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Finite-temperature magnetism of Ni monolayers: Interplay between flips and amplitude fluctuations of the local moments

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The temperature dependence of the magnetization and spin fluctuation energies in Ni bulk and monolayers are determined in the framework of a functional-integral itinerant-electron theory. The electronic structure is obtained in the static approximation from a realistic *spd*-band model by using a real-space recursive expansion of the local Green's functions. The statistical averages of the spin fluctuations are performed by treating disorder within the coherent potential approximation. Results for the magnetization $M(T)$ and spin fluctuation energies of (001) fcc monolayers are presented for several values of the nearest-neighbor distance. Local environmental effects are discussed by comparison with bulk results. An interesting transition from simple spin flips to amplitude fluctuations of the local exchange fields is revealed as a function of dimensionality and bond length. Important qualitative differences in the spin fluctuation energies of Ni are observed as compared to Fe, which reflect different mechanisms of the dominant spin excitations in these itinerant-electron ferromagnets. The effects of *sp* electrons and *sp*-*d* hybridizations are discussed.

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I. INTRODUCTION

The magnetic properties of transition metals (TMs) at the nanoscale are of central importance from both fundamental and technological standpoints. At temperature $T = 0$, it is well known that the local environment of the atoms and the dimensionality of the system play crucial roles in defining the most important magnetic properties such as the magnetic moments, magnetic order, and magnetic anisotropy.[1–4] However, very little is still known about the microscopic origin of the magnetic behavior of TM nanostructures at finite temperatures. This is quite remarkable since the understanding of the temperature dependence of magnetism in low-dimensional systems and nanostructures is crucial for the development of new materials with potential applications in magnetic recording and magnetic memory devices.[4–11]

Previous experimental[12–21] and theoretical studies[22–36] have demonstrated that it is very difficult to infer *a priori* simple general trends on the stability of magnetism at finite temperature T, for example, as a function of local coordination number z_l and nearest-neighbor (NN) distance d. In order to derive reliable conclusions concerning the environment dependence of the magnetization curve and Curie temperature T_C, it is necessary to develop an electronic theory that takes into account both the fluctuations of the magnetic moments and the itinerant character of the d-electron states. Simple spin Hamiltonians, for example, based on the Heisenberg, xy, or Ising models, are not expected to be very predictive, unless they incorporate the electronic effects responsible for the environmental dependence of the effective interactions between the magnetic moments.[37,38] Furthermore, the magnetic properties of TMs are known to depend qualitatively on the d-band filling. For example, in Fe bulk the spin fluctuation energies (SFEs) show a double-minimum structure as a function of the exchange fields ξ, which indicates that the dominant magnetic excitations are flips of the magnetic moments keeping their amplitude approximately constant.[34–36,39–41] In contrast, the dominant spin excitations in bulk Ni involve mainly amplitude fluctuations of the local moments around the $T = 0$ values. This corresponds to a a single minimum in the SFE[34,41] and is probably related to the fact that the magnetic moments at $T = 0$ are much smaller in Ni than in Fe. Whether these trends hold at the nanoscale is unclear at present. In fact, one expects that for low coordinated sites l (small local coordination number z_l), Ni could also show a Heisenberg- or Ising-like behavior due to the decrease of the kinetic energy E_K relative to the exchange energy E_{ex}.[42] Moreover, the perturbation introduced by flipping an exchange field ξ_l at atom l is in general less important when z_l is small. It is one of the goals of this paper to investigate this problem.

The electronic structure effects associated to the itinerant character of the magnetic d states are therefore particularly important in Ni since the physics behind the ferromagnetic order is qualitatively different from what is observed in Fe. In this context, the purpose of this work is to extend our previous study[35] of Fe to low-dimensional Ni systems and to investigate the role of the local environment on the electronic and magnetic properties by taking the (001) fcc free monolayer as a representative example. In Sec. II we recall the theoretical method used for the calculations, which is based on Hubbard and Hasegawa's spin fluctuation theory for the periodic solid,[41,43] as recently extended in the context of cluster magnetism[34] and thin films.[35] The temperature dependence of the magnetization and spin fluctuation energies of a fcc (001) monolayer are presented and discussed in Sec. III. Comparison is made with the corresponding bulk results in order to quantify the role of dimensionality. Finally, Sec. IV summarizes the main conclusions.

