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Magnetic instabilities along the superconducting phase boundary of Nb/Ni multilayers

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We report vibrating reed and superconducting quantum interference device magnetometer data that exhibit prominent dips or oscillations of the superconducting (SC) onset temperature, $\Delta T_C(H) \approx 0.01-0.7$ K, for a $[\mathrm{Nb}(23\ \mathrm{nm})/\mathrm{Ni}(5\ \mathrm{nm})]_5$ multilayer (ML) in dc magnetic fields applied nearly parallel to the ML plane. The vibrating reed data exhibit reproducible structures below T_C that may reflect multiple SC transitions, but they are sensitive to ac field amplitude and dc field orientation. This striking behavior poses challenges for theoretical and experimental investigations of interfaces between SC and ferromagnetic layers that involve magnetic pair breaking effects, "pi phase shifts" of the SC order parameter, and exotic ("LOFF") pairing states. Alternatively, the anomalies may mark dynamical instabilities within a confined, strongly anisotropic Abrikosov vortex lattice.
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Multilayers (MLs) composed of alternating superconducting (SC) and ferromagnetic (FM) thin films exhibit remarkable properties related to the destabilizing effect of magnetic interactions on SC pairing.[1,2] This work was motivated by evidence of oscillations of $T_C(y)$ of order of 0.1 K for $[\mathrm{Nb}(23\ \mathrm{nm})/\mathrm{Ni}(y)]_5$ ML (Nb layer thickness $x=23$ nm and Ni layer thickness $2.5 \leqslant y \leqslant 6.0$ nm).[3,4] Similar nonmonotonic decreases of T_C with FM layer thickness have been observed in other SC/FM bilayer, trilayer, and ML systems, and attributed to a magnetic exchange coupling between FM layers that oscillates in magnitude and sign as a function of the FM layer thickness.[1,5–8] The SC proximity effect and magnetic pair breaking interactions within the Ni layers alter the magnitude and phase of the complex SC order parameter that determines the stability of the SC state of the ML, and may even induce "pi phase shifts" that can completely suppress superconductivity.[8]

A recent letter[9] has reported the resistively measured T_C of $\mathrm{Ni}(7\ \mathrm{nm})/\mathrm{Nb}(x)/\mathrm{Ni}(7\ \mathrm{nm})$ trilayers ($16\ \mathrm{nm} \leqslant x \leqslant 52\ \mathrm{nm}$) as a function of applied magnetic field that controls the relative orientation of the magnetizations of the two Ni layers. Although very small, the observed shifts ($2 \leqslant \Delta T_C \leqslant 41$ mK) between parallel and antiparallel Ni layer orientations were ten times larger than predicted by existing theories for FM/SC/FM trilayers, using measured normal state parameters.[10]

We wish to point out that interpreting such shifts $\Delta T_C \approx 10$ mK is problematic because the experimental definition of $T_C(H)$ is not precise in applied magnetic fields that couple the equilibrium of the SC state to the weak stability of the Abrikosov vortex lattice. Indeed, modest probe currents can initiate dissipative motion or nonlinear dynamics of vortices[11,12] in materials with few defects to "pin" them against Lorentz forces. Consequently, resistive determinations of $T_C(H)$ require an arbitrary "voltage criterion" that defines when dissipation due to vortex motion under the applied current drive has dropped to negligible levels.[9] The temperature interval between the initial decrease of resistance and apparent T_C defined by the voltage criterion can be substantial and very sensitive to the probe current amplitude,[13] and inferred T_C's do not necessarily reflect the equilibrium phase boundary between normal and SC states.

