Heat Induced Antiferromagnetic Coupling in Multilayers with Ge Spacers

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We report on heat induced antiferromagnetic exchange coupling in a new system: ferromagnetic Fe films separated by a spacer of amorphous Ge. Antiferromagnetic coupling occurs at spacer thicknesses between 20 and 25 Å. It exhibits a striking temperature dependence which has a positive temperature coefficient and is fully reversible in the temperature range between 40 and 230 K. Our findings about the importance of the interfaces support the interpretation that resonant tunneling through localized states in the gap of the spacer mediate the magnetic exchange. [S0031-9007(98)05484-2]

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As early as in the 1950s a question arose, stimulated by the huge success of semiconductor physics and some open questions about the mechanism of itinerant ferromagnetism: Is there a material in which a ferromagnetic interaction can be switched on by thermal excitation of charge carriers [1]? The answer, so far, has been no, in spite of extended experimental search. Also theoretical modeling has yielded that heat induced long range order cannot be obtained in a doped semiconductor containing magnetic ions under realistic assumptions [2]. However, the discovery of oscillatory exchange coupling in magnetic multilayers [3] and its explanation in terms of RKKY coupling [4] brought about a new idea. In those layered heterostructures the symmetry is such that all but one component of the RKKY coupling, the one perpendicular to the layer plane, cancel out. Then it is the absence of frustration in only one direction that makes long range interlayer exchange coupling possible. Maybe this particular symmetry would make it possible, too, to realize an artificial solid which exhibits heat induced or semiconducting magnetic exchange coupling. By "heat induced" or "semiconducting" magnetic behavior we mean the magnetic analog to the electrical semiconductor in the sense that the effective exchange interaction increases via thermal excitation of charge carriers.

In 1992 Toscano and co-workers [5] were the first to give evidence for such a coupling. They studied Fe/a-Si/Fe trilayers prepared by evaporation at temperatures around 40 K. In these heterostructures the two iron layers are exchange coupled across the amorphous silicon (a-Si) spacer, the coupling showing two sign changes and, most importantly, indeed heat induced behavior [6]. Other interesting candidates for thermally induced coupling are sputtered multilayers of iron and silicon, which, due to interdiffusion at room temperature, tend to form polycrystalline silicides [7–9]. Those multilayers have originally been reported to show a strong antiferromagnetic alignment at room temperature and weaker coupling after cooling. However, in recent publications those observations are reinterpreted to be due to a predominance of pinhole coupling at low temperatures [9,10] or to be based on a pronounced biquadratic coupling with a strong but conventional temperature dependence [11], respectively. Therefore, to our knowledge, Fe/a-Si/Fe trilayers remain the only system so far reported to show thermally induced effective exchange coupling. The above mentioned reports, on the other side, have provoked the general impression that heat induced exchange interaction does not exist at all. This situation calls for further clarification. In this Letter we set out to demonstrate that the phenomenon indeed does occur more generally. We present evidence for a new system exhibiting this most unusual coupling behavior: Fe layers separated by a-Ge spacers. It is demonstrated that the coupling with increasing spacer thickness switches from ferromagnetic to antiferromagnetic and back to ferromagnetic. We unambiguously show that in the antiferromagnetic region the effective exchange interaction between the two iron layers has a positive temperature coefficient.

The samples consist of Fe/a-Ge/Fe trilayers grown by molecular beam evaporation onto ferromagnetic amorphous ribbon $Fe_5Co_{75}B_{20}$ substrates. Sample preparation and magnetic measurements are performed in a UHV chamber with a base pressure of less than 10^{-10} Torr. We emphasize that a key to the present finding is the enhanced temperature control during sample preparation. It is decisive to hold the substrate at $T = 30$ K in order to avoid interdiffusion during the evaporation process and it is likewise important to anneal the trilayers at moderate temperatures prior to the magnetic study presumably to form the appropriate interfaces.

