

# Antiferromagnetic interlayer coupling in Fe/*c*-SiFe/Fe sandwiches and multilayers

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We report an *ab initio* study of the interlayer exchange coupling in Fe/*c*-FeSi/Fe sandwiches and Fe/*c*-FeSi multilayers. We perform several structural studies, which show the stability of the CsCl arrangement seen experimentally for the spacer. We study the effect of pinholes, interface roughness, or structural misconfigurations of the spacer on the sign and magnitude of the exchange constant  $J$ . We have extended Bruno's theory to account for the effect of different effective masses on the exchange constant, leading to an excellent description of its asymptotic behavior, as compared with experiments. The results presented here allow us to identify Fe/*c*-FeSi sandwiches as an example of a new class of magnetic multilayer devices, where the spacer material is semimetallic.

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## I. INTRODUCTION

Magnetic multilayer devices (MMD) can be roughly classified according to the conducting nature of the spacer material, which leads to markedly different behavior of the exchange-coupling constant between the magnetic layers  $J$ , as a function of the thickness of the spacer  $z$ .<sup>1</sup> The first class comprises those systems whose spacer is a metal. In the case,  $J(z)$  shows oscillatory behavior, whose period is typically of a few Angstrom; superimposed to these oscillations,  $J(z)$  also decays as  $1/z^2$ , becoming negligible after several tens of Angstrom. The second class is composed of those devices where the spacer is a semiconductor. Now,  $J(z)$  is frequently antiferromagnetic (AF) and its magnitude decreases exponentially with a decay length of at most two or three Angstrom. (Fe/*c*-FeSi) MMD stand out among all such structures as an example of a new class of MMD in which the spacer is semimetallic. Indeed, the exchange-coupling constant of these devices is always AF, but has a fairly large decay length, becoming negligible at spacer thicknesses more proper of a metallic than of a semiconducting spacer.<sup>2-10</sup> From a theoretical point of view, the class is particularly interesting, since it shares a number of features with critical phases in statistical physics. The unique behavior of  $J(z)$  might also make MMD in this class strongly attractive for manufacturers interested in the design of spin-polarized transport devices.

The details of growth and the crystal structure of (Fe/*c*-FeSi) MMD also make them especially suitable for manufacturing purposes. The silicide spacer, which possesses the CsCl structure (*c*-FeSi),<sup>7,9,11</sup> can be formed by interdiffusion of Fe and Si slabs of appropriate thicknesses grown epitaxially.<sup>10,12</sup> Low-energy electron-diffraction (LEED) and auger electron spectroscopy experiments, which have been applied successfully to study epitaxial Fe/Si/Fe (001), have shown that the perpendicular interlayer distance in this bcc-like structure remains constant at  $\approx 1.43$  Å, very close to the values for pure bulk bcc Fe.<sup>10</sup>

Despite the large amount of experimental information, and the plausible technological relevance of these MMD, no systematic *ab initio* studies of their structural stability, elec-

tronic structure, and related magnetic properties have been performed up to now to the best of our knowledge. Only the asymptotic region of exponentially decaying AF exchange has been qualitatively analyzed in a paper by de Vries *et al.*<sup>9</sup> using both Bruno's ideas<sup>1</sup> and a model by Shi and co-workers.<sup>13</sup> We report in this paper a theoretical study of the magnetic and structural properties of Fe/*c*-FeSi/Fe (001) trilayer (TD) and Fe/*c*-FeSi multilayer devices. We separate our results into two parts, depending on the thickness of the spacer. *Ab initio* methods are best suited to deal with devices having thin spacers. We have therefore used density-functional theory<sup>14</sup> to study the structural stability, the exchange constant  $J$ , and the magnetic-moment distribution of such devices. We have also studied how interstitial defects such as pinholes or interface roughness affect the sign and magnitude of  $J$ , by studying several mixed interfaces. We have used, on the other hand, a slightly modified version of Bruno's model<sup>1</sup> to study the exchange constant of TD with thick spacers. We find that the asymptotic behavior of  $J$  is determined by a flat band of the *c*-FeSi spacer, located at the  $M$  point in the Brillouin zone and not at the  $X$  point, as speculated previously.<sup>9</sup>

