Magnetization and AC Susceptibility Study of Nb/Ni/Nb Thin Films

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A study on the behavior of the magnetization and the ac susceptibility in Nb(40 nm)/Ni(x nm)/Nb(40 nm) thin films was carried out. It was observed that the hysteresis loops of these heterostructures show oscillations of the magnetization in regions of magnetic field where the irreversible reversal of the magnetization of the ferromagnet occurs. This behavior appears to show a coupling between the magnetic fields generated by the vortex in the Nb and the magnetization of the Ni nanograins. The analysis of the real and imaginary components of the ac susceptibility in the tri-layers shows no uniform results in relation to Ni layer thickness. However, the critical temperatures calculated from both components at zero applied field show an oscillation with respect to the Ni layer thickness.

***Index Terms*—AC susceptibility, niobium-nickel films, superconductor-ferromagnet hybrids.**

I. Introduction

ONE of the main characteristics of nanostructured systems that exhibit new phenomena or show unique properties is that they are heterogeneous. This means that are formed by two or more materials with different properties which have a surface contact or are separated by a thin layer of another material. Exchange-spring magnets where two ferromagnetic phases, one hard and one soft, are in contact through the surface are an example of the previously mentioned. Bilayers formed by ferromagnetic and antiferromagnetic phases, which leads to unidirectional anisotropy, constitute another example extensively studied.

A heterogeneous system that attracts a lot of interest and has been the subject of intense study in recent years is that formed by a ferromagnetic material and a superconductor [1]–[3]. The physical phenomena that arise when two solid materials with mutually excluding states, such as ferromagnetic and superconducting, are placed in contact, offer a wide range of research and potential technological applications. In these systems, sometimes referred to as Superconductor-Ferromagnet Hybrid systems (FSH), a strong mutual interaction between both subsystems can dramatically alter the properties of the constituent materials. Interaction of superconductivity with ferromagnetism has been extensively studied, both in experimental and theoretical form, in several systems. However, most of the research has been centered on electric transport properties and relatively little work has been conducted to understand the magnetic behavior of these heterostructures. Furthermore, the magnetic phenomena involved in the interaction between ferromagnetism and superconductivity are abundant and poorly understood, so that their study is fertile ground to gain a more complete understanding of these physical systems.

In a previous work by Mattson *et al.* [4], the authors showed, for multilayers of Nb/Ni, the superconducting element is highly sensitive to the presence of the ferromagnetic layer. They displayed measurements in which the superconductivity was destroyed at ferromagnet-thicknesses of the order of 1 nm and this was explained as an effect of the onset of the exchange field in the superconducting. Lee *et al.* [5] found a monotonic decrease in the critical temperature with increasing thickness of the ferromagnet in Nb/Co/Nb tri-layers and highlighted trends of suppression of the dominant superconducting contribution to the magnetization by the Co films [5]. Shortly after, they also reported interaction between the Nb layers through a very thin Co film [6]. More recently, nuclear magnetic resonance spectroscopy has been used to look for the spin screening effect in F/S/F arrangements [7], [8] and exotic phenomena like oscillations of the induced magnetization have been proven [9]. The observation of these features depends crucially on precise magnetic measurements.

In this paper we present a study on the behavior of the magnetization and the ac susceptibility in thin films of Nb/Ni/Nb. For sufficiently low temperatures, the hysteresis loops of these heterostructures show oscillations of the magnetization in regions of magnetic field consistent with the irreversible reversal of magnetization of the ferromagnetic. Then, the reversal of magnetization of the Ni nanoparticles can have effects on the vortex distribution of Nb, modifying the overall magnetic behavior of the tri-layer. It was also conducted an analysis of the behavior of the ac susceptibility as a function of temperature and strength of the applied field to the different samples.

II. Experiment

Tri-layers of nominal composition Nb(40 nm)/Ni(x nm)/Nb(40 nm) were deposited on Si(100) substrates by dc magnetron sputtering from high purity commercial targets of Nb and Ni. The substrates were maintained at room temperature and the depositions were carried out only when the vacuum in the chamber was better than 2×10^{-7} Torr. The samples were covered by a thin layer of Ag (~4 nm) as a protection to avoid oxidation. The different samples prepared in this study correspond to the ferromagnetic layer thicknesses of 5, 10, 15, and 30 nm. A sample consisting of a monolayer of Nb and protected by a thin layer of silver was also prepared under identical conditions to the other, for the purpose of comparing the magnetic properties.