II. THEORY

In the following we review the functional-integral theory used for the calculations of the magnetic properties of Ni bulk and monolayers. Since the effects of the hybridization between the d and sp electrons are expected to be important in Ni films, we extend the theory for a d-band Hamiltonian[34–36] to the case of an spd-band model. Our starting point is the Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_I. \tag{1}$$

The first term

$$\hat{H}_0 = \sum_{l,\alpha,\sigma} \varepsilon^0_{l\alpha}\, \hat{n}_{l\alpha\sigma} + \sum_{\substack{l\neq m\\ \alpha,\beta,\sigma}} t^{\alpha\beta}_{lm}\, \hat{c}^\dagger_{l\alpha\sigma}\hat{c}_{m\beta\sigma} \tag{2}$$

describes the single-particle electronic structure of the valence spd electrons in the tight-binding approximation.[44] As usual, $\hat{c}^\dagger_{l\alpha\sigma}$ ($\hat{c}_{l\alpha\sigma}$) refers to the creation (annihilation) operator of an electron with spin σ at the orbital α of atom l where $\alpha \in \{s,p_x,p_y,p_z,xy,yz,zx,x^2-y^2,3z^2-r^2\}$. The corresponding number operator is given by $\hat{n}_{l\alpha\sigma} = \hat{c}^\dagger_{l\alpha\sigma}\hat{c}_{l\alpha\sigma}$. The parameters $\varepsilon^0_{l\alpha}$ stand for the bare energy levels of the isolated atom ($\alpha \equiv s,p,d$) and $t^{\alpha\beta}_{lm}$ for the hopping integrals between atoms l and m. The second term

$$\hat{H}_I = \frac{1}{2} \sum_{\substack{l,\alpha,\beta\\ \sigma,\sigma'}}{}' \, U^{\alpha\beta}_{\sigma\sigma'}\, \hat{n}_{l\alpha\sigma}\hat{n}_{l\beta\sigma'} \tag{3}$$

describes the interactions among electrons by means of an intra-atomic Hubbard-like spd-band model. The prime in the sum indicates that self-interactions are to be excluded [i.e., $(\alpha\sigma) \neq (\beta\sigma')$]. The parameters $U^{\alpha\beta}_{\sigma\sigma'}$ are the effective Coulomb repulsion integrals between electrons at the orbitals α and β having spin projections σ and σ'. For example, for d electrons we have $U^{dd}_{\uparrow\downarrow} - U^{dd}_{\uparrow\uparrow} = J_{dd}$, where $J_{dd} > 0$ is the average d-electron exchange integral. Notice that Eq. (3) does not respect spin-rotational symmetry since the exchange terms of the form $H_{xy} = -\sum_{l,\alpha<\beta} J_{\alpha\beta}(S^-_{l\alpha}S^+_{l\beta} + S^+_{l\alpha}S^-_{l\beta})$ have been dropped. Nevertheless, this is not expected to be a serious limitation in the present work because we are interested in studying the spin fluctuations on top of broken-symmetry ferromagnetic ground states.

