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Stoner enhanced paramagnetic influence on superconductivity in a superconductor/metallic heterostructure

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ABSTRACT

The unusual magnetic behaviour of a Pd/$Nb_{67}Ti_{33}$/Pd heterostructure was investigated using polarised neutron reflectivity technique. On application of a large in-plane magnetic field, *Stoner enhanced paramagnet* Pd was found to influence the magnetic state of the $Nb_{67}Ti_{33}$ layer both above and below the superconducting transition temperature T_c significantly. Unlike the case of a conventional *proximity effect* for a superconductor/metallic heterostructure, the pair correlation in the superconducting state has been found to be more stable in the higher field limit compared to its low field counterpart, possibly signaturing a 'novel' coupled state in the system. The superconducting state in $Nb_{67}Ti_{33}$ has been found to be diamagnetic in nature at all the fields that can be fitted using a Meissner kind of behaviour in the high field limit. The magnetic properties of Pd and $Nb_{67}Ti_{33}$ are in excellent agreement with those measured using bulk magnetisation measurements.

1. Introduction

The magnetic behaviour of superconducting thin films are of huge interest mostly in multilayered structures in combination with other magnetic layers (ferromagnetic, normal metal, etc.) [1,2]. In the thin film geometry, the behaviour of a superconducting material can be significantly different from its bulk counterparts because of reduced dimensionality. The 3-dimensional superconducting behaviour often changes to 2-dimensional nature in a superconducting thin film when the thickness is reduced below a certain critical limit [3,4]. In the presence of a normal metal (N) near a superconductor (S), the physical properties of both materials change near the interface. At the superconductor (S)/normal (N) metal interface, Cooper pairs penetrate and decay into the normal metal over a characteristic distance $\sim L_T = \sqrt{D/T}$, called as the thermal diffusion length where D is the diffusion constant and T is the absolute temperature [1]. This has been experimentally observed in the case of a S/N bilayer, where T_c decreases monotonically with an increase in the normal layer thickness (d_N) due to enhanced Cooper pair breaking [5]. This is called as the *proximity effect* [6] in the case of a S/N structure. In the case of proximity effect for a superconductor (S)/ferromagnet (F) heterostructure, the decay of the pair correlation will be modulated in the F layer under the influence of the ferromagnetic exchange field and will show a damped oscillatory behaviour [1].

In the recent times, continued theoretical and experimental interests have grown in this area of research to understand the behaviour of superconducting thin films and multilayered S/N systems. While typically macroscopic experimental probes like magnetisation and transport measurements [7,8,5] are used for these investigations, less has been done using microscopic probes like neutrons in these systems. The technique of polarised neutron reflectivity (PNR) is an ideal method to probe the structural and magnetic behaviour of superconducting thin films and S/F multilayers [9–15,4]. In a PNR experiment, spin polarised neutrons in their 'up (↑)' and 'down (↓)' spin eigen states (with respect to the direction of the applied magnetic field) arrive at the sample surface and specular reflection of the neutrons are detected which in the case of a multilayered structure provide information regarding the structural and magnetic behaviour of the heterostructure. The PNR technique provides large spatial sensitivity (~10 Å) to measure microscopic variation of the magnetic flux profile in a multilayered structure perpendicular to the plane of the film which have been successfully used to measure the magnetic behaviour of thick [10,11] and thin [12,13] superconducting films, influence of ferromagnetism on superconductivity in a F/S heterostructure [14,15] for an applied field in the plane of the film.

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In the present case, we have investigated the behaviour of a *N*/*S*/*N* trilayered system consisting of $Nb_{67}Ti_{33}$ as the *S* layer sandwiched between two thin Pd layers using PNR technique. $Nb_{67}Ti_{33}$ is a type-II superconductor ($\lambda \geqslant \xi$, where λ is the penetration depth and ξ is the coherence length of the superconductor) with a superconducting transition temperature $T_c \sim 8.5$ K which is widely used commercially in superconducting tapes and magnets. The magnetic behaviour of these films were investigated in the low and high magnetic field limits to investigate the influence of the Pd layers on the magnetic state of $Nb_{67}Ti_{33}$ in the normal and the superconducting states.

2. Experimental details

Thin films of $Nb_{67}Ti_{33}$ were grown at the industrial research facility located at Hitachi Global Storage Technology, California using a DC sputtering system in an UHV atmosphere on 3″ Silicon {1 0 0} wafers. The sample used in the present measurement has the following structure Ta (1.5 nm)/Pd (3 nm)/$Nb_{67}Ti_{33}$(200 nm)/Pd (3 nm)/Si (substrate). Ta was used as a capping layer to protect the other layers from oxidation. For the PNR measurements, a square piece of 1″ × 1″ size was cut from the central region of the wafer while the magnetisation measurements were performed using the off-cuts. The PNR measurements were performed at the AMOR reflectometer located within the SINQ beamline at Paul Scherrer Institut, Switzerland. AMOR uses pulsed neutron source that can be used both in time of flight (TOF) and ($\theta - 2\theta$) geometry. For the present measurements, the TOF mode was used with a typical instrumental resolution of 4–10%. The magnetic field was applied using a pair of Helmholz coil electromagnets in the plane of the film and the base temperature of operation was 2.5 K. The PNR measurements were performed under field cooled conditions at two different temperatures 15 K (normal state, $T > T_c$) and 3 K ($T < T_c$) for each value of the applied magnetic fields. The bulk magnetic behaviour of this film was measured using a commercial MPMS SQUID magnetometer that confirmed the superconducting nature of these films with a sharp transition at a temperature of 7.8 K.

