Search for stable ferromagnets among A^{IV}/Fe digital alloys (A^{IV} = Si, Ge) using first-principles calculations

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Using first-principles electronic structure calculations we investigate the existence of stable ferromagnets among the A^{IV}/Fe digital alloys (A^{IV} = Si,Ge), modeled as periodic sequence of Fe monolayers in the A^{IV} host. Total-energy calculations and the magnetic force theorem are exploited for accurate determination of the magnetic ordering. To estimate the critical temperatures, Monte Carlo simulations are employed, while the renormalization-group analytical expressions are applied to assess the impact of the interlayer exchange on the critical temperature values. According to our results, among the systems under consideration only the Ge-based alloys feature a stable ferromagnetic ordering at nonzero temperature. The critical temperatures of these systems were found to be strongly dependent on the underlying crystal structure.

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I. INTRODUCTION

Digital alloys (DAs) of magnetic transition metals (TMs) with nonmagnetic semiconductors have, for many years, attracted substantial attention.[1–6] A DA is an example of δ doping,[7–9] which represents a periodic sequence of the ultrathin TM layers embedded within a semiconductor host. Such a way of preparation allows tailoring the magnetic properties of the samples freely by changing the TM atoms' concentration or semiconductor spacer thickness and can yield to high Curie temperature.[8,9] In combination with high-mobility modulation-doped heterostructures[10–13] ferromagnetic (FM) DAs would become attractive for spin-transport applications,[14] provided that they remain magnetically ordered above room temperature.

Si/Mn DAs, being the most natural candidates for the integration of spintronics with standard Si electronics, have attracted attention since 2008 when room-temperature ferromagnetism was observed in these systems.[15] However, the nature of this FM ordering remains unclear. Complex atomic structure of real DAs, close to amorphous,[15] with smeared TM layers,[6,16] clusters, and secondary phases,[3] hinders direct comparison with *ab initio* calculations. From the theoretical side, however, even the simple model of atomically flat TM monolayers (MLs) can be very useful.[17–20] For instance, *ab initio* calculations predict A^{IV}/Mn DAs (A^{IV} = Si,Ge), in which Mn MLs formed substitutionally, to be half metallic ferromagnets.[21–29]

Recently, the critical temperatures of the intralayer magnetic ordering in A^{IV}/Mn DAs were estimated for the case of easy-axis anisotropy by means of Monte Carlo (MC) simulations.[27,28] The most realistic estimates, based on the disordered local moment (DLM) picture,[30,31] yielded temperature values well below room temperature (≈200 K) for FM DAs, contrary to expectations.[23] Nevertheless, the results are encouraging, since some *ferrimagnetic* alloys were predicted to be ordered up to room temperature,[27] revealing the potential of the digital doping for fabrication of magnetic semiconductors with high enough transition temperatures.

If a DA has the easy-plane magnetic anisotropy, the issue of the long-range ordering becomes a delicate problem. In this case, the system will feature no magnetic order at any temperature except 0 K,[28] since, according to the Mermin-Wagner theorem, the ordering will be destroyed by long-range spin fluctuations.[32,33] In the limit of strong enough easy-plane anisotropy such systems were shown to undergo relatively sharp phase change, so-called Berezinskii-Kosterlitz-Thouless (BKT) transition.[34–36] The crucial role in the stabilization of the magnetic ordering across this transition is played by the interlayer exchange coupling.[37] Two questions arise in this context: (i) Do other A^{IV}-based FM DAs, except A^{IV}/Mn, exist? (ii) What are the critical temperatures for the easy-plane anisotropy case provided the interlayer exchange coupling is taken into account?