In order to reduce the number of parameters and to simplify the notation we neglect in the following the minor differences in the Coulomb repulsion between s and p electrons, as well as the exchange integrals among them. Mathematically, this implies $U^{\alpha\beta}_{\sigma\sigma'} = U^{ss}_{\sigma\sigma'}$ for α and $\beta \in \{s,p_x,p_y,p_z\}$ and for σ and $\sigma' \in \{\uparrow,\downarrow\}$. The interactions involving the d orbitals are characterized by the average dd repulsion $U_{dd} = (U^{dd}_{\uparrow\downarrow} + U^{dd}_{\uparrow\uparrow})/2$, the sp-d repulsion $U_{sd} = (U^{sd}_{\uparrow\downarrow} + U^{sd}_{\uparrow\uparrow})/2$, and the average d-electron exchange integral $J_{dd} = U^{dd}_{\uparrow\downarrow} - U^{dd}_{\uparrow\uparrow}$. It is then meaningful to introduce the operator

$$\hat{N}^s_l = \sum_{\sigma,\,\alpha\in sp} \hat{n}_{l\alpha\sigma} \tag{4}$$

giving the number sp electrons at atom l, and the z component of the local sp spin operator

$$\hat{S}^s_{lz} = \frac{1}{2}\sum_{\alpha\in sp}(\hat{n}_{l\alpha\uparrow} - \hat{n}_{l\alpha\downarrow}), \tag{5}$$

where $\alpha \in \{s,p_x,p_y,p_z\}$. Similarly, the d-electron number operator is given by

$$\hat{N}^d_l = \sum_{\sigma,\,\alpha\in d} \hat{n}_{l\alpha\sigma} \tag{6}$$

and the z component of the d spin operator by

$$\hat{S}^d_{lz} = \frac{1}{2}\sum_{\alpha\in d}(\hat{n}_{l\alpha\uparrow} - \hat{n}_{l\alpha\downarrow}). \tag{7}$$

With these definitions the interaction Hamiltonian can be written as

$$\hat{H}_I = \hat{H}_{dd} + \hat{H}_{ss} + \hat{H}_{sd}, \tag{8}$$

where

$$\hat{H}_{dd} = \sum_l \left\{ \frac{U_{dd}}{2}\left(\hat{N}^d_l\right)^2 - J_{dd}\left(\hat{S}^d_{lz}\right)^2 \right\}, \tag{9}$$

$$\hat{H}_{ss} = \frac{U_{ss}}{2}\sum_l \left(\hat{N}^s_l\right)^2, \tag{10}$$

and

$$\hat{H}_{sd} = U_{sd}\sum_l \hat{N}^s_l \hat{N}^d_l. \tag{11}$$

Notice that we have neglected for simplicity the orbital dependence of the Coulomb integrals within each irreducible s, p, and d band as well exchange integrals involving the more delocalized s and p orbitals. Strictly speaking, Eqs. (8)–(10) are equivalent to Eq. (3) only if the self-interaction terms present in Eqs. (8)–(10) are canceled out by redefining the single-particle energy levels. This has no consequences on the magnetic properties to be discussed below.[45]

The finite-temperature properties are determined by applying the functional-integral method developed by Hubbard and Hasegawa for periodic solids.[41,43] Within this formalism, a linearization of the two particle operators of the intra-atomic Hubbard-like model [Eqs. (9)–(11)] is performed by means of a three-field Hubbard-Stratonovich transformation within the static approximation. Thus, a charge field η_{ls} for the sp electrons, a charge field η_{ld} for the d electrons, and an exchange field ξ_l for the d electrons are introduced at each atom l, which describe the finite-temperature fluctuations of the s, p, and d energy levels and of the local exchange splittings. Using the notation $\vec{\xi} = (\xi_1,\ldots,\xi_n)$, $\vec{\eta}_d = (\eta_{1d},\ldots,\eta_{nd})$, and $\vec{\eta}_s = (\eta_{1s},\ldots,\eta_{ns})$, the partition function Z is given by

$$Z \propto \int d\vec{\xi}\, d\vec{\eta}_d\, d\vec{\eta}_s\, \exp\{-\beta F'(\vec{\xi},\vec{\eta}_d,\vec{\eta}_s)\}, \tag{12}$$

where the free energy F' associated with the charge and exchange fields $\vec{\eta}_s$, $\vec{\eta}_d$, and $\vec{\xi}$ is given by

$$F' = \frac{1}{2}\sum_l \left(U_{dd}\,\eta^2_{ld} + U_{ss}\,\eta^2_{ls} + 2\,U_{sd}\,\eta_{ls}\,\eta_{ld} + \frac{J_{dd}}{2}\xi^2_l \right) - \frac{1}{\beta}\ln\{\mathrm{Tr}[\exp\{-\beta(\hat{H}' - \mu\hat{N})\}]\}. \tag{13}$$

