the interface, ε is the static dielectric constant of the depleted insulating material and e is the electron charge. With reasonable numbers for $\Delta\Phi = 1$ eV and a fairly large value for $\varepsilon = 15$ one expects a depletion layer of ~ 1nm. Consequently, charge transfer is not the dominant mechanism. In the case of SRO charge carrier transfer will not affect T_{Curie} at all.

An interesting feature is the different behavior of the evolution of magnetism in SL's with LCMO as compared to SRO. The two FM materials are different in two aspects. First FM in LCMO is based on the double-exchange mechanism whereas SRO is an itinerant bad metal ferromagnet. Second, the saturation magnetization in LCMO is around $3.5\mu_B$ whereas in SRO bulk as well as thin films a value of $1.5\mu_B$ has been reported [27]. Consequently, a much more pronounced effect of the ferromagnetic layer is expected for the LCMO as spacer. Fig.4 and Fig.5 suggest, that LCMO and SRO have quite similar effects in the SL's, this however holds only for the critical temperatures. The evolution of the magnetic state is quite different for the two materials. Fig. 7 compares the temperature dependence of magnetization for two SL's of similar composition [20nmYBCO/20nm FM]. The different magnetic spacers cause comparable reduction of T_C – partially masked in the lower part of Fig.7 by the spontaneous vortex state in the YBCO – but the influence of the SC layer on the evolution of the ferromagnetic state in the magnetic spacer is completely different. Whereas in the case of the LCMO the magnetic moment below 160K is much lower, but still comparable to that of single layer films, the ferromagnetic signal in the YBCO/SRO sample can barely be identified above the superconducting transition.

In the literature there are several papers dealing with magnetic coupling in ferromagnetic/normal metal multilayers. The prevailing experimental evidence indicates that the exchange coupling with metal spacers is short range (1-5nm) and a thickness dependent crossover from FM to AFM coupling occurs. These arguments cannot simply be transferred to oxide SL's. The short-range spin diffusion length up to several nm in metallic FM systems will confine the interaction effect due to the neighboring metal layer to a region of less than 3nm close to the interfaces. In the case of the oxide SL's the interaction length must be apparently long range (10–30nm). In recent experiments carried out on LCMO/YBCO bilayer structures of different thickness the spin diffusion length has been determined to 30 nm [28]. To qualitatively explain the reduction of T_{Curie} two different scenarios can be invoked. One is based on the role of the interface roughness that affects T_{Curie} by thickness fluctuations of the superconducting film and the influence of lateral fluctuations on the magnetic coupling as demonstrated by Sa de Melo [30]. The other is the argument that the coupling of the ferromagnetic layers via the YBCO is affected by peculiarities of the electronic properties of YBCO above T_C. The formation of the pseudogap and/or the spin gap is essential in this context. The close vicinity of T_{Curie} with the temperature for the spin gap opening in the YBCO normal state suggests the interrelation of the two temperatures. A reduced polarizability of the charge carrier spins below

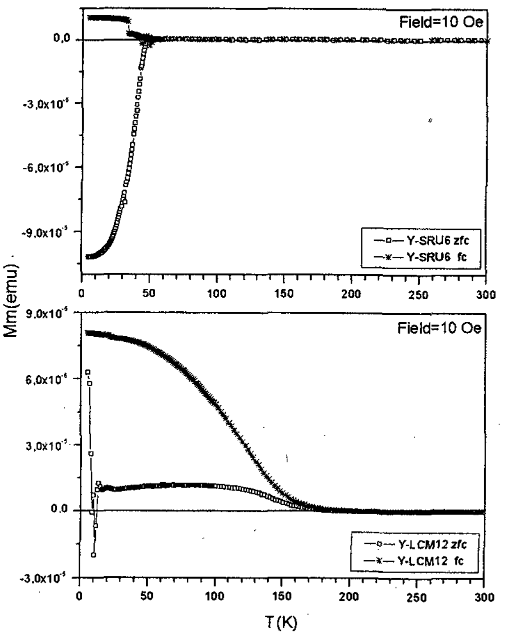

Fig.7 Zero field cooled (□) and field cooled (ξ) magnetization vs. temperature plots for an YBCO/SRO $(20nm/20nm)_{10}$ (top) and an YBCO/LCMO $(20nm/20nm)_5$ superlattice (bottom).

the T_C reduction. A certain analogy to the model of Sa de Melo [30] can be seen which predicts a modification of the density of states and a weakening of the coupling via the appearance of a superconducting gap. Probably the spingap plays that role in the oxide SL's. A suggestive argument to support this view would be a simple relation between the pseudogap and the T_{Curie} reduction. Determining the pseudogap temperature for the SL's investigated from the generic phase diagram T vs. doping and plotting these data vs. the measured reduction of T_{Curie} a simple linear relation is found [c.f. Fig. 8].

The reduced evolution of the ferromagnetic state in the SL's as ferromagnetic spacer – much more pronounced in the case of SRO compared to LCMO - is tentatively explained by the mechanism leading to ferromagnetic order. In the itinerant band ferromagnet the relative shift of the DOS parabola for spin-up and spin-down population of the electrons cause the spin polarization. Coupling via a system with a spin gap the driving force to establish magnetic order is much more reduced as compared to materials where the double exchange mechanism causes ferromagnetism.

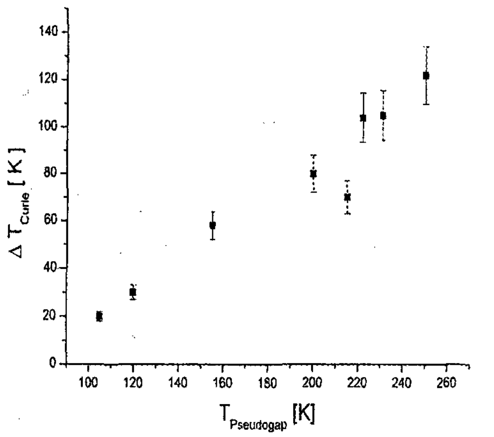

Fig.8. Relation of the T_{Curie} reduction and the pseudogap temperature in symmetric YBCO/LCMO superlattices.

4.SUMMARY

High quality cuprate/manganite superlattices have been prepared which show simultaneously the occurrence of superconductivity and ferromagnetism. The tempera-tures for the phase transitions can be varied systematically by tailoring the SL composition. The physical origin of the T_C reduction is seen in interplay of spin correlation and charge localization due to magnetic correlation giving rise to a long-range magnetic proximity effect. For the reduction of T_{Curie} the role of the pseudogap temperature should be considered. Furthermore, the role of the structural and electronic properties of the interfaces should be studied in more detail.

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