Magnetic State Modification Induced by Superconducting Response in Ferromagnet/Superconductor Hybrids

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Magnetization measurements in superconductor/ferromagnet Nb/Co superlattices show a complex behavior as a function of temperature, applied field and sample history. In base to a simple model it is shown that this behavior is due to an interplay between the superconductor magnetization temperature dependence, the ferromagnet magnetization time dependence, and the stray fields of both materials. It is also shown that the magnetic state of the Co layers is modified by the Nb superconducting response, implying that the problem of a superconductor/ferromagnetic heterogeneous sample has to be solved in a self-consistent manner.

The interaction between a superconductor, SC, and a ferromagnet, FM, in close contact at an interface, as in a superlattice, has attracted attention in the last years due to the possibility of fabricating SC/FM hybrid devices. These engineered materials originate the appearance of interesting physical phenomena due to the different scales and mechanisms of interaction, like SC pair breaking effects related to exchange interaction at the interface, or electromagnetic interaction with the stray fields of the FM both at the mesoscopic and macroscopic level.

Most of the research has focused on the ways in which the FM affects the SC response. For example, in the Domain Wall Superconductivity effect observed in SC/FM bi-layers, superconductivity nucleates in those places where the perpendicular component of the inhomogeneous FM domain structure’s stray field is close to zero. In the Spin Switch effect, observed in FM/SC/FM trilayers, the Cooper pair, due to its finite size, experiments different average values of the exchange field when the FM layers are ferro- or antiferromagnetically oriented. As a consequence, the SC order parameter is more depressed when the FM layers are ferro-magnetically oriented. This allows the control of the SC order parameter value through an external macroscopic parameter.

In contrast, very little work has been done in exploring in which way the SC affects the magnetic state of the FM layer. Recently, we have shown the importance of the FM stray fields in the overall magnetic response of Nb/Co superlattices. In that work we also hinted to the possibility that the SC response may modify the magnetic state of the FM layers. In this letter we show that, indeed, the SC response modifies the magnetic state of the FM layers. The system global electromagnetic response is determined by an interplay between the SC magnetization temperature dependence and the FM magnetization time evolution.

We present data on the temperature, T, dependence of the magnetic flux expulsion, \( \Delta \phi \), directly proportional to the SC magnetization, for Nb/Co superlattices. Data is presented for a \([Nb(44 nm)/Co(10 nm)]x19\) and a \([Nb(44 nm)/Co(7.5 nm)]x19\) superlattice. The results on these samples are representative of the measurements we performed on a collection of Nb/Co FM/SC superlattices. Since our experimental setup measures magnetic flux variations, all \( \Delta \phi \) values are measured with respect to the first data point, always above the superconducting critical temperature, \( T_{CS} \). Sample preparation, characterization method and measurement details are described in reference. For all flux expulsion data,
the applied field, $H_a$, is parallel to the sample surface. In the normal state, both superlattices present FM behavior with Curie temperatures, $T_C$, above 300 K. Flux expulsion in the SC state was measured as a function of $T$ in field cooling experiments, for two different Co layer’s initial FM states, -FC, with the Co layers initially saturated in the negative $H_a$ direction, and +FC, with the Co layers initially saturated in the positive $H_a$ direction. A detailed explanation of these measurement protocols is also included in reference 5.

Figure 1 is a composite that summarizes the experimental results. In the main panel we show the FM hysteresis loop of the Co layers at $T = 7 K$, close but above $T_{CS}$ of the Nb layers. In the superimposed panels, we show the $T$ dependence of $\Delta \phi$ for +FC and -FC measurements. For each initial state, several $T$ cycles were measured, each cycle sweeping $T$ down from 7 K to 5.5 K and up to 7 K again. The solid dots in the hysteresis curve connected with arrows to the panels indicate the initial magnetic state for each experiment.