The effective Hamiltonian

$$\hat{H}'(\vec{\xi},\vec{\eta}_d,\vec{\eta}_s) = \hat{H}_0 + \sum_l \{U_{dd}\, i\eta_{ld}\hat{N}^d_l + U_{ss}\, i\eta_{ls}\,\hat{N}^s_l + U_{sd}\left(i\eta_{ls}\hat{N}^d_l + i\eta_{ld}\hat{N}^s_l\right) - J_{dd}\,\xi_l\,\hat{S}^d_{lz}\}, \tag{14}$$

describes the dynamics as if the electrons were independent particles moving in a random alloy with fluctuating spin-dependent energy levels. The thermodynamic properties of the system are obtained as a statistical average over all possible distributions of the charge and exchange fields. The underlying static approximation is exact in the atomic limit ($t_{lm}^{\alpha\beta} = 0, \forall l \neq m$) where no fluctuations are present and in the noninteracting limit ($U_{\sigma\sigma'}^{\alpha\beta} = 0$).

For $T \to 0$ the dominating field configuration corresponds to the saddle point in the free energy $F'(\vec{\xi},\vec{\eta}_d,\vec{\eta}_s)$. The partial derivatives of F' are given by

$$\frac{\partial F'}{\partial \xi_l} = \frac{J_{dd}}{2}\big(\xi_l - 2\big\langle \hat{S}_{lz}^d \big\rangle'\big), \tag{15}$$

$$\frac{\partial F'}{\partial \eta_{ld}} = U_d\big(\eta_{ld} + i\big\langle \hat{N}_l^d \big\rangle'\big) + U_{sd}\big(\eta_{ls} + i\big\langle \hat{N}_l^s \big\rangle'\big), \tag{16}$$

and

$$\frac{\partial F'}{\partial \eta_{ls}} = U_{ss}\big(\eta_{ls} + i\big\langle \hat{N}_l^s \big\rangle'\big) + U_{sd}\big(\eta_{ld} + i\big\langle \hat{N}_l^d \big\rangle'\big), \tag{17}$$

where $\langle \cdots \rangle'$ indicates average with respect to the single-particle Hamiltonian $\hat{H}'$. The average *sp* occupations, *d* occupations, and spin moments entering Eqs. (15)–(17) can be readily obtained from Eqs. (4)–(7) by noticing that

$$\langle \hat{n}_{l\alpha\sigma} \rangle' = \int_{-\infty}^{+\infty} \rho_{l\alpha\sigma}(\varepsilon)\, f(\varepsilon)\, d\varepsilon\,, \tag{18}$$

where $f(\varepsilon)$ refers to the Fermi function and $\rho_{l\alpha\sigma}(\varepsilon)$ to the local density of states (DOS) at the orbital $l\alpha\sigma$. Notice that by setting Eqs. (15)–(17) equal to zero one recovers the usual mean-field equations for η_{ld}, η_{ls}, and ξ_l corresponding to the self-consistent *spd*-band theory at $T = 0$.[1,46]