As our main experimental tool we use surface magnetometry by spin polarized secondary electron emission (SPSEE). A 1–5 keV primary electron beam produces a cascade of secondary electrons on the sample surface. A subsequent spin analysis of the emitted secondary electrons with reference to the two in plane quantization axes is carried out in a 100 keV Mott detector. The spin polarization P, defined as $P = (N\uparrow - N\downarrow)/(N\uparrow + N\downarrow)$, is proportional to the magnetization of the sample at the surface [12,13]. $N\uparrow$ and $N\downarrow$ are the number of electrons with spin parallel and antiparallel to the chosen quantization axis, respectively. The high surface sensitivity allows us to directly probe the magnetization of the outermost layer and therefore makes SPSEE well suited for *in situ* analysis of magnetic multilayers. We monitor the response of an exchange coupled surface layer with respect to the magnetization of a substrate and in this way study the exchange coupling across a particular spacer material between surface layer and substrate.

For the preparation of the samples we first evaporate at 30 K a 15 Å iron layer onto the sputter cleaned substrate. Next the Ge spacer is prepared as a wedge in order to allow measurements within a certain thickness range in one experimental run. A 15 Å Fe overlayer completes the sample. Auger electron spectroscopy is used to determine the thickness and check the cleanliness of the evaporated films. The entire structure is evaporated at 30 K which makes us confident that we are dealing with clean amorphous Ge as spacer material. Short annealing of the samples at 200 K prior to the magnetic measurements is important, evidently for interface formation. Studies of growth and annealing of Ge on Fe and Fe on Ge as well as coevaporation of Fe during the growth of the Ge wedge let us rule out that interdiffusion plays an important role in sample preparation.

First we address the thickness dependence of the exchange coupling across amorphous Ge. To do so we perform SPSEE measurements of the polarization P at remanence along the wedge. This signal P as shown in Fig. 1 can be considered, at least in the limit of weak exchange coupling and constant temperature, to be roughly proportional to the coupling strength J. This is the case for the following reasons: because of the high surface sensitivity of SPSEE the signal reflects but the magnetization of the Fe overlayer. In the case of antiferromagnetic coupling, for instance, this magnetization closely follows the one of the substrate, but with opposite sign. To illustrate this, typical hysteresis loops of Fe overlayer and substrate, respectively, are shown in Fig. 2. The fact that the Fe overlayer adopts the coercivity of the substrate lets us conclude that magnetic coupling does exist across the spacer, and the sign of the P signal of the Fe overlayer at remanence with respect to the one of the substrate unambiguously reveals the sign of the coupling J. The uncoupled Fe overlayer, as grown under the present conditions, shows virtually no remanence and an approximately linear response of P to a small magnetic field, be it an external field or an effective coupling field. Therefore, for a constant Fe overlayer

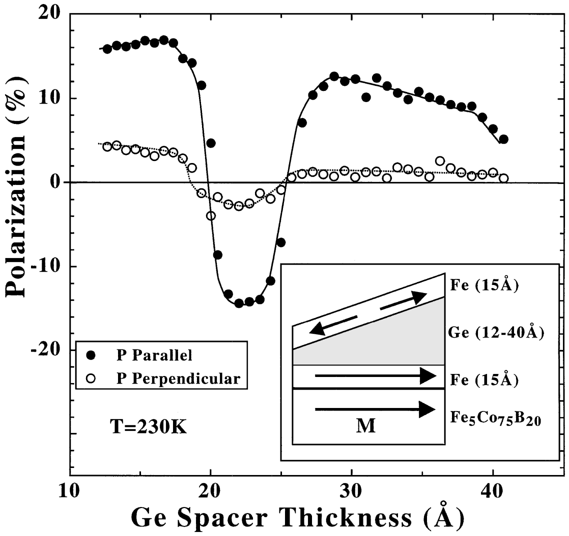

FIG. 1. Spin polarization P of secondary electrons at remanence of the Fe overlayer of an Fe/a-Ge wedge/Fe sandwich deposited on an $Fe_5Co_{75}B_{20}$ substrate versus Ge spacer thickness. In-plane components parallel and perpendicular to the magnetizing field are shown.

