Interplay between magnetism and superconductivity in Nb/Co multilayers

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We have used torque magnetometry to investigate the influence of magnetic spacer layers on the superconductivity in Nb/Co multilayers. Compared to similar systems which have nonmagnetic spacer layers, the samples exhibit two dimensional superconductivity for relatively thin magnetic layer thicknesses d_{Co} , reflecting the effects of magnetic pair breaking. Moreover, the transition temperature to the superconducting state T_c shows a nonmonatonic dependence on d_{Co} , which we examine within the framework of recent theories.

Over recent years there has been considerable interest in multilayer systems composed of alternating magnetic (M) and superconducting (S) layers. These systems are of interest as they are model systems in which to investigate the interplay of the competing superconducting and magnetic order parameters. The superconducting proximity effect brings the superconducting electrons into intimate contact with the local moments in the ferromagnetic layers, which can exert a pair breaking effect on the superconducting charge carriers via the exchange interaction. This in turn usually results in a rapid suppression of the superconducting transition temperature T_c . One controversial area which has attracted attention concerns the theoretical prediction of an *oscillation* of T_c when the thickness of the ferromagnetic layer d_M is increased. There have been a number of experimental works on S/M multilayers $[V/Fe (Ref. 1)$ and Nb/Gd $(Refs. 2$ and 3) showing a rapid decrease in T_c as a function of thickness followed by a local maximum. This was taken as a manifestation of an oscillation of T_c in these experiments. In other works [Nb/Co and V/Co, 4 V/Fe (Ref. 5)], however, the decrease of T_c was either monotonic or followed by a plateau showing no signs of oscillation.

A theoretical account of an oscillation in T_c has been suggested by Radovich et al.,⁶ who developed a dirty-limit version of a model due to Usadel, 7 in which Eilenberger's transportlike equations⁸ are applied to S/M interfaces. It was shown that when diffusing into a ferromagnetic layer the Cooper pair is subject to an interaction from the local exchange field. The result of this interaction is a phase shift in the superconducting wave function. When considered for the case of multilayers a phase shift $\Delta \phi = \pi$ between two neighboring superconducting layers can be energetically more favorable than the usual $\Delta \phi = 0$ found for an *s*-wave superconductor. The $\Delta \phi = 0 \leftrightarrow \pi$ phase shift occurs periodically as a function of the ferromagnetic layer thickness d_M , resulting in the oscillation of T_c . However, a nonmonatonic dependence of T_c is also observed in some bilayer and trilayer systems,^{9,10} where the π -switching effect is not applicable. Several explanations for these observations have been proposed, including the influence on T_c of changing interface transparency³ and a spatial modulation of the pair density in the ferromagnetic layer.¹¹

In this paper we present experimental results of a systematic study on Nb/Co multilayers, to investigate the interplay between magnetism and superconductivity in this system. We use our determination of the critical temperature T_c as a function of increasing thickness d_{Co} of the magnetic spacer layer to demonstrate a $T_c(d_{Co})$ dependence which has a clear *oscillatory* behavior in a way not previously observed in this or other systems. We use these results to speculate on the possible origin of the effect in our system. We also consider the enhanced effects of magnetic spacer layers on the decoupling of the superconducting layers compared to nonmagnetic systems.

The Nb/Co multilayers consisted of five superconducting layers of thickness 50 nm interspaced with ferromagnetic layers of thickness d_{Co} . Angular-dependent torque measurements were performed in the nonsuperconducting state to verify that for all values of d_{Co} the magnetic layers are ferromagnetic. The Nb/Co multilayers were prepared by MBE at the University of Leeds using $Si(111)$ substrates. Si wafers were cleaned at an estimated temperature of 880 °C to remove the native oxide layer. The growth chamber base pressure was maintained at better than 2×10^{-10} mbar throughout. On cooling a clear 7×7 reconstruction was routinely observed, indicating that the starting surfaces were clean and well ordered. A nominal 2.5 nm thick Nb film was first grown, followed by five bilayers of Co/Nb, where the Nb layer was 50 nm thick, whilst the Co thickness varied. The growth temperature of Co/Nb layers was about 200 °C. The samples were then cooled to about $100\degree$ C for the deposition of a 2.0 nm capping layer of Au. During growth, reflection high energy electron diffraction (RHEED) was employed to monitor the film structures. For all the samples grown, sharp concentric ring-shaped patterns were observed, indicating that these films are of a polycrystalline nature.