In this work we are interested in the temperature dependence of the magnetic properties which are dominated by the low-lying spin fluctuations. Moreover, since $J_{dd} \ll U_{dd}$ the energy involved in local charge fluctuations is much larger than the spin-fluctuation energies. Therefore, it is reasonable to neglect the thermal fluctuations of the charge fields $\eta_{l\alpha}$ by setting them equal to the values $\overline{\eta}_{l\alpha}$ yielding the saddle point of the free energy $F'(\vec{\xi},\vec{\eta}_d,\vec{\eta}_s)$ for the given exchange-field configuration $\vec{\xi}$. From Eqs. (16) and (17) and the saddle-point condition $\partial F'/\partial \eta_{ld} = \partial F'/\partial \eta_{ls} = 0$ it follows that $i\overline{\eta}_{l\alpha} = \nu_{l\alpha} = \langle \hat{N}_l^\alpha \rangle'$ for $\alpha = s$ and d. Physically, this means that the charge distribution $\nu_{l\alpha}$ is calculated self-consistently for each exchange-field configuration $\vec{\xi}$. Taking into account that the self-consistent *sp* and *d* occupations $\nu_{l\alpha}$ (or equivalently the charge fields $\overline{\eta}_{l\alpha}$) are implicit functions of $\vec{\xi}$, we may regard the free energy $F'(\vec{\xi}) = F'(\vec{\xi},\vec{\overline{\eta}}_d,\vec{\overline{\eta}}_s)$ as a function of $\vec{\xi}$ alone. Finally, we have

$$Z \propto \int d\vec{\xi}\, \exp\{-\beta F'(\vec{\xi})\}, \tag{19}$$

where

$$F'(\vec{\xi}) = \frac{1}{2}\sum_l \left(\frac{J_{dd}}{2}\xi_l^2 - U_{dd}\nu_{ld}^2 - U_{ss}\nu_{ls}^2 - 2U_{sd}\nu_{ld}\nu_{ls}\right) - \frac{1}{\beta}\ln\left(\mathrm{Tr}[\exp\{-\beta(\hat{H}'(\vec{\xi}) - \mu\hat{N})\}]\right). \tag{20}$$

The integrand in Eq. (19) is proportional to the probability density $P(\vec{\xi}) = \exp\{-\beta F'(\vec{\xi})\}/Z$ for the exchange-field configuration $\vec{\xi}$.

The thermodynamic properties of the system can be obtained as a statistical average over all the possible distributions of the exchange fields with $\exp\{-\beta F'(\vec{\xi})\}$ as weighting factor. For instance, the local magnetization at atom l, which measures the long-range order, is given by

$$M_l(T) = 2\big\langle\!\big\langle \hat{S}_{lz}^s + \hat{S}_{lz}^d \big\rangle\!\big\rangle = \frac{2}{Z}\int d\vec{\xi}\, \big\langle \hat{S}_{lz}^s + \hat{S}_{lz}^d \big\rangle'_{\vec{\xi}}\;\; e^{-\beta F'(\vec{\xi})}\,. \tag{21}$$

Besides $M_l(T)$ it is interesting to compute the local magnetic moment

$$\begin{aligned}\mu_l &= 2\big\langle\!\big\langle (\hat{S}_{lz}^s + \hat{S}_{lz}^d)^2 \big\rangle\!\big\rangle^{\frac{1}{2}} \\ &= 2\left[\frac{1}{Z}\int d\vec{\xi}\, \big\langle (\hat{S}_{lz}^s + \hat{S}_{lz}^d)^2 \big\rangle'_{\vec{\xi}}\;\; e^{-\beta F'(\vec{\xi})}\right]^{\frac{1}{2}}\,,\end{aligned} \tag{22}$$

which provides information on the local spin correlations in particular above T_{C}.

