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Anomalous Critical Field Dependence of Cr-Nb-Cr Trilayers on Niobium Thickness

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Cr-Nb-Cr trilayer thin films were deposited using DC/RF sputtering with Cr layer thickness maintained at 7.5 nm and Nb thickness varying from 30 nm to 229 nm, in order to study the interplay of antiferromagnetism and superconductivity. Various viewpoints have been expressed in the literature with respect to analyzing the anomalous properties that have been identified in superconductor/magnetic thin films. The study of Cr-Nb-Cr thin films were taken up to identify the existence of similar anomalous superconducting properties on account of antiferromagnetic-superconductor interfaces. X-ray reflectivity measurements showed high quality of thin film structure with minimum surface and interface roughness. Transport measurements down to 2K were carried out in the presence of magnetic fields up to 2 T applied perpendicular to the trilayers to deduce T_c. The suppression of T_c due to Cr layer is larger than proximity effect of similar metal films. It could be explained by proximity effect using antiferromagnetic Cr layer. Upper critical field measurements show a distinct non monotonic dependence of upper critical magnetic field and the slope on Nb layer thickness. The analysis due to WHH theory to deduce upper critical fields for the trilayer thin films studied did not match with experimental values. Although studies performed on Cr-Nb-Cr trilayers did not show any anomaly in T_c, it clearly showed a depression of T_c much larger than proximity effect, non monotonic behavior in $B_{c2}(0)$ and dBc_2/dT_c behavior with Nb layer thickness.

Keywords Superconductivity, Thin films, Magnetoresistance, Upper critical field, Proximity effect, Antiferromagetism

1 Introduction

Thin film heterostructures involving superconductivity and magnetism is exciting because of several interesting phenomena associated with it. Due to proximity effect, superconductivity can be influenced into the ferromagnetic layer because of the penetration of cooper pairs and gives an opportunity to study superconductivity in the presence of large exchange field. In thin film form, when a conventional spin singlet superconductor is brought into contact with a ferromagnetic thin film, the Cooper pair correlations extend over a length of only a few nanometers across the S-F interface. The exchange field in the ferromagnet creates a spatially varying order parameter which results in an oscillatory variation of the critical temperature as a function of ferromagnetic layer thickness[1]. This also leads to other interesting effects like realization of π junctions in S/F/S systems, spin valve behavior in complex S/F systems, localized domain wall superconductivity and inverse effect of superconductivity on ferromagnetism[1-7]. The superconducting transition temperature oscillations in S/F systems have been interpreted as arising due to stabilization, in certain ranges of thickness, of a higher T_c π phase[8]. Experiments performed on Fe/Nb/Fe trilayers[9] showed similar non monotonic dependence of T_c as a function of thickness of Fe layer. In such a system π phase cannot be invoked because of geometrical reasons, since the superconducting layer is sandwiched between two Fe layers, the explanation of onset of ferromagnetism was invoked for the observation of non monotonic dependence of T_c. Hence, in general when non-monotonic variation of T_c was observed it is attributed either to the π phase or to the onset of ferromagnetism. This leads us to a question whether anomalous behavior is found only in superconducting transition temperature or are there any other physical properties which could also become anomalous. Further, the study of intricacy and complexity of superconductor/magnetic structures is important and is promising for potential applications such as spin valve behavior. The developing field of superconducting spintronics offers a plenty of exciting phenomena and the π phase shifters based on S/F/S junctions are predicted to be the promising candidates as basic elements in the field on quantum

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computation[10,11]. The basic building block for all these experiments are thin film heterostructures involving superconductivity and magnetism. In general, there are very few studies on the proximity effect between superconducting and antiferromagnetic (AFM) orderings because the resultant internal field is not significant enough to affect superconductivity. Earlier report of Nb-Cr multilayers did not report any anomaly in T_c or upper critical field[12] and explained based on proximity effect. In this report we have observed depression of T_c much larger than proximity effect[13-14] in similar metal films. This larger depression of T_c could be explained by proximity effect with antiferromagnetic Cr thin film[15]. We also report non monotonic variation of upper critical magnetic field $B_{c2}(0)$ and the slope of critical field with temperature dB_{c2}/dT for different Nb thickness in Cr-Nb-Cr trilayer thin films. It has also been observed that the dependence of critical field with temperature does not follow WHH model at low temperatures.