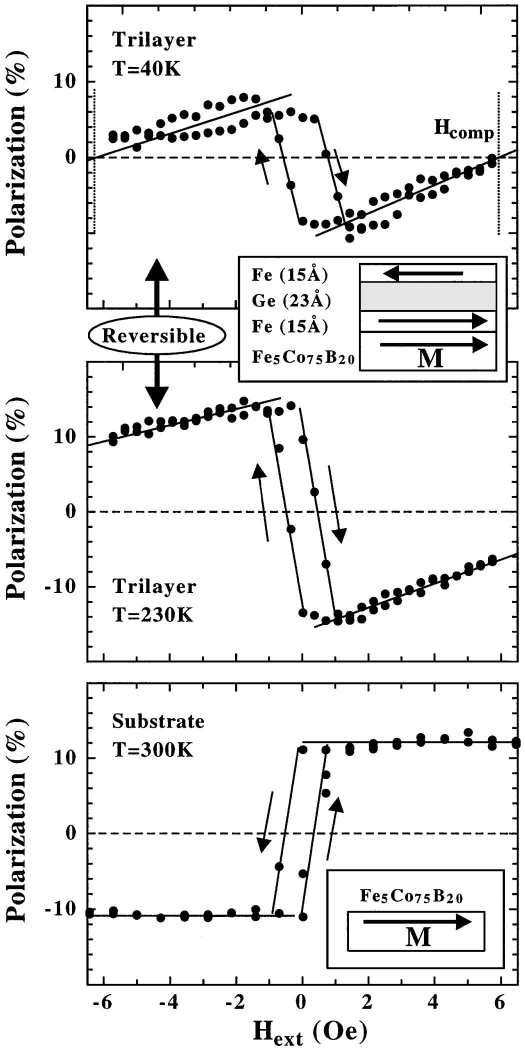

FIG. 2. Spin polarization P of secondary electrons versus applied magnetic field at $T = 40$ K (top panel) and at $T = 230$ K (center panel) of an antiferromagnetically coupled Fe/a-Ge/Fe trilayer deposited on an $Fe_5Co_{75}B_{20}$ substrate. The dotted lines in the top panel indicate the compensation field H_{comp} at $T = 40$ K. At $T = 230$ K H_{comp} is about twice as large as at 40 K. The bottom panel depicts a hysteresis loop of the substrate at room temperature.

thickness the polarization P of the coupled Fe overlayer at remanence can be taken as a relative measure of the coupling strength J. We note that each data point in Fig. 1 is obtained from the difference between two polarization measurements during which the substrate is magnetized in the opposite easy directions, respectively, which rules out an unidirectional shift of the hysteresis as a possible cause of the sign change.

As a result the dependence of the two in-plane components of P of the sample at remanence on the spacer thickness is shown in Fig. 1. With increasing spacer thickness the sign of the P signal changes from positive to negative and back to positive. This clearly demonstrates the occurrence of antiferromagnetic coupling in an intermediate thickness range, in striking similarity to the observations on a-Si spacers [5,6]. At Ge thicknesses below 19 Å the coupling is ferromagnetic and strong enough to saturate the magnetization of the Fe overlayer. In a region between 20 and 25 Å the coupling becomes antiferromagnetic, whereas at larger thicknesses it is again ferromagnetic but clearly weaker than in the first ferromagnetic region. Further we comment that the nonzero perpendicular component of P in Fig. 1 is due to a slight misalignment of the easy magnetization direction of the amorphous ribbon substrate with respect to its axis.

The positive temperature coefficient of the coupling strength, however, is certainly the most crucial aspect of exchange coupling in multilayered structures with semiconducting spacers. In the thickness range of antiferromagnetic coupling we identify heat induced effective exchange interaction. We strongly emphasize that the term heat induced in the the present Letter always refers to a fully reversible temperature dependence. It is not to be confused with the annealing procedures for sample formation as discussed further below.