The configurational averages over the random alloy are determined by computing the average Green's function within the single-site coherent-potential approximation (CPA).[47,48] In the CPA, one considers an effective medium characterized by a complex self-energy $\hat{\Sigma}(\varepsilon)$. This effective medium is self-consistent in the sense that for each energy ε it matches the average of the perturbed Green's function associated to the fluctuation of the exchange field ξ at any given site l with respect to the effective medium. The average Green's function $\hat{\mathbf{G}}(\varepsilon)$ associated to the self-energy $\hat{\Sigma}$ is expressed in terms of the effective-medium Hamiltonian $\hat{H}_{\mathrm{eff}} = \hat{H}_0 + \hat{\Sigma}$ as $\hat{\mathbf{G}}(\varepsilon) = [\varepsilon - \hat{H}_{\mathrm{eff}}]^{-1}$. The perturbed Hamiltonian is given by $\hat{H}_{\mathrm{p}} = \hat{H}_{\mathrm{eff}} + \hat{V}_l$ with the perturbation

$$\begin{aligned}\hat{V}_l(\xi,\varepsilon) = \sum_\sigma \big\{&(U_{dd}\,\nu_{ld} + U_{sd}\,\nu_{ls})\hat{N}_l^d + U_{sd}\,\nu_{ld} + U_{ss}\,\nu_{ls})\hat{N}_l^s \\ &- J_{dd}\,\xi_l\,\hat{S}_{lz}^d\big\} - \hat{\Sigma}_l(\varepsilon),\end{aligned} \tag{23}$$

where $\hat{\Sigma}_l$ is the self-energy operator reduced to the site l. Finally, the self-consistent equation for $\hat{\Sigma}_l$ reads[47,48]

$$\hat{\Sigma}_l = \langle\!\langle (1 - \hat{\mathbf{G}}_l\,\hat{V}_l)^{-1}\,(\hat{V}_l + \hat{\Sigma}_l) \rangle\!\rangle. \tag{24}$$

The CPA configurational average is analogous to the case of a *d*-band model considered in Ref. 35, where further details and references may be found.

III. RESULTS

In this section we present and discuss our results for bulk fcc Ni and for Ni (001) monolayers. Local environmental effects are investigated by varying the nearest-neighbor (NN) distances. The parameters used for the calculations are determined as follows. The hopping integrals $t_{ij}^{\alpha\beta}$ between atoms are fitted to the band structure of the fcc Ni solid.[49] The dominant *d*-electron exchange integral J_{dd} is chosen to yield the proper bulk magnetic moment $\mu_{lb} = 0.6\,\mu_{\mathrm{B}}$ at $T = 0$ [$J_{dd}(\mathrm{Ni}) = 0.96$ eV]. The Green's functions of the effective medium and for the perturbation are computed by using

Haydock-Heine-Kelly's recursion method[50] and the Dyson equation,[47] respectively.

A. Ni bulk

In this section, as a reference for further discussion, we present the results of bulk fcc Ni. The temperature dependence of the magnetization $M(T)$ and of the local magnetic moment $\mu = 2\sqrt{\langle \hat{S}_z^2 \rangle}$ are shown in Fig. 1(a). First of all, one observes that the shape of the $M(T)$ curve is qualitatively similar to the experimental one. The dependence of μ is in qualitative agreement with previous calculations,[51] in particular concerning the increase of μ with increasing T for $T > T_C$. The value $T_C = 610$ K obtained for the Curie temperature is quite close to the experimental result[52] value $T_C(\text{exp.}) \simeq 630$ K. One of the reasons for the good agreement between our results and experiment is probably related to the nature of the spin-fluctuations in bulk Ni and to the ability of the CPA to cope with them. In contrast to Fe, the probability distribution for the exchange field ξ shows in Ni a single peak, which resembles a Lorentz distribution with a temperature-dependent mean value and width. Actually, it is more closely a Gaussian-like distribution, with some degree of asymmetry for $T < T_C$, since the free energy is approximately quadratic as a function of ξ [see Fig. 1(b)]. In this context it is important to recall that the CPA yields the exact average Green's function if the energy levels of the alloy follow a Lorentz probability distribution.[48,53] Consequently, the CPA should be very good approximation for describing the temperature dependence of $M(T)$ when the fluctuations of the modulus of the local moments dominate. Concerning the comparison with previous works[43] it should be also noted that we do not assume a rigid model density of states (DOS) but rather perform a realistic self-consistent determination of the electronic structure, both in the ground state and in the presence of spin fluctuations. Furthermore, we have taken into account explicitly the contributions of the more delocalized *sp* electrons and *sp*-*d* hybridizations, which are important for the electronic structure, despite being almost negligible for the magnetic moments. The *sp* local moments are quite small, typically $|\mu_{sp}| \simeq 0.04\mu_B$, and are in most cases antiparallel to the *d* moments. The quantitative description of the electronic structure and single-particle Green's functions has certainly also contributed to the good agreement with experiment.