## II. DEVICES WITH THIN SPACERS

We have determined the spin-polarized electronic structure using a scalar-relativistic version of the  $k$ -space tight-binding (TB) linear muffin-tin orbital (LMTO) method<sup>15</sup> in the atomic spheres approximation. We have used two different versions of the generalized gradient approximation for the exchange and correlation potential: the Perdew-Wang<sup>16</sup> (PW) and the Langreth-Mehl-Hu (LMH),<sup>17</sup> and benchmark them against preliminary results obtained in the local spin-density approximation (LSDA).<sup>18</sup> We model the system according to the existing LEED structural analysis of molecular-beam-epitaxy-grown Fe/SiFe/Fe (001) sandwiches briefly mentioned above.<sup>10,12</sup> For both the experimental interlayer distance (1.43 Å) and that obtained from a total-energy minimization of *c*-FeSi bulk (1.42 Å) we obtain overall qualitative agreement, so that we discuss the results

obtained using the experimental distance, unless otherwise stated.

For trilayer structures, we repeat the sequence  $\text{Fe}_7/c\text{-Fe}_{(n-1)}\text{Si}_n/\text{Fe}_7$  in the (001) direction, with  $n = 1 - 6$ . Two successive trilayer sandwiches are then separated by enough layers of atomic empty spheres to have each individual trilayer decoupled from the rest. The electronic and magnetic structures are calculated using an increasing number of  $k$  points in the irreducible Brillouin zone. Convergence is obtained for 90  $k$  points.

We have first made structural analyses to test whether the experimental result of interdiffusion of a thin Si film into the Fe slabs, which creates the  $c\text{-FeSi}$  structure, can be understood in terms of the energetics of the different plausible atomic arrangements in the spacer. We have therefore computed the total energy for  $\text{Fe}_7/\text{Si}_3\text{Fe}_2/\text{Fe}_7$  TD with different layer configurations in the spacer, which modify the  $c\text{-CsCl}$  structure. We have checked, for instance, that the  $\text{Fe}_7/\text{Si}/\text{Fe}/\text{Si}_2/\text{Fe}/\text{Fe}_7$  arrangement is 300-mRy higher in energy than the  $\text{Fe}_7/\text{Si}/\text{Fe}/\text{Si}/\text{Fe}/\text{Si}/\text{Fe}_7$  configuration, and that in the former case the ferromagnetic (FM) alignment is slightly more stable than the AF one.

We have further performed molecular-dynamics simulations, relaxing the atomic positions in the  $z$  direction, for  $\text{Fe}_9/c\text{-}(\text{Fe}_1\text{Si}_2)/\text{Fe}_9$  and  $\text{Fe}_7/c\text{-}(\text{Fe}_2\text{Si}_3)/\text{Fe}_7$  TD using the SIESTA code.<sup>19</sup> SIESTA is a density-functional theory code based in pseudopotentials, which allows structure optimizations of the whole system that are beyond the capabilities of the all-electrons TB-LMTO method, particularly when surfaces are present. We have used a minimal basis set and the PW functional for exchange correlation.<sup>16</sup> Pseudopotentials were generated with the Troullier-Martins method,<sup>20</sup> with  $4s^13d^7$  and  $3s^23p^2$  valence configurations for Fe and Si, respectively. We have found that the forces in the theoretical bcc-like configuration just make the interstitial Fe atoms move slightly into the FeSi spacer, and expand a bit the Si-Si distances, the structure being very stable otherwise. The obtained exchange constants  $J$  are also similar within 10% to those obtained with the TB-LMTO method in the unrelaxed structure, therefore lending further support to our theoretical study.