On the other hand, shifts of order 10 mK are known to result from confinement of supercurrents and quantized magnetic flux by mesoscopic boundaries such as thin-film cross sections[4,14] or submicron holes lithographically patterned in SC films.[11,15] In these cases, the T_C shifts reflect an equilibrium phase boundary when carefully measured at "vanishing" drive current.[15,16]

Magnetometry techniques offer alternative definitions of the SC onset, such as an initial diamagnetic change of the real part (m') or an abrupt increase in the dissipative imaginary part (m'') of the ac magnetic moment in field-cooling (FC) experiments.[11] Vibrating reed (VR) magnetometry is particularly well suited for measurements of thin-film or anisotropic samples[17] and is essentially a transverse ac susceptibility technique that employs very low ac fields generated perpendicular to the dc applied field.[18,19] The high sensitivity of the VR to the entire sample bulk (which is not necessarily the case with resistivity measurements) at relatively low ac drives has been exploited to detect subtle transitions between equilibrium vortex lattice phases in bulk FM superconductors[17] and the onsets of plastic deformation and flow of the vortex lattice.[12]

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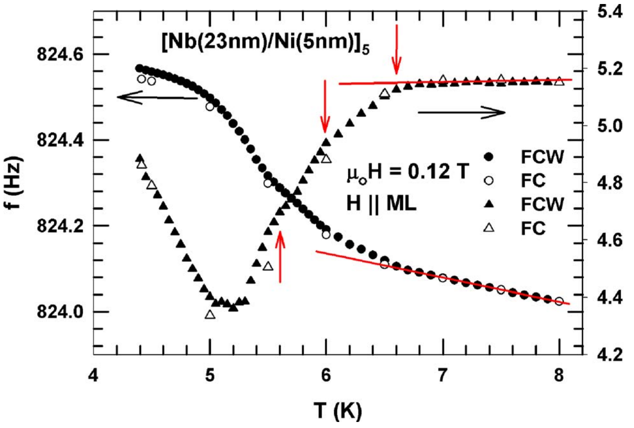

FIG. 1. (Color online) VR frequency f and amplitude A vs temperature T for a $[\mathrm{Nb}(23\ \mathrm{nm})/\mathrm{Ni}(5\ \mathrm{nm})]_5$ ML in parallel magnetic field $\mu_o H=0.12$ T with VR drive $V=100$ mV. Open symbols denote initial field-cooled (FC) data, and solid symbols denote subsequent FC-warming cycle (FCW) data. Red lines indicate the assumed normal-state base lines in $f(T)$ and $A(T)$, and vertical arrows denote the temperatures of the SC onset near 6.6 K, and slope changes apparent near 6.0 and 5.6 K.

The $[\mathrm{Nb}(23\ \mathrm{nm})/\mathrm{Ni}(5\ \mathrm{nm})]_5$ ML was fabricated by dc magnetron sputtering on a Si (100) substrate and exhibited textured growth of Nb (110) and Ni (111) layers[3,4] with only small Nb–Ni interdiffusion and interface roughness.

In order to carry out vibrating reed experiments, a sample ML was glued onto one end of a rectangular reed ($2.5\times 7\times 0.05\ \mathrm{mm}^3$, cut from a Si(100) wafer), the opposite end of which was mounted onto one side of a piezoelectric transducer to form a cantilever beam arrangement.[17] The transducer was driven by a synthesizer tuned to the fundamental cantilever mode (frequency $f=650-865$ Hz), and the reed amplitude was detected capacitively using a resonant rf (327 MHz) cavity technique.[17,20] The Si reed was oriented with its long dimension and the ML plane parallel to the dc magnetic field, which results in a transverse ac field (viewed in the moving VR frame) oriented perpendicular to the applied dc field.[18,19] Shifts of the VR frequency f (comparable to transverse m') measure a magnetic restoring torque exerted on supercurrents that screen the ac field. The inverse VR amplitude A^{-1} measures ac losses (comparable to transverse m'') generated primarily by vortex motion.[18]

In order to complement the transverse VR response perpendicular to the applied dc field, we also carried out conventional longitudinal measurements (with both dc and ac fields applied along the ML plane) using a Quantum Design MPMS5 supreconducting quantum interference device (SQUID) magnetometer operated at frequencies $0.1\leqslant f\leqslant 10$ Hz and drive amplitudes $0.01\leqslant \mu_o h_o \leqslant 0.3$ mT. The real part (m') of the ac moment measures the supercurrent (circulating perpendicular to the dc field) that screens the ac field, and the imaginary part (m'') mainly measures ac losses generated by SC vortex motion.[18]