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TABLE I

Sample ID	Thickness Nb (nm)	Thickness Ni (nm)
A	40±2	0
B	40±2	4±1
C	40±2	9±1
D	40±2	15±1
E	40±2	29±1

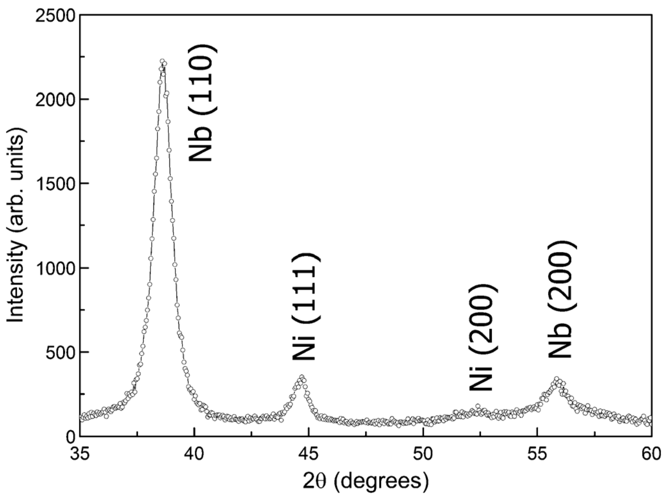

Fig. 1. XRD pattern of the sample E.

Structural characterizations of the samples were made through Rutherford Backscattering analysis (RBS) to determine layer thicknesses and X-ray diffraction (XRD) to obtain grain size and possible crystal texturing.

The magnetic characteristics of the films were obtained from hysteresis loops at different temperatures and ac susceptibility measurements by using a Quantum Design's SQUID magnetometer. The ac susceptibility measurements were carried out on samples of lateral dimensions of roughly $5 \times 10\ \mathrm{mm}^2$, in the temperature range between ~5 to 9 K with and without a dc magnetic applied field (parallel to the plane of the films). In order to generate the real (χ') (in-phase) and the imaginary (χ'') (out of phase) components of the differential magnetic susceptibility an ac magnetic field of 3 Oe and 71 Hz was applied.

III. Results and Discussion

A. Structural Characteristics

Table I shows the thicknesses of the Ni layer in each one of the samples studied in this paper. Sample A corresponds to a monolayer of Nb.

A typical result of XRD of one of the films is shown in Fig. 1. This pattern corresponds to a tri-layer with 29 nm of Ni (sample E) and it shows the two main peaks of Nb and the two main peaks of Ni.

Using the angular position of the peaks, the lattice parameter of each element was calculated, and the results are $a_{\mathrm{Nb}} = (3.300 \pm 0.005)$ Å and $a_{\mathrm{Ni}} = (3.512 \pm 0.015)$ Å, respectively. Compared with the corresponding values of bulk materials ($a_{\mathrm{Nb}} = 3.300$ Å and $a_{\mathrm{Ni}} = 3,520$ Å), it was observed that there was good crystallinity in the samples. Also, there was no texturing on these films. The application of the Scherrer formula for the grain size in the layer of Nb resulted in approximately (8 ± 1) nm, while the result for Ni is very similar, (9 ± 1) nm. Although the crystallite size of Ni is small, the ferromagnetic characteristic of this layer is guaranteed since all the effects that are going to be analyzed occur at low temperatures.