We now discuss the behavior of  $J(z)$ , defined as the difference between the total energies of FM and AF alignments of the Fe slabs, for thin TD with perfect interfaces. Figure 1 shows the exchange constant as a function of the number  $n$  of Si monolayers, which is a measure of the thickness of the spacer. We always find a positive  $J$  of the order of some millirydberg which decreases with  $z$  in a nonmonotonic fashion, has a bump for  $n=3$ , and vanishes for  $n \approx 5-6$ . The bump might correlate with a small protuberance, which is seen experimentally at exactly the same spacer thickness.<sup>9</sup> It is not an artifact produced by the TB-LMTO code because we also find it when we use SIESTA. Its height depends, on the other hand, on the choice of the interlayer distances, becoming smaller when we use theoretical or relaxed values for them (see the inset in Fig. 1). The experimental peak seen for  $n=4$  seems to be somewhat sample dependent,<sup>21</sup> and we find no traces of it in our simulations. The exchange constant becomes negligible for spacer thicknesses larger than five or

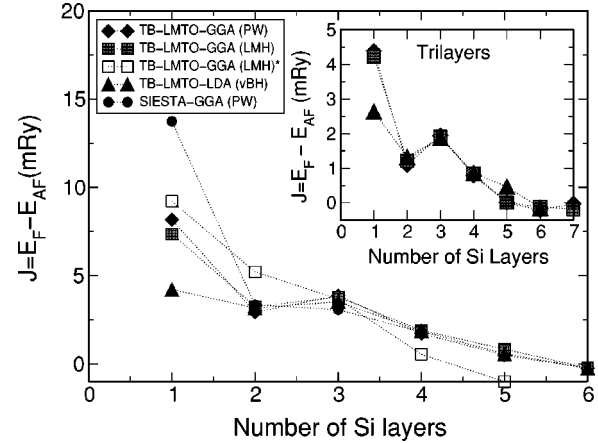


FIG. 1. Total-energy difference  $J = E_F - E_{AF}$  as a function of the number  $n$  of Si monolayers in the spacer for  $\text{Fe}_7/c\text{-Fe}_{(n-1)}\text{Si}_n/\text{Fe}_7$  (001) multilayers. The inset shows  $J$  for trilayers. Open squares denote values of  $J$  obtained using the theoretical lattice constant and the LMH functional.

six Si monolayers, which correspond to a distance of about 15 Å. Because the error bars coming from our calculations are of the order of the values obtained, we can only honestly conclude that  $J$  does not have any large oscillation for such thicknesses. Experiments actually show that for  $z$  larger than  $\approx 13$  Å,  $J(z)$  follows the asymptotic behavior of a semiconductor, with a large decay length of about 3.6 Å. This overall theoretical behavior for thin trilayer devices, that was also obtained within the LSDA,<sup>18</sup> is therefore in nice qualitative agreement with the experimental results by de Vries *et al.*<sup>9</sup>

We find that the exchange constant obtained for multilayers follows the same trend as that found for trilayers, its magnitude being roughly a factor of 2 larger (see inset of Fig. 1). This fact can be qualitatively explained in terms of simple Heisenberg-like physics, making use of an analogy with spin chains: the energy to flip a spin in a molecule composed of two atoms is half that required to do so in an infinite chain. We also find that for given  $z$ ,  $J$  slightly oscillates as a function of the thickness of the iron slabs, as seen in Co/Cu MMD.<sup>22,1</sup>

de Vries and co-workers observe FM-type contributions superimposed on top of the AF-like behavior in their Kerr hysteresis loops for TD with spacer thicknesses smaller than about 6–7 Å.<sup>9</sup> We present now results for several TD where we simulate a variety of atomic misconfigurations inside the spacer or at its interfaces with the iron slabs, because we wish to test whether they can induce such ferromagnetic coupling. The first one consists of the  $\text{Fe}_7/\text{Si}/\text{Fe}/\text{Si}_2/\text{Fe}/\text{Fe}_7$  TD, which was introduced a few paragraphs before. It is supposed in this case that the Si layers have not diffused completely into the Fe slabs to form the  $c\text{-FeSi}$  structure. We find that  $J$  is ferromagnetic, with  $J = -3.49$  mRy. We present next the case of dense arrays of thin pinholes or rough interfaces for TD with  $n = 1, 2$ , and 3 (e.g., up to about 8 Å). We simulate them by doubling the cross section of our unit cell, which now contains two atoms per atomic plane. For  $n = 1$ , we exchange one atom in the Si layer by another in the last layer of one of the iron slabs. We create this way a periodic

