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Heat-activated magnetic exchange coupling across Ge barriers and Ge/Si heterostructures

B. Briner,* U. Ramsperger, and M. Landolt
Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule, CH-8093 Zürich, Switzerland
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We report the observation of thermally activated exchange coupling across amorphous germanium barriers. At $T=50$ K the coupling persists up to $d_{Ge}=60$ Å. Heat input reversibly enhances the coupling strength. The exchange across *a*-Ge always is ferromagnetic. Experiments with Ge/Si heterostructures show that the occurrence of antiferromagnetic coupling requires the presence of *some* *a*-Si in the barrier and a *total* spacer thickness of ~ 20 Å. Results indicate that the density of defect states determines the nature of the coupling through amorphous semiconductors.

Exchange coupling in layered metallic structures has attracted a lot of scientific interest in recent years,[1] because the controlled growth of crystalline multilayers allows us to study in detail the implications of the conduction electron morphology (density, wave vector) on the magnetic ordering. We have extended this research field by investigating *a*-Si, and we have shown that even an amorphous semiconductor barrier where localized electronic states dominate the charge transport exhibits oscillatory coupling.[2,3] Due to the semiconducting nature of the spacer material, the coupling across *a*-Si always is thermally activated.[4] Heat-induced ferromagnetic coupling has been found for the amorphous insulator SiO.[5] Recent investigations of light- and heat-induced exchange coupling across a crystalline narrow-band-gap silicide[6] and model calculations for semiconductor barriers[8,9] have treated the implications of a small gap height on the magnetic interaction.

The question of which physical process governs the coupling across nonmetals still is a matter of debate. In order to improve our understanding of this phenomenon it is important to assess the influence of gap height, defect states, and metal/semiconductor interface structure on the sign and strength of the coupling. Our decision to investigate amorphous germanium and combined Ge/Si spacers is motivated by the desire to clarify the nature of the exchange across amorphous semiconductors. The present results are unexpected from earlier experiments with *a*-Si barriers. We find that heat activation is a common feature of coupling across semiconductors while the occurrence of oscillatory coupling strongly depends on the choice of the spacer material. Experiments indicate that the height of the mobility gap does not govern the coupling sign in a manner predicted by theory,[8,9] Results on Ge/Si barriers and annealing studies suggest that the observed coupling is a bulk property of the whole semiconductor barrier, and does not sensitively depend on the morphology of the ferromagnet/semiconductor interfaces. On the other hand, these experiments present strong evidence that defect states play a decisive role for the exchange across amorphous semiconductors. In a qualitative explanation we propose that the occurrence of oscillatory coupling hinges upon a sufficiently high defect density. If defects indeed are the key ingredient for coupling through nonmetallic spacers we expect that controlled doping of crystalline semiconductors will develop into a powerful tool for tailoring the strength and sign of magnetic exchange coupling.

The samples which we investigate all have the same structure. An amorphous ferromagnetic $FeNiB_{0.5}$ ribbon serves as substrate for film growth and as magnetic driver. On the substrate we evaporate a semiconductor barrier of variable thickness and composition. Finally a 15–20-Å-thick Fe capping layer is deposited. Samples are prepared and magnetically characterized in an UHV system (base pressure 10^{-10} Torr) equipped with a He cryostat which allows us to maintain a temperature of 50 K during all stages of the experiment. To separately probe the magnetic state of the thin capping layer we use a surface-sensitive magnetometer based on spin polarized secondary electron emission (SPSEE) which has been described before.[4]

In our setup the detection of exchange coupling relies on the particular choice of the magnetic driver and overlayer materials. The $FeNiB_{0.5}$ ribbon has a square hysteresis with near 100% remanence and a small coercive field of ~ 0.5 Oe. The polycrystalline Fe capping layer, on the other hand, is magnetically soft. In the absence of an external field it breaks up into domains to lower its magnetostatic energy. Due to low lateral resolution SPSEE averages over several domains and yields no remanent polarization for an Fe overlayer which is decoupled from the substrate. Exchange coupling forces the overlayer to follow the square low-coercivity substrate hysteresis. As long as the coupling is not able to saturate the capping layer the remanent overlayer SPSEE polarization (P_{rem}) monotonically increases with the coupling strength. P_{rem} serves as a relative measure of the coupling strength for comparison of samples with different barrier thicknesses and for temperature dependences. We have shown for an antiferromagnetically (AF) coupled Fe/Si/Fe trilayer that $P_{rem}=5\%$ corresponds to an exchange field of $H_{exch}\approx 15$ Oe.[4] Obviously SPSEE allows us to detect very weak coupling fields in a convenient way at the expense of the fact that the exact relation between H_{exch} and P_{rem} is unknown.