The samples were measured using a torque magnetometer constructed at St. Andrews. The sensor consists of a phosphor bronze cantilever at the end of which is placed the sample. On application of a uniform external magnetic field the sample experiences a torque $\tau = \mu_0 M \times H$ per unit volume of sample. This causes a deflection of the cantilever which may be determined by measuring the differential change in capacitance between the cantilever and surrounding electrodes. The change in capacitance is thus a measure of the elastic strain and hence of the torque exerted on the moment by the field. The technique is sensitive to compo-

FIG. 1. Torque as a function applied field for multilayer samples consisting of 5 layers of 50 nm thick Nb interlayered with Co layers. ~a! A sample with Co layer thickness 1.8 nm for the geometry with the field applied parallel to the planes. The filled circles were taken at 7.6 K, below T_c in the superconducting state. The open circles were measured at 10 K above T_c . For this orientation there is no contribution to the signal from the ferromagnetic layers (see text). The inset shows the point at which the superconducting contribution to the signal vanishes, which is used to determine $H_{c,\parallel}(T)$. (b) Similar measurements on a sample with Co layer thickness 4.2 nm, with the field directed at 20 \degree to the plane of the film. The filled circles were taken at 7.0 K, below T_c , and the open squares were measured at 10 K above T_c . For this geometry and layer thickness the contribution to the torque signal from the Co layers is significant. The value of $H_{c_2\perp}(T)$ may be determined from the point at which the contribution from the superconducting signal vanishes.

nents of the magnetization \tilde{M} which are *perpendicular* to the

applied magnetic field \vec{H} . This component may thus be determined from the measured torque and the value of the applied field. The measurement of superconducting films of isotropic materials is possible due to the strong shape anisotropy effect, where the ''demagnetization'' gives rise to significant components perpendicular to the applied field. The sensitivity of torque magnetometry clearly increases with applied field, which restricts its application at fields close to zero. For small moment systems such as the multilayers investigated here, it is thus particularly difficult to measure at low fields. However, as we discuss below, in the present study the temperature dependences of the measured upper critical fields follow very well defined functions for all orientations, so extrapolation to zero field may be performed with high precision.

The upper critical fields of the samples were measured for the field applied parallel to the plane H_{c} and perpendicular to the plane $H_{c_2\perp}$ of the films. For a given temperature the determination of the upper critical field was taken to be the point at which the field dependence of the torque signal $|\tau|$ $= \mu_0 |\tilde{M}| |\tilde{H}| \sin \theta$ changes from a rapidly varying to a slowly varying function of field. This H_c ₂ point is found very close to where the torque signal becomes reversible, and at fields above this point one simply observes that signal which is present in the normal state $T>T_c$. Examples of this type of measurement for two orientations are given in Fig. 1, which show the field-dependent torque signal both above and below the superconducting transition temperature T_c . For the case of the field oriented within the plane of the film there is no contribution to the torque signal from the ferromagnetic layers, since the moments are always confined to the plane and hence parallel to the applied field [Fig. $1(a)$]. This follows from the fact that the in-plane orientation is the easy direction for the ferromagnetic moments due to the effective demagnetizing field arising from the shape anisotropy of the thin film. We note that this orientation corresponds to the point of *maximum* sensitivity to the torque signal from the superconducting layers, since the shape anisotropy acts as an effective ''magnetizing'' field in a diamagnetic system, which together with the intrinsic anisotropy of the multilayer holds the magnetization vector close to the perpendicular direction. For the field applied at an angle to the plane of the film for $T < T_c$ there are contributions from both the superconducting and magnetic layers below $H_{c_2}(T)$, so that above this field the torque signal coincides with that from the ferromagnetic layers observed above T_c , which varies only slowly [Fig. $1(b)$]. For each orientation of the field relative to the superconducting layers, it was only possible to determine H_c , over a range of temperatures. The restriction at low temperature arises from critical field values in excess of our maximum available field (about 1 T). The high temperature limit is set by the point where the falling sensitivity with