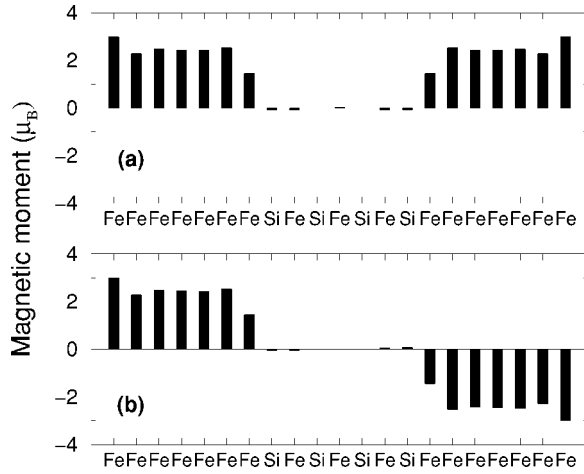


FIG. 2. Distribution of magnetic moments for  $\text{Fe}_7/c\text{-Fe}_3\text{Si}_4/\text{Fe}_7$  (001) sandwiches in (a) ferromagnetic and (b) antiferromagnetic configurations. Filled and empty bars correspond to magnetic moments in Fe and Si, respectively.

array of thin pinholes, which leads to a strongly ferromagnetic  $J$  of  $-73$  mRy (about 30 times larger in magnitude than the corresponding one for a pure  $c\text{-FeSi}$  spacer). We subsequently look at two different configurations for  $n=2$ . In the first one, the Fe and Si atoms are arranged again in such a way that there are thin bridges along the spacer as before. The exchange constant is then also negative, with a value of  $-33$  mRy. We make the second configuration such that one of the Si layers has no iron atoms, which means that one of the interfaces is rough while the other one is perfect. We find in this largely asymmetric case that  $J$  is equal to  $-0.7$  mRy, which is of the same order of magnitude as its antiferromagnetic counterpart. We finally present for  $n=3$  the case where both interfaces are rough while the central Si layer has no defects. We find that the exchange constant is again largely ferromagnetic,  $J = -5.1$  mRy. We therefore conclude that atomic misconfigurations generically give rise to very strong ferromagnetic couplings. A comparison of our results with those presented by de Vries<sup>9</sup> lets us infer some further conclusions about the structure of these sandwiches, namely, (a) it is very likely that for TD with thicknesses smaller than about  $3 \text{ \AA}$  there is a process of diffusion of iron into the Si layer (or vice versa), so that the Si layer is disrupted; (b) for TD with spacer thicknesses larger than about  $4 \text{ \AA}$ , diffusion takes place but leads to the formation of a  $c\text{-FeSi}$  spacer with interfaces of high quality.

We come to comment now on the profiles that we find for the magnetic moments, which we show in Fig. 2 for TD with  $n=4$ . First, we have that the  $c\text{-FeSi}$  spacer displays tiny magnetic moments of order  $0.05\mu_B$  at the interface with the iron slabs, with whom they couple antiferromagnetically. Second, the absolute values of these magnetic moments are almost identical for both alignments. Third, iron atoms in the vicinity of the interface with the spacer have reduced magnetic moments ( $\sim 1.46\mu_B$ ) due to the hybridization of their orbitals with those belonging to  $c\text{-FeSi}$  atoms. Surface effects, on the other hand, induce enhanced magnetic moments at the Fe external layers ( $M \sim 2.97\mu_B$ ). Finally, magnetic

moments at the center of the Fe slabs slightly oscillate around the bulk value ( $\sim 2.20\mu_B$ ). We find similar behavior for multilayer devices, apart from the obvious fact that no surface effects exist for them.