To explain these problems, we performed a detailed *ab initio* study of the magnetic ordering in A^{IV}/Fe DAs, with A^{IV} = Si,Ge. In the spirit of our previous works,[27,28] we investigated magnetism depending on the structure of the DA considering two cases: the substitutional Fe ML and the interstitial Fe ML. Since epitaxial growth is a nonequilibrium process, both structural situations are in principle possible and should be considered. Indeed, according to the experimental data on A^{IV}/TM DAs, available mostly for the Si/Mn DAs, the TM dopants in Si were found in both substitutional and interstitial positions, depending critically on the growth technique used. For instance, under suitable growth conditions, Mn can form a two-dimensional rowlike structure, which could be useful for achieving δ layer doping of silicon by manganese.[38] In this study, Mn was found to be incorporated into Si as an interstitial dopant in good agreement with recent first-principles calculations.[39–41] Meanwhile, molecular-beam epitaxy produces samples with mostly substitutional Mn in

the Si matrix.[15] In our work, for each morphological scenario, the electronic structure is calculated for numerically relaxed atomic positions. Next, the magnetic ordering is investigated based on the total-energy calculations and the analysis of calculated parameters of the interatomic exchange coupling. The main attention was paid to the search for stable ferromagnets with high Curie temperatures. Additionally, the issue of the interlayer exchange coupling effect on the transition temperatures was addressed.

II. METHODOLOGY

A. Computational details

To simulate the A^{IV}/Fe DAs (001)-oriented diamond-structure tetragonal supercells containing 7 MLs of A^{IV} and one substitutional (Fe_S) or interstitial (Fe_I) iron ML were used. The moduli of the basis vectors **a** and **c** were equal to a_0 and $2a_0$, respectively, where $a_0 = 5.46$ Å (5.77 Å) is the optimized lattice constant of bulk Si (Ge). The chosen spacer thickness is big enough in the sense that it is close to the minimal experimental thickness of 10 MLs,[1] while it is small enough to explore a sizable effect of the interlayer exchange coupling on the critical temperature.

For the electronic structure calculations we employed density functional theory within the generalized gradient approximation[42] to the exchange-correlation potential. To optimize the atomic positions the projector augmented-wave method[43] in the VASP implementation[44,45] was used. The structural relaxations were performed until the forces on each atom were less than 10^{-2} eV/Å. The energy cutoff for the plane-wave expansion of wave functions was set to 500 eV and a Γ-centered special k-point grid of $6 \times 6 \times 3$ was used to sample the Brillouin zone.

For the relaxed geometry further *ab initio* calculations were performed using the Korringa-Kohn-Rostoker method within the atomic sphere approximation to the crystal potential.[46–48] We took an angular momentum cutoff of $l_{max} = 3$ for the Green's function and a k-point mesh of $12 \times 12 \times 6$ for the Brillouin-zone integration.

In order to circumvent the well-known shortcoming of the generalized gradient approximation, which cannot correctly describe the band gap in Si and Ge and even yields metallic behavior of bulk Ge, the Si and Ge valence p states were treated with an additional Coulomb potential U.[49] As a result, calculated electronic spectra of the Si and Ge were in good agreement with the band structures obtained with the GW approximation and recent hybrid functional method, which are known to reproduce well experimental spectra of semiconductors.[29,50,51] With $U_{Si} = 1.38$ eV for Si and $U_{Ge} = 1.53$ eV for Ge the calculated band gap in Si and Ge was found to be close to the experimental values of 1.12 and 0.65 eV, respectively.

Our approach to determine the magnetic order involves both total-energy calculations and the estimation of the interatomic exchange coupling parameters J_{ij} within the magnetic force theorem.[52] Both types of computations were performed for FM, AFM (i.e., antiferromagnetic) and DLM reference states. The latter models a system in the paramagnetic phase and is simulated within the framework of the coherent-potential approximation.[53,54] The reason to consider this state is twofold. First of all, it allows us to approach the finite-temperature properties of the system. Second, if the total energy in a DLM calculation is lower than that of corresponding FM calculation, one can safely assume the ground state of the system to not be FM.

