Controlling exchange coupling strength in Ni\textsubscript{x}Cu\textsubscript{100−x} thin films

B. Nagy · Yu. N. Khaydukov · L. F. Kiss · Sz. Sajti · D. G. Merkel · F. Tanczikó · A. S. Vasenko · R. O. Tsaregorodtsev · A. Rühm · T. Keller · L. Bottyán

Received: date / Accepted: date

Abstract Thickness (d\textsubscript{F}) and concentration (x) dependence of the Curie temperature of Ni\textsubscript{x}Cu\textsubscript{100−x} ferromagnetic (F) alloy layers (x = 55, 65, d\textsubscript{F} = [3 nm ÷ 12 nm]) being in contact with a vanadium layer was studied. The Curie temperature of the ferromagnetic layers depends on the thickness when it is comparable with the interface layer between the F and the vanadium layers, which is attributed to the proximity coupling of the interface region with the rest of the F layer. The present study provides valuable information for fabrication of samples with controlled exchange coupling strength for studies of superconductor/ferromagnet (S/F) proximity effects.

Keywords Curie temperature · NiCu alloys · Proximity effects · magnetic interface · magnetic thin films

PACS 75.70.-i · 75.30.Kz · 75.50.Cc · 74.70.Ad

1 Introduction

Proximity effects occurring at the interface of superconducting and ferromagnetic phases have attracted considerable attention in recent years [1]. Extensive theoretical work has been exerted to understand the nature of these mixed states. For weak ferromagnets, with Curie temperatures below T\textsubscript{C} ∼ 100 K a transition of the homogeneous F layer into a non-homogeneous domain-like (“cryptoferromagnetic”) phase is predicted [2], [3], [4] with domain sizes less than the coherence length of the superconductor. Conventional 3d ferromagnets, like Fe, Co and Ni, exhibit Curie temperatures above 300 K. In order to study S/F proximity effects with a weak ferromagnet, a control of the Curie temperature of the F layer is required. In this work we use the dilution of a conventional ferromagnetic material with non-magnetic atoms, namely nickel with copper. Ni and Cu are fully miscible in the bulk [5] with almost linear dependence of the Curie temperature (and, hence, the exchange coupling strength) on the Ni concentration [6].

In addition to the concentration dependence of the Curie temperature, the dependence on the thickness of thin F layer was reported in several works, e.g. T\textsubscript{C}(d\textsubscript{Gd}) for Nb/Gd structures [7]. A linear dependence on d\textsubscript{Gd} was observed for small d\textsubscript{Gd}-s, then it is saturated for larger Gd thicknesses, a phenomenon, which was ascribed to the finite-size effects [8]. In [9] the T\textsubscript{C}(d\textsubscript{F}) was studied in thin Ni and Ni\textsubscript{60}Cu\textsubscript{40} layers being in contact with a Nb layer, and only a linear thickness dependence was observed.

Here we study the magnetic properties of Ni\textsubscript{x}Cu\textsubscript{100−x} films (x = 55 and 65 and d\textsubscript{F} = [3 nm ÷ 12 nm]) being in contact with a 80 nm thick vanadium layer. These systems were prepared to reveal proximity effects between the superconducting vanadium and the ferromagnetic Ni\textsubscript{x}Cu\textsubscript{100−x} and study their dependence on the exchange coupling strength of the F layer by means of polarized neutron reflectometry.
To enhance the contribution of the V/Ni$_{x}$Cu$_{100-x}$ interface to the scattered neutron intensity the V layer was covered with a copper layer to form a neutron waveguide structure [10]. We found the Curie temperature dependence on the F layer thickness, similarly to [7], a linear dependence at small enough thicknesses and a saturation at larger thicknesses. This behavior is ascribed to a proximity coupling of the V/Ni$_{x}$Cu$_{100-x}$ interface transition region (“dead layer”) with the rest of the F layer. Although the transition layer is non-magnetic, proximity to the main (non-mixed) part of the ferromagnetic layer increasingly suppresses the Curie temperature with decreasing the F layer thickness.

2 Sample preparation and experiment

Samples with a nominal composition of Cu(31 nm)/V(80 nm)/Ni$_{x}$Cu$_{100-x}$/MgO were prepared using Molecular Beam Epitaxy in the Wigner Research Centre for Physics (Budapest, Hungary). The MgO substrate surface was cleaned by rinsing it in the isopropanol in an ultrasonic cleaner and by heating it to 600°C in ultrahigh vacuum for 30 minutes. The metallic layers were deposited at room temperature in a base pressure of $5 \times 10^{-11}$ mbar. All samples were rotated during deposition to increase lateral homogeneity of the films. Mixing of Ni and Cu was achieved by co-evaporation with deposition rates $v_{Ni} = 0.11\text{Å/s}$ and $v_{Cu} = 0.1\text{Å/s}$ for $x = 55$ and $v_{Ni} = 0.17\text{Å/s}$, $v_{Cu} = 0.1\text{Å/s}$ for $x = 65$.