### III. DEVICES WITH THICK SPACERS

The asymptotic behavior of the exchange constant can be better understood by using a phenomenological theory developed by Bruno.<sup>1</sup> The model assumes that electrons in the ferromagnetic and spacer slabs are free. The corresponding dispersion relations are parabolae with different band edges and effective masses. The exchange constant

$$J(z) = -\frac{\hbar^2}{2\pi^2 m_{sp}} \times \text{Im} \left[ \int_0^\infty dx \frac{x(k_F+x)(2k_F+x)\Delta r^2 e^{-2(k_F+x)z}}{1-2\bar{r}^2 e^{-2(k_F+x)z} + r_\uparrow^2 r_\downarrow^2 e^{-4(k_F+x)z}} \right], \quad (1)$$

where  $\Delta r = (r_\uparrow + r_\downarrow)/2$ ,  $\bar{r} = (r_\uparrow - r_\downarrow)/2$ , can then be written in terms of the following effective reflection coefficients:

$$r_{\uparrow,\downarrow} = \frac{(k_F+x)i - k_{\uparrow,\downarrow}}{(k_F+x)i + k_{\uparrow,\downarrow}}, \quad (2)$$

where

$$k_F = \sqrt{\frac{m_{sp}}{m} E_{g,sp}},$$

$$k_{\uparrow,\downarrow} = \sqrt{\frac{m_{\uparrow,\downarrow}}{m} (E_{g,sp} - E_{g,\uparrow,\downarrow}) - \frac{m_{\uparrow,\downarrow}}{s_{sp}} (k_F+x)^2} \quad (3)$$

are effective wave vectors and  $E_{g,i}$ ,  $m_i$  are the band edges and effective masses of the parabolae under scrutiny in the spacer and the ferromagnet, respectively. Equation (1) is usually evaluated at the saddle point, which is equivalent to evaluating its integrand exactly at the Fermi energy. A thorough study of the Fermi surface then allows us to identify the relevant callipers and hence determine the decay length of  $J(z)$ . For a semimetallic spacer the Fermi surface and its relevant callipers vanish, though, naively providing an infinite decay length. Equation (1) must therefore be evaluated exactly. This is what we have actually done for (Fe/ $c\text{-FeSi}$ ) MMD in the present work. To do so, we have previously, and independently, obtained all band edges and effective masses from band-structure calculations. Such a procedure has therefore left us no room for fitting.

We plot in the middle panel of Fig. 3 a plot of the bands of the spacer in the neighborhood of the Fermi energy, with the relevant one drawn as a dashed line. Such a band has, on the one hand, an extended flat portion around the  $X$  point and slightly above the Fermi energy, which gives rise to a strong peak in the density of states, as shown in the right panel of the figure. The band also has an edge at the  $M$  point located almost exactly at the Fermi energy, which gives rise to the three-dimensional band edge in the density of states shown in the right panel. This is the feature that actually accounts for the semimetallic character of the spacer. Since the corre-

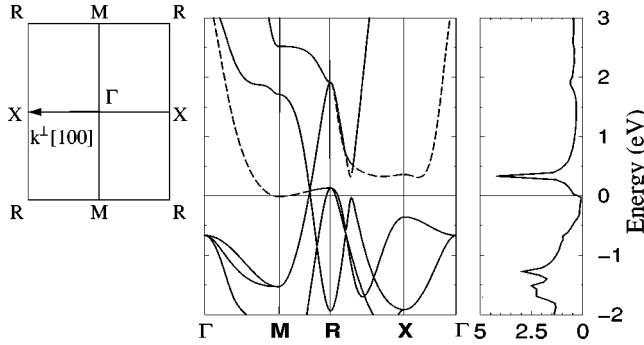


FIG. 3. Details of the band structure (central panel) and total density of states in units of states/electron volts (right panel) of bulk *c*-FeSi. The band drawn with a dashed line has a band edge at the *M* point and a large flat section around the *X* point. The two-dimensional Brillouin zone is plotted in the left panel.

sponding piece of the Fermi surface shrinks to zero here, the calliper vanishes. There are also a few bands crossing the Fermi energy in the MR direction, but their contribution to the density of states is very small.