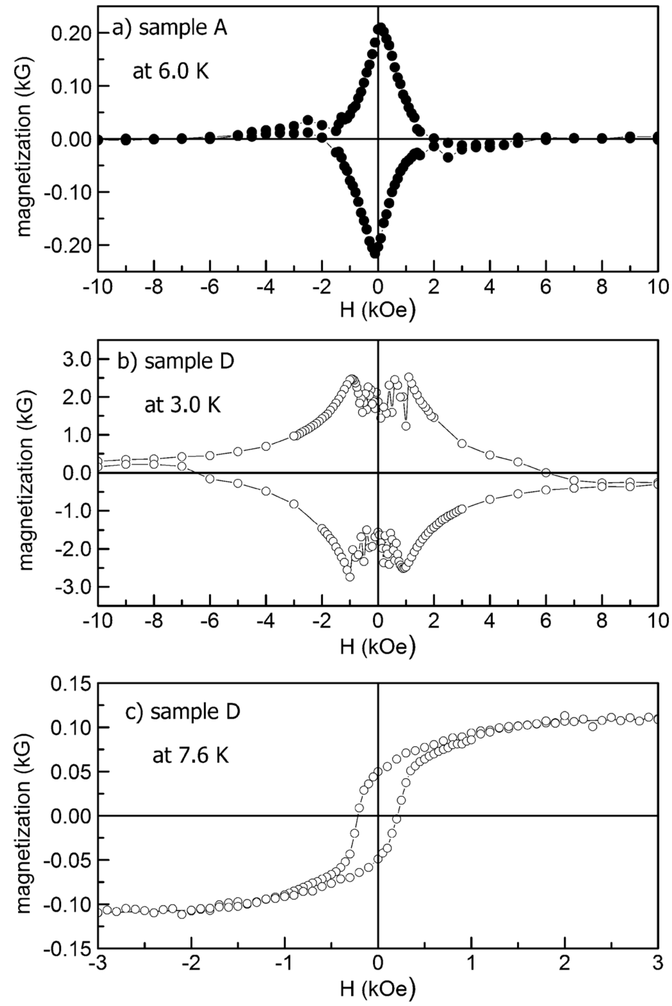

Fig. 2. (a) Hysteresis cycle of sample A. (b) Hysteresis loop of sample D at 3.0 K and (c) at 7.6 K.

B. Magnetic Properties

The effect of the presence of the layer of Ni on the hysteresis loop of Nb can be seen, in the way of example, by the behavior shown in Fig. 2. Sample A is superconducting at temperatures less than 7.7 K, as discussed later. Fig. 2(a) shows the hysteresis loop of the film of Nb obtained at 6.0 K. Similar curves were obtained for temperatures ranging between 3 and 7 K.

In all cases it was observed that the normal increment of the magnetizing is near to the zero field, and a smooth second quadrant. However, this picture is completely different to those samples which have a layer of Ni. For example, in Fig. 2(b) it is shown that the hysteresis loop was obtained at 3.0 K for the sample D. In this image are notable the magnetization oscillations that occur when the applied field strength is in the range ± 1 kOe, approximately. This region of magnetic fields appears to correspond to the region where irreversible rotations occur in the magnetization of the nanoparticles in the layer of Ni. This statement can be corroborated by observing the magnetic behavior of this tri-layer at 7.6 K [Fig. 2(c)], a temperature just above the critical temperature of superconducting Nb layers in the sample. In Fig. 2(c), the magnetization corresponds exclusively to the layer of Ni, which exhibits a coercivity of approximately 220 Oe. One can observe also that the hysteretic behavior of this layer is approximately restricted to the region of ± 1 kOe. For fields applied above this value, the ascending and descending branches of the hysteresis loop are overlapped, so that only reversible rotation events are expected to more intense fields. It is possible to think that the change in orientation of magnetization in the Ni nanograins produces variations in the vortex distribution in the layers of Nb, from the contact surfaces between both materials. The exchange length of Ni ($\sim$8 nm) is sufficiently long to encompass a substantial part of the layers of Nb, making the effect visible. This appears to agree with that mentioned in [10] where it is stated that vortex currents in the superconducting generate magnetic fields that interact with the magnetization of the ferromagnet, which leads to the formation of topological defects coupled superconductor-ferromagnet. Several recent studies in hybrid S/F have reinforced this view. For example, the strong coupling between vortices in the S film and magnetization in F films with perpendicular anisotropy results in a combined domain structure where vortices reside on top of magnetic domains of the same polarity, as was experimentally observed and theoretically calculated by Vlasko-Vlasov *et al.* [11]. Also, it was shown that through the interaction between the stray magnetic field from nanostructured magnetic vortices and a superconducting thin film, it is possible to induce significant changes in the transport properties of S [12]. Similar results were presented in [13] and [14].

In order to observe the effect of the Ni-thickness on the critical temperature of superconductivity of Nb, ac susceptibility measurements were performed in the samples. The measurements were performed as a function of temperature and for various applied field strengths. Initially, tests were performed varying the frequency of the ac wave, for frequencies between 1 and 800 Hz, approximately. However, there were no important differences, so that the final tests were done at an intermediate frequency of 71 Hz, as aforementioned. The general form of the real and the imaginary components of susceptibility are shown in Fig. 3 for two different samples (A and C) and three field intensities (zero, 500 Oe, and 1000 Oe).

