J. Phys. D: Appl. Phys. 35 (2002) 2398-2402

Nanostructures and the proximity effect

M I Montero¹, Kai Liu², O M Stoll¹, A Hoffmann³, J J Åkerman⁴, J I Martín⁵, J L Vicent⁶, S M Baker⁷, T P Russell⁸, C Leighton⁹, J Nogués¹⁰ and Ivan K Schuller¹

¹ Physics Department, University of California-San Diego, La Jolla, CA 92093-0319, USA

² Physics Department, University of California, Davis, CA 95616, USA

³ Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA

⁴ Motorola Labs, Physical Sciences Research Labs, 7700 South River Parkway, Tempe,

AZ 85284, USA

⁵ Depto. Física, F. Ciencias, Universidad de Oviedo, 33007 Oviedo, Spain

⁶ Depto. Física de Materiales, F. Ciencias Físicas, Universidad Complutense, 28040 Madrid, Spain

⁷ Department of Chemistry, Harvey Mudd College, Claremont, CA 91711, USA

⁸ Polymer Science and Engineering, University of Massachusetts, Amherst, MA 01003, USA

⁹ Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455, USA

¹⁰ Institució Catalana de Recerca i Estudis Avançats (ICREA) and Departament de Física, Universitat Autonoma de Barcelona, 08193 Bellaterra (Barcelona), Spain

Received 11 April 2002 Published 13 September 2002 Online at stacks.iop.org/JPhysD/35/2398

Abstract

Nanostructures have interesting properties due to the confinement of electrons in small structures. In addition, since an appreciable fraction of the electronic wave-function resides outside the physical extension of the nanostructure, proximity effects become more important with decreasing size. Moreover, in magnetic nanostructures the magnetic fields also extend a considerable distance outside the physical extent of the nanostructure. Thus, the interplay between size confinement and proximity effects become particularly interesting in magnetic nanostructures. We give two interesting experimental examples of proximity effects with magnetic nanostructures. In one, small magnetic dots radically modify the magnetotransport properties of superconducting films. In the other, the properties of a ferromagnet with nanoscopic antidots are considerably changed because of proximity with an antiferromagnet.

1. Introduction

Magnetic nanostructures are currently intensively investigated because of the interesting modification of their properties due to the confinement in size to length scales comparable to interesting magnetic length scales (dipolar, RKKY, exchange, spin diffusion length, etc) [1–7]. The presence of many surfaces and interfaces, as well as interplay among constituents, often lead to novel and enhanced properties over their bulk counterparts [8–11]. Technologically, these structures are becoming ever more important as they provide new functionality (e.g. magnetic memory) and miniaturization (less power consumption, ultra-high packing density, etc), most notably in the sensor and storage industries [12, 13].

Proximity effects have been investigated for many years in the field of superconductivity [14]. In magnetism there are theoretical predictions (not yet firmly established experimentally) claiming the existence of similar effects [15, 16]. In both cases the proximity effects have been investigated in various configurations of heterostructures and superlattices [17–19].

When materials are confined further in all dimensions, the electronic wave function or the magnetic field arising from the nanostructure extends considerably beyond its physical extent in all directions. Thus, the proximity effects must be considerably enhanced, due to dimensionality effects. Moreover, nanostructures are very seldom isolated from the environment and often consist of multiple components. Generally, they are deposited over a substrate or embedded in a matrix which may be in some sense 'active' (i.e. metallic, magnetic, superconducting, etc). Because of these reasons the proximity effect becomes very important for nanostructures that are in contact with other materials. Thus, the combination of nanostructuring and the proximity effect are both interesting scientifically and important technologically.

There are two ways in which the proximity effect may become important in the context of nanostructures. The properties of the surrounding medium are affected in a substantial way or the properties of the nanostructure itself are changed due to the proximity between the medium and the structures. This generally occurs as length scales become comparable to relevant sizes that determine given physical properties. We show, here, two examples that clearly illustrate this idea. In one, the magnetotransport properties of a superconducting thin film are radically modified by the proximity with an array of nanostructured magnetic dots. In the other, the hysteresis of a thin magnetic film containing antidots is considerably changed by the proximity with an antiferromagnet. The relevant length scales in these cases are the superconducting coherence length and the domain size.