The layer structure of the samples was characterized by neutron reflectometry at angle-dispersive neutron reflectometers GINA (Budapest Neutron Center, Hungary) and NREX (research reactor FRM-II, Garching, Germany). The measurements at the GINA reflectometer were conducted in $H = 500\text{ Oe}$ external field at room temperature in a momentum transfer range of $Q = [0.08 \div 0.95]\text{ nm}^{-1}$. At room temperature the spin-up and spin-down reflectivities were identical and the sum of reflectograms in the two polarizations was fitted. A typical neutron reflectivity curve measured on the sample Cu(31 nm)/V(80 nm)/Ni$_{35}$Cu$_{65}$(8.5 nm)/MgO is shown in Fig. 1a.

The reflectivity curve is characterized by Kiessig oscillations due to the interference on different interfaces. Fit of the data by the program FitSuite [11] allowed to restore the nuclear scattering length density (SLD) profiles (Fig. 1b). From these profiles the actual thicknesses of F layers were derived (Table 1). In addition, the analysis of neutron reflectometry data suggests the existence of non-abrupt interface between vanadium and F layers, with a typical thickness between 2.5-2 nm (see inset in the Fig. 1b).

A set of low-temperature PNR measurements have been performed on the Cu(31 nm)/V(80 nm)/Ni$_{35}$Cu$_{65}$(6 nm)/MgO sample at the NREX neutron reflectometer. Magnetic field $H = 200\text{ Oe}$ was applied parallel to the surface. Spin-up and spin-down reflectivities $R^+ (Q)$ and $R^- (Q)$ were measured in a momentum transfer range of $Q = [0.17 \div 0.5] \text{ nm}^{-1}$ (Fig. 2b). Those reflectivities could also be well described using the SLD profiles obtained from the fitting of the room temperature data.

In order to quantify the magnetic moment of the F layer, the spin asymmetry [the normalized difference of the spin-up and spin-down reflectivities $(R^+ (Q) - R^- (Q))/(R^+ (Q) + R^- (Q))$], Fig. 2b] was modeled. The full line in Fig. 2b corresponds to the above-described SLD profile and a $4M = 1.4 \pm 0.2 \text{ kGs}$ magnetization in the Ni$_{35}$Cu$_{65}$ layer, which is slightly higher than the 1.1 kGs measured by Rusanov et al. [12] for a similar Ni concentration.

The Curie temperature of the samples was determined from data collected by the (Quantum Design) SQUID magnetometer at the Wigner Research Center, Budapest. Following a cooling down in zero external magnetic field, the temperature dependence of the magnetic moment of the samples was measured in 10 Oe magnetic field applied parallel with the sample surface (Fig. 3). The Curie temperature was defined as the temperature of the minimum in the derivative of the magnetic moment curve (Fig. 3) and Table 1.

| Sample        | Fitted F-layer thickness [nm] | Curie temperature [K] |
|---------------|-----------------------------|-----------------------|
| Ni$_{35}$Cu$_{65}$ 3 nm | 4.5                      | 86                    |
| Ni$_{35}$Cu$_{65}$ 6 nm | 7.0                      | 215                   |
| Ni$_{35}$Cu$_{65}$ 8.5 nm | 8.5                      | 280                   |
| Ni$_{35}$Cu$_{65}$ 12 nm | 12.7                     | 280                   |
| Ni$_{35}$Cu$_{45}$ 8.5 nm | 6.0                      | 130                   |
| Ni$_{35}$Cu$_{45}$ 12 nm | -                        | 160                   |