We determined the SC onset temperatures using either the VR frequency f or amplitude A for a $[\mathrm{Nb}(23\ \mathrm{nm})/\mathrm{Ni}(5\ \mathrm{nm})]_5$ ML sample in parallel dc fields (see Fig. 1). The data define two reversible phase boundaries that coincide remarkably well, except where they exhibit large downshifts at 0.22, 0.36, and 0.62 T (and smaller shifts at 0.1 and 0.58 T), as shown in Fig. 2. Additional peaks or abrupt slope changes in $f(T)$ and $A(T)$ were also identified well below the upper SC onset (see Fig. 1) and were generally found (almost independent of dc field) near characteristic temperatures of 5.1, 5.6, and 6.0 K. These anomalies could be grouped into two other trajectories that closely paralleled the behavior of the SC onsets, as shown in Fig. 2.

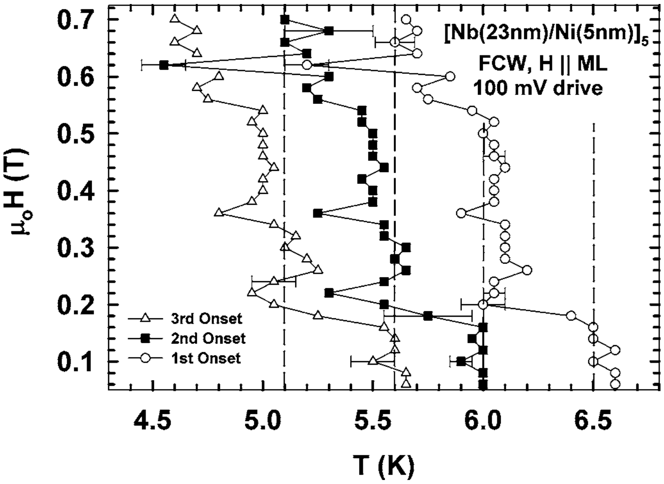

FIG. 2. Magnetic field (H)-temperature (T) "phase boundary" between the SC and normal states for a $[\mathrm{Nb}(23\ \mathrm{nm})/\mathrm{Ni}(5\ \mathrm{nm})]_5$ ML, defined by an abrupt decrease in the VR amplitude A (open circles). Data were taken in field-cooled-warming (FCW) experiments with VR drive $V=100$ mV and dc field H nearly parallel to the ML plane. The left (open triangles) and middle (solid squares) trajectories mark temperatures of two anomalies in the slope of $A(T)$ (see Fig. 1). Solid lines are guides to the eye, and error bars denote uncertainties in extracting the points from data. Dashed lines denote possible phase transition lines between SC states of the entire ML or SC transitions within individual Nb layers.

The complementary $m''(H,T)$ boundary determined from longitudinal SQUID data for field parallel to the ML plane exhibits strong oscillations (60–180 mK), and these anomalies persist to fields of at least 0.65 T with an average period $\mu_o \delta H \approx 80$ mT, as shown in Fig. 3. Additional SQUID measurements using ac field amplitudes $\mu_o h_o=0.02$, 0.05, 0.1, and 0.2 mT yield different trajectories—probably a result of

