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Magnetic interactions in amorphous spin-glass–superconducting multilayers

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We have investigated the spin-glass transition of some amorphous multilayers where a spin-glass layer of $GdAl_2$, with a bulk freezing temperature T_f of ≈ 11 K, is intercalated with a non-magnetic layer of Mo_3Si. The transition temperature, as defined by the dc susceptibility maximum, has been investigated with a superconducting quantum interference device as a function of thickness. For thin $GdAl_2$ layers (≈ 9 Å), T_f depends dramatically on the spacing between the magnetic layers, i.e., the thickness of Mo_3Si. This is a signature of a long-range magnetic interaction across the nonmagnetic layers. Such an effect is confirmed by the absence of any superconductivity in the Mo_3Si layers up to thicknesses of 60 Å, whereas the bulk phase has a T_c around 6 K.

Materials which lie at the border of magnetism and superconductivity typically display unusual physical properties. Notable examples are the Kondo systems, the heavy-fermion compounds,[1,2] and Chevrel superconducting phases.[3,4] More recently, the high-T_c superconductivity of the copper oxides has also been suggested to be related to antiferromagnetic correlations. All of the above phenomena are a consequence of strong electronic correlations.[5,6]

In the present work, the basic idea is to force the coexistence of magnetism and superconductivity by building an artificial layering of a superconducting phase (Mo_3Si, with a bulk T_c of 6 K) and of a spin-glass phase ($GdAl_2$, with a bulk T_f of 11 K). We investigate the magnetic and transport properties in order to elucidate the interplay of superconductivity with magnetism. The choice of a weakly magnetic layer is believed to be crucial in order to avoid a too-strong depairing of the Cooper pairs at the magnetic interfaces. Thus, we have selected a spin-glass phase with a low-freezing temperature (11 K at the $GdAl_2$ concentration). In the frozen spin-glass phase, there exists the possibility for a larger cancellation of magnetic polarization out of the layers, as a result of the directional randomness in the exchange interactions. Actually, the coexistence of spin-glass order with superconductivity was discovered earlier in ternary $Nd_{1-x}Th_xRu_2$ alloys.[7]

We selected compositions of the two phases for which amorphous alloys easily form. This avoids any uncontrolled epitaxial growth at the interfaces. The magnetic and electric properties are reported for different thicknesses of $GdAl_2$ ($x = 9$, 18, and 27 Å) and of Mo_3Si ($y = 10$–60 Å). The spin-glass temperature dramatically depends on the two thicknesses whereas no superconductivity has been detected in the composite structures.

The multilayer films were prepared by rf-diode sputtering using argon gas. The substrates were etched prior to deposition and then film growth was achieved at room temperature. Sapphire, kapton, or Si wafers were used. The present results represent materials grown on Si. The alternate layering was controlled by a shutter driven in front of the two half-targets (75 mm diameter) by an electronic switch. Composite targets were used to ensure a constant composition as a function of the sputtering time. The composition of the two phases were determined by electron microprobe analysis. Note that the exact compositions of the GdAl (respectively, MoSi) layers were $Gd_{0.33}Al_{0.67}$ (respectively, $Mo_{0.73}Si_{0.27}$) which we name $GdAl_2$ (respectively, Mo_3Si). The individual thicknesses of the layers were controlled via the sputtering time and they were checked afterwards by x-ray diffraction. Good compositional homogeneity was found over the entire surface of the samples (typical size 2×10 mm). Indeed, no evidence for any composition gradients was detected by the electron microprobe analysis. The total thickness of the multilayer was typically of 1500 Å. The periodicity of the stacking was proved by small-angle x-ray scattering. For periods ranging from 19 to 78 Å, Bragg peaks were observed at least up to fourth order and even up to sixth order in some cases. Thus, the layer period could be accurately determined by including the correction due the complex refraction index of the film.[8] A typical small-angle x-ray scan is shown in Fig. 1 for one $GdAl_2/Mo_3Si$ multilayer. At larger angles, a halo confirmed the amorphous nature of the multilayers.

The magnetic properties were investigated using a SHE superconducting quantum interference device and sweeping the temperature between 1.5 and 100 K in a constant external field. All of the samples show behavior typical for spin glasses, i.e., a pronounced maximum in the dc susceptibility after cooling in zero applied field (ZFC), applying a field and determining the magnetization with increasing temperature. Upon cooling in a field of 150 Oe (FC), parallel to the plane of the films, the susceptibility maximum nearly disappears, and a plateau forms below T_f. This behavior is illustrated in Fig. 2.