FIG. 2. Angular-dependent torque signal for a sample with d_{Nb} =50 nm and d_{Co} =4.2 nm below T_c at a temperature of 7.7 K and an applied field of 2.72 kOe. The contribution to the signal from the ferromagnetic Co layers can be seen by reference to the inset which shows the same measurement taken above T_c at 10 K. The sharp peaks in the central region reflect pinning of vortices in the superconducting state, and their proximity to 180° indicates the high effective magnetic anisotropy of the superconducting layers $(see text).$

decreasing field prevents an unambiguous determination of the transition to the normal state. As we explain further below, however, this does not adversely effect the determination of the temperature dependence of the upper critical field nor the consequent determination of T_c .

In highly anisotropic superconducting systems such as the high- T_c cuprate materials, torque magnetometry is often used to estimate the superconducting anisotropy. This is usually performed above the so-called irreversibility line which exists in these materials, where hysteretic effects due to vortex line pinning are no longer present. The angulardependent reversible magnetization can then be fitted to yield a parameter reflecting the superconducting anisotropy $\gamma = \lambda_{\perp}/\lambda_{\parallel}$, where λ_{\perp} , λ_{\parallel} are the superconducting penetration depths from currents flowing perpendicular and parallel to the superconducting planes respectively.^{12,13} Figure 2 is a typical angular-dependent torque signal for one of the Nb/Co multilayers, close to T_c . It is apparent from the large peaks in the central region of the plot that the superconducting signal is irreversible. Furthermore, with reference to the inset, which shows the ferromagnetic signal for $T>T_c$, it can be seen that below T_c there is a very significant contribution to the signal from the Co layers. The combination of these two factors precludes the application of the above type of analysis.^{12,13} Moreover, one cannot ignore in these thin film samples the large contribution from shape anisotropy which is usually neglected in experiments on single crystals of high- T_c material. It is nonetheless worth noting that the close proximity of the two central peaks of Fig. 2 to 180° indicates that the total effective magnetic anisotropy of the supercon-

FIG. 3. Temperature dependences of the upper critical field $H_{c2\parallel}$ for the field applied parallel to the plane of the film. Each sample consists of 5 layers 50 nm thick Nb interlayered with Co layers of thickness given in the plot. Markers indicate experimental points; lines are fits to Eq. (1) (for the square-root dependence) and Eq. (3) (for the linear dependence).

ducting layers is high. 12 To gain insight into the microscopic superconducting anisotropy of these multilayer systems we thus adopt another approach, discussed below.

For all available samples, with the exception of that with a Co layer thickness of zero, the variation of H_{c} _n near T_c is well described by the formula for a two-dimensional superconductor¹⁴ (see Fig. 3):

$$
H_{c_2\parallel}(T) = H_{c_2\parallel}(0)(1 - T/T_c)^{1/2}.
$$
 (1)

In contrast, the sample with zero Co thickness showed a linear dependence near T_c which is typical of threedimensional superconducting behavior (Fig. 3). For a $3D$ system $H_{c_2}(T) \propto \xi^{-2}(T)$ where $\xi(T) = \xi(0)(1 - T/T_c)^{-1/2}$ is the superconducting coherence length, which leads to

$$
H_{c_2}(T) = H_{c_2}(0)(1 - T/T_c). \tag{2}
$$

Deviations from this linear Ginzburg-Landau dependence near T_c are well known from superconducting multilayers systems with non-magnetic spacer layers.^{14,15} At high temperatures the system behaves as an anisotropic 3D superconductor, which is reflected in a linear dependence of H_{c} _c_i. As the temperature falls the superconducting coherence length in a direction perpendicular to the planes ξ_{\perp} approaches the separation of the superconducting layers d ^{14,15} The system then becomes a 2D superconductor, as the stack of thin superconducting layers are coupled only by the proximity effect. The temperature dependence of H_{c} _{il} $\propto (\xi_{\perp} \xi_{\parallel})^{-1}$, which at lower temperatures depends only on the in-plane coherence length and is given by H_{c_2} \propto $[d \xi |(T)]^{-1}$, thus giving rise to the form of Eq. (1) .^{14,15}