2 Experimental details

Trilayer thin films of Cr-Nb-Cr were deposited by DC/RF magnetron sputtering on Si and glass substrates at ambient temperature. Before depositing the films, substrates were initially cleaned ultrasonically in acetone and then in isopropyl alcohol for 10 min each; it was further cleaned by plasma cleaning procedure using RF sputtering in the presence of 99.9995% pure Ar gas at a pressure of 5 mbar at 300 V for 8 min. The Cr and Nb targets were presputtered for 45 min with the shutter closed by passing pure Ar gas at a pressure of 5 mbar to clean the targets. Thin films of Cr (antiferromagnet) and Nb (superconductor) were deposited using RF sputtering and DC magnetron sputtering respectively on Si and glass substrates. Trilayer Cr-Nb-Cr thin films were deposited on the whole wafer by sequential sputtering by flowing 90 sccm of 99.9995% pure Ar gas at a pressure of 8 mbar after achieving a base pressure of 1×10^{-5} Pa in the chamber. The pressure is maintained by adjusting the throttle valve between the cryopump and the chamber. The thickness of each layer in the trilayer is estimated based on prior calibration of the deposition rates using DEKTAK surface profiler. The thickness of the Cr layer was maintained at 7.5 nm for all the three trilayers, while the Nb thickness was varied as: [Cr (7.5 nm) –Nb (30 nm)-Cr (7.5 nm) named S1], [Cr (7.5 nm) –Nb (45 nm)-Cr (7.5 nm) named S2] & [Cr (7.5 nm) –Nb (229 nm)-Cr (7.5 nm) named S3]. The total thicknesses of these films were measured using DEKTAK surface profiler for corroboration. The crystal structure and orientation of the thin films were characterized by X-ray diffraction (GIXRD) measurements in a STOE diffractometer using CuK_α radiation. X-ray reflectivity (XRR) measurement was performed to analyze thin film parameters such as thickness, interface roughness etc. Transport measurements were performed by Magnetoresistance system, M/S Cryogenic UK. Superconducting transition temperature, T_c was measured resistively down to 1.8 K in a standard four probe geometry for different magnetic fields from 0 T to 2 T applied perpendicular to the thin film.

3 Results and Discussion

Room temperature GIXRD patterns of (Cr-Nb-Cr) trilayer thin films for two different thicknesses of Nb has been shown in Fig.1. Bragg peaks pertaining to (110), (211) and (200) of Nb is clearly seen and no peaks corresponding to Cr could be detected. Fig. 2. shows X-ray reflectivity scan measured with Cu K_α radiation for S1 film. The XRR spectrum is fitted with Parratt formalism[16] which showed surface and interface roughness of less than 0.8 nm manifesting high structural quality of films. The film thickness estimated from the fit procedures agreed within 11%