When the Nb layers in the heterostructure are in the superconducting state, it is expected that the real component of susceptibility is negative. As the temperature increases, this component increases and eventually vanishes when the material becomes normal metal. So, it is typical to define the critical temperature for this phase transformation as the value for which χ' vanishes. This temperature, which will be named T^*, is shown in Fig. 3(a) for sample A. An important difference between the component χ' of sample A and that corresponding to the other samples is that for tri-layer χ' becomes positive for temperatures above T^* [see Fig. 3(c)]. This is because there is a positive contribution to χ' from the Ni layer. As expected, with the increase of the intensity of the applied field, the field penetration into the superconductor causes the intensity of the χ' component to decrease and become zero to increasingly smaller values of T^*. It is shown in Fig. 4(a) for the different samples.

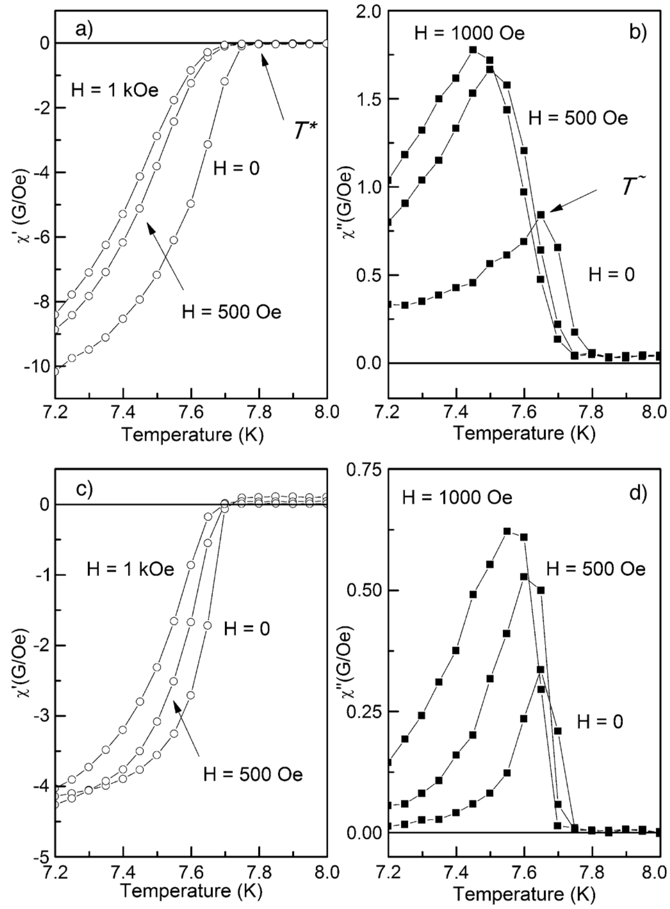

Fig. 3. AC susceptibility for samples A and C at 0, 500, and 1000 Oe. Panels (a) and (b) correspond to the real and the imaginary components of sample A, respectively. (c) and (d) display the corresponding components of sample C.

The temperature dependence of the imaginary part of the ac susceptibility, χ'', is shown in Fig. 3(b) for the sample A and Fig. 3(d) for the sample C. χ'' reaches a maximum at a point that coincides, approximately, with the maximum of the derivative of χ'. The peak at χ'', symbolized here by $T^{\sim}$, is produced by highly metastable state in which the Nb is found when taking place the phase transformation. In fact, this temperature has been considered an appropriate value to characterize the transition [9]. The change of $T^{\sim}$ with the field strength, for all samples studied, is shown in Fig. 4(b).

The behavior of the critical temperatures T^* and $T^{\sim}$, as a function of the intensity of the applied field, is shown in Fig. 4. It can be observed that both temperatures do not vanish even for fields as intense as 14 kOe. Similar results for multilayer stacks of Nb(23 nm)/Ni(5 nm) were reported by De Long *et al.* [15]. However, the behavior of these critical temperatures for the different samples is somewhat puzzling. As shown in Fig. 4, the curves corresponding to different thicknesses of Ni do not follow a uniform pattern, and there are some intersections among curves. This seem to be more evident in the low field region. For high fields, on the other hand, Fig. 4(a) shows that the critical temperature of the Nb monolayer is substantially