Neutrons are spin-1/2 particles and in a multilayered architecture, the potential energy of a neutron in the i^{th} layer can be written as: $V_i = (\hbar^2/2\pi m_n)\rho_i b_i - \boldsymbol{\mu_n} \cdot \boldsymbol{B_i}$ where ρ_i is the atomic number density, b_i is the scattering length density, m_n is the mass of a neutron, $\boldsymbol{\mu_n}$ is the magnetic moment of a neutron and $\boldsymbol{B_i}$ is the magnetic flux density (due to an applied field $\boldsymbol{H_i}$). The first energy term represents the nuclear potential (describing the chemical contrast of different elements in the layered structure) which stays almost constant for a specific sample structure and is independent of moderate temperature fluctuations while the second (magnetic) term depends on the mutual alignments of the neutron magnetic moment relative to the applied magnetic field direction.

In a specular PNR experiment, reflectivities corresponding to the '↑' and '↓' spin eigen states of the neutrons (relative to the applied magnetic field) are measured and the magnetic information can be most efficiently obtained from the spin asymmetry parameter defined by $S_{\uparrow,\downarrow} = (R_\uparrow - R_\downarrow)/(R_\uparrow + R_\downarrow)$ where $R_{\uparrow,\downarrow}$ are the reflectivities corresponding to the '↑' and '↓' spin eigen states of the neutrons respectively. The structural and magnetic information can be obtained by fitting the reflectivity data which in the present case was modelled using an optical reflectivity model as described by Blundell and Bland [9] and fitted to the experimental data using a Levelberg–Mardquardt minimisation algorithm [16,17]. The reflectivities corresponding to the '↑' and '↓' spin states measured at a specific magnetic field and temperature were fitted simultaneously as fitting only spin asymmetry might lead to systematic errors when the background contributions are different in $R_\uparrow$ and $R_\downarrow$.

3. Results and discussion

In Fig. 1a, the reflectivities measured in the normal state for an in-plane applied field of 800 G were plotted. The periodicity of the principal fringes appearing on the reflectivities correspond to the thickness of the $Nb_{67}Ti_{33}$ layer and other parameters like number densities, scattering length densities, surface roughness of different layers describing the nuclear profile of the multilayered structure were obtained by fitting the reflectivity data in the normal state. The thickness and RMS roughness (within the bracket) of different layers estimated from this are as follows – Ta: 14.6(1) Å, Pd (top): 30.2(1.5) Å, $Nb_{67}Ti_{33}$: 2016(23) Å, Pd (bottom): 29.3(1.1) Å. The absence of significant roughness on different layers confirms the presence of smooth interfaces in this sample. In order to describe the magnetic behaviour of the sample in the most simplest way, the magnetisation in different layers were described by magnetic potential steps of finite heights and the magnetisation in an individual layer has been assumed to be uniform across the thickness of that specific layer (see Profile – A in Fig. 1b). Using this model, minimal evidence of magnetism was found in the normal state which on cooling below $T < T_c$ did not show any significant change. This can be viewed from the reflectivity data in Fig. 1a where the difference between $R_\uparrow$ and $R_\downarrow$ is negligible indicating minimal magnetic contrast in the system, also supported by the absence of pronounced spin asymmetry as illustrated in the Fig. 2a. It is to be

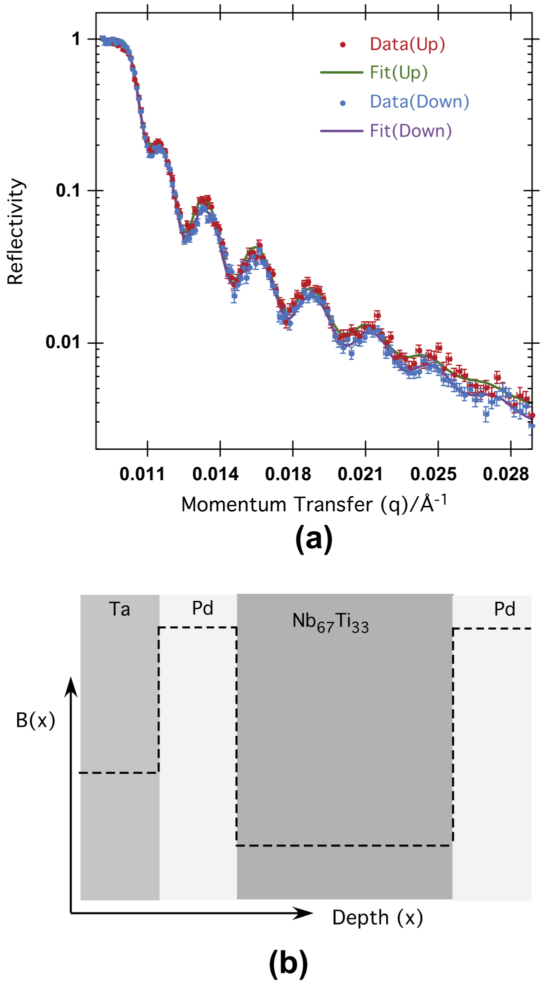

Fig. 1. (a) Polarised neutron reflectivity data measured in the normal state of $Nb_{67}Ti_{33}$ layer for an in-plane applied field of 800G. The points are the experimental data and the solid lines are the fits. (b) A cartoon describing the magnetic behaviour as described in Profile – A.