Spin dynamics in the spacer of an interlayer-coupled Fe/$Fe_{0.57}Si_{0.43}$/Fe trilayer probed with nuclear resonant scattering of synchrotron radiation

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Using nuclear resonant scattering of synchrotron radiation, we investigated the origin of the interlayer coupling in Fe/$Fe_{0.57}Si_{0.43}$/Fe multilayers. The sensitivity of the technique to dynamical processes on a nanosecond timescale and the choice of different isotopes during the growth of the heterostructure allowed us to selectively probe the temperature dependence of the spin dynamics in the spacer layer. Combining these results with macroscopic magnetization measurements, it is shown that the static magnetic ordering in the $Fe_{0.57}Si_{0.43}$ spacer layer leads to the suppression of the interlayer coupling at low temperatures. Above 150 K, fluctuating magnetic Fe moments in the $Fe_{0.57}Si_{0.43}$ spacer with characteristic frequencies in the MHz regime give rise to a biquadratic interlayer coupling between the iron layers.

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I. INTRODUCTION

The mechanism of interlayer exchange coupling in magnetic multilayers has received considerable attention. By now, the nature of the bilinear interlayer coupling in 3*d* metallic multilayers is known to be well understood.[1–3] In contrast, the progress in the field of multilayers where the nonmagnetic metallic spacer layer has been replaced by a semiconductor is limited.[4,5] Due to interdiffusion at the semiconductor/metal interface, the formation of metalliclike spacers with unconventional coupling properties is observed. The major obstacle to understand the experimental results is the difficulty to characterize the magnetic properties of the buried spacer layer.

In the field of metal/semiconductor exchange-coupled systems, the Fe/Si system has been most actively studied. Experimental work on Fe/Si multilayers[6–8] was initiated shortly after the discovery of interlayer exchange coupling.[9,10] Initially, it was considered to be problematic that silicides tend to form at the Fe/Si interface. Soon, however, it was recognized that epitaxial Fe/FeSi multilayers with rather unique magnetic properties may be stabilized on various substrates.[11,12] Despite the major efforts to elucidate the nature of the interlayer coupling mediated by a silicide spacer, the reported results are still controversial and indicate a dramatic dependence of the exchange coupling properties on the exact stoichiometry and composition of the trilayer structure.[13–15] A more detailed study of the Fe/FeSi heterostructures, and of the local spin configuration in the silicide spacer layer in particular, may help to understand the nature of this complex coupling behavior.

From experimental point of view, however, it remains challenging to sensitively probe weak local ferromagnetism, paramagnetism, or induced spin polarization in thin films. In particular, experimental techniques that provide local spin information, originating from selective parts of the exchange-coupled system such as the interface or the spacer layer, are scarce. While x-ray resonant magnetic scattering allows one to selectively probe the microscopic magnetic properties of buried spacers, the technique requires a multilayer structure composed of different chemical species. Hyperfine interaction techniques such as perturbed angular correlation spectroscopy (PAC) and low energy muon spin rotation (μSR), on the other hand, cannot be performed without introducing foreign probe atoms in the region of interest.[16–18]

In this paper, we demonstrate how *nuclear resonant scattering of synchrotron radiation* offers a unique possibility to directly probe the local magnetic spin dynamics. Based on the Mössbauer effect, the technique benefits from its isotopic selectivity and provides direct access to the local magnetic structure in selective parts of a heterostructure.[19,20] For instance, by selectively enriching the spacer layer of an interlayer-coupled Fe/FeSi heterostructure with the nuclear resonant ^{57}Fe isotope, and the other layers with the nonresonant ^{56}Fe isotope, the actual spin structure in the ^{57}FeSi spacer can be directly investigated without altering the trilayer composition. Additionally, the technique is sensitive to dynamical processes on a time scale comparable to the lifetime of the nuclear excited state (144 ns in the case of ^{57}Fe). This will be an additional advantage as will be illustrated below.