These length scales are nanoscopic and thus the effects are considerably enhanced when the material's physical size becomes comparable or smaller to these length scales. The changes are quite unusual and/or surprising; regular dips appear in the magnetotransport of superconducting films and the hysteresis mode becomes non-symmetric in the magnetic films. To date the understanding of these phenomena is only partial and, therefore, much more research is needed. They also hold the promise of important applications such as Josephson type detectors and enhanced magnetic storage.

2. Sample preparation

The preparation of 'nanostructures in proximity' relies on a combination of state-of-the-art thin film techniques (sputtering, MBE, etc) combined with novel nanolithography methods (e-beam, diblock copolymer, alumina self assembly, etc). The thin film techniques have been described extensively elsewhere so we will not do it here. Nanolithography techniques generally rely on the preparation of a mask with lift-off. Figure 1 shows the most important steps in such a process. The key to this process is the formation of holes of the appropriate geometry in the masks. These can be written in PMMA using electron beams or produced by selfassembly. While e-beam lithography allows the preparation of almost any desired geometry, it is very limited in overall size (typically $100 \,\mu\text{m} \times 100 \,\mu\text{m}$). Self-assembly is less flexible since it relies on chemical and or physical interactions, which cannot be manipulated completely at will. However, these latter techniques allow preparation of macroscopic ($cm \times cm$) samples. Here we give an example of each one of these techniques applied to a specific example.

3. Modification of the medium by the proximity with nanostructures (arrays of nanostructured magnets in proximity with a superconductor)

The penetration of the magnetic field in a type II conventional superconductor occurs in a vortex lattice ('Abrikosov' lattice)



Figure 1. Sketch of (*a*) the e-beam lithography process and (*b*) the diblock copolymer lithography process.

in most of the H-T phase diagram. Each superconducting vortex has a normal core which is roughly equal to the superconducting coherence length and the separation of the vortices (*a*) is given by the simple formula

$$a \cong \sqrt{\frac{\Phi_0}{H}} \tag{1}$$

where Φ_0 is a flux quantum and H is the applied field. A correction factor of the order of one arises depending on the particular geometry of the vortex lattice. Typical vortex lattice distances in the 50–1000 nm range are present in conventional superconductors such as Nb, at easily accessible temperatures and magnetic fields. In the absence of pinning, when a current flows through the superconductor the vortices move, giving rise to the so-called 'flux-flow resistance'. The vortices are usually pinned by defects or impurities in the superconductor which lowers the flux-flow resistance and enhances the critical current in the superconductor. For instance, figure 2(a) shows the fluxflow resistance of a Nb (100 nm) film as a function of magnetic field H, just below the superconducting transition temperature $T_{\rm C}$. The flux-flow resistance is a smooth featureless curve as expected for a random distribution of pinning sites.

The properties of a thin superconducting film can be drastically changed by the proximity with an underlying array of magnetic dots. Arrays of nanostructured magnetic dots can be used as periodic pinning sites and thus may match the periodic vortex lattice. The competing of the two periodicities produces very interesting singularities in the flux flow resistance and the critical current of the superconductors. Such a system is illustrated conceptually in figure 3(a) which shows the side view of a Nb film with an underlying array of magnetic (Ni, Fe or Co) dots. Figure 3(b) shows the scanning electron microscope top view image of a square array of Ni dots prepared by electron beam lithography, with typical dot size ~300 nm and separation ~700 nm. The pinning produced



Figure 2. Resistance vs applied magnetic field for (*a*) a uniform Nb film (100 nm thick), (*b*) Nb film (100 nm thick) with a square (400 nm) array of Ni dots, (*c*) Nb film (100 nm thick) with a rectangular (400 nm \times 900 nm) array of Ni dots and (*d*) Nb film (100 nm thick) with a triangular (400 nm) array of Ni dots.