The freezing temperature T_f denoted by the maximum in the dc susceptibility is shown versus layer thicknesses x ($GdAl_2$) and y (Mo_3Si) in Fig. 3. An important result is that T_f is always well below the corresponding value for the bulk phase (11 K), even for $GdAl_2$ layers as thick as 27 Å. At constant Mo_3Si thickness, T_f dramatically drops when the spin-glass layer thickness x is reduced from 27 to 9 Å. This is clear evidence that the two-dimensional character degrades the spin-glass order.[9]

Another interesting feature is the T_f dependence on the spacing y between the magnetic layers. Here a strong decrease of T_f is observed upon increasing y, in particular for the 9 Å thick spin-glass layers (Fig. 3). This indicates that the magnetic layers are strongly exchange-coupled across the nonmagnetic films, probably via Ruderman-Kittel-Kasuya-Yosida (RKKY)-type interactions. Some oscillations of T_f even seem to exist at large spacings y, viz. beyond 30 Å. Although this phenomenon requires further confirmation, particularly for other x values, it is striking that it is not observed at small thicknesses of y. We suggest, pending further experimentation, a tentative explanation of the above effects, which relies on the existence of an uniaxial anisotropy.[10] The source of this anisotropy could be either the interfaces[11] or the nearly two-dimensional character of the magnetic layers. Then the magnetic coupling between the layers can be represented by a sum of a spin-glass-like polarization and a ferromagneticlike component, oriented along the uniaxial anisotropy direction. The first term, which is highly incoherent (random) from site to site, both in magnitude and in direction, is believed to dominate at short distances from the magnetic layers. However, this component decays very rapidly at larger distance due to random cancellations in direction and magnitude. On the other hand, the small term resulting from the uniaxial anisotropy will decay more slowly. Indeed these contributions to the RKKY polarization have the same direction (e.g., perpendicular to the layers) and all begin with the same phase at the $GdAl_2/Mo_3Si$ interfaces. Thus, these coherent oscillations can produce a long-range ferromagnetic interaction across the Mo_3Si layers for certain values of its thickness. Accordingly, we argue that, at small values of y, T_f decreases monotonously upon increasing y as a result of the weakening of the dominant spin-glass interaction. Oppositely, at larger interspacings y, the remaining "ferromagnetic" interactions between the layers can be strong enough to increase T_f. At other y distances, this coherency is ineffective and a reduced T_f results. The fact that a ferromagnetic component to the spin polarization propagates over distances as large as 30 Å, is fully consistent

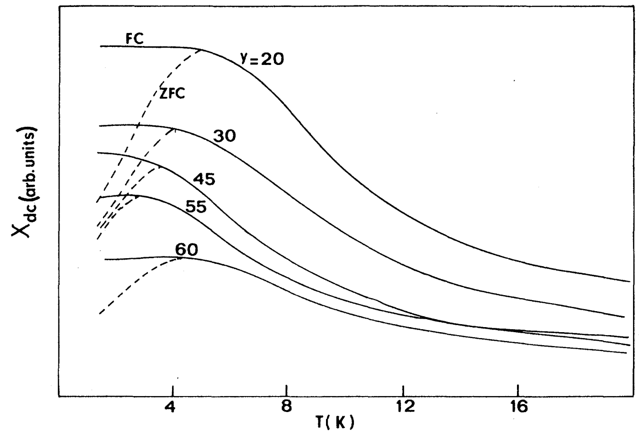

FIG. 2. dc susceptibility vs temperature for multilayer of different thickness, either after zero-field cooling (ZFC, dashed lines) or cooled in a field (FC, solid lines) of 150 Oe.

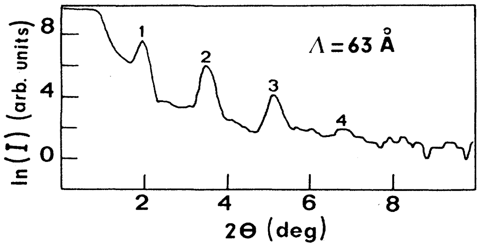

FIG. 1. X-ray diffraction profile (logarithmic scale) at small angles for a $GdAl_2/Mo_3Si$ amorphous multilayers with $x = 18$ Å and $y = 45$ Å. The radiation is Co $K\alpha_1$. The order of the satellites are indicated.

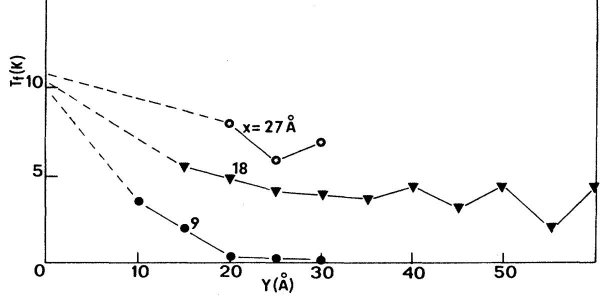

FIG. 3. Plot of the spin-glass transition temperature vs the $GdAl_2$ thickness (x) and the Mo_3Si thickness (y). The lines are guides to the eyes.