II. EXPERIMENTAL

The viability of the nuclear resonant scattering technique for the direct study of the local magnetic spin behavior in buried spacers will be demonstrated for an Fe/$Fe_{0.57}Si_{0.43}$/Fe trilayer deposited onto a thick Au buffer.[21] This system, with its specific composition, is an important representative of the group of Fe/FeSi heterostructures with complex coupling properties. The multilayer structure was grown by molecular-beam epitaxy (MBE) onto a polished MgO(001) substrate. First, a ^{56}Fe(60 Å) base layer was deposited, followed by a 1500 Å thick Au(001) buffer.[22] Both layers were grown at 180 °C. Subsequently, an ^{56}Fe(40 Å)/$^{57}Fe_{0.57}Si_{0.43}$(34 Å)/^{56}Fe(20 Å) trilayer structure was evaporated and capped with a 45 Å thick Au(001) layer to protect the sample against oxidation. The bottom ^{56}Fe film of the trilayer was prepared in a two-stage mixed temperature deposition sequence, involving the evaporation of the first 10 Å at room temperature and the remaining 30 Å at 150 °C. The ^{57}FeSi

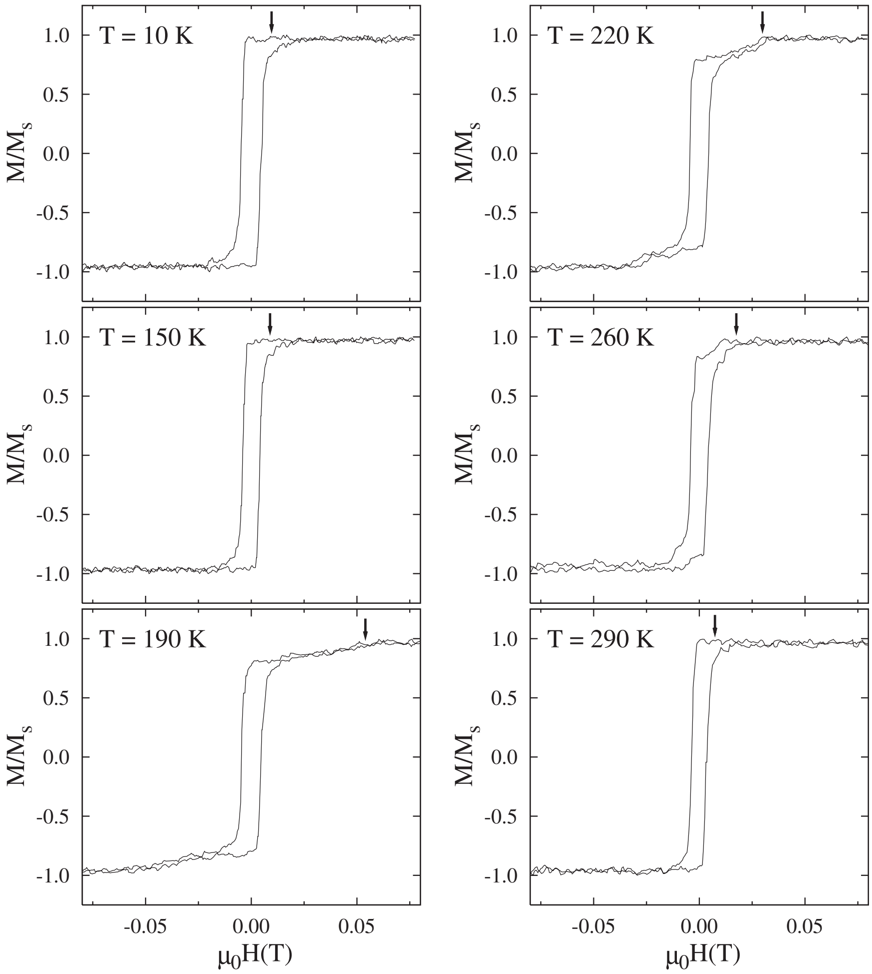

FIG. 1. Easy-axis magnetization loops of the $Fe/Fe_{0.57}Si_{0.43}/Fe$ structure as a function of temperature. The black arrows indicate the saturation field $\mu_0 H_{sat}$, defined as the external field value at which the magnetization reaches 95% of its saturation value, averaged over the increasing and decreasing field branch.

layer and the second ^{56}Fe film were deposited at 150 °C. The silicide spacer in the epitaxial trilayer adopts the metastable cubic CsCl crystal structure.[12,23–25] The epitaxial growth and the quality of the sample were monitored *in situ* with reflection high-energy electron diffraction (RHEED) measurements, and *ex situ* with x-ray diffraction. Conversion-electron Mössbauer experiments on samples with an identical composition, but where the ^{57}Fe and ^{56}Fe atoms were replaced by the natural Fe isotope, have shown that the magnetization in the different Fe layers is oriented in the plane of the multilayer.[21] This excludes the possibility of perpendicular anisotropy and stripe domains in the trilayer structure.[26]