Figure 3. (*a*) Schematic of a Nb film with a regular array of magnetic dots. (*b*) Top view SEM image of a square (700 nm) array of Ni dots.

by such a square array is shown in figure 2(b). Deep (note the logarithmic scale) periodic minima appear, in some cases up to the 35th order [20]. When a triangular array geometry is used, a series of periodic minima (not as deep) still exist, but only to about 4th order (figure 2(d)). There is also a radical change in the background. The position of the *n*th minimum H_n is (to within 5%) given by the simple formula

$$H_n = \frac{n\Phi_0}{a^2} \tag{2}$$

with n an integer. Clearly the implication is that the various matching peaks correspond to n vortices per dot.

Figure 2(c) shows the interesting dependence of the fluxflow resistance for a rectangular array, with current applied along the long direction of the rectangle. In this case, as for the square and triangular arrays, the first two minima correspond to n vortices per dot, i.e.

$$H_n = \frac{n\Phi_0}{ab} \tag{3}$$

where a and b are the short and long sides of the rectangle, respectively. However, after the second order peak, the periodicity changes to

$$H_n = \frac{n\Phi_0}{a^2} \tag{4}$$

with a being the periodicity along the direction of vortex motion perpendicular to the applied current. In addition, at low fields, several fractional minima can be found which correspond to a non-integer number of vortices per dot [21].

Many interesting questions arise from this work and have been the subject of intense experimental and theoretical investigations. For instance, the location of the vortices [22, 23], the role of the dot size [24], the array geometry [25–27], the magnetic state [28], the nature of the pinning [29–31], the origin of the background [32, 33], the importance of the corrugation, the apparent stronger pinning by the square vs triangular array, and the effect on the critical current, etc are just some issues that have been raised and investigated. Moreover, these systems have given origin to the design of devices such as rf frequency detectors [34] similar to microwave-frequency Josephson-effect detectors. In addition, the connection to many other areas of physics such as epitaxial growth [35, 36], two dimensional melting [37] and even plasma physics [38] where either matching effects between two periodicities or vortex physics is essential, have been noticed and are being currently investigated.

4. Modification of the nanotructures by the proximity with a medium (antidots in a ferromagnet in proximity with an antiferromagnet)

A ferromagnetic thin film in proximity with an antiferromagnet produces interesting results even in the absence of the nanostructuring. Figure 4(a) shows the hysteresis of a single Fe film. The hysteresis loop is centred around H = 0, has a small coercivity and is square. When an identical Fe film is in contact with FeF₂ (an antiferromagnet), due to the proximity effect, the hystheresis loop shifts to negative H, broadens and becomes sheared, as shown in figure 4(b). This is the so-called exchange bias effect and has been the subject of extensive research for the last forty years [39, 40].

The properties of a nanostructured material are also drastically affected by the proximity effect. An example of this is the effect on the magnetic properties of a ferromagnet (Fe) thin film filled with nanosized 'antidots' (holes) in proximity with an antiferromagnet (FeF₂) [41, 42]. Figure 5(a) shows the side view design of such a structure. Figure 5(b) shows the top view of such a structure prepared by lithography using self-assembly of diblock copolymers [43]. The reason for using this preparation technique is that macroscopic areas are needed, to produce sufficient materials that can be easily measured using standard SQUID magnetometry. The dark spots show the 'antidots' in the Fe film.

A comparison and the importance of the proximity effect are shown in figures 4(c) and (d). If the antidots are introduced



Figure 4. Magnetic hysteresis loops of (*a*) a uniform single Fe layer (15 nm thick), (*b*) a uniform Fe (15 nm)/FeF₂(20 nm) film, (*c*) an Fe-network (15 nm) film, and (*d*) a Fe-network (15 nm)/uniform-FeF₂ (20 nm) film at 10 K, after field cooling in



Figure 5. (*a*) Schematic of a nanoporous Fe-network/uniform-FeF₂ structure. (*b*) Top view AFM phase image of a nanoporous Fe network with about 20 nm pore sizes. The dark and bright areas are pores and networks, respectively.