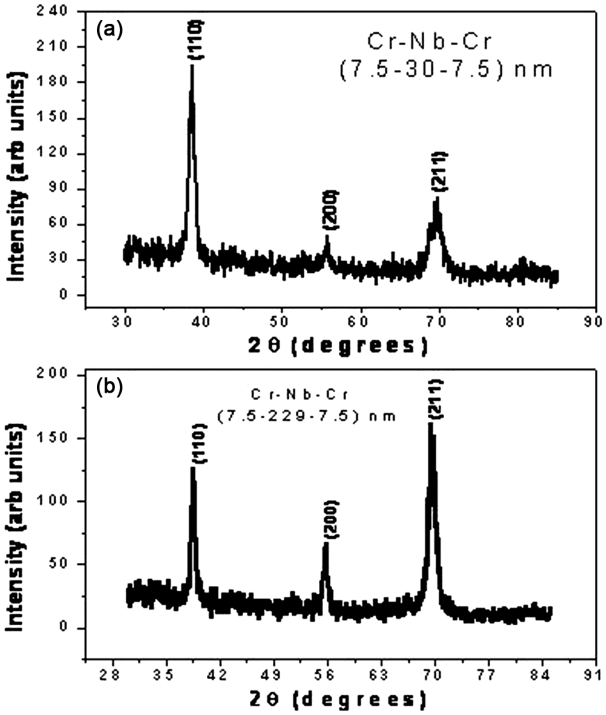

Fig. 1 — GIXRD pattern of (Cr-Nb-Cr) trilayer thin films deposited on borosilicate glass substrates; (a) S1 and (b) S3.

of the thickness determined using DEKTAK surface profile measurement system.

Superconducting transition temperature for all three samples measured as a function of magnetic field and temperature are shown in Fig. 3. For all the trilayers, superconducting transition temperature, T_c is defined as 90% value of the normal state resistance values. The T_c of these three samples were analyzed based on proximity effect formalism[13,14]. The data from Cheng *et al*[12] were used to fit the variation of T_c with thickness of superconducting thin film (d_s) for constant Cr thickness of 3 nm (d_n). The thickness of Cr thin film (d_n) was changed from 3 nm to 7.5 nm and the revised variation of T_c with d_s is obtained. The results of these dependences and the experimental values provided by Cheng *et al.* and our results are plotted in Fig. 4. It is observed that the T_c of our films have enhanced depression than the proximity effect induced depression indicating the possibility of Cr having magnetic moments. In such a situation the T_{cns} has to be taken to be zero.

$$\frac{1}{\sqrt{2}\,\xi_S}\sqrt{\left(\frac{T_{CNS}}{T_C}-1\right)}\tan\left[\frac{d_S}{\sqrt{2}\,\xi_S}\sqrt{\left(\frac{T_{CNS}}{T_C}-1\right)}\right]=$$
$$\frac{2}{\xi_{AF}}\frac{D_{AF}N_{AF}(\varepsilon_F)}{D_S N_S(\varepsilon_F)}\tanh\left[\frac{2d_{AF}}{\xi_{AF}}\right] \qquad \ldots (1)$$

To evaluate the T_{cns} of the SC/AF bilayers, one frequency approximation results[15] has been utilized. The results of T_{cns}(AF) also shown in Fig.4 which almost matches with the experimental result indicating the AF of deposited Cr film. It is to be noted that T_c for the sample S1 at zero magnetic field was observed at 7.07 K and the transition temperature shifts to lower temperatures with increase in magnetic field as can be seen in Fig. 5(a). The application of magnetic field and its dependence on T_c can be very well seen in the case of S2 thin film, where T_c of 8 K could be measured at zero magnetic field and the decrease of T_c with magnetic field up to 1.8 T is plotted in Fig. 5(b). Again for the case of S3 thin film, monotonic decrease of T_c with increase in magnetic fields were observed which is shown in Fig. 5(c). From the measurement of superconducting transition temperature, T_c as a function of magnetic field B, critical magnetic field needed for the suppression of superconductivity defined as $B_{c2}(T)$ can be measured. The upper critical field at 0 K is calculated by taking the slope $\left[\frac{dB_{c2}}{dT}\right]_{T_C}$ near $T_c(0)$ and using Ginzburg-Landau-Abrikosov-Gor'kov (GLAG) equation(1)[17].