The macroscopic coupling properties of the $Fe/Fe_{0.57}Si_{0.43}/Fe$ trilayer were investigated by means of easy-axis hysteresis loops, measured between 10 K and room temperature with a vibrating sample magnetometer. The evolution of the saturation field $\mu_0 H_{sat}$, which is a good measure for the interlayer coupling strength, is indicated with black arrows in Fig. 1. The small values of the saturation field at low temperatures ($\mu_0 H_{sat} \leq 10$ mT) indicate the absence of antiferromagnetic or biquadratic interlayer coupling. Above 150 K, however, the interlayer coupling appears abruptly and reaches a maximal strength around 190 K ($\mu_0 H_{sat} = 54$ mT). The high remanent magnetization relative to the saturation magnetization ($M_r/M_s = 0.78$) indicates the presence of a biquadratic interlayer coupling. Above 190 K, the biquadratic coupling falls off quickly with temperature. A complete overview of the temperature dependence of the interlayer coupling strength is given in Fig. 3(a). Both the biquadratic character of the interlayer coupling and its unusual temperature behavior cannot be explained by the conventional theories for exchange interlayer coupling. Since the coupling is mediated through the silicide spacer, a detailed study of the spacer layer properties is indispensable to understand the magnetic coupling behavior.

The nuclear resonant scattering experiments were performed at beamline ID-22N of the ESRF in Grenoble, France.[27] A 14.4 keV photon beam with a spectral width of 6 meV was reflected by the sample at a grazing angle of 4.050 mrad. The time dependence of the scattered intensity was recorded at 13 different temperatures in the range of 10–350 K, as shown in Fig. 2. Essentially, the time evolution of the radiation that is emitted from isolated ^{57}Fe nuclei after excitation by the synchrotron beam can be described by an exponential decay, determined by the lifetime of the excited nuclear level. In a magnetic environment, however, the energy levels of the ^{57}Fe nuclei are hyperfine-split and an oscillatory inter-resonance interference behavior will be superimposed onto the exponential time decay. This so-called quantum beat pattern can be clearly seen in the 10 K measurement and indicates the presence of a static magnetic ordering in the silicide spacer at low temperatures. With increasing temperature, the beat pattern fades out. At elevated

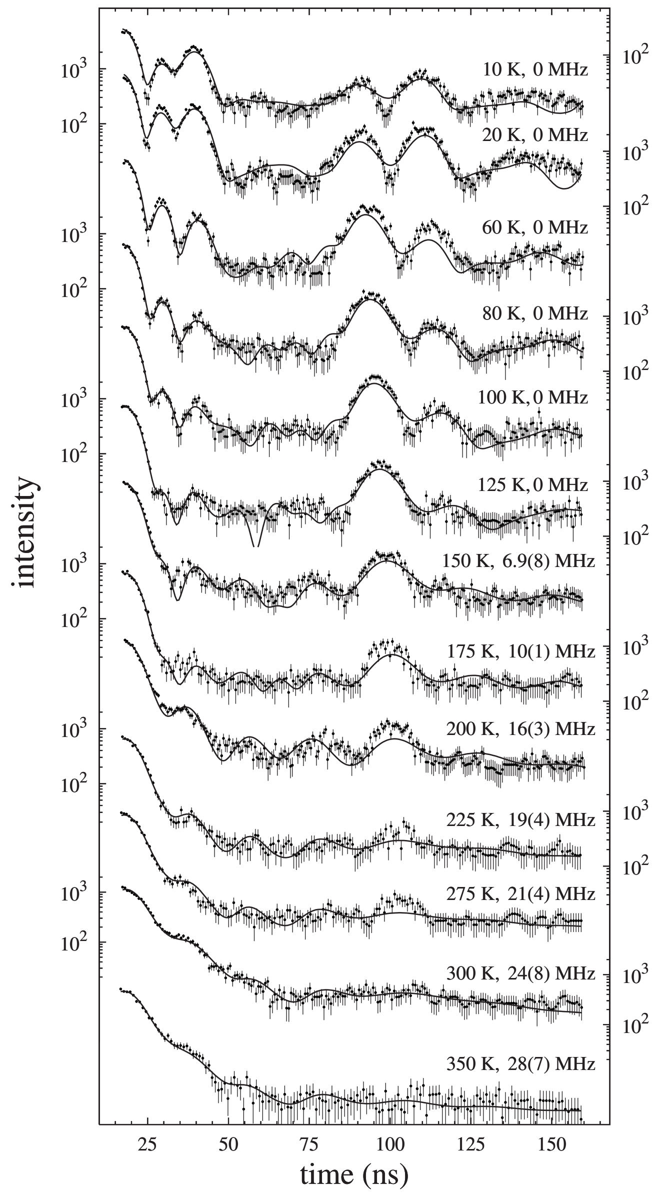

FIG. 2. Time spectra of the $^{56}Fe/^{57}Fe_{0.57}Si_{0.43}/^{56}Fe$ structure, as a function of temperature. The solid lines through the data are the fits obtained by the fitting program CONUSS (Ref. 29). The temperature and the weighted fluctuation frequency $\bar{\nu}$ are indicated for each spectrum.