The data for the first $T$ down-sweep in each panel shows the behavior already discussed in our previous work.[5] As discussed there, the SC response is proportional to the effective field, $H_{eff}$, originated by the superposition of the applied field, $H_a$, and the Co layers’ stray field, $H_s$. For the -FC measurements (lower branch of the Co hysteresis loop), at low $H_a$ and negative Co magnetization, the SC layers sense a positive $H_{eff}$ due to the Co’s $H_a$, see panel (d). At higher $H_a$, the Co magnetization becomes positive, $H_s$ becomes negative and larger than $H_a$, and the SC senses a negative $H_{eff}$, see panel (b). Panel (c) shows an intermediate case, where the magnetization is already reversed, but $H_s$ is smaller than $H_a$ and the SC still senses a positive $H_{eff}$. Panel (a), +FC initial state (upper branch of the Co hysteresis loop) is the mirror experiment from panel (d).

The novel feature observed in these data is present in the dependence of the Co magnetization in the normal state with the number of $T$ sweeps, i.e. cycles. This dependence is observed as a non-repeatability of the normal state magnetization value after a cycle is completed. This behavior is not due to an experimental artifact related to an instrumental drift, since this instrumental drift has been subtracted from the data. A systematic behavior is observed in spite of the seemingly complex dependence. The direction of the variation follows the sign of the applied field, and is independent of the Co magnetization direction, i.e. the stray field, compare data in panel (b) and (d), for example. The difference between the first and second normal state $\Delta \phi$ values in panel (d) of 360 superconducting flux quanta is equivalent to a change of 1.1 emu cm$^{-3}$ in the Co layer’s magnetization, which shows that this effect is small but not negligible.

In order to understand this behavior, we have constructed a simple “toy model” to qualitatively simulate the experimental data. Although the model is very simple, a careful consideration of its hypothesis should be made to fully understand the implications of the results. The first requirement is that an electromagnetic stray-field mediated interaction should exist between the FM and the SC components. This is not achievable if the materials are modelled as nearly infinite slabs parallel to the applied field, since the stray field of this geometry is negligible. Consequently, both materials are modelled as ellipsoids with one of the principal axis parallel to the field. The “toy model” sample consists, then, of a FM and a SC ellipsoids, located side by side. Figure 2 depicts the main ideas of the model. Panel (a) shows the situation at $T > T_{CS}$ where the $H_{eff}$ sensed by the SC ellipsoid is composed by $H_a$ (straight lines) and the FM $H_s$ (dipolar lines arising from the FM ellipsoid). When $T$ is reduced below $T_{CS}$, as depicted in panel (b), the flux expulsion from the SC ellipsoid modifies the $H_{eff}$ sensed by the FM ellipsoid and consequently its magnetization. The solution of the problem has now to be found in a self-consistent way.

The ellipsoid shape or eccentricity, $\epsilon$, was selected as to maximize the stray field effects. That an optimum value exists is clear from the fact that in the $\epsilon \rightarrow \infty$ “needle” limit, the stray fields approach zero due to the negligible demagnetizing effects, and that in the $\epsilon \rightarrow 0$ “disk” limit, the stray fields also approach zero since the ellipsoid is being magnetized along the shape anisotropy “hard axis”. The optimal $\epsilon$ value actually depends on the material’s magnetization, but since it is weakly dependent on it, a value of 10 was found to maximize the stray field effects in nearly all the $T-H_a$ range. Also, since an exact three dimensional spatial solution of this electromagnetic problem is beyond the scope of this work, and would only obscure the results of the model, the spatial dependence of the stray fields is neglected, and $H_a$ due to

![Figure 2: Schematics of the “toy model” behavior. Panel (a): at $T > T_{CS}$ the normal Nb ellipsoid experiences an effective field due to the applied field (straight lines) and the ferromagnetic Co ellipsoid stray field (dipole like lines). Panel (b): As $T$ is reduced below $T_{CS}$ the magnetic flux expulsion from the Nb ellipsoid modifies the effective field over the Co ellipsoid.](image-url)
each ellipsoid is evaluated only at the center of the other ellipsoid.

The second ingredient in the model is a “time” dependence. This dependence cannot be ascribed to the superconducting material since we have shown that no vortices are present in the \( T - H_a \) range of these experiments. Consequently, it must be arising from the creep in the FM material. Following this idea, the magnetization of the SC Nb ellipsoid is modelled by a \( T \) dependent, time independent Meissner state. As a further simplification of the model the \( T \) dependence is forced to follow that of a parallel slab with a two fluid \( T \) law. On the other hand, the FM Co ellipsoid magnetization does not present a \( T \) dependence since its \( T_{CS} \) is much higher than the measurement range. It only shows a time dependence which must be numerically simulated, as described in the next paragraph.