Sample preparation starts by sputter cleaning the substrate and cooling it down to 50 K. For film evaporation

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we heat the high purity sources (Fe: 99.998% wire; Ge: 99.999% grains in a tungsten crucible) by electron bombardment. Deposition rates of about 4 Å/min are monitored with a quartz microbalance and by Auger electron spectroscopy (AES). The low growth temperature has been chosen to avoid diffusion of Fe into the semiconductor barrier. An AES study at $T=50$ K clearly confirms the absence of appreciable interdiffusion under the present preparation conditions. In Fig. 1 we plot the relative intensities of the Fe *LMM* Auger line at 650 eV and the Ge *LMM* Auger line at 1147 eV versus the evaporation time. Data are well fitted with exponential curves and accepted values for the mean free paths.[7] Similar AES results are also obtained for the low-temperature growth of Fe on Ge (Fig. 1, lower panel).

Exchange coupling through 47 Å of *a*-Ge is illustrated in the top panel of Fig. 2. The overlayer SPSEE hysteresis (right side) exhibits the same shape and coercivity as the hysteresis obtained from the bare substrate (left). In the bottom panel of Fig. 2 we give an overview of the thickness dependence of exchange coupling across *a*-Ge. The remanent SPSEE overlayer polarization P_{rem} is plotted versus the thickness of the Ge barrier. Low-temperature ($T=50$ K) results are depicted by dots and indicate the persistence of ferromagnetic (FM) coupling for Ge thicknesses up to 60 Å. Note that for all spacer thicknesses the coupling is ferromagnetic. The data in Fig. 2 exhibit considerable scatter in spite of the fact that the statistical error for the determination of P_{rem} is $<1\%$. This scatter must entirely be ascribed to inhomogeneity in the semiconductor barriers and reflects a pronounced sensitivity of the exchange coupling to sample quality. Such a behavior is an important feature of semiconductor spacers and can readily be understood if defect states are considered the key ingredient of the coupling process.

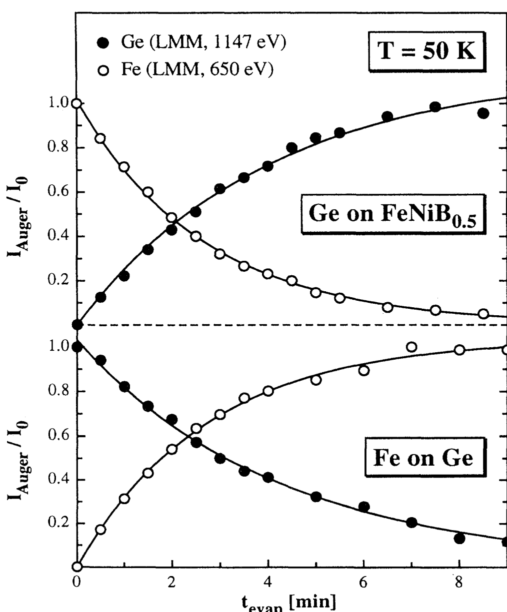

FIG. 1. AES growth study for *a*-Ge evaporated onto $FeNiB_{0.5}$ (top panel) and for Fe deposited onto *a*-Ge (bottom panel). Exponential fits use mean free paths of 13 Å for the Fe *LMM* line at 650 eV and of 20 Å for the Ge *LMM* line at 1147 eV. No signs of interdiffusion are visible.