We observe heat induced effective exchange interaction directly by measuring the temperature dependence of the compensation field H_{comp}. H_{comp} is the external magnetic field necessary to compensate the negative exchange field, i.e., to cancel the antiferromagnetic coupling. Thus H_{comp} is strictly proportional to the coupling strength J. In the present SPSEE measurements H_{comp} is reached at the point where the signal P of the complete sample becomes zero, since P reflects the magnetization of the Fe overlayer only which has no remanence. Figure 2 depicts two hysteresis loops of a sample at a given spacer thickness within the antiferromagnetic coupling range measured at different temperatures. A hysteresis loop of the substrate recorded at room temperature is included in Fig. 2 for comparison. We find that at $T = 40$ K an external field of about 6 Oe is sufficient to compensate the coupling, whereas at $T = 230$ K the compensation field can be estimated to be twice as large. This is a definite evidence for a positive temperature coefficient of the effective exchange interaction, which we name heat induced or semiconducting magnetic behavior. We note that the remanent polarization in Fig. 2 obviously is larger at 230 than at 40 K thus demonstrating that an increase in the coupling strength indeed produces an increase of the Fe overlayer polarization signal. As further observation we mention that the zero crossings of P in Fig. 1 do not, within the accuracy of our thickness determination, depend on the temperature.

The measurement of compensation fields also allows a crude quantitative estimate of J. When M_{S} denotes the saturation magnetization of the iron layer and t_{Fe} its thickness then $J = t_{\mathrm{Fe}} M_{\mathrm{S}} H_{\mathrm{comp}}$. For $H_{\mathrm{comp}} = 6$ Oe at $T = 40$ K we now get $J = 1.5\ \mathrm{merg/cm^2}$, a value that is a few orders of magnitude smaller than for metallic multilayers but comparable to the coupling strength across a-Si.

So far the exchange across Ge, either evaporated at 50 K [14] or prepared at room temperature [15], has been reported to be always ferromagnetic. The difference between those observations and the present findings arises from varying preparation conditions leading to quite different spacer materials. We observe that the existence of heat induced antiferromagnetic coupling as described here subtly depends on the details of preparation. Only for samples evaporated at 30 K and only after short annealing at 200 K does antiferromagnetic coupling with a fully reversible temperature dependence and positive temperature coefficient occur. We emphasize that this preparation and annealing procedure cannot be replaced by just evaporating the layers at 200 K; on the contrary, evaporation of the trilayers at 200 K or above always leads to strong ferromagnetic coupling only.

Next we discuss some details of the annealing procedure with the aim to gain some information on the coupling mechanism. The trilayers as grown, prior to annealing, exhibit ferromagnetic coupling for all spacer thicknesses up to 50 Å. During the first heating cycle an irreversible change of the coupling behavior sets in rather abruptly at around 190 K which leads to the occurrence of antiferromagnetic coupling as shown in Fig. 1. After the drastic changes in the coupling behavior during the first annealing process, however, the heterostructure remains stable against further annealing below room temperature. In particular, the temperature dependence of the coupling strength as shown in Fig. 2 is fully reversible. To test the possible influence of Fe-Ge interdiffusion during the annealing process we also treat both interfaces separately. First we grow a thin Ge-on-Fe wedge structure and anneal it at above 200 K. The Fe and Ge Auger intensities do not change upon annealing which rules out substantial interdiffusion across the first interface. Then, a regular Ge-on-Fe wedge is annealed at 200 K and, after cooling to 30 K, the second Fe layer is added to form the trilayer. No difference to the trilayers as grown at 30 K can be seen in the coupling, and the complete heterostructure has to be annealed again to 200 K to finally provoke the antiferromagnetic coupling. Next we address the influence of the interdiffusion at the second interface between the Ge wedge and the Fe top layer. We evaporate a wedge shaped Fe overlayer on a Ge layer and carefully monitor Auger intensities and the spin polarization during an annealing process. No changes of these signals can be detected. Therefore substantial interdiffusion of Fe into the Ge spacer again can be ruled out. As a further test a few percent of Fe impurities are implanted in the Ge spacer by low-rate coevaporation. This again leads to pure ferromagnetic coupling after preparation and, without any heat treatment, does not produce antiferromagnetic interaction. The latter would be expected if interdiffusion upon annealing was the reason for the occurrence of antiferromagnetic coupling across a-Ge. We also perform experiments with a Ge wedge grown directly on the substrate without prior Fe evaporation. No heat induced antiferromagnetic coupling can be observed in these samples. From all these observations we conclude that the heat treatment after evaporation at low temperature provokes irreversible changes at both interfaces. Evidently this interface formation is necessary for the occurrence of heat induced antiferromagnetic coupling across a-Ge.