temperatures, e.g., at 350 K, the time spectrum is close to an exponential decay which indicates the absence of static magnetic hyperfine interactions.

III. RESULTS AND DISCUSSION

Since the shape of the quantum beat pattern is very sensitive to slight variations of the hyperfine interaction parameters,[28] both the magnitude and the orientation of the ^{57}Fe hyperfine fields, and thus of the local magnetization in the isotopically tagged silicide spacer, can be determined from the spectra. The software package CONUSS (Ref. 29) was used to analyze the experimental time spectra. Table I shows the parameters obtained for the 10 K measurement. The pronounced beat pattern in the spectrum originates from four different magnetic hyperfine fields, which are associated with different local ^{57}Fe environments. All hyperfine fields are aligned with the Fe[100] easy axis. A Lorentzian distribution (full width at half maximum Γ) was included for each hyperfine field.

TABLE I. Low-temperature (10 K) fit parameters for the $Fe_{0.57}Si_{0.43}$ structure. For each spacer site, the magnetic hyperfine field B_{hf} and its corresponding Larmor frequency ν_L, the width Γ of the Lorentzian distribution, the weight, and the isomer shift δ relative to δ_1 are shown.

Site	B_{hf} (T)	ν_L (MHz)	Γ (T)	Weight	$\delta-\delta_1$ (mm/s)
1	−28.1(2)	22.2(2)	6.8(3)	0.34(2)	0
2	−10.6(1)	8.4(1)	4.6(5)	0.37(4)	0.4(2)
3	−5.4(3)	4.3(2)	4.5(3)	0.23(3)	0.9(2)
4	−24.2(1)	19.1(1)	2.6(9)	0.06(2)	0.1(1)

Up to 125 K, a consistent analysis of the spectra could be obtained by only slightly varying the fit parameters. At higher temperatures, the damped time spectra could not be analyzed by applying the model as described above. So far, we assumed the magnetic state in the silicide spacer to be static. However, since it is known that dynamical effects may cause a fading out of the magnetic quantum beat signal, we introduced the stochastic theory of Blume-Tjon[30] in our analysis. This dynamical model accounts for fluctuations of the magnetic hyperfine fields in the silicide spacer between two opposite in-plane directions, parametrized by a fluctuation frequency ν.

Assuming dynamical effects in the FeSi spacer, all spectra in Fig. 2 could be analyzed consistently, with only the four hyperfine field values and their fluctuation frequencies as free parameters. The magnitudes of the hyperfine fields were found to slightly decrease with increasing temperature. In accordance with the low-temperature measurements, the easy axis of the silicide spacer was chosen as the in-plane fluctuation direction. The weighted fluctuation frequency $\bar{\nu}$, defined as the weighted average of the fluctuation frequencies by which the hyperfine fields in the silicide spacer fluctuate, is indicated in Fig. 2. Within the error bars, the fluctuation frequencies for the measurements below 150 K were found to be zero. Above 150 K, the hyperfine fields—hence the magnetic moments in the silicide spacer—fluctuate with increasing frequency as the temperature increases. Around room temperature, the fluctuation frequency exceeds the characteristic Larmor frequency ν_L, which is, in the case of ^{57}Fe, proportional to the magnitude of the hyperfine field by a factor 0.79 MHz/T. For these very fast fluctuations, the ^{57}Fe nuclei experience a superparamagnetic environment, resulting in an exponential-like nuclear resonant resonantly scattered time spectrum.