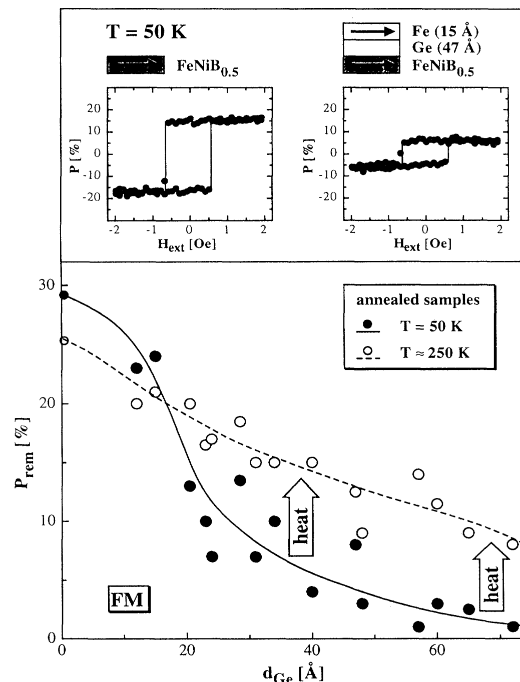

FIG. 2. Top panel: SPSEE hystereses of the bare substrate (left) and of an $Fe/Ge/FeNiB_{0.5}$ trilayer (right) which indicates ferromagnetic (FM) coupling across 47 Å of *a*-Ge. Bottom panel: remanent overlayer SPSEE polarization vs Ge thickness at $T=50$ K (dots) and $T\approx250$ K (open circles), respectively. Lines and arrows are drawn as guides to the eye.

In Fig. 2 we also illustrate the positive temperature coefficient of the coupling across *a*-Ge. Open circles represent the overlayer SPSEE signal measured when samples are heated to about 250 K by means of either a halogen lamp or a small filament mounted beneath the substrate. In comparison to the data obtained for $T=50$ K we find a substantial and fully reversible increase of P_{rem} for $d_{Ge}\geq 20$ Å which is caused by an increased coupling strength. This observation is completely analogous to the heat-activated exchange across *a*-Si and *a*-SiO barriers[4,5] and suggests a close relationship between the coupling processes in these materials. The positive temperature coefficient confirms the semiconducting nature of the Ge barrier and excludes a metal-insulator transition in the spacer as possible origin of the observed coupling.

Annealing effects are also seen for *a*-Ge and further help to understand the coupling mechanism. They occur on the first heating cycle after sample preparation and lead to an irreversible reduction of the coupling strength at $T=50$ K. A reduced coupling strength after annealing is readily explained if we assume that defects mediate the exchange and annealing reduces the defect density. On the other hand, Fe diffusion would lead to increased coupling because it would reduce the effective barrier thickness. Successive temperature dependences of P_{rem} are reversible as long as the former annealing temperature is not exceeded. Dots in Fig. 2 represent low-temperature

results obtained after a short annealing to 250 K. All thermally driven coupling changes depicted in Fig. 2 are fully reversible.

We also want to mention that exchange across *a*-Ge is not light sensitive. We find that all coupling changes provoked by illumination with the halogen lamp must be attributed to sample heating. The reason for the decreased overlayer remanence with thin (< 20 Å) Ge spacers probably is the nearly complete saturation of the Fe overlayer due to a strong coupling already at $T=50$ K. As a consequence the thermal dependence of the overlayer magnetization is dominated by the usual decrease in the spontaneous magnetization of substrate and overlayer.

Now we turn to the intriguing question of why *no AF exchange* is observed for *a*-Ge whereas *a*-Si exhibits a sign change of the coupling. Such a behavior is unexpected on theoretical grounds. A model proposed by Slonczewski[8] relates the coupling sign to the gap height of the semiconductor. This theory relies on spin-dependent conduction electron reflections at the metal-semiconductor interfaces. It predicts AF coupling for small gaps at variance with the present observation where *a*-Ge exhibits FM coupling in spite of its smaller gap as compared to *a*-Si. Recently Bruno has proposed an extension of the barrier model to finite temperatures which describes heat-induced coupling for sufficiently small barriers.[9] To test the alternative proposition that interfaces may have a dominant influence on the coupling we use the following setup. The *a*-Ge barrier is replaced by a sequence of two or three Ge and Si sublayers which together form a heterostructure barrier. Across these barriers interface-controlled coupling should become evident by characteristic differences between samples with interchanged order of the sublayers.