For an anisotropic superconducting film with the orientation of the applied field perpendicular to the planes the coherence length is unrestricted. It is thus a common feature

FIG. 4. Temperature dependence of the upper critical field for both the parallel $H_{c_2\parallel}(T)$ and perpendicular $H_{c_2\perp}(T)$ orientation of the field for a sample with d_{Nb} =50 nm and d_{Co} =1.8 nm. Lines are fits to the points using Eq. (1) and Eq. (3) , respectively. Critical temperatures extracted from the fits are $T_c = 7.97(2) \text{ K } [H_{c_2 \perp}(T)],$ T_c =7.98(2) K $[H_{c} (T)]$.

that the upper critical field $H_{c_2\perp} \propto \xi_{\parallel}^{-2}(T)$ is lower than that for the parallel orientation and follows a linear dependence with temperature:

$$
H_{c_2\perp}(T) = H_{c_2\perp}(0)(1 - T/T_c),\tag{3}
$$

as in the case for a 3D superconductor [Eq. (2)]. Figure 4 shows the temperature dependence of both $H_{c2\parallel}$ and $H_{c2\perp}$ for a Nb/Co multilayer with $d_{Co} = 1.8$ nm. The experimental points are fitted with Eqs. (1) and (3) , respectively. The two values of T_c extracted from fitting agree very well within the small experimental uncertainty: $T_c = 7.97(2) \text{ K } [H_{c_2 \perp}(T)];$ T_c =7.98(2) K $[H_{c₂||}(T)]$. This demonstrates the similarity of behavior between our samples containing magnetic spacers and previous studies on nonmagnetic superconducting multilayers. Furthermore the reliability of determining T_c by this method is demonstrated. We note one difference between the present study and those concerned with nonmagnetic spacers. In the former a crossover in behavior with temperature between 3D and 2D coupling is often observed as the coherence length increases such that $\xi_{\perp} \gtrsim d$, so that for sufficiently high temperatures the dependence $H_{c_2\parallel}$ $\propto (d\xi_{\parallel})^{-1}$ crosses over to a linear dependence $H_{c_2\parallel}$ $\propto (\xi_1 \xi_0)^{-1}$ close to T_c . Such behavior has been observed in Nb/Co multilayers in a previous study, but only for *very thin* magnetic layers $d_{Co} \le 0.3$ nm.⁴ For thicker Co layers the 2D behavior is effectively observed all the way to T_c .⁴ In our samples, with the exception of the $d_{Co}=0$ nm sample, the smallest magnetic layer spacing is $d_{Co} = 1.8$ nm, so for the field oriented parallel to the magnetic layers a temperature dependence described by Eq. (1) is always applicable. We are therefore justified in extrapolating fits to our data using Eq. (1) and Eq. (3) for the two extremes of field orientation, as attested to by the agreement obtained for values of T_c . More interestingly, the suppression of true 3D superconducting behavior occurs for much thinner nonsuperconducting spacer layers than would be the case if these were nonmag-

FIG. 5. Angular dependence of the upper critical field for samples with different Co layer thickness, measured at 6.8 K. Lines are fits to the experimental points with Eq. (5) . The strong anisotropy observed in the magnetic samples reflects the 2D nature of the superconductivity due to the weaker coupling between the layers, and is most pronounced in the samples with the largest value of d_{Co} .

netic. For the case of nonmagnetic spacers the crossover to 2D behavior occurs when ξ_1 falls below some characteristic length which is of the order of *d*. In the Nb/Co samples the relevant effective length scale $d_{eff} \geq d_{Co}$ which reflects the pair breaking influence of the ferromagnetic layer. To illustrate this further we consider the case of uniaxial anisotropy such that $H_{c_2\parallel} = \phi_0/2\pi \xi_\perp \xi_\parallel$ and $H_{c_2\perp} = \phi_0/2\pi \xi_\parallel^2$. We define a quantity

$$
d_{\perp} = \left(\frac{\phi_0}{2\pi H_{c_2 \perp}}\right)^{1/2} \frac{H_{c_2 \perp}}{H_{c_2 \parallel}},
$$
(4)