The similarities of the coupling behavior as well as of the general material properties between a-Si and a-Ge suggest that the exchange mechanism is the same in both cases. It has been proposed [6] that tunneling through localized defect states in the semiconducting spacer could be responsible for the observed exchange effects and their spectacular temperature dependence. The interface sensitivity observed in the present study supports this hypothesis. A tunneling probability generally depends on the conditions at the interface and therefore is likely to be sensitive to its specific composition. More experiments, perhaps with an interface-chemistry sensitive method, are required to describe the transitions at the interfaces upon annealing. However, the fully reversible temperature dependence of the effective exchange interaction between the two ferromagnets with its positive temperature coefficient, i.e., the heat induced or semiconducting magnetic behavior, proposedly comes about because the number of participating channels increases with temperature via thermal excitation of charge carriers, like a magnetic analog to the electrical semiconductor.

In conclusion, we present evidence for heat induced antiferromagnetic exchange interaction in a new system: Fe/a-Ge/Fe trilayers. The fact that the intriguing coupling phenomenon is a general one and not restricted to only one set of materials raises confidence that building layered structures of ferromagnets and semiconductors is a way to produce semiconducting exchange coupling in an artificial solid. To quantitatively explain the mechanism, however, still is a supreme task in the future which must be accompanied by further basic experiments on crystalline or amorphous model systems.

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[1] G. Busch, Helv. Phys. Acta **40**, 812 (1967).

[2] J. Bernasconi and S. Strässler, Phys. Kondens. Mater. **10**, 224 (1969).

[3] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, Phys. Rev. Lett. **57**, 2442 (1986).

[4] W. Baltensperger and J. S. Helman, Appl. Phys. Lett. **57**, 2954 (1990); P. Bruno and C. Chappert, Phys. Rev. Lett. **67**, 1602 (1991).

[5] S. Toscano, B. Briner, H. Hopster, and M. Landolt, J. Magn. Magn. Mater. **114**, L6 (1992).

[6] B. Briner and M. Landolt, Phys. Rev. Lett. **73**, 340 (1994).

[7] E. E. Fullerton, J. E. Mattson, S. R. Lee, C. H. Sowers, Y. Y. Huang, G. Felcher, and S. D. Bader, J. Magn. Magn. Mater. **117**, L301 (1992).

[8] K. Inomata, K. Yusu, and Y. Saito, Phys. Rev. Lett. **74**, 1863 (1995).

[9] J. Kohlhepp and F. J. A. den Broeder, J. Magn. Magn. Mater. **156**, 261 (1996).

[10] F. J. A. den Broeder and J. Kohlhepp, Phys. Rev. Lett. **75**, 3026 (1995).

[11] E. E. Fullerton and S. D. Bader, Phys. Rev. B **53**, 5112 (1996).

[12] M. Landolt, Appl. Phys. A **41**, 83 (1986).

[13] H. C. Siegmann, J. Phys. Condens. Matter **4**, 8395 (1992).

[14] B. Briner, U. Ramsperger, and M. Landolt, Phys. Rev. B **51**, 7303 (1995).

[15] J. J. de Vries, Ph.D. thesis, Eindhoven University of Technology, 1996.