Performing magnetic force theorem calculations, the noncollinear spin configurations are always involved, since the J_{ij} are computed through the changes of the energy upon infinitesimally small deviations of the local spins from the quantization axis. This opens a way to judge the stability of the particular spin structure with respect to deviation from the collinear order without performing explicit self-consistent electronic structure calculations in the noncollinear mode. The exchange coupling constants can be used to obtain spin-wave spectra by the diagonalization of the Heisenberg Hamiltonian,

$$H = -\sum_{i \neq j} J_{ij} \mathbf{e}_i \mathbf{e}_j - \Delta \sum_i \left(e_i^z\right)^2, \qquad (1)$$

where i and j label magnetic atoms, $\mathbf{e}_i$ is a unit vector in the direction of the magnetic moment of the ith atom, and Δ is the magnetic anisotropy parameter, which is positive (negative) for the case of the easy-axis (easy-plane) anisotropy. In the framework of our scalar-relativistic approach we accounted for the anisotropy of the system phenomenologically. Since the value and even the sign of the magnetic anisotropy energy Δ is not known for A^{IV}/Fe DAs, it was used as a parameter in our simulations. Note that the anisotropy term can be neglected when computing magnon spectra, since the anisotropy modifies them weakly, being much smaller than the typical exchange energy. However, it is essential for the estimations of critical temperatures. The latter were found by means of the MC simulation using the model Hamiltonian, Eq. (1). It is well known that the dependence of the critical temperature on the value of the magnetic anisotropy parameter Δ has a weak logarithmic nature.[55] Therefore the variation of Δ in a physically reasonable interval does not change the qualitative picture of the phase transition. The details of our MC scheme can be found in Refs. 27 and 56. Note that the scheme can be reliably applied only to the case of easy-axis anisotropy.

B. Renormalization-group analytical expressions

Apart from the MC method, to estimate the critical temperatures, in particular in the case of easy-plane anisotropy, we resorted to the renormalization group (RG) analysis expressions.[37,57,58] For the reader's convenience, the equations are explicitly reproduced below.

We begin with the Heisenberg model for a ferromagnet with a weak interlayer coupling and easy-axis anisotropy, considering only nearest-neighbor interaction,

$$H = -\frac{J}{2} \sum_{i,j} \mathbf{S}_i \mathbf{S}_j - \frac{J'}{2} \sum_{i,j} \mathbf{S}_i \mathbf{S}_j - J\zeta \sum_i \left(S_i^z\right)^2. \qquad (2)$$

In this equation $\mathbf{S}_i$ is a vector of the spin moment of the ith atom, $J > 0$ is the in-plane nearest-neighbor exchange parameter, $J' = \alpha J$ is the nearest-neighbor interlayer coupling parameter ($\alpha \ll 1$), j denotes nearest neighbors within a

single plane and different planes in the first and second sum, respectively; $\zeta > 0$ is a single-ion anisotropy parameter. Since the formulation of the Heisenberg model in Refs. 57 and 58 differs from that in our implementation of magnetic force theorem and the MC method [cf. Eqs. (1) and (2)], redefining the parameters is necessary.

According to Ref. 57, the system described by the Hamiltonian Eq. (2) undergoes magnetic phase transition at $T_c = 4\pi J S^2 t_c$, t_c being defined as

$$t_c \approx \left[\ln\frac{8\pi t_c S}{\theta(f_c,\alpha_c)} + 2\ln\frac{1}{t_c} + 1\right]^{-1}, \quad (3)$$

where S is the spin of the magnetic ion, $f = 2\zeta(1 - 1/2S)$, $\theta(f_c,\alpha_c) = f_c + \alpha_c + \sqrt{f_c^2 + 2\alpha_c f_c}$; at that $f_c/f = (\alpha_c/\alpha)^2 = t_c^2$, and the subscript c means that the value is taken at $t = t_c$. To estimate T_c for the case of zero interlayer exchange ($\alpha = 0$), an alternative expression is used:

$$t_c \approx \left[\ln\frac{4\pi t_c S}{f} + 4\ln\frac{1}{t_c} + 1\right]^{-1}. \quad (4)$$