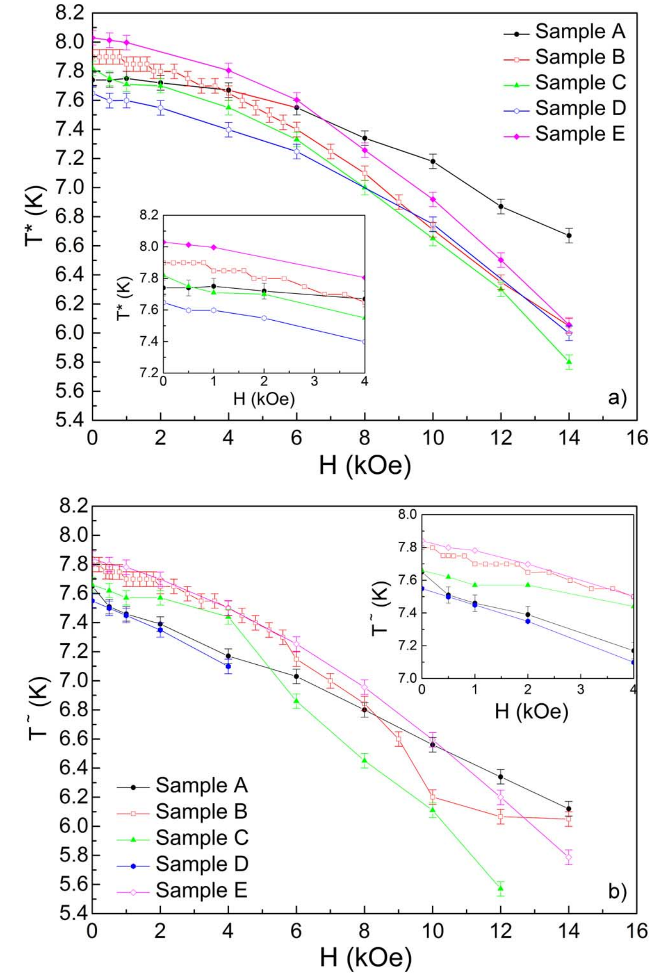

Fig. 4. Critical temperatures T^* (a) and $T^\sim$ (b) as a function of the field strength for the samples studied. Insets in panels show the low field region.

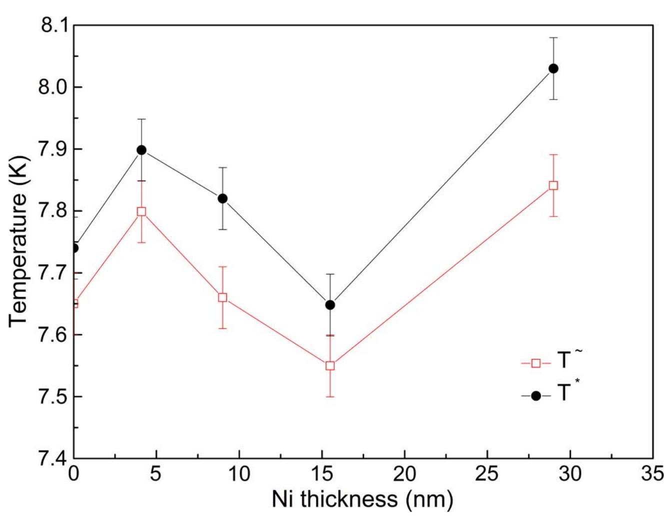

Fig. 5. Dependence of the critical temperatures T^* and $T^\sim$, to zero field, with the thickness of Ni layer.

greater than those samples having Ni. Although the behaviors of both critical temperatures are not exactly in unison, as might be expected, it is interesting to note that for low field strengths, the sample E (largest thickness of Ni) shows higher values of T^* and $T^\sim$.

Finally, the results of T^* and $T^\sim$ to zero field, as a function of the Ni layer thickness, are shown in Fig. 5. We can observe from the figure that the critical temperatures obtained from the ac susceptibility exhibit an oscillation with a minimum value for the thickness of 15 nm, approximately. Similar oscillations of the critical temperature as a function of the thickness of the F layer were observed by Jiang *et al.* [16] in Nb/Gd, and Cirilo and co-workers [17] in Nb/PdNi.

IV. Conclusion

A study on the behavior of the magnetization and the ac susceptibility in thin films of Nb/Ni/Nb was carried out. The main result of this investigation shows the effects of the Ni layer on the magnetization curve of the heterostructure. In fact, when the temperature is sufficiently low and the Nb is in the superconducting state, the hysteresis loops show oscillations of the magnetization in regions of magnetic field where the irreversible reversal of the magnetization of the ferromagnet occurs. This behavior appears to show a coupling between the magnetic fields generated by the vortex in the Nb and the magnetization of the Ni nanograins. The analysis of the real and imaginary components of the ac susceptibility in the tri-layers shows no uniform results, in relation to Ni layer thickness. Particularly, the critical temperatures calculated from both components at zero field show an oscillation with respect to the aforementioned thickness. This suggests that a more comprehensive analysis should be performed for this type of system.

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