# Suppression of ferromagnetism due to superconducting proximity effects

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**Abstract**

Superconducting proximity effects in a ferromagnetic metal are directly observed by means of $^{59}$Co NMR in Co/Al bilayers. Below the superconducting transition temperature, the resonant frequency decreases and the nuclear spin-lattice relaxation rate divided by temperature, $1/T_1T$, increases. These results indicate the reduction of magnetic moment in the Co layer. We discuss the spatial variation of the local magnetic moment and its temperature dependence by analyzing the resonance line shapes.

*Key words:* superconductivity;proximity;ferromagnetism;NMR

The superconducting proximity effects in a ferromagnetic metal are difficult to detect, because the penetration depth, $\xi_F$, of the Cooper pairs into the ferromagnet is limited to several nm for iron-group metals. To clarify the penetration of superconductivity, the ferromagnetic layer thickness dependence of the superconducting transition temperature, $T_c(d_F)$, has been extensively investigated, but the results are not conclusive [1],[2]. In this paper we report the direct and local observation of the superconducting proximity effects in a ferromagnetic metal by means of NMR at the ferromagnetic sites.

The sample is Co(20Å)/Al(500Å) bilayers fabricated by the vacuum deposition. On Mylar film SiO, Co, Al and SiO were successively deposited from electrically heated crucibles made of alumina-coated tungsten filament. The pressure during the deposition was kept below $2 \times 10^{-6}$ Torr for Co and below $1 \times 10^{-6}$Torr for other materials. The film was cut and piled with varnish and set into the sample coil so that rf field was parallel to the film. The characterization of the sample by the atomic force microscopy showed the interface and the crystal structure were rather disordered. Nevertheless, we expect that these small imperfections are not problematic on our purpose to identify the existence of the superconducting proximity effects. The superconducting coherence length in the Al is $\xi_S \sim \sqrt{\xi_{BCS}\ell} = 400$Å, and the superconducting penetration depth in the Co is $\xi_F \sim \sqrt{\hbar D/k_B T_{Curie}} = 12$Å, where $\ell$ is a mean free path and $D$ is a diffusion constant determined by the transport measurements. Since these lengths are comparable to the thicknesses of the Al and Co layers, respectively, we expect that the proximity effect clearly appears. We measured $^{59}$Co-NMR in a zero magnetic field, and hence the NMR signal is attributed to the nuclei located in magnetic domain walls of the Co layer because of the signal enhancement effect [3]. By means of a conventional spin echo method, we measured the resonance spectrum, $I(f)$, and the nuclear spin-lattice relaxation rate, $1/T_1$, at the peak frequency of $I(f)$. The relaxation is nonexponential because it is caused by the wall excitation [4]. We define $1/T_1$ as the fastest component of the relaxation.

The temperature dependence of the peak frequency, $f_0$, and of the nuclear spin-lattice relaxation rate divided by temperature, $1/T_1T$, is shown in Fig. 1. For a Co single layer these quantities should be constant at low temperatures [3],[4], whereas for the Co/Al bilayer $f_0$ decreased and $1/T_1T$ increased below $T_c$. These results suggest that the magnetic moments in

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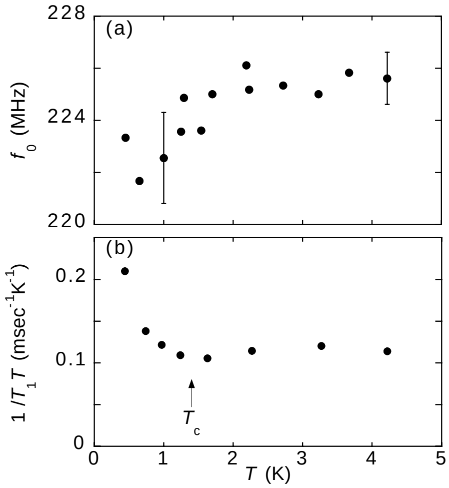


Fig. 1. Temperature dependence of peak frequency (a) and nuclear spin-lattice relaxation rate divided by temperature (b). The superconducting transition temperature, $T_c$, is shown by the arrow.

the Co layer reduce across the superconducting transition. The peak frequency decreases in keeping with the moments because the resonance frequency is determined by the mean hyperfine field, which is linear with the magnetic moment. On the other hand, $1/T_1T$ increases with the decrease in the moments, since the excitation of spin waves in the domain walls is facilitated [4]. The reduction in the magnetic moments can be explained by the interaction between the superconducting property of the conduction electrons and the magnetic property of $3d$ electrons. We assume that the free energy in the Co layer consists of the G-L model with the superconducting wavefunction $\psi(x)$, the Stoner model with the local magnetization $M(x)$, and their lowest coupling energy $\frac{1}{2}\eta|\psi(x)|^2M(x)^2$, where $x$ is the distance from the interface. The last term phenomenologically means the suppression of penetration of $\psi(x)$ in the presence of $M(x)$. As the result, the equilibrium magnetization to minimize the free energy is given by $M(x) = \sqrt{M_0^2 - \eta|\psi(x)|^2}$, where $M_0$ is the saturation magnetization for bulk Co.

To confirm the above discussion, we inspect the resonance line shapes. Since the resonance frequency is proportional to the local magnetization, $I(f)$ shows the distribution of magnetization in such an inhomogeneous system as our sample. Supposing that $M(x)$ monotonically increase with $x$, we can translate $I(f)$ to $M(x)$ as follows. Since the area of resonance spectrum, $I(f)df$, is proportional to the amount of Co nuclei located between $x$ and $x + dx$; *i.e.* , $dx \propto I(f)df$, we can relate the frequency to the nuclear position by $x/d_{\mathrm{Co}} = \int_0^f I(f')df' / \int_0^\infty I(f')df'$, where $d_{\mathrm{Co}}(= 20\text{Å})$ is the Co layer thickness. The result is shown in Fig. 2. The reduction in $M(x)$ near the interface is shown even

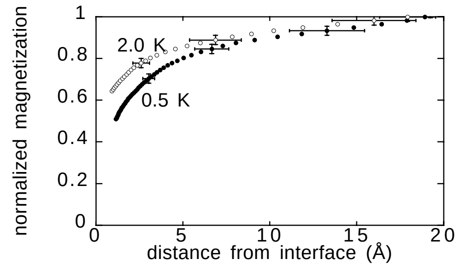


Fig. 2. Spatial dependence of local magnetization normalized to bulk value at 2.0 K (above $T_c$, open symbols) and 0.5 K (below $T_c$, closed symbols). Error bars results from the distribution of magnetization in domain walls, which is estimated from the width of resonance spectrum for the Co single layer.

in a normal-conducting state. This is attributed to the defects at the interface. Nevertheless we see further decrease in the moments all over the sample in the superconducting state. This result proves that $M(x)$ decreases with the penetration of superconductivity according to the above equation.

In conclusion, we profiled the spatial dependence of magnetization in the atomic scale and clarified that the ferromagnetism is suppressed by the superconducting proximity. This effect has been neglected so far, because the energy scale of the ferromagnetic order, $T_{\mathrm{Curie}}$, is about $10^3$ times larger than that of the superconducting order, $T_c$. To explain our results we suggest two possible causes. Since the regions of the small magnetization have low $T_{\mathrm{Curie}}$ effectively, they will be affected by the superconducting proximity significantly. For example, the magnetization is small near the interface with the non-magnetic layer or in the domain walls where the moments are inhomogeneously aligned. In addition, $3d$ electrons in the iron-group metals are not fully localized. It is possible that the itinerant properties of $3d$ electrons are altered by the superconducting proximity, and thereby the magnetization reduces.

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