$$B_{c2}(0) = -0.693T_C\left[\frac{dB_{c2}}{dT}\right]_{T_C} \qquad \ldots (2)$$

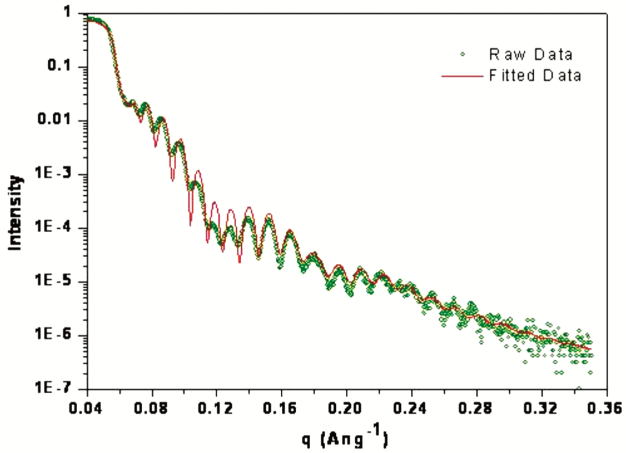

Fig. 2 — X-ray reflectivity (XRR) studies measured with Cu K_α radiation for (Cr-Nb-Cr) thin films; Symbols denote experimental values and the solid line is due to fit by Parratt formalism for S1 trilayer.

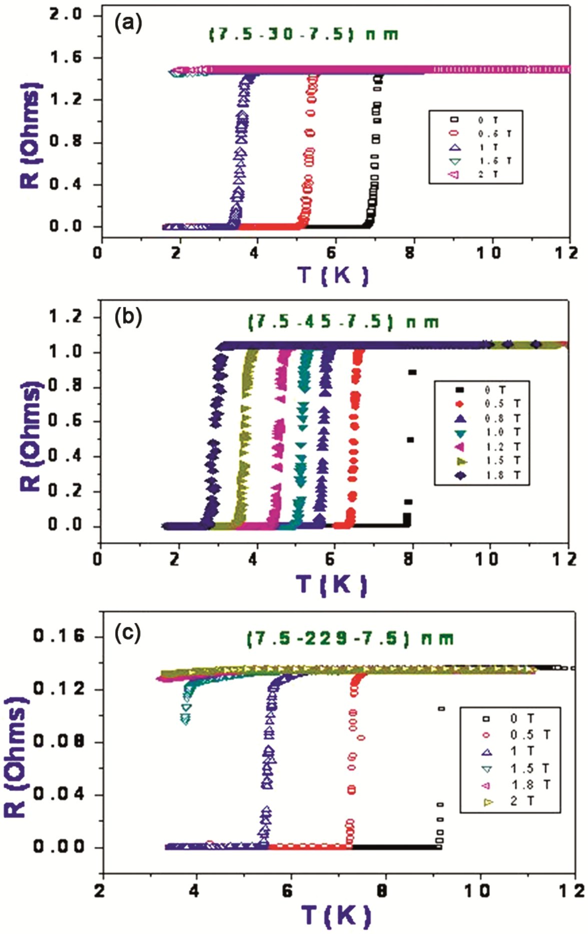

Fig. 3 — Resistance *vs*. Temperature behavior of (Cr-Nb-Cr) films for various magnetic fields ranging from 0 T to 2 T for (a) S1; (b) S2; and (c) S3 trilayer thin films showing superconducting transition temperature.

While the $\left[\frac{dB_{c2}}{dT}\right]_{T_C}$ near T_C (0) values were 0.28 T/K and 0.29 T/K for S1 and S3 are nearly the same, for S2 whose Nb layer thickness having a value in between these two showed 0.36 T/K. This non-monotonic dependence of dB_{C2}/dT_c vs Nb layer thickness was clearly observed and shown in Fig. 5 as insert. $B_{C2}(0)$ calculated using (1) is shown in Fig. 6 which also shows non-monotonic dependence on thickness.

Further, the analysis of upper critical field with temperature was performed based on the formalism of WHH for disordered Type II superconductor[18]. An implicit relation between the upper critical field $B_{C2}(T)$ (represented in terms of a parameter $\bar{h}$ and reduced temperature (represented in terms of the parameter t) was derived by solving the linearized Gorkov's equations. This is reduced to the closed form of equation (2) under dirty limit $2\pi T\tau \ll 1$.

$$\ln(1/t) = \sum_{n=1}^{\infty}\left\{\frac{1}{|2n+1|} - \left[|2n+1| + \frac{\bar{h}}{t} + \frac{\left(\alpha\bar{h}/t\right)^2}{|2n+1|+([\bar{h}+\lambda_{so}]/t)}\right]^{-1}\right\} \ldots (3)$$

where $t=T/T_c$ and $\bar{h} = \frac{B_{c2}(t)}{\left[\frac{dB_{c2}}{dt}\right]_{T_c}}$. The variation of $B_{C2}(T)$ with T can be obtained from the above WHH equation.