In the case of the easy-plane ferromagnet, it is convenient to replace $-J\zeta$ in Eq. (2) by $D = 4Jd/(1 - 1/2S)$ (condition $J \gg D > 0$ must be fulfilled).[58] Then, the BKT transition occurs at temperature $T_{\mathrm{BKT}} = 2\pi J S^2 t_{\mathrm{BKT}}$, with

$$t_{\mathrm{BKT}} \approx \left[\ln\sqrt{\frac{2\pi t_{\mathrm{BKT}} S}{d}} + 2\ln\frac{2}{t_{\mathrm{BKT}}} - \frac{1}{2}\right]^{-1}. \quad (5)$$

Once the interlayer exchange is switched on (i.e., $\alpha \neq 0$), the nonzero $t_c = T_c/(2\pi J S^2)$ appears,

$$t_c \approx \left[\ln\sqrt{\frac{2\pi t_c S}{d}} + 2\ln\frac{2}{t_{\mathrm{BKT}}} - \frac{12.25}{\ln(d/\alpha)} - \frac{1}{2}\right]^{-1}. \quad (6)$$

The latter relation is qualitatively valid for $\alpha \ll d$. The estimations were performed in a quantum regime, i.e., $T_c/JS \ll 32$ is assumed to hold.[57]

III. RESULTS AND DISCUSSION

A. Magnetic ordering

First we discuss of the *intralayer* magnetic ordering in $A^{\mathrm{IV}}/\mathrm{Fe_S}$ DAs. Figure 1(a) shows the exchange coupling parameters calculated for the FM and DLM reference states in $A^{\mathrm{IV}}/\mathrm{Fe_S}$ DAs. The exchange interactions depend strongly on

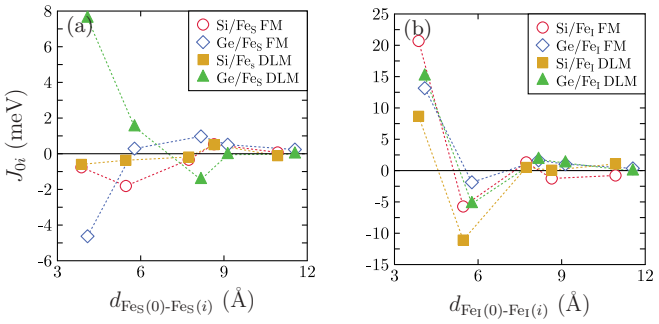

FIG. 1. (Color online) Exchange coupling constants J_{0i} (in meV) as a function of the Fe-Fe distance for $A^{\mathrm{IV}}/\mathrm{Fe_S}$ (a) and $A^{\mathrm{IV}}/\mathrm{Fe_I}$ (b) DAs in FM (empty markers) and DLM (filled markers) states.

TABLE I. The total-energy differences, $E_{\mathrm{AFM}} - E_{\mathrm{FM}}$ and $E_{\mathrm{DLM}} - E_{\mathrm{FM}}$, and FM state local magnetic moments m_{loc} as a function of host and type of ML. Energies are in meV per Fe atom, the magnetic moments in μ_{B}.

	Si/Fe$_S$	Si/Fe$_I$	Ge/Fe$_S$	Ge/Fe$_I$
$E_{\mathrm{AFM}} - E_{\mathrm{FM}}$	21.22	40.82	74.13	148.44
$E_{\mathrm{DLM}} - E_{\mathrm{FM}}$	116.81	−3.50	27.29	57.56
m_{loc}	1.66	1.84	2.50	2.27