3 Results and discussion

Dependence of the Curie temperature on the Ni concentration for samples with $d_F = 8.5$ nm is shown in Fig. 4 along with data from [6]. This dependence is linear and agrees with the behavior of bulk Ni$_x$Cu$_{100-x}$ alloys for $x > 55$ [6]. The linear fit to the present data provides $T_C = 0 \text{ K}$ for a Ni concentration $x = 40 \pm 5\%$. This allows us to conclude that samples with $d_F > 8.5$ nm behave like the bulk alloy. However, we observed a dependence of the Curie temperature on the F layer thickness (Fig. 5), similar to the one reported in [7] for Nb/Gd structures. Namely, the Curie temperature increases with increasing the F layer thickness for samples with $d_F < 8.5$ nm and then approaches a saturation corresponding to the bulk value. While in [7] this behavior was attributed to the manifestation of the finite-size effects, we ascribe the thickness dependence of the Curie temperature to the influence of the interface of vanadium and Ni$_{35}$Cu$_{65}$.
Fig. 1 Experimental (dots) and model (solid line) reflectivity curves measured on sample Cu(31nm)/V(80nm)/Ni_{35}Cu_{65}(8.5nm)/MgO. (a). Extracted depth-profile of nuclear scattering length density of entire structure, (b). Inset: SLD profile around the V/Ni_{100-x} interface can be characterized by the presence of a transition layer with a thickness of the order of 4 nm.

Fig. 2 $R^+$ reflectivity from the sample Cu(31nm)/V(80nm)/Ni_{35}Cu_{65}(6nm)/MgO measured at $T = 10$K in $H = 200$ Oe external field, (a). The solid line corresponds to the model explained in the text. Experimental (open circles) and model (full line) spin asymmetries $(R^+ - R^-)/(R^+ + R^-)$ corresponding to the nuclear SLD profile and magnetization $4M = 1.4$ kGs in the Ni_{35}Cu_{65}(6nm) layer.

Ni_{100-x}Cu layers. Indeed, neutron reflectometry gives estimation for the thickness of the transition (T) layer between vanadium and Ni_{100-x} of the order of $d_T \sim 2-5$ nm. This transition layer, due to its vanadium content is mostly non-magnetic, but proximity to the main part of the F layer (M) may lead to an induced magnetization in it. In other words we model the F layer as a proximity-coupled T/M bilayer.

Following Bergeret et al. [13] we can write the expression for the effective exchange field $H_{\text{eff}}$ of the T/M bilayer as,

$$H_{\text{eff}} = \frac{H\nu_M d_M}{\nu_M d_M + \nu_T d_T},$$  \hspace{1cm} (1) 

where $H$ is the exchange field of the bulk Ni_{100-x}, $\nu_M, T$ and $d_M, T$ are the densities of states (DOS) and thicknesses of the main part of the F layer and the transition layer, respectively ($d_T \equiv d_M + d_T$). Eq. (1) is valid provided the thicknesses of M and T layers are smaller than the spin diffusion length, i.e. the spin is conserved in the samples. Postulating proportionality between exchange field and the Curie temperature we can rewrite (1) as,

$$\frac{T_C(d_T)}{T_{C\text{bulk}}} = \frac{d_M}{d_M + c d_T},$$  \hspace{1cm} (2) 

where $T_{C\text{bulk}}$ is the bulk Curie temperature for the given concentration [5] and $c = \nu_T/\nu_M$ is the ratio of DOS of the T and M layers.

Eq. (2) can qualitatively explain the thickness dependence of the Curie temperature. Indeed, when the thickness of the transition layer is comparable with the total thickness of the F layer the dependence $T_C(d_T)$ is almost linear. When the thickness of the F layer is much larger than the thickness of the transition layer, the Curie temperature of the layer is close to the bulk value. Fit of the data to the expression (2)
Fig. 3 Temperature dependence of the derivative of the magnetic moment of the samples measured by SQUID. Curie temperatures identified as the temperature of the minimum of the derivative magnetic moment are listed in Table 1.

Fig. 4 Ni-concentration dependence of the Curie temperature for Ni$_x$Cu$_{100-x}$ layers with F-layer thickness of $d_F = 8.5$ nm (open squares) along with literature data from Bakonyi et al. [6].

provides $d_T = 4$ nm, a value comparable with the thickness of the transition layer estimated from the neutron data, and $c = 0.1$. Similar values of the non-magnetic transition layer (or the “dead layer” as it is often called in the literature) for Ni$_x$Cu$_{100-x}$ alloys were reported in other experiments like measurements of oscillations of the critical current in S/F/S Josephson junctions [14] or direct measurements of the DOS [15]. Since the transition layer is non-magnetic, it
does not play a role in the “oscillating” superconductivity [16]. The presence of a transition layer of the same order of 1 nm was similarly reported for Nb/PdNi structures [17]. The relatively small value $c = 0.1$ indicates that the transition layer exhibits a much smaller DOS than the one in the ferromagnetic layer. Such a large decrease in the DOS indicate a change in the character of the interface region from a majority $d$-like to a majority $s$-like, i.e. to a non-magnetic one.