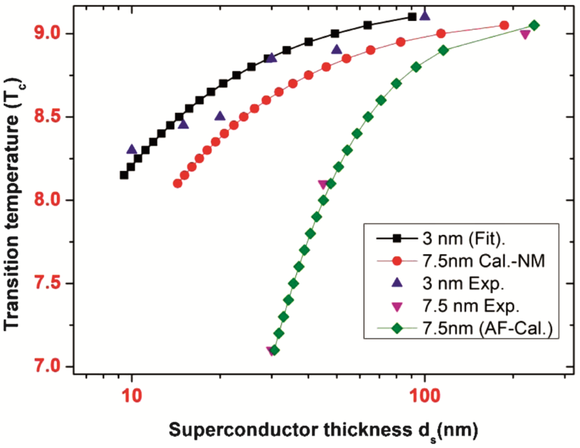

Fig. 4 — Plot of Tc variation with thickness.

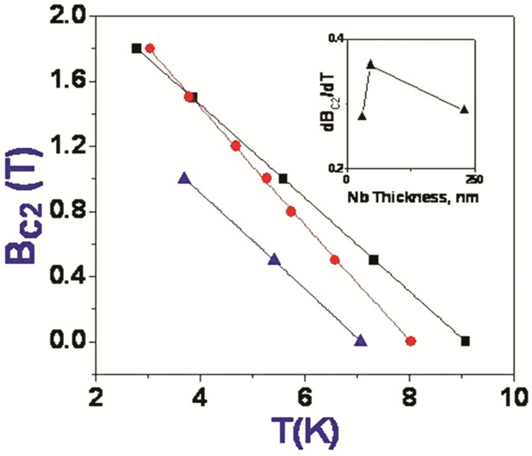

Fig. 5 — Critical magnetic field vs Temperature for Cr-Nb-Cr trilayer thin films; ▲- S1; ●- S2; and ■ – S3 trilayer thin films. Inset shows non-monotonic dependence of dBc_2/dT vs Nb layer thickness for Cr-Nb-Cr thin films.

The calculated variation of uppercritical field with temperature using WHH theory and experimentally observed values for all three Cr-Nb-Cr thin films studied were plotted in Fig.7. It can be seen that WHH theory does not fit well to the experimental data even if the paramagnetic limiting parameter(α) and spin-orbit parameter (λ_{so}) set equal to zero. Several experiments on Pnictides[19-21] show WHH theory does not follow strong linear increase of critical field with temperature and the authors attribute it to emergence superconducting phase, FFLO phase[22,23].

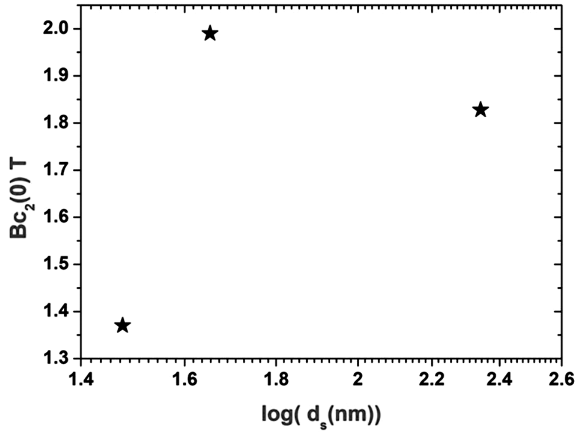

Fig. 6 — Variation of $B_{C2}(0)$ with Nb thickness.

