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X-ray scattering study of interfacial roughness in Nb/PdNi multilayers

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ABSTRACT

Specular and diffuse X-ray scattering are used to study interfacial roughness in $Nb/Pd_{0.81}Ni_{0.19}$ multilayers deposited by dc UHV sputtering. The data are analyzed to extract information about the correlated behavior of interface roughness in both the lateral and vertical directions. X-ray reflectivity is treated quantitatively by computer-aided simulation and modelling in order to extract values also for the layers thickness. From the analysis of the diffusive spectra of the reflectivity maps the roughness correlation has been evaluated.

1. Introduction

In recent years much activity has been devoted to the study of the interplay between superconductivity (S) and ferromagnetism (F) in artificial S/F hybrids in which these two antagonistic orderings are present in spatially separated layers, interacting via the proximity effect [1,2]. Such great interest is due to the peculiar effects emerging from the inhomogeneous nature of the superconducting order parameter induced in the F layer. Signatures of this inhomogeneous state are: nonmonotonic dependence of the superconducting transition temperature as a function of the ferromagnetic layer thickness, $T_c(d_F)$, (for an exhaustive review see [3]), nonmonotonic behavior of the anisotropy coefficient in S/F/S trilayers [4], negative critical current [5,6] and reversed density of states [7] in Josephson and tunnel S/F/S π-junctions. At the beginning of this research the experimental investigation was performed on S/F multilayers [3,8–11]. However, also due to the lack of sample reproducibility, the results of the measurements were often not conclusive and a clear comparison between the theoretical predictions and the experimental results could be controversial [12–14]. Even correlations between the observed $T_c(d_F)$ dependencies and the preparation methods (Molecular Beam Epitaxy or sputtering technique) were suggested [3,15]. Nowadays, it is well known that the superconducting properties of S/F hybrids are strongly influenced by certain aspects of their structure, for example the thickness control of each layer [16,17], the flatness [18–20], and the presence of alloying or interdiffusion at the interfaces [21,22]. Fortunately, the continuous developments in the fabrication techniques made recently possible the reliable realization of complex heterostructures consisting of layers of different materials only few nanometers thick, coupled through high-quality contacts. Lately, even so called stepped 0–π Josephson junctions, namely junctions with a tailored step in the thickness of the ferromagnetic layer, were successfully fabricated [23]. Very recently a complex multilayered structure consisting of up to more than ten single layers has been employed to observe spin-triplet superconductivity in ferromagnetic Josephson junctions [24,25]. These technological progresses are crucial, since the peculiar effects related to the S/F coexistence are observable in a narrow range of ferromagnetic thickness of a few nanometers, where structural and magnetic properties are also critical. Due to the small ferromagnetic thickness required in these structures, in fact, the thickness and the roughness of these layers has to be controlled on a sub-nanometer scale. It has been demonstrated that, for instance, small fluctuation in the layers parameter, especially in the region where the π-phase takes place, could affect the shape of the resistive transitions of S/F/S trilayers [26–28]. Moreover, study of interfaces between metals or oxides is important in view of modification of ultra-thin films grown on different templates [29] and for optimizing the low-dimensional devices [30,31]. For all these reasons the preparation of high-quality multilayers should be

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supported by a robust characterization of these key physical parameters. In this sense X-ray Reflectivity (XRR) provides a nondestructive and accurate characterization of thin films and multilayers. XRR probes the intensity of scattered X-rays at small angles, providing information about film thickness, roughness, and density. These analyses, pursued for the case of S/N systems (here N stands for normal metal) [32–34], is lacking in the case of S/F ones. To our knowledge XRR has been employed on hybrid S/F structures only in the specular condition, to evaluate the thickness and the roughness of the layers [9,11,13–15,35–38]. Occasionally Rutherford Backscattering analysis (RBS) has been also used to determine the layers thickness [15–17] with no particular focus, however, on the elemental area density or impurity distribution. We believe that a more systematic study of the structural properties of heterostructures employed in experiments aimed at the investigation of the interplay between superconductivity and magnetism is fundamental.

In this paper X-Ray reflectivity (XRR) has been employed in the characterization of a series of high-quality superconducting Nb/$Pd_{0.81}Ni_{0.19}$(PdNi) multilayers (containing up to nineteen single layers) deposited by dc UHV-sputtering. Reflectivity data are treated quantitatively by computer-aided simulation and modeling in order to extract values for the interface roughness and the layers thickness. From the analysis of the diffusive spectra of the reflectivity maps also the roughness correlation has been evaluated.