Moreover, interface mixing considerably increases the resistance of the V/Ni$_x$Cu$_{100-x}$ interface, similar to what has been reported for Nb/Ni$_{60}$Cu$_{40}$ structures [9]. Indeed, both V and Nb represent strong scattering centers in both Cu and Ni, resulting in specific residual resistivities 5 to 6 times higher than does Cu in Ni or Ni in Cu [18]. The interface resistance and its relation to the interface transparency in S/weak-F hybrid structures was elaborated in [19], [20].

4 Conclusions

In conclusion, thickness and concentration dependencies of the Curie temperature of Ni$_x$Cu$_{100-x}$ ferromagnetic layer being in contact with a vanadium layer was studied. The Curie temperature was found to increase with the thickness of the F layer when the thickness is comparable to that of the mixed V/F interface region and it is saturated at larger thicknesses to the bulk value. The dependence is explained by the proximity coupling of the mixed transition layer with the rest of the F layer. The present study provides valuable information for fabrication of samples with controlled exchange coupling strength for studies of the superconductor/ferromagnet proximity effects.

Acknowledgements The authors are grateful to Mr. G. Kertész for his help in the sample preparation and to Drs. J. Major, I. Bakonyi and A. A. Golubov for the fruitful discussions. Financial support by the National Office of Innovation of Hungary under contract NAP-VENEUS, and by the European Commission under the 7th Framework Programme through the ‘Research Infrastructures’ action, NMI3 are gratefully acknowledged.

References

1. A. I. Buzdin: Rev. Mod. Phys. 77, 935 (2005)
2. P. W. Anderson and H. Suhl: Phys.Rev. 116, 898 (1959)
3. A. I. Buzdin and L. N. Bulaevskii: Sov. Phys. JETP 67, 576 (1988)
4. F. S. Bergeret, K. B. Efetov, and A. I. Larkin: Phys. Rev. B 62, 11872 (2000)
5. D. J. Chakrabarty, D. E. Laughlin, S. W. Chen and Y. A. Chang: Phase Diagrams of Binary Nickel Alloys. P. Nash ed., ASM International Materials Park OH, 85-95 (1991)
6. I. Bakonyi, E. Toth-Kadar, J. Toth, T. Becsei, T. Tarnoczi, P. Kamasa: J. Phys. - Condens. Matter 11, 963 (1999)
7. J. S. Jang, D. Davidovic, D. H. Reich, and C. L. Chien: Phys. Rev. Lett. 74, 314 (1995)
8. M. E. Fisher and M. N. Barber: Phys. Rev. Lett. 28, 1516 (1972)
9. J. Kim, J. H. Kwon, K. Char, H. Doh, H.-Y. Choi: Phys. Rev. B 72, 014518 (2005)
10. Yu. N. Khaydukov, Yu. V. Nikitin, L. Bottyan, A. Rühm, V.L. Akseenov: Cryst.Rep. 55,1235 (2010)
11. http://www.ft.kfki.hu/
12. A. Rusanov, R. Boogaard, M. Hesselberth, H. Sellier, J. Aarts: Physica C 369, 300 (2002)
13. F. S. Bergeret, A. F. Volkov, and K. B. Efetov: Phys. Rev. Lett. 86, 3140 (2001)
14. V. A. Oboznov, V. V. Bol’ginov, A. K. Feofanov, V. V. Ryazanov, and A. I. Buzdin: Phys. Rev. Lett. 96, 197003 (2006)
15. L. Cretinon, A. K. Gupta, H. Sellier, F. Lefloch, M. Fauré, A. Buzdin, and H. Courtois: Phys. Rev. B 72, 024511 (2005)
16. Th. Mühle, N. N. Garif’yanov, Yu. V. Goryunov, G. G. Khaliullin, L. R. Tagirov, K. Westerholt, I. A. Garifullin, and H. Zabel: Phys. Rev. Lett. 77, 1857 (1996)
17. T. Kontos, M. Aprili, J. Lesueur, and X. Grison: Phys. Rev. Lett. 86, 304 (2001)
18. Landolt-Börnstein New Series Group III Vol.15a p. 166, Springer-Verlag Berlin, Heidelberg, New York (1982)
19. A. Alija, D. Pérez de Lara, E. M. Gonzalez, G. N. Kakazei, J. B. Sousa, J. P. Araujo, A. Hierro-Rodriguez, J. I. Martin, J. M. Alameda, M. Vélez, and J. L. Vicent: Phys. Rev. B 82, 184529 (2010)
20. D. Mancusi, E. A. Ilyina, V. N. Kushnir, S. L. Prischepe, C. Cirillo, and C. Attanasio: J. Appl. Phys. 110, 113904 (2011)