into the uniform Fe layer, the effect is minimal, i.e. a small decrease of the coercivity. On the other hand, when the antidots are introduced into the Fe layer which is in proximity with the antiferromagnetic FeF_2 , the effect is very dramatic. In addition to the shift and broadening of the hysteresis loop, a dramatic

change occurs in the reversal mode of the magnetization. The two sides of the hysteresis curve are different (unlike any other magnetic material), indicating that the reversal mode is different on the two sides. Earlier neutron scattering [44] and transport measurements [45] indicated that in an exchange biased system this is in fact the case. The left-hand side of the loop occurs by domain rotation (Stoner-Wohlfarth), whereas the right-hand side is by domain wall motion. These effects have only been discovered recently and thus are under intense investigation. The asymmetry in the reversal for exchange biased systems is correlated with the presence of a threefold anisotropy. A detailed explanation, the universality of these effects for different materials systems and their connection to the magnitude of the exchange bias is not yet available. Nevertheless, these effects have important implications within the context of the work presented here (i.e. connected to the proximity effect with nanostructures). As shown in figure 4(d), this affects drastically the behaviour of the Fe film; on the left-hand side of the loop the change is small whereas on the right-hand side the magnetization remains almost constant and then suddenly jumps. The behaviour on the right-hand side is consistent with a domain wall mechanism which is pinned in the antidots. Since the reversal mode on the left-hand side is due to domain rotation, the antidots have no effect. Many interesting questions arise in this context which are presently under investigation, such as the lateral size and vertical etching depth dependence of exchange bias, effects of patterning on magnetic anisotropy, thermal stability, nanopatterning processes, etc.

5. Conclusion

We highlighted here an important aspect of nanostructures which make their properties even more interesting and perhaps will be important in improving their application in devices. Due to their small sizes, the electronic wave-function and the magnetic field extends considerably beyond their physical extent. Since nanostructures in most cases are in contact with other materials ('intervening medium'), proximity effects As a consequence, either the may become important. properties of the intervening medium may be drastically affected or the property of the nanostructure itself changes. Two classical examples of this type are the change in the magnetotransport of a superconducting film in contact with an array of magnetic dots and the changes in the hysteresis loop of a ferromagnetic film containing nanoscopic antidots in contact with an antiferromagnet. This opens up an extra degree of freedom by which one can tailor the material properties.

This paper was written in celebration of Prof. T Shinjo's retirement from Kyoto University. Prof. Shinjo is an important and active contributor to the field of magnetism and nanostructured materials for many years and was a great organizer of many conferences in the field. IKS has been very fortunate to interact with him in this field and thankful for many discussions about Sumo.

Work supported by the US DOE, AFOSR and NSF.

References

Shinjo T, Okuno T, Hassdorf R, Shigeto K and Ono T 2000 Science 289 930

M I Montero et al

- [2] Himpsel F J, Ortega J E, Mankey G J and Willis R F 1998 Adv. Phys. 47 511
- [3] Martín J I, Nogués J, Liu K, Vicent J L and Schuller I K J. Magn. Magn. Mater. submitted
- [4] Demokritov S O, Hillebrands B and Slavin A N 2001 *Phys. Rep.* 348 441
- [5] Castaño F J, Hao Y, Hwang M, Ross C A, Vogeli B, Smith H I and Haratani S 2001 Appl. Phys. Lett. 79 1504
- [6] Wittborn J, Rao K V, Nogués J and Schuller I K 2000 Appl. Phys. Lett. 76 2931
- [7] Martín J I, Nogués J, Schuller I K, Bael M J V, Ternst K, Haesendonck C V, Moshchalkov V V and Bruynseraede Y 1998 Appl. Phys. Lett. 72 255
- [8] Ono T, Miyajima H, Shigeto K, Mibu K, Hosoito N and Shinjo T 1999 Science 284 468
- [9] Sellmyer D J, Zheng M and Skomski R 2001 J. Phys.: Cond. Matter 13 R433
- [10] Yu C, Li D, Pearson J and Bader S D 2001 Appl. Phys. Lett. 79 3848
- [11] Liu K, Chien C L, Searson P C and Yu-Zhang K 1998 Appl. Phys. Lett. 73 1436
- [12] Prinz G A 1995 Physics Today 48 58
- [13] Wolf S A and Treger T 2000 IEEE Trans. Magn. 36 2748
- [14] Gilabert A 1977 Ann. Phys. 2 203
- [15] Kiwi M and Zuckermann M I 1973 AIP Conf. Proc. 18 347
- [16] Åkerman J J, Guedes I, Leighton C, Grimsditch M and Schuller I K 2002 Phys. Rev. B 65 104432
- [17] Schuller I K, Kim S and Leighton C 1999 J. Magn. Magn. Mater. 200 571
- [18] Jin B Y and Ketterson J B 1989 Adv. Phys. 38 189
- [19] Shinjo T 1999 Surf. Sci. 438 329
- [20] Hoffmann A 1999 *PhD Thesis* La Jolla: University of California, San Diego
- [21] Stoll O M, Montero M I, Guimpel J, Åkerman J J and Schuller I K 2002 Phys. Rev. B 65 104518
- [22] Harada K, Kasai H, Matsuda T, Tonomura A and Moshchalkov V V 1996 *Science* **274** 1167
- [23] Reichhardt C, Olson C J and Nori F 1998 Phys. Rev. B 57 7937
- [24] Hoffmann A, Prieto P and Schuller I K 2000 Phys. Rev. B 61 6958