Figure 3 compares the temperature dependence of the biquadratic interlayer coupling strength [panel (a)] with the temperature dependence of the spin dynamics in the silicide spacer [panel (b)]. At low temperatures, the static magnetic

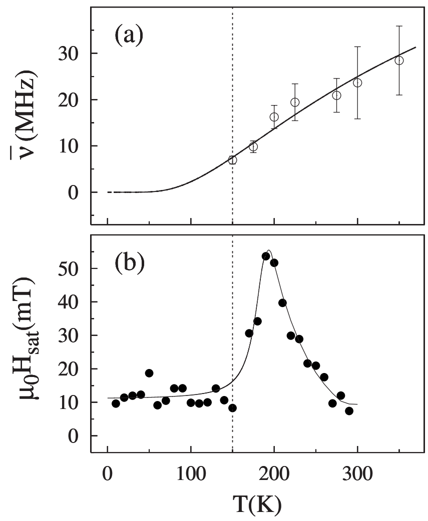

FIG. 3. Temperature evolution of (a) the interlayer coupling strength in the $Fe/Fe_{0.57}Si_{0.43}/Fe$ multilayer and (b) the weighted fluctuation frequency $\bar{\nu}$ of the magnetic fields in the silicide spacer. The solid line in panel (a) is a fit to the data, describing the temperature dependence of the spin fluctuation frequency by a thermal activation process with an activation energy E_a=31(3) meV. The solid line in panel (b) is a guide for the eyes.

ordering in the FeSi layer leads to the vanishing of the interlayer coupling. Above 150 K, the magnetic moments in the silicide spacer start to fluctuate in the MHz regime, thereby mediating the interlayer coupling between the two ferromagnetic layers. This dynamical effect can be well described by a thermal activation process with an activation energy E_a of 31(3) meV. Around 190 K, the paramagnetic entities in the spacer layer achieve a maximal efficiency in mediating the interlayer coupling. For higher temperatures, when the fluctuation frequency of the magnetic moments exceeds the characteristic Larmor frequency, the coupling strength drops again to very small values.

The observed appearance of the biquadratic coupling above 150 K is related to a perturbation of the static magnetic ordering in the silicide spacer at low temperatures. This agrees well with the experimental observations of Fullerton *et al.*,[31] where an enhanced remanence at 10 K was attributed to regions of the spacer layer that become ferromagnetic. A different point of view was taken in the work of Gareev *et al.*:[14] here, the temperature behavior of the interlayer coupling strength in Fe(50 Å)/$Fe_{0.56}Si_{0.44}$(26 Å)/Fe(50 Å) trilayers is related to a significant reduction in the number of carriers in the spacer near the Fermi level below 80 K. However, these results cannot explain resistivity measurements by our group,[21] showing a strong decrease in the resistivity upon cooling. Again, freezing of the magnetic moments in the silicide spacer can explain the temperature dependence of the interlayer coupling.

The idea of fluctuating spins in the spacer layer as a means to mediate an exchange coupling between two ferromagnetic layers has been suggested before for Fe/Cr(100) superlattices,[32] and is theoretically supported by the loose-spin model of Slonczewski.[33] Indeed, the overall temperature behavior of the biquadratic interlayer coupling in our work is similar to the temperature dependence of the biquadratic coupling discovered in the Fe/Cr multilayers. However, in contrast to the Fe/Cr work, the huge maximal value of the biquadratic coupling strength and its rapid decrease above 190 K cannot be fully reproduced by the loose-spin model. This is not surprising, since this model does not take into account the possibility of long-range spin ordering in the spacer layer at low temperatures. Clearly, theoretical models that fully account for these effects need to be further developed.

IV. CONCLUSIONS

In conclusion, we demonstrate that local spin dynamical processes can be probed directly on a nanosecond time scale by means of nuclear resonant scattering of synchrotron radiation. The isotopic selectivity and the time sensitivity of this technique offer a unique possibility to study the buried interfaces and spacers of hybrid multilayered systems, which is demonstrated in this paper for a relevant system, i.e., an $Fe/Fe_{0.57}Si_{0.43}/Fe$ trilayer epitaxially grown on a thick Au buffer layer. In order to clarify the origin and the unconventional temperature dependence of the macroscopic interlayer coupling that was discovered in the sample, we selectively investigated the microscopic magnetic state in the $Fe_{0.57}Si_{0.43}$ spacer layer as a function of temperature. Using nuclear resonant scattering of synchrotron radiation, we were able to demonstrate that high frequency dynamical effects in the buried FeSi spacer give rise to the biquadratic coupling at elevated temperatures. We are convinced that our approach opens the door to more systematic studies of the local magnetic properties in interlayer-coupled heterostructures with poorly understood macroscopic coupling properties. To a broader extent, the technique could be especially important for the study of all types of strongly coupled nanosystems involving high frequency dynamics, such as exchange biased systems and magnetic-tunnel junctions, which are an active field of research due to their broad range of technical applications.

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