Several authors<sup>9,13</sup> have speculated that the main contribution to  $J(z)$  should come from the *X* point. But a brief inspection of the middle panel shows that such a contribution must be oscillatory, due to the combined effect of the semi-conducting character of the band and its negative effective mass at such a point. This suspicion is indeed confirmed after evaluating numerically Eq. (1). Having discarded the *X* point as the source of the behavior of  $J(z)$ , we subsequently turn our attention to the *M* point. The overall behavior of the exchange constant is in this case in excellent agreement with the experimental results available to us.<sup>9</sup> Moreover, we obtain exactly the same value of the decay length (3.6 Å) as in experiments. The layout of Fig. 4 purposely mimics that of Fig. 5 in Ref. 9 with which it should be compared. Incidentally, we find that the quantitative and even the qualitative behavior of  $J(z)$  depends very sensitively on the exact position of the edge of the band in the spacer. Shifting the edge downward by a few tens of meV makes  $J(z)$  behave in a metallic fashion.

MMD with semimetallic spacers have some analogies with critical phenomena,<sup>23,24</sup> since they can be viewed as the critical point between an ordered phase with gapless excitations and power-law decaying correlation functions (e.g., MMD with metallic spacers) and a disordered one with a gap in the excitation spectrum and exponentially decaying correlation functions (e.g., MMD with semiconducting spacers). The critical point is then characterized by the divergence of a

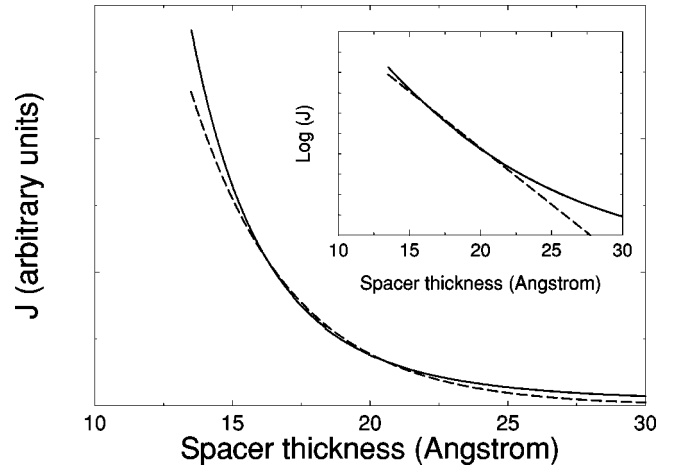


FIG. 4. Asymptotic behavior of the exchange constant as a function of the thickness of the spacer. Solid line is the result from our amended version of Bruno's theory; dashed line is a fit to  $e^{-z/3.6}$ . The inset shows the logarithm of  $J$  as a function of  $z$  to ease comparison with experiments.

suitably defined coherence length, which for MMD is the inverse of some relevant calliper at the Fermi surface.

#### IV. CONCLUSIONS

To summarize, we have identified a new class of MMD, which we term critical since they share some features with critical phases in statistical physics. These MMD are characterized by their spacer, which is semimetallic. Critical MMD have exponentially decaying antiferromagnetic exchange constants  $J(z)$  with large decay lengths.<sup>25</sup> We have found that (Fe/*c*-FeSi) MMD are examples in this class. We have then performed a thorough study of their structural and magnetic properties, using density-functional theory methods and also an amended version of Bruno's phenomenological theory. Our work is in excellent agreement with known experimental results.

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