

Spin-like susceptibility of metallic and insulating thin films at low temperature - Supporting material

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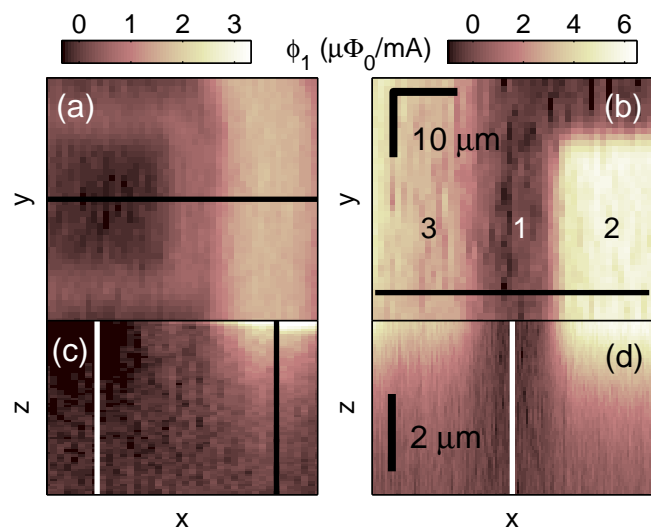
This supplementary document discusses the height dependence of the susceptibility signal over various sample regions. The results provide an estimate of the spin response from the bare sample substrate with or without thermally grown SiO_x and confirm that the strongest response originates from the deposited, patterned structures. Furthermore, a verification that the observed signal is not an instrumental artifact using superconducting rings is presented.

HEIGHT DEPENDENCE AND SUBSTRATE RESPONSE

Because the sensor background is much larger than the sample response, the lateral scans discussed in the Letter can only detect *variations* of the sample response. Information about the absolute value can be obtained from the height dependence of Φ_{SQUID} , shown in Supplementary Figs. 1 and 2 for ϕ_1 . ϕ_2 and nonlinear components are too small to measure their height dependence with sufficient accuracy, but it is most plausible that correlated lateral variations in the different components have the same origin. Far from the sample, ϕ_1 exhibits a linear variation with height of unknown but presumably instrumental origin, which was subtracted from the data shown. Upon approaching a metal film, we find a deviation from this linear trend, which can be attributed to the sample response and whose magnitude is approximately equal to the signal in the lateral scans.

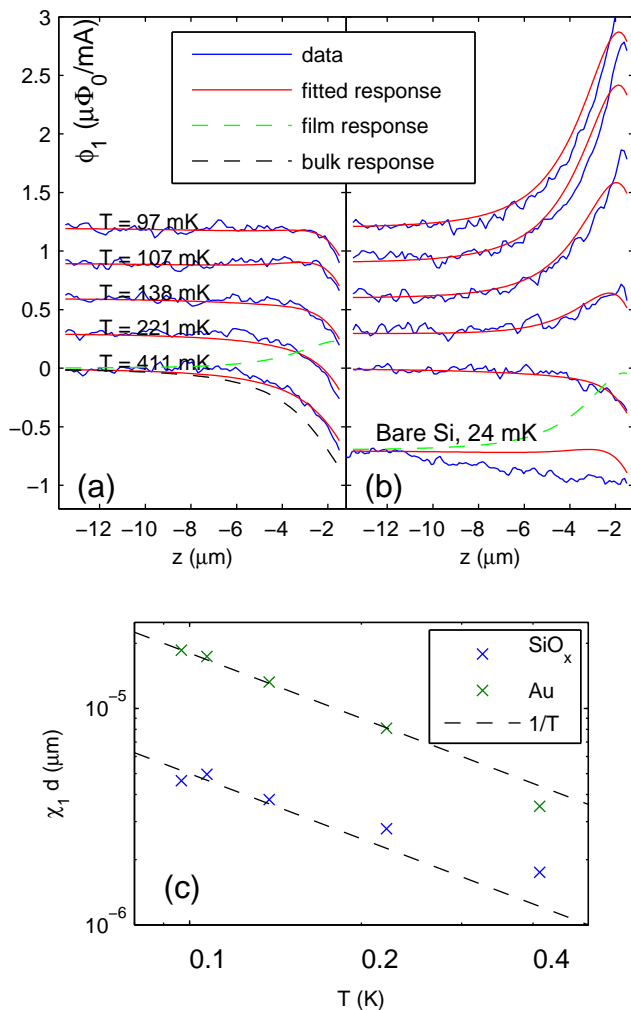
To quantitatively analyze the height dependence, both the bulk susceptibility of the Si substrate and the surface contributions have to be considered. By matching the boundary conditions for the magnetic field at the interface between vacuum and a material with susceptibility $\chi \ll 1$, one can show that the response field \mathbf{B} of the sample to a field \mathbf{H} applied from the vacuum side is $\mathbf{B} = \mu_0 \chi \tilde{\mathbf{H}}/2$, where $\tilde{\mathbf{H}}$ is defined as in the Letter. The film response given in the Letter can then be obtained by considering two nearby interfaces with an equal but opposite discontinuity in χ . We modeled the field coil as a thin loop in order to compute \mathbf{H} , and used the measured pickup-loop-field-coil inductance, which reflects the direct response of the sensor to \mathbf{H} , in order to convert the measured flux into the z -component of \mathbf{B} .