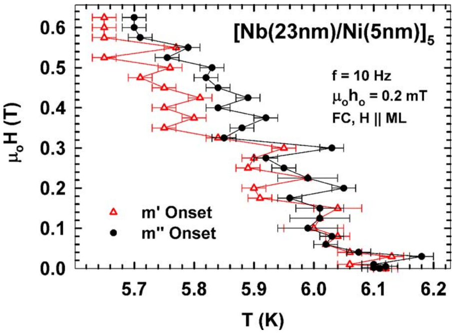

FIG. 3. (Color online) Magnetic field (H)-temperature (T) "phase boundary" between the SC and normal states for a $[\mathrm{Nb}(23\ \mathrm{nm})/\mathrm{Ni}(5\ \mathrm{nm})]_5$ ML, determined from SQUID magnetometer experiments at ac frequency $f=10$ Hz and amplitude $\mu_o h_o=0.2$ mT, with applied dc field H parallel to the ML plane. Solid symbols denote the SC onset temperature indicated by an abrupt increase in the imaginary part m'', and open symbols by an abrupt diamagnetic shift in the real part m', of the ac magnetic moment during FC. The divergence of the m' and m'' data near $\mu_o H=0.2$ T reflects the onset of vortex depinning with increasing dc field.

vortex depinning and nonequilibrium disorder of the vortex lattice. Nevertheless, these data consistently exhibit oscillations with the same average spacing and magnitudes shown in Fig. 3.

Small oscillations (of order of 10–30 mK and rough period of 70 mT) were previously observed in lower-precision SQUID measurements[4] on Nb/Ni ML. These variations were interpreted as "matching anomalies" at applied fields where the vortex density is equal to an integral multiple of the average density of strong vortex pinning centers.[4,15] Matching anomalies are most prominent if pinning centers have a characteristic size $\xi(T) < D < \lambda(T)$ and are located on a periodic lattice.[21] The SC/FM ML is essentially a one-dimensional periodic array of thin SC slabs spaced by Ni layers that strongly confine vortices when their planar cross sections are oriented perpendicular to an applied dc magnetic field **H**. A related matching effect has been predicted[22] and observed in ML (Ref. 23) when N_L vortices enter in successive chains aligned parallel to the ML and **H** (an average oscillation period of 70 mT implies[4] $N_L \approx 2 \times 10^3$). Moreover, calculations[22,24,25] predict that a succession of vortex lattices will form within a thin SC slab in an increasing parallel field, and these phases will have different packing topologies whose ranges of stability may not be strictly periodic in applied field.

However, matching analyses of parallel dc field data (Fig. 3) do not clearly discriminate between cases for which there is either a nonzero SC order parameter within the Ni layers or isolated SC Nb layers (having distinct T_C's) separated by Ni layers having zero SC order parameter.[4]

Additional VR data were acquired at piezoelectric drives of 5, 10, 26, and 50 mV at selected dc fields of 0.08, 0.10, 0.36, and 0.62 T, at which thermal hysteresis was observed. The SC onsets exhibited extraordinary downshifts of 0.5–1.0 K with increasing VR drive, indicating weak vortex pinning and nonequilibrium vortex lattice disorder. The line of onsets near 6.5 K was found to extend to at least 0.62 T for drives of 5–26 mV, but disappeared at $\mu_o H \approx 0.2$ T for drives $V \geqslant 50$ mV, whereas a line of anomalies near 6.0 K was found to terminate at 0.62 T for $V \geqslant 26$ mV (see Fig. 2). The downshift of the SC onset near 0.2 T (0.62 T) therefore marks a hysteretic jump from the 6.5 to 6.0 K (6.0–5.5 K) "transition line," possibly due to an increase in SC currents that drive the vortex lattice and have the potential (for critical current densities $J_C \approx 10^7$ A/cm^2) to switch the magnetization direction of Ni domains.

Both VR and ac SQUID magnetometry sensitively probe the pinning and dynamics of Abrikosov vortices,[18] and the anomalies of Figs. 1–3 are clearly affected by vortex dynamics, but since the local vortex pinning force is proportional to the spatial gradient of the squared sc order parameter, the anomalies nevertheless reflect unusually strong variations in the stability of the sc state. Since both the real and imaginary parts of the VR signal behave similarly, the anomalies of Fig. 2 may well reflect SC phase transition lines for the entire ML or for individual Nb layers. The possibility remains open that they are caused by changes in the Ni-layer magnetizations that alter the phase and coupling between the SC order parameter of adjacent Nb layers with applied dc field.

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