2. Fabrication and preliminary characterization of the multilayers

2.1. Growth of Nb/PdNi multilayers

The samples consist of weakly ferromagnetic PdNi and superconducting Nb layers. The Nb/PdNi multilayers analyzed in this work have the following structure: Si/(PdNi/Nb)$_{\times m}$/PdNi, where m is the number of bilayers in the stack. The samples have nominal Nb and PdNi layers thickness of $d_{Nb}=190$ Å and $d_{PdNi}=20$ Å, while different for the number of bilayers, $m=5$, 7, 9. In the following we will call these samples KL5, KL7, and KL9, respectively. The PdNi cap layer has the same nominal thickness of the internal one. Great care was paid to samples fabrication in order to provide identical deposition conditions for all the multilayers in the series. This concern pertains in particular to the quality of the interfaces and of the layering, as well as to the thickness control through the whole structures. The multilayers were deposited by dc magnetron sputtering on Si(100) substrates at room temperature. The sputtering is a multi-target UHV vacuum system, equipped with a load-lock chamber. The base pressure in the main chamber was in the 10^{-10} mbar range. The load-lock, with a base pressure of the order of 10^{-8} mbar, can house up to 6 substrates. In each deposition run they are transferred one at a time and positioned exactly in the center of the deposition chamber. The Argon pressure during the deposition is precisely fixed and monitored to a value of 4×10^{-3} mbar. The Nb and PdNi layers have been deposited at typical power of $W_{Nb}=77$ Watt and $W_{PdNi}=105$ Watt, which correspond to the rates $r_{Nb}=1$ Å/s and $r_{PdNi}=2$ Å/s, respectively. This careful deposition procedure is necessary to exclude fluctuations of the samples parameters.

2.2. Compositional analysis. Superconducting and magnetic properties

Since the ferromagnetic properties of the PdNi alloy strongly depends on the Ni content a careful characterization of this quantity is necessary. To this purpose the Ni concentration in the samples was checked by Energy Dispersive Spectroscopy (EDS). In particular, to determine the Ni content and the uniformity of the composition the samples were analysed using a LEO-EVO 50 scanning electron microscope equipped with an energy dispersive spectrometer (EDS, Oxford INCA Energy 300). The EDS analysis was performed over different areas for each film. This investigation pointed out a uniform percentage of Ni in PdNi alloy. The result of both the methods gives the value of 19% for Ni in PdNi alloy which is well above the critical percentage [39].

The superconducting and magnetic properties of the Nb/PdNi multilayers have been studied in great details in previous papers [40–42]. Here in Fig. 1 we show the resistive transition curve for the sample KL5, which becomes superconducting at $T=5.5$ K.

Using a Vibrating Sample Magnetometer (Cryogenics Ltd.), magnetization curves have been measured to observe if magnetism was established in the hybrids. Due to the small signal coming from the thin layers of the weakly ferromagnetic alloy a multilayer with $m=14$ has been deliberately fabricated in order to increase the magnetic signal. In Fig. 2 is plotted the *M(H)* curve measured on this multilayer which shows that at $T=4$ K the sample is ferromagnetic. From that, we calculated the saturation magnetization $M_{sat}=0.33\ \mu_B$/atom. This value agrees with the one obtained using a SQUID magnetometer on a single PdNi film (19.2 nm thick) with the same Ni content [40].

3. Low angle X-ray reflectometry

3.1. The scattering geometry

A Philips X'Pert-MRD high resolution analytic diffractometer equipped with a four-circle cradle was used to study the X-ray reflectivity of the samples. A Cu $K_{\alpha1}$ ($\lambda=1.5406$ Å) source was used at 40 kV and 40 mA. Low angle X-ray measurements were carried out by using a monochromatic radiation obtained equipping the diffractometer with a four crystal Ge 220 Bartels asymmetric monochromator and a graded parabolic Guttman mirror positioned on the primary arm. On the secondary arm, a parallel thin film collimator guaranteed the alignment of the reflected beam from the samples to the detector.

In Fig. 3 is shown a schematic drawing of coplanar X-ray reflectivity measurements performed in the present experiment. The incident angle, ω, is the angle at which the X-ray beam falls on the sample surface. The angle between the incident beam and the diffracted beam is indicated as 2θ. The scattering process involves the wave vectors of the incident and the scattered wave, $\mathbf{q}_i$ and $\mathbf{q}_f$ respectively, defining the wave vector transfer $\mathbf{q}=\mathbf{q}_i-\mathbf{q}_f$. The angular coordinates are related to wave vector transfer ones according to:

$$q_x = \frac{2\pi}{\lambda}(\cos\omega - \cos(2\theta-\omega))$$

$$q_z = \frac{2\pi}{\lambda}(\sin\omega + \sin(2\theta-\omega))$$

representing the components of the wave vector transfer parallel and perpendicular to the scattering plane. In this way it separates regions

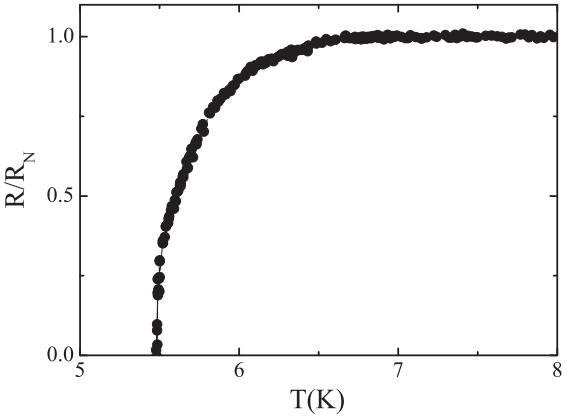

Fig. 1. Superconducting transition curve for the sample KL5.