the host: for the FM state in Si/Fe$_S$ DA the second-neighbor exchange dominates, $J_{02} = -1.8$ meV, while in Ge/Fe$_S$ DA the nearest-neighbor exchange stands out clearly with $J_{01} = -4.6$ meV. In both cases the leading exchange parameters are negative, while differences $E_{\mathrm{AFM}} - E_{\mathrm{FM}}$ and $E_{\mathrm{DLM}} - E_{\mathrm{FM}}$ are positive (see Table I), suggesting the ground state is probably noncollinear. In the DLM reference state of Fe$_S$ MLs the exchange constants differ from those obtained for the FM case. For the Si/Fe$_S$ DA the magnetic structure is dominated by the competition between interactions, described by J_{0i}, $i = 1, \ldots, 4$, while for the Ge/Fe$_S$ DA exchange parameters point clearly to the stable FM alignment. The large deviation in the exchange constants of FM and DLM reference states in Ge/Fe$_S$ DA indicates a substantial difference between the electronic structures of both reference states. This difference of the exchange parameters can be attributed to the temperature dependence of the effective interatomic exchange interactions.

In the case of the interstitial Fe ML in Si and Ge hosts, the FM nearest-neighbor exchange interactions dominate introducing a strong trend towards FM ordering, since $J_{01} > 0$ [Fig. 1(b)]. The interactions with atoms in the other coordination spheres are much weaker. As shown in Table I, the FM state energy is lower than that of the AFM state for both alloys. However, for the Si/Fe$_I$ DA the energy of the DLM state is lower, which points to a possible instability of the FM order. To gain deeper insight into the spin structure of the A^{IV}/Fe DAs the adiabatic spin-wave spectra were calculated. For the Si/Fe$_I$ DA the obtained dispersion curve shows that, despite that the FM state is energetically favored compared to the AFM one, the ground state turns out to be noncollinear [see the solid red curve shallow minima with negative energies in Fig. 2(a)]. However, with increasing the spin disorder, simulated via the DLM method, this noncollinear spin structure develops into the collinear AFM[110] structure, as shown by mustard dot-and-dash line. In the case of the Ge/Fe$_S$ DA, the FM ordering is stabilized upon increasing the spin disorder, which is manifested in the relocation of the dispersion curve minimum from the $\bar{\mathrm{M}}$ point to the $\bar{\Gamma}$ point. As far as the Ge/Fe$_I$ DA is concerned, it appears to be a stable ferromagnet with an exchange coupling strength weakly depending on the spin disorder (cf. dispersion curves for FM and DLM states). The local moments in the FM state are 1.84 μ_{B} and 2.27 μ_{B} for Si/Fe$_I$ and Ge/Fe$_I$ DAs, respectively. The difference between the magnetic moments in the Si and Ge hosts can be explained by the degree of the hybridization between the Fe 3*d* orbitals and *sp* band of the semiconductor. In the Si matrix, the interatomic distances are shorter then in Ge. This leads to a stronger *d*-*d* and *sp*-*d* hybridization. While the majority spin channel of Fe is mostly occupied, the number of 3*d* minority

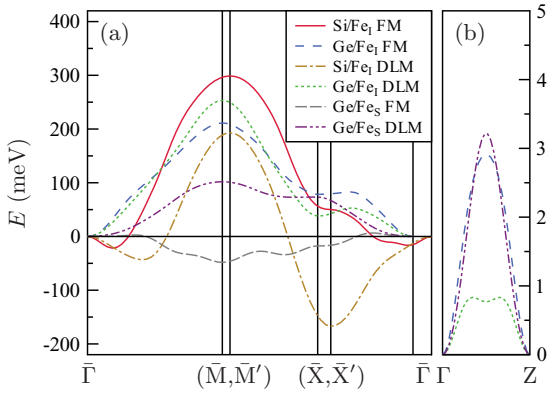

FIG. 2. (Color online) Magnon dispersions for A^{IV}/Fe DAs, as calculated in FM and DLM states over the first two-dimensional (2D) Brillouin zone (a) and three-dimensional (3D) Brillouin zone (b). The high-symmetry points have the following coordinates: $\bar{\Gamma}$: (0, 0); $\bar{M}$: (π/a_0^{Ge}, π/a_0^{Ge}); $\bar{M}'$: (π/a_0^{Si}, π/a_0^{Si}); $\bar{X}$: (π/a_0^{Ge}, 0); $\bar{X}'$: (π/a_0^{Si}, 0); Γ: (0, 0, 0); and Z: (0, 0, $\pi/2a_0^{Ge}$). Note that spectra were calculated using supercells with $|\mathbf{a}| = a_0/\sqrt{2}$.