To simulate the \( T \) sweeps at constant \( H_a \), the self-consistent equilibrium state of the magnetized ellipsoids is solved at a given \( T \). After this, the magnetization change for the Co ellipsoid, is reduced by a given percentage, and the magnetization of the Nb ellipsoid is recalculated for this, now fixed, value of the Co ellipsoid’s magnetization, i.e. stray field. This algorithm results in an effective exponential time dependence for the Co magnetization. The sample’s magnetization, \( M_T \), is defined as the total magnetic moment divided by the total sample volume. In order to compare the results to the experiments, the simulation data is presented as \( \Delta M_T = M_T - M_o \), where \( M_o \) is the value for the first simulated point, always at \( T > T_{CS} \).

Panels (a) and (b) in figure 3 show the prediction of the model for situations similar to panels (c) and (a) in figure 1, i.e. opposite direction of \( H_a \) and same value of \( M_o \). It is clear that the principal features of the experimental data are qualitatively reproduced. First, there is a dependence of the normal state magnetization with the number of cycles. This dependence follows the sign of the applied field and is not correlated to the magnetization direction. Second, there is an irreversibility between cooling-down and warming-up sweeps. Third, a non-monotonic \( T \) dependence is observed for cooling-down sweeps.

An interesting feature not actually observable in the data in figure 1 but presented in reference 8 is a non-monotonic \( T \) dependence that develops for applied fields near the coercive field of the Co layers. The main panel in figure 4 shows a comparison between experimental and simulated data, where the simulation parameters have been selected as to maximize this non-monotonic \( T \) dependence. The origin of this behavior becomes clear when examining separately the Nb and Co magnetization response in the simulated data. The inset shows the SC ellipsoid magnetization, \( M_{SC} \), the FM ellipsoid magnetization, \( M_{FM} \), and the sample’s magnetization, \( M_T \), as a function of simulated data point number, i.e. “time”, while \( T \) is swept down from above \( T_{CS} \). The \( T \) sweep is linear with this “simulated time”. The time dependence of \( M_{SC} \) is that arising from the \( T \) sweep, given that the Meissner state does not present an intrinsic time dependence. The time dependence of \( M_{FM} \), on the other hand, has a twofold origin. First, the flux expulsion in the SC originates an increase of local magnetic field in the FM material, as schematized in panel (b) of figure 2. Second, the \( M_{FM} \) presents an intrinsic time dependence in its response to the magnetic field changes. In this light, the origin of the non-monotonic \( T \) dependence becomes clear.

As \( T \) is swept down from above \( T_{CS} \), the \( T \) dependence of the SC ellipsoid magnetization produces a flux expulsion in the sample. This originates a field increase in the FM material increasing its \( M_{FM} \). At lower temperatures, the \( T \) dependence of the SC material is relatively weak, and the time dependence of the FM material emerges as a “paramagnetic” like signal, resembling a paramagnetic Meissner effect.

The results and the toy model presented here clarify the response of SC/FM hybrid structures and, at the same time, raise an interesting question. We have demonstrated that the electrodynamic response of these hybrid
systems involves a combination of two separate phenomena. In the first place, the diamagnetic response of the SC layers expels the magnetic flux into the FM layers. As a consequence, the FM material responds with a time dependence, clearly in the direction of the applied field. Both materials affect each other with their respective stray fields. In this process, the magnetic domain structure of the FM seems to play an important role, since the stray fields of an infinite slab are negligible. Clearly, in order to observe stray field effects, a non slab geometry has to be present in the samples. In this picture, an interesting point arises. Given that the response of the hybrid material is affected, and in some $T$ and $H$ range, dominated, by the intrinsic time dependence, the effects described here may be important if the device operation is based on magnetization changes and designed to work at frequencies similar to the creep of the FM.

In summary, we have demonstrated that the electrodynamic response of SC/FM hybrid materials is determined by an interplay between the temperature dependence of the SC magnetization, the time dependence of the FM magnetization, and the effective interaction between them mediated by the stray fields.

Work partially supported by ANPCyT PICT2003-03-13511, ANPCyT PICT2003-03-13297 and Fundación Antorchas. CM acknowledges financial support from and JG is a member of CONICET, Argentina.

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