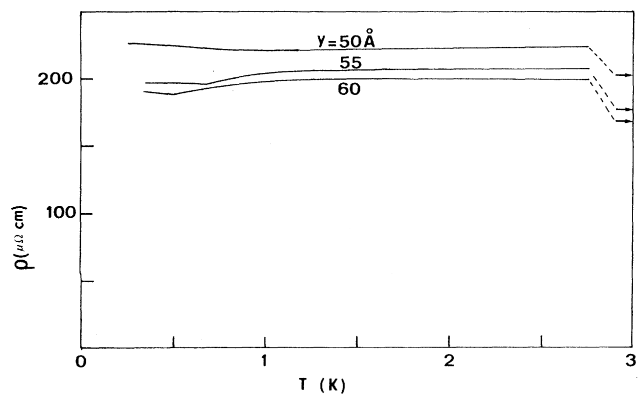

FIG. 4. Temperature dependence of the resistivity for three multilayers. The arrows represent the room-temperature values.

with the lack of any superconductivity down to 0.3 K in all our samples, even those with a thickness of $y=60$ Å. Figure 4 shows the electrical resistivity as a function of temperature for several multilayers. Indeed, no anomaly in the resistivity could be detected neither is there any Meissner shielding observed from magnetic measurements. The temperature dependence of the resistivity is weak up to 300 K because the samples are amorphous.

To conclude, we would like to emphasize that this first study of multilayers involving a spin glass and a superconductor reveals the depression of the spin-glass ordering upon reducing the dimensionality of the magnetic layers. No evidence is found for the coexistence of magnetic order and superconductivity, at least in the range of layers thicknesses under study here. Rather surprisingly, we observe a strong magnetic interaction of the spin-glass layers across the superconducting ones. In addition, this interaction is possibly coherent and ferromagnetic at certain large spacings, typically beyond 30 Å. Such coherent magnetic interactions have already been found in metallic superlattices involving a nonmagnetic Yttrium and various heavy magnetic rare-earth elements.[12–14] In our case, a residual magnetic anisotropy creating a ferromagnetic-like interaction between spin-glass layers would mediate the coherency at large interspacings. Further experiments along these lines are presently in progress.

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[1]F. Steglich, J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, W. Franz, and H. Schafer, Phys. Rev. Lett. **43**, 1892 (1979).
[2]G. R. Stewart, Z. Fisk, J. O. Willis, and J. L. Smith, Phys. Rev. Lett. **52**, 679 (1984).
[3]R. Chevrel, M. Sergent, and J. Prigent, J. Solid State Chem. **3**, 515 (1971).
[4]O. Fischer, Appl. Phys. Lett. **16**, 1 (1978).
[5]P. Thiry, G. Rossi, Y. Petroff, A. Revcolevschi, and J. Jegoudez, Europhys. Lett. **5**, 55 (1988).
[6]P. Burlet, C. Vettier, M. J. G. M. Jurgens, J. Y. Henry, J. Rossat-Mignot, H. Noel, M. Potel, P. Gougeon, and J. C. Levet, Physica (Utrecht) **153–155**, 1115 (1988).
[7]D. Huser, M. J. F. Rewiersma, J. A. Mydosh, and G. J. Nieuwenhuys, Phys. Rev. Lett. **51**, 1290 (1983).
[8]A. M. Kadin and J. E. Keem, Scr. Metall. **20**, 443 (1986).
[9]G. C. Kenning, J. M. Slaughter, and J. A. Cowen, Phys. Rev. Lett. **59**, 2596 (1987).
[10]D. Sherrington, D. M. Cragg, and D. J. Elderfield, J. Magn. Magn. Mater. **31–34**, 1417 (1983).
[11]H. J. G. Draaisma and W. J. M. de Jonge, J. Appl. Phys. **62**, 3318 (1987).
[12]J. Kwo, E. M. Gyorgy, D. B. McWhan, M. Hong, F. J. DiSalvo, C. Vettier, and J. E. Bower, Phys. Rev. Lett. **55**, 1402 (1988).
[13]C. F. Majkrzac, D. Gibbs, P. Böni, A. I. Goldman, J. Kwo, M. Hong, T. C. Hsien, R. M. Fleming, D. B. McWhan, Y. Yafet, J. W. Cable, J. Bohr, N. Grimm, and C. L. Chien, J. Appl. Phys. **63**, 3447 (1988).
[14]Y. Yafet, J. Kwo, M. Hong, C. F. Majkrzak, and T. O'Brien, J. Appl. Phys. **63**, 3453 (1988).