The four different barrier types used for this study are sketched in the top of Fig. 3. Below we plot the remanent overlayer polarization versus the total spacer thickness. Three observations in Fig. 3 are unexpected and indeed striking. First we find that AF coupling occurs also for combined barriers and not only for a pure *a*-Si spacer. Furthermore, the appearance of AF coupling does not depend on the detailed barrier structure. It indeed is observed for all four sample types. Finally the nature of the coupling definitely correlates with the *total thickness* of the barrier.

A positive temperature coefficient is found also for coupling across heterostructure barriers. Dots in Fig. 3 indicate data observed at 50 K, open circles represent thermally activated coupling at $T\approx200$ K. We find a strong enhancement of the coupling at 200 K with respect to the low-temperature results which appear for both FM and AF coupling in full analogy to the temperature dependence of coupling across *a*-Si. This finding further supports the view that heat-activated coupling is an intrinsic property of amorphous semiconductor barriers.

The observation that AF coupling depends on the total spacer thickness but not on the size and relative position of the *a*-Si layer is intricate. From it we infer that the *a*-Ge sublayer plays an active role in the determination of the coupling sign as soon as some *a*-Si, more than 4 Å in this study, is present in the barrier. In addition we conclude that the reversal of the coupling sign cannot result from an exchange process which relies only on the interference of carrier reflections at the boundaries of the *a*-Si sublayer. Figure 4 further supports this conclusion. Here we plot the data of Fig. 3 versus the sum of the Ge- and Si-layer thicknesses. There is no correlation between either Σd_{Ge} or Σd_{Si} and the sign of the exchange coupling.

In conclusion, we present the observation of heat-activated ferromagnetic exchange coupling across *a*-Ge. By comparing these results with the heat-induced oscillatory coupling found for *a*-Si (Refs. 3 and 4) we infer that a positive temperature coefficient of the coupling strength is a common distinctive feature of semiconducting barriers while the existence of AF coupling strongly hinges upon the choice of the spacer material. Experiments with Ge/Si heterostructures clearly reveal that the occurrence of AF exchange is predetermined neither by the interface structure nor by the height of the mobility gap. Thus two major theoretical approaches to coupling across nonmetals are shown to be inappropriate for the present amorphous barriers. As an alternative approach we now propose that the impurity or defect density is the decisive quantity that governs magnetic interaction across amorphous semiconductors. This view is corroborated by the observed heat sensitivity of the coupling which indicates that low-energy excitations strongly influence the magnetic exchange. Shallow excitations occur in an amorphous material because of randomly distributed defect energies around the Fermi level, which in-

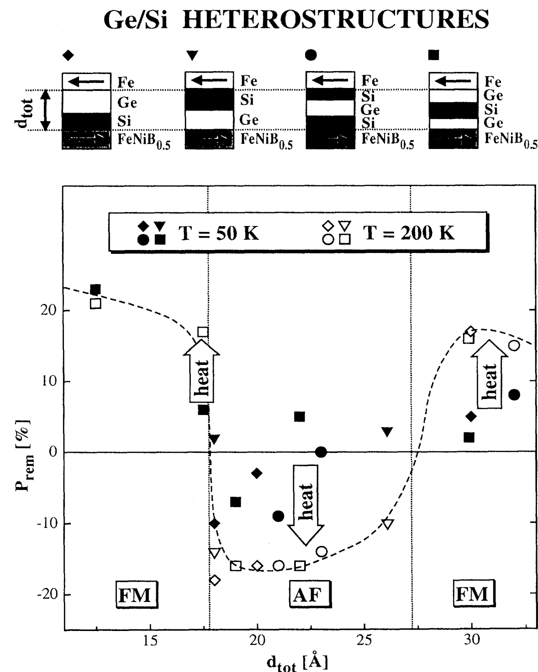

FIG. 3. Remanent overlayer polarization vs *total* spacer thickness for samples with combined Ge/Si barriers measured at 50 K (black symbols) and at 200 K (open symbols), respectively. Antiferromagnetic (AF) coupling is found for $d_{tot}\approx20$ Å regardless of the detailed sequence of the individual Ge and Si layers. Line and arrows are drawn as guides to the eye.