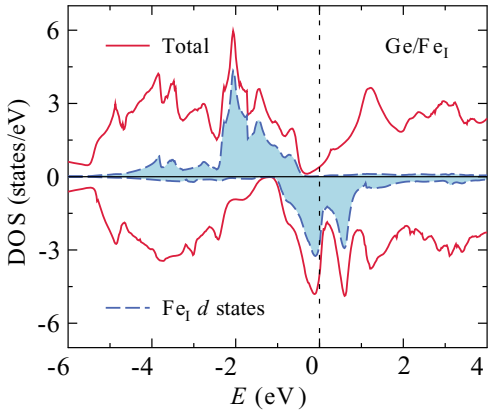

FIG. 3. (Color online) Calculated spin-polarized total and partial d DOS for the Ge/Fe$_I$ DA in the FM state. Majority DOS is plotted as positive values, minority DOS as negative ones. The vertical dashed line indicates the Fermi energy.

electrons is strongly affected upon this hybridization. As the result, the Fe magnetic moments in the Si are substantially smaller then in Ge.

Let us now discuss of the *interlayer* magnetic ordering in the DAs with a stable intralayer ferromagnetism. The most simple and at the same time reliable approach for the investigation of the interplane magnetic ordering is provided by the computation of the spin-wave spectra over the 3D Brillouin zone. Using this approach we found that in both Ge-based alloys under consideration the neighboring Fe MLs are ferromagnetically coupled to each other, as follows from the results of calculations, performed for DLM and FM (only for Ge/Fe$_I$) reference states [see Fig. 2(b)].

Thus among the considered compounds only the Ge/Fe$_S$ and Ge/Fe$_I$ DAs are stable ferromagnets at nonzero temperature. Therefore, further, we restrict our discussion to these two systems because of possible applications in spintronic devices. The Ge/Fe$_I$ DA will be discussed below in more detail than the Ge/Fe$_S$ one, because the former alloy possesses a FM ground state.

B. Electronic structure of Ge/Fe$_I$

In Fig. 3 we show the density of states (DOS), resolved into majority and minority spin components (positive and negative values, respectively), for the FM Ge/Fe$_I$ DA. According to our calculations, the Ge/Fe$_I$ DA is metallic, which is typical for the A^{IV}/TM DAs.[21,23–25,59,60] In the majority spin channel the Fe$_I$ d states are almost fully occupied, and in the vicinity of the valence-band maximum their contribution is insignificant, while in the minority spin channel the Fermi level cuts a narrow d states peak leading to the high spin polarization $P(E_F) = 82\%$. The exchange splitting of Fe$_I$ d states is around 2 eV. According to Bader analysis,[61,62] performed for the Ge/Fe$_I$ DA, the Fe$_I$ ion carries 7.7 electrons, i.e., there is no significant charge transfer between Fe$_I$ and its surrounding Ge ions. Thus one can assume that Fe$_I$ is almost in the neutral (^{0}Fe$_I$) configuration. At the same time, based on analysis of the Fe$_I$ partial charges values within the Wigner-Seitz sphere, we found a $4s \rightarrow 3d$ electron transfer (and also a tiny $4s \rightarrow 4p$ transfer) leading to the $3d^{6.8}4s^{0.45}4p^{0.45}$ configuration. Assuming complete $4s \rightarrow 3d$ electron transfer, we approximately consider this charge configuration as $3d^8$, giving rise to a spin state $S = 1$, as this takes place for an isolated interstitial ^{0}Fe$_I$ in Si.[63] A similar situation was also observed in the Si/Mn$_I$ DA,[64] where a Mn$_I$ $3d^7$ configuration gives rise to the spin state $S = 3/2$. In the next section we use these results to estimate the critical temperature for Ge/Fe$_I$. Note that a similar analysis cannot be performed for the Ge/Fe$_S$ DA since its exact ground state is not known (see Sec. III A).