which for the simplest case where both $H_{c_2\parallel}$ and $H_{c_2\perp}$ are linear in temperature (3D behavior) would correspond to $d_+(T) = \xi_+(T)$. For the case of a crossover to 2D behavior in a superconducting multilayer with a nonmagnetic spacer d_{\perp} $= d$, the layer thickness, which is temperature independent. The current measurements are performed in a similar 2D regime, with the two critical fields described by Eqs. (1) and (3) and with d_{\perp} independent of temperature, so we make the identification $d_{\perp} = d_{eff}$. Taking as an example the data on the sample with $d_{Co} = 1.8$ nm, we obtain a value $d_{eff} = d_{\perp}$ = 12 nm, so that $d_{eff} \ge d_{Co}$. The effective length scale d_{eff} which is responsible for the crossover from $3D$ to $2D$ (proximity coupled) superconductivity, which occurs when ξ_{\perp} $\langle d_{eff}$, is thus much larger than the physical thickness of the magnetic spacer layers *d* . This is not surprising, since one would expect the ferromagnetic layer to suppress the superconducting order parameter some distance into the Nb layer.

To explore further the two-dimensional nature of the multilayers we have also measured the dependence of the H_{c2} on the angle θ between the film plane and the direction of the applied field. Figure 5 shows representative curves for samples having different Co layer thicknesses. The essential feature for all the curves is that they have a cusplike peak around $\theta=0^{\circ}$ and follow the dependence typical for 2D superconductors 16,4 given by

FIG. 6. The dependence of T_c on the thickness of the magnetic spacer layers d_{Co} . The size of the points reflects the uncertainty of the measurement. The line is used as a guide to the eye.

$$
\left| \frac{H_c(\theta)\sin\theta}{H_{c\perp}} \right| + \left(\frac{H_c(\theta)\cos\theta}{H_{c\parallel}} \right)^2 = 1.
$$
 (5)

For the samples with a magnetic spacer layer the data can be very well described by the fits to Eq. (5) . Also note that for larger values of d_{Co} the anisotropy is much more pronounced. For comparison, Fig. 5 also shows the case of the sample with the zero thickness of Co. For this sample the slope of $H_{c2}(\theta)$ dependence at $\theta=0^{\circ}$ is close to zero which is an indication of the superconducting system being in the 3D regime $15-17$.

Summarizing the results from the $H_{c2}(\mathcal{T})$ and $H_{c2}(\theta)$ dependencies, all multilayers with nonzero Co are in the partially decoupled state, which we label as 2D, in which the superconducting layers are coupled via the proximity effect across the magnetic layer. The effective thickness d_{eff} of the magnetic layer is much larger than its physical thickness d_{Co} , presumably due to the pair breaking¹⁴ influence of the ferromagnetic layers. This is to be contrasted with the case of nonmagnetic spacer layers, where these two length scales are comparable. This enhanced suppression is so great that in all samples except those with extremely thin magnetic layers $(d_{Co} < 0.3$ nm), the crossover with increasing temperature to a 3D state is never in fact observed experimentally.⁴

In order to determine the dependence of T_c on thickness d_{Co} , H_{c2} _[(*T*) was measured for all samples. The experimental points were fitted to Eq. (1) from which the values of T_c and $H_{c2\parallel}(0)$ were extracted. Figure 6 shows the dependence of T_c on the thickness of the Co interlayer d_{Co} . There are two significant features of this dependence. First, the values of T_c for the samples with nonzero amount of Co are significantly reduced compared to that of pure Nb film. Secondly, following the initial rapid fall of $T_c(d_{Co})$ there is an oscillation of T_c with increasing layer thickness.

The initial overall reduction of T_c can be attributed to the pair-breaking influence of the ferromagnetic layers.⁶ Due to the proximity effect between the superconducting layers superconducting electrons diffuse into the ferromagnetic layers and are subjected to the exchange field. The partial polarization of electrons therefore inhibits the Cooper pair formation. As a result the transition temperature of the superlattice T_c is much lower than that of the bulk material. The thicker the ferromagnetic layer the lower the T_c . According to Ref. 6, the rapid fall of T_c slows when d_{Co} is of the order of ξ_M , the characteristic penetration length of Cooper pairs into the ferromagnetic layers.