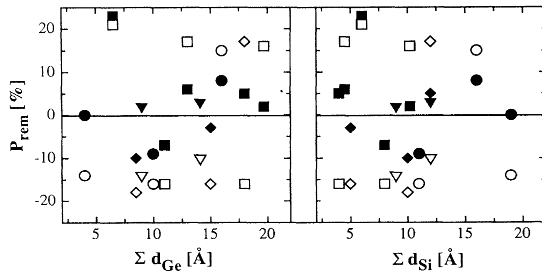

FIG. 4. Same data as in Fig. 3 but plotted vs the total Ge thickness (left panel) and the total Si thickness (right panel), respectively. No correlation exists between the coupling sign and Σ_{Ge} or Σd_{Si}.

troduce a small effective gap even in the presence of a large mobility gap.

A full theory should be able to consistently explain the occurrence of exchange coupling through Ge, Si, SiO, and Ge/Si barriers. It should clarify why AF coupling depends on the presence of Si in the barrier and on the total barrier thickness. To put such a wealth of observations under one common roof is a formidable task far beyond the scope of this study. A quantitative description depends on detailed information about the density and spectral distribution of resonant states in the semiconductor which is not accessible in our experiments. Here we wish to present a qualitative picture of resonant tunneling which may serve as a first step towards a thorough theoretical understanding of the phenomenon. We adopt the premise that tunneling through single-defect states mediates FM coupling and that AF coupling results from tunneling through defect pairs. This assumption is based on the fact that for a sufficiently high defect density the localized electronic states hybridize due to substantial overlap, and it is supported by general considerations[10] as well as by numerical simulations.[11] It is a nontrivial problem to determine the balance between single-defect (FM) and pair-defect (AF) coupling paths in a semiconductor with randomly distributed defects. For a simplified barrier structure we have shown[12] that a crossover from FM to AF coupling does indeed occur for a spacer thickness comparable to the average defect separation. The occurrence of a second AF coupled region in *a*-Si (Ref. 3) can also be explained if we assume that all coupling paths across an *even number* of defects mediate AF coupling. Heat-activated coupling is expected to result from the increased importance of phonon-assisted tunneling at higher temperatures and from the spread of the resonance energies around the Fermi level. At elevated temperatures the Fermi distribution function of the conduction electrons in the ferromagnets smears out and as a consequence more defect paths become accessible.

As an estimate of the actual defect densities we take values obtained by Knotek *et al.*[13] for room-temperature UHV-evaporated Si and Ge of $\rho = 3\times10^{19}$ and 1.5×10^{18} $eV^{-1}\,cm^{-3}$, respectively. Note that in *a*-Ge ρ is lower by more than an order of magnitude compared to the density in Si. Because we evaporate at $T=50$ K these values should be understood as a lower bound for the actual defect density which in the case of Si may exceed 10^{20} $eV^{-1}\,cm^{-3}$. Such a high defect concentration leads to substantial overlap between localized states due to an average defect separation of only 20 Å. Therefore we expect the balance between single-defect and multiple-defect tunneling to be important for the coupling sign across *a*-Si. On the other hand, the lower defect concentration in *a*-Ge corresponds to a larger separation of at least 60 Å between individual resonant states which prevents substantial hybridization. The observation that Ge/Si heterostructures exhibit a sign change of the coupling can be interpreted along the same lines. As soon as a high defect density is present in a part of the barrier (the Si layer) the remaining defects in the low-density region of the barrier will most probably find a partner defect with which to hybridize. For Ge/Si barriers a Si sublayer thickness of 4 Å is sufficient to provoke AF coupling while for the case of pure Si spacers[4] the crossover from FM to AF occurs around $d_{Si}=14$ Å. Experiment and the qualitative model both indicate that adding a high defect density region to the barrier is sufficient to change its coupling behavior.

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*Present address: IBM, T. J. Watson Research Center, Yorktown Heights, NY 10598.

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