C. Critical temperatures

To treat the effect of the interlayer exchange on the value of the critical temperature of FM DAs Ge/Fe, the analytical expressions of the RG approach were applied.[37,57,58] Using our MC method, we failed to detect numerically the 3D ordering, because of too weak coupling between Fe MLs. Therefore, we used the MC simulations in order to estimate the critical temperatures for the intralayer ordering only. The obvious advantage of the MC method lies in the account of interactions with a large number of coordination spheres, while the RG expressions in the formulation of Refs. 37, 57, and 58 are based on the account of the nearest-neighbor exchange only (both intra- and interlayer). Nevertheless, the latter approach had been proven to yield T_c estimations in good agreement with experimental values.[37,57,58] Note that the RG approach employed is only applicable to the systems with FM ground state and therefore the effect of 3D ordering was examined only for the Ge/Fe$_I$ DA case. As far as the Ge/Fe$_S$ DA is concerned, its critical temperature was estimated within MC method using DLM state exchange coupling parameters.

In relation to DAs, the magnetic ordering in a purely 2D regime was discussed in our previous work.[28] Here we only recall that in the case of the easy-axis anisotropy in the 2D Heisenberg system the magnetic ordering is possible at nonzero temperature, while in the easy-plane case the ordering will only be present at 0 K. In the former case, the account of the 3D exchange mode is expected to slightly enhance

the critical temperature. In the latter case, however, owing to the existence of a quasi-long-range order for $T < T_{\mathrm{BKT}}$, the introduction of an arbitrarily weak interplane exchange leads to the occurrence of a magnetic transition at $T_c > T_{\mathrm{BKT}}$.[37,58] In this case, the BKT transition does not disappear, but precedes the second-order phase transition.

For consistency reasons, the magnetic anisotropy energy Δ was chosen to be equal to 0.5 meV as in previous calculations.[28] This value, in fact, is of the same order of magnitude as the band contribution to the magnetic anisotropy energy $E_{\parallel} - E_{\perp} = 0.374$ meV for Ge/Fe_I DA, as resulting from a relativistic calculation including spin-orbit coupling, as implemented in the VASP code. Moreover, we have found that the Ge/Fe_I alloy is not an easy-plane magnet, since for the different directions of magnetic moments in the ML plane different energies were obtained. Regarding the RG estimation, the specific parameters of the Ge/Fe_I DA are the spin $S = 1$ (see previous section) and the in-plane nearest-neighbor exchange coupling integral $J_{01} = 13.153$ meV, calculated for the ground state (i.e., the FM state, but not the DLM one). The T_c estimation performed with these parameters in the easy-axis situation without interlayer exchange yields the value of 243.4 K. The nearest-neighbor *interlayer* exchange parameter, evaluated by means of the magnetic force theorem, turns out to be FM, $J'_{01} = \alpha J_{01} = 0.057$ meV, where $\alpha = 0.0043$. The account of this interlayer exchange leads to the enhancement of the critical temperature up to 270 K. For the Ge spacer thickness of 11 layers $J'_{01} = 0.003$ meV, i.e., the order of magnitude weaker than that in the case of a seven-layer-thick spacer. This causes only 1.3% enhancement of the Curie temperature upon switching on the interlayer exchange, while in the latter case the effect is almost 11%. Finally, starting from the spacer thickness of 15 atomic layers the interlayer exchange coupling does not influence the value of the Curie temperature for the easy-axis case any more.