There have been several theoretical models proposed to account for the periodic variation of T_c on d_M . In the model for a dirty-limit superconductor/ferromagnet multilayer, which has been suggested by Radovic et al.,⁶ the oscillation of T_c results from a change of the coupling between the superconducting layers. According to the model, the typical phase difference $\Delta \phi$ between two neighboring superconducting layers can not only be $\Delta \phi = 0$, as is the case for superconductor/nonmagnetic metal systems, but can also be $\Delta \phi = \pi$ depending on the superlattice parameters and, primarily, on the thickness of the ferromagnetic layer. The $\Delta \phi = \pi$ solution may give a higher T_c for certain ranges of d_M , such that the ground state switches rapidly from the $\Delta \phi = 0$ to the $\Delta \phi = \pi$ state with increasing d_M . This results in a nonmonatonic dependence of T_c on d_M , characterized by a sharp increase of T_c as the π -phase becomes energetically favorable. Several works on *S*/*M* multilayers have reported a single peak in $T_c(d_M)$, which have been attributed to " π -switching" (e.g., Refs. 2,4,3, and 10). However, similar observations have also been made in trilayer *M*/*S*/*M* systems, where the phase switching argument cannot be invoked.⁹ In Ref. 9 the rise of T_c in Fe/Nb/Fe multilayers was found to coincide with the crossover from paramagnetic to ferromagnetic ordering of the magnetic layer. It was shown by Arts *et al.*⁵ that this and other effects could result from considerations of the interface transparency of the layers, which in the particular cases of Refs. 9 and 10 would result from the decrease of the transparency due to the onset of static exchange splitting in the ferromagnetic layers. A general approach by Khusainov and Proshin 11 includes the transparency of the interface as a parameter, and they show that the earlier model by Radovich *et al.*⁶ is a special case in which the transmissivity of the interface is very high. It is found that the quasiparticle motion in the ferromagnetic layer has a mixed diffusionlike and *spin-wave-like* character. As the clean limit is approached the spin-wave contribution dominates and the penetration depth of the pair amplitude into the ferromagnetic layer grows larger than the oscillation period. Even without considering phase switching, a periodic variation of $T_c(d_M)$ is possible in this model, due to the periodic compensation of the exchange field due to oscillations of the pair amplitude inside the magnetic layer. A maximum in T_c occurs when the period of the Usadel function describing the order parameter matches the thickness of the ferromagnetic layer.

Comparing the data of Fig. 6 with the various theoretical models, a good qualitative agreement is found with the predictions of Khusainov and Proshin¹¹ in the limit of strong exchange and approaching high transparency of the *S*/*M* boundary. In the model of Ref. 11 this would be characterized by typical parameters $\sigma_s \approx 1$, $2I\tau_f \sim 3$, where σ_s characterizes the transparency of the interface, *I* is the exchange field and τ_f^{-1} is the frequency of scattering by nonmagnetic impurities. This model is characterized by an initial fall in $T_c(d_M)$, followed by a plateau region and then a slowly damped oscillation, as is seen in the experimental data of Fig. 6. This in contrast to the sharp and strongly damped oscillations of the Radovich model, 6 valid only in the dirty ferromagnetic metal limit $2I\tau_f \ll 1$.¹¹ It is possible that both pair amplitude oscillations and π -switching could play a role in the observed oscillation of T_c in this system, and the influence of the latter can only be ruled out by comparison of the multilayer data with that from bilayers and trilayers of the same system.

In conclusion, we have measured the temperature and angular dependence of $H_{c_2}(T)$ for Nb/Co multilayer and found the behavior to be highly consistent with an anisotropic su-

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perconducting system in the 2D limit, where the superconducting layers are coupled via the proximity effect. The effect of magnetic spacer layers has been shown to be much more effective at inducing the 2D behavior than nonmagnetic layers of similar thickness, due to the enhanced pair breaking effect of the local exchange field on the superconducting pairs. The dependence of T_c on the thickness of the magnetic layer d_{Co} is nonmonatonic and oscillatory. These results are in qualitative agreement with recent models, although the relative importance of π -phase switching and pair amplitude oscillations cannot be determined at present.

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