The fits shown in Supplementary Fig. 2 were obtained as follows. First, the linear trend was fitted for large heights, where the sample response is negligible, and subtracted from the data. Supplementary Figs. 1 and 2 show the results of this subtraction. We then fitted a linear combination of the the models for the bulk and surface response to the 410 mK dataset over Si with thermal SiO_x in Supplementary Fig. 2(a), using only the two prefac-



Supplementary Fig. 1: Comparison of the lateral and height dependence of the linear in-phase response over different parts of the samples. (a) Susceptibility scan at $T = 200$ mK of an area analogous to the right half of Fig. 1(f), (g) from a sample similar to sample I, however with a thermally grown oxide on the substrate. (b) Susceptibility scan at $T = 24$ mK of an area of sample II as shown in the top portion of Fig. 1(c)-(e), but rotated clockwise by 90° . The numbers indicated the nature of each region, as explained in the caption of Fig. 1. (c), (d) Dependence of the signal on height and lateral position along the black line in panels (a), (b) at $T = 100$ mK and 24 mK, respectively. The vertical lines indicate the positions at which the line cuts shown in Supplementary Fig. 2 were taken. A fitted linear background with a slope of about $0.25 \mu\Phi_0/(\text{mA } \mu\text{m})$ along the z -direction was subtracted from the raw data.

tors as fit parameters. The two dashed lines show the two individual contributions to this fit. The value of the bulk coefficient corresponds to a diamagnetic susceptibility of 2.3×10^{-6} . The deviation from the literature value of 3.2×10^{-6} is reasonable, given the approximations and uncertainties of our analysis. In particular, there may be an offset in the height between in-plane and height scans due to the effect of scanner hysteresis in the latter, and the fit is poorly conditioned because the bulk and film



Supplementary Fig. 2: Temperature dependence of the surface contribution. (a) Line cuts along the white line in Supplementary Fig. 1(c) over thermally oxidized Si at a range of temperatures, and fits to the response (see text). (b) Corresponding line cuts along the black line over a 100 nm thick Au wire. The lowest curve is a line cut along the white line in Supplementary Fig. 1(d) over bare Si with only a native oxide on sample II. The height above the sample surface is given by $-z$. The linear background subtracted from the data was fitted for $-13.5 \mu\text{m} < z < -7.5 \mu\text{m}$. The offset of each curve is arbitrary and has been adjusted for clarity.

contributions have a similar overall shape.

For all other curves from this sample (lower temperatures and over the Au wire), we kept the bulk coefficient fixed at the value obtained from the 410 mK fit and only varied the surface contribution. The temperature dependence of its coefficient is shown in Supplementary Fig. 2(c). The dashed line approximating the Au

data points corresponds to $\chi T = 1.8 \times 10^{-5}$ K (assuming a film thickness $d = 100$ nm), in reasonable agreement with the values extracted from the lateral scans of other samples discussed in the Letter. The line through the SiO_x data points is a factor 3.6 lower. Its value could be explained with a surface density of 6×10^{16} spins/ m^2 , or a volume spin density of 6×10^{22} spins/ m^3 in the 1 μm thick oxide layer, which was grown at 1000°C with a wet process.

Over bare Si with only a native oxide [bottom curve in Supplementary Fig. 2(b)], no variation is apparent near the sample surface. The dashed green line on top of this curve corresponds to the surface response with $\chi_1 d = 5 \times 10^{-6} \mu\text{m}$, which results in the red curve when adding the same bulk response as used in the other fits. At $T = 24$ mK, this value corresponds to an area density of 1.5×10^{16} spins/ m^2 , more than an order of magnitude smaller than the values typically found for our deposited films. We estimate the uncertainty of this value to be a factor 2 to 3. In any case, this small value confirms that it is reasonable to define the bare substrate signal as zero when analyzing the lateral variation of the response, as done in the Letter.

CHARACTERIZATION OF INSTRUMENTAL PHASE SHIFT

To check for measurement artifacts in ϕ_2 , we took the same measurement as on the Au rings on several superconducting Al rings, whose Meissner response can be assumed to be instantaneous. For a predominantly linear response, the phase lag in the combined sample and instrumental response, determined by ϕ_2/ϕ_1 , can be expected to be independent of the sample volume, magnitude of the susceptibility, or the amplitude of H_a . As seen in Fig. 2(c), ϕ_2/ϕ_1 is larger for the Au structures than for the superconducting ring, which shows that ϕ_2 is not due to an instrumental phase shift alone. Nevertheless, the finite value for the superconducting ring indicates that the contribution of the other samples to ϕ_2 is somewhat smaller than the raw values displayed in Fig. 2(b). The instrumental part of ϕ_2 is slightly different for sample II (not shown), but also clearly smaller than the total value.

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