- [25] Jaccard Y, Martín J I, Cyrille M-C, Vélez M, Vicent J L and Schuller I K 1998 Phys. Rev. B 58 8232
- [26] Morgan D J and Ketterson J B 1998 Phys. Rev. Lett. 80 3614
- [27] Martín J I, Vélez M, Hoffmann A, Schuller I K and Vicent J L 1999 Phys. Rev. Lett. 83 1022
- [28] Van Bael M J V, Temst K, Moshchalkov V V and Bruynseraede Y 1999 *Phys. Rev.* B **59** 14674
- [29] Nozaki Y, Otani Y, Runge K, Miyajima H, Pannetier B, Nozières J P and Fillion G 1996 J. Appl. Phys. 79 8571
- [30] Otani Y, Pannetier B, Nozières J P and Givord D 1993J. Magn. Magn. Mater. 126 622
- [31] Martín J I, Vélez M, Hoffmann A, Schuller I K and Vicent J L 2000 Phys. Rev. B 62 9110
- [32] Vélez M, Jaque D, Martín J I, Montero M I, Schuller I K and Vicent J L 2002 Phys. Rev. B 65 104511
- [33] Reichhardt C, Zimanyi G T and Grønbech-Jensen N 2001 Phys. Rev. B 64 014501
- [34] Gilabert A, Schuller I K, Moshchalkov V V and Bruynseraede Y 1994 Appl. Phys. Lett. 64 2884
- [35] Materials Science & Engineering B (Solid-State Materials for Advanced Technology) 1999 First Lawrence Symp. on Critical Issues on Epitaxy (Mesa, AZ, USA, 1999)
- [36] Pashley D W 1999 Mater. Sci. Technol. 15 2-B
- [37] Sinha S K (ed) 1980 Ordering in Two Dimensions (Amsterdam: North-Holland)
- [38] Fine K S, Class A C, Flynn W G and Driscoll C F 1995 *Phys. Rev. Lett.* **75** 3277
- [39] For a recent review see e.g. Nogués J and Schuller I K 1999 J. Magn. Magn. Mat. 192 203
- [40] Fraune M, Rüdiger U, Güntherodt G, Cardoso S and Freitas P 2000 Appl. Phys. Lett. 77 3815
- [41] Liu K, Baker S M, Tuominen M, Russell T P and Schuller I K 2001 Phys. Rev. B 63 060403
- [42] Sun L, Ding Y, Chien C L and Searson P C 2001 Phys. Rev. B 64 184430
- [43] Park M, Harrison C, Chaikin P, Register R A and Adamson D H 1997 *Science* **276** 1401
- [44] Fitzsimmons M R, Yashar P C, Leighton C, Nogués J, Dura J A, Majkrzak C F and Schuller I K 2000 Phys. Rev. Lett. 84 3986
- [45] Leighton C, Song M, Nogués J, Cyrille M C and Schuller I K 2000 J. Appl. Phys. 88 344