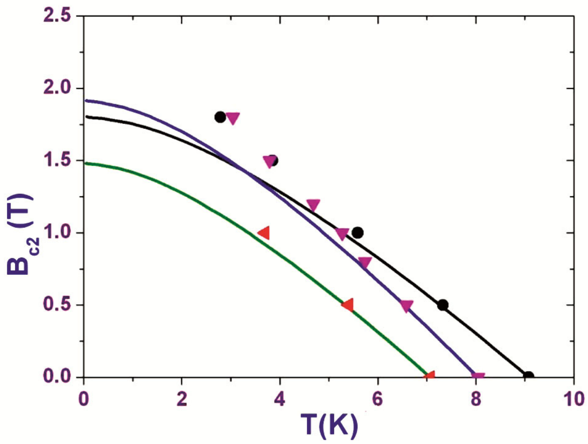

Fig. 7 — Variation of $B_{c2}(T)$ with temperature.

Studies on Nb/Gd multilayers showed nonmonotonic or oscillatory behavior of T_c with progressive increase of layer thickness of Gd [1]. These results were explained based on π junction coupling across two superconducting layers mediated by a magnetic layer. Interface effects in Si/Nb/Si trilayers[24] was stated to be the reason for the observation of anomalous Hall coefficient for ultrathin Nb layer thickness in Si/Nb/Si. The non monotonic dependence of Tc behavior was also seen in Fe/Nb/Fe trilayers[9]. Here based on geometrical reasons, π junction coupling is ruled out and instead the authors give an explanation based on the presence of dead magnetic layer at the interface of Fe/Nb; this separates superconducting and ferromagnetism in real space and the influence of the intermixed layer between superconducting Nb layer and ferromagnetic Fe layer was assumed to be the reason for non monotonic T_c behavior. One interesting aspect of this model is that it could explain the absence of any difference in the results of T_c with Nb layer thickness and also $B_{c2}(T)$ behavior both for non magnetic and ferromagnetic samples. However, our experiments are based on antiferromagnetic/superconductor trilayers *viz* Cr-Nb-Cr and were performed to look for obscure anomalies in superconducting properties on account of antiferromagnetic-superconductor interfaces. Although measurements on Cr-Nb-Cr trilayers did not show any anomaly in T_c, it clearly showed larger depression in T_c than proximity effect, non monotonic behavior in variation of $B_{c2}(0)$ and dB_{c2}/dT_c with Nb layer thickness. We also think that the influence of boundary scattering across the interfaces plays a vital role on spin fluctuations and its concomitant effect on superconductivity. Hence, the interesting finding is that although resistive transitions did not show any anomaly, $B_{c2}(0)$ and dB_{c2}/dT_c behavior showed an anomalous behavior with Nb layer thickness in Cr-Nb-Cr trilayers and antiferromangetism has been established for Cr layer based on proximity effect calculations. It has also been observed that the dependence of critical field with temperature does not follow WHH model at low temperatures.

4 Conclusions

Cr-Nb-Cr trilayers were prepared using DC/RF magnetron sputtering at ambient temperatures for three different thicknesses of Nb with Cr layer thickness kept constant at 7.5 nm. X-ray reflectivity measurements revealed high quality of thin film structure with low surface and interface roughness. Transport measurements with magnetic fields applied perpendicular to the trilayer film geometry and temperatures down to 2 K revealed monotonic decrease of superconducting transition temperature, T_c. and clearly showed larger depression in T_c than proximity effect, non monotonic behavior in variation of $B_{c2}(0)$ and dB_{c2}/dT_c with Nb layer thickness. The variation of T_c with Nb thickness could be fitted well with the one frequency approximation of modified de-gennes theory of proximity effect indicating that a 7.5 nm Cr thin film behaves as an antiferrormagnetic metal. To our knowledge this is the first experimental result that matches the variation of T_{CNS} in AF-SC-AF trilayers to the proximity effect formalism. The variation of $B_{c2}(T)$ with temperature obtained by solving the WHH equations in dirty limit did not conform to the experimentally measured $B_{c2}(T)$ variation with temperature at low temperatures.

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