Despite that the Ge/Fe_I has been shown to demonstrate the magnetic anisotropy of the easy-axis type, we also discuss the easy-plane anisotropy situation just as an illustration. In the case of the easy-plane anisotropy without interlayer exchange the estimated BKT temperature is equal to 241.8 K, i.e., it has the same order as T_c in the easy-axis case. As we already mentioned, switching on the 3D interaction mode must lead to the appearance of a nonzero Curie temperature T_c, which is typically 3−5% higher than T_{BKT}. However, the chosen anisotropy parameter $d = -\Delta/16J_{01} = 0.0023$ turns out to be almost 2 times smaller than the interlayer exchange parameter $\alpha = 0.0043$, which hinders the straightforward applicability of the employed equations, since Eq. (6) is valid only if $\alpha \ll d$. One can circumvent this by going to the case of thicker spacers, where this condition is met: choosing $\alpha = 10^{-4}$, which corresponds to the Ge spacer thickness of 15 atomic layers, one obtains $T_c = 282.5$ K.

As far as the MC simulations are concerned, the estimations performed for the Ge/Fe_I DA and the easy-axis case with the above given parameters J_{ij} and Δ, but taking into account interaction with 20 coordination spheres, give the value of 304 K (Table II), which is approximately 25% higher than the corresponding RG result. If the J_{ij} from the DLM calculation are taken, then the estimated critical temperature for the Ge/Fe_I alloy is equal to 240 K, which can be regarded as a more realistic value within the employed model of the DAs, since the exchange parameters obtained for the DLM state provide a better basis for the T_c estimation.[65] Regarding the Ge/Fe_S DA, our estimations yield a significantly lower T_c value of 98 K, since the exchange interactions in this system are much weaker than those in the Ge/Fe_I one. Note that, although we failed to detect the 3D ordering by means of the MC simulations, it is logical to assume that the value of T_c for the Ge/Fe_I (Ge/Fe_S) DA is somewhat higher than 240 K (98 K) due to the interlayer exchange. As far as the easy-plane case is considered, the estimations of the BKT temperatures by means of the MC method were not performed since their values were shown to be very close to the values of the critical temperatures obtained for the case of the easy-axis anisotropy.[28]

TABLE II. Critical temperatures (T_c and T_{BKT}, in K) estimated for the Ge/Fe_I DA by means of RG and MC methods for cases of easy-axis and easy-plane anisotropy in 2D and 3D regimes. The column "Reference state" indicates the state for which the J_{ij} employed for the estimation were calculated.

Method	Regime	Reference state	Easy axis	Easy plane
RG	2D	FM	243.4	241.8
	3D	FM	270	282.5
MC	2D	FM	304	–
	2D	DLM	240	–

IV. CONCLUSION

We applied first-principles electronic structure calculations within density functional theory to search for stable ferromagnetic systems among the A^{IV}/Fe digital alloys, where A^{IV} = Si or Ge. To take into account possible structural variations in the A^{IV}/Fe digital alloys two different types of Fe monolayers—substitutional (Fe_S) and interstitial (Fe_S) ones—were considered. It was shown that only the germanium-based alloys, Ge/Fe_I and Ge/Fe_S, demonstrate stable ferromagnetic ordering at nonzero temperature. The Curie temperatures for these systems were estimated from exchange coupling constants calculated for a ferromagnetic and a paramagnetic reference state. The critical temperature of the Ge/Fe digital alloys was found to depend strongly on the underlying crystal structure. Upon the choice of the reference state and the type of the magnetic anisotropy the Curie temperature for the Ge/Fe_I alloy in the absence of the interlayer exchange coupling was found to be in the range between 240 and 304 K. The more realistic value of 240 K is provided by the DLM method, since the corresponding exchange constants are estimated in the paramagnetic state. As far as the Ge/Fe_S alloy is concerned, the estimated Curie temperature turned out to be equal to 98 K, which is significantly lower than in the case of the Ge/Fe_I. We believe these values of 98 and 240 K to be increased by approximately 5–10% due to the introduction of the interlayer exchange coupling, similarly to what we have obtained for Ge/Fe_I alloy using RG approach expressions.

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