In Fig. 1(b) the local spin fluctuation energy $\Delta F(\xi)$ is shown as a function of the exchange field ξ for several representative temperatures ($t = T/T_C$). First of all one observes that the form of $\Delta F(\xi)$ is qualitatively different from what is found in bulk Fe.[35] In Ni $\Delta F(\xi)$ has a single-minimum at $\xi = \xi^0$, which implies that the low-temperature spin excitations involve mainly amplitude fluctuations of the local exchange fields around a temperature-dependent average value $\langle \xi \rangle \simeq \xi^0$. $\Delta F(\xi)$ shows a slight asymmetry around ξ^0 which favors fluctuations towards smaller values of ξ and thus leads to the decrease of $\langle \xi \rangle$ and of $M(T)$ as T increases. For $T \geqslant T_C$ we have $\xi^0 = 0$ and $\Delta F(\xi)$ becomes symmetric. The trends in Ni differ strikingly from bulk Fe, where $\Delta F(\xi)$ is far from parabolic to the point that a second minimum develops out of a saddle point at $\xi = \xi^{0-} \simeq -\xi^{0+} < 0$ as T increases and approaches T_C. At the same time, the fluctuations of the size of the exchange fields yield a small decrease of ξ^{0-}. One concludes than in the present framework the spin-flip excitations are essentially absent in Ni, which is probably due to the fact that the magnetic moments at $T = 0$ are much smaller in Ni than in Fe. As a result the energy required to fluctuate the amplitude of the moments is much smaller than the spin-flip-like fluctuations observed in Fe.[35] This behavior reflects the strong itinerant character of Ni magnetism, which contrasts with the simpler fluctuations of the direction of Heisenberg- or Ising-like localized magnetic moments. As is discussed below, low-dimensional systems and in particular ultrathin films show somewhat different behavior with a stronger tendency to favor localized spin excitations.[34]

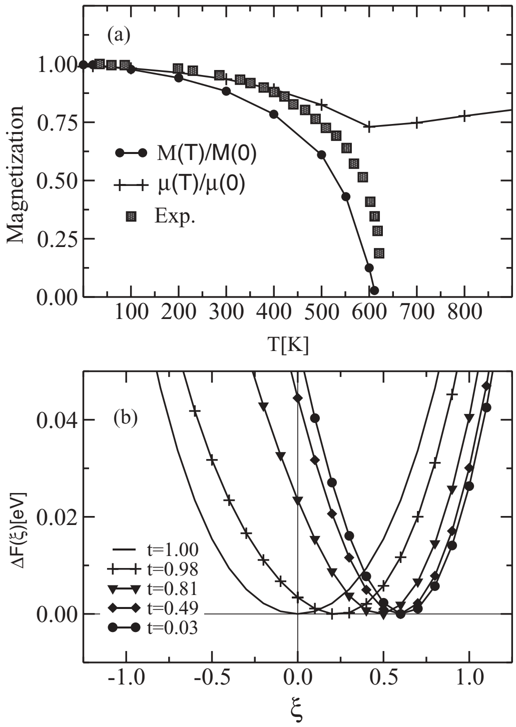

FIG. 1. (a) Temperature dependence of the magnetization $M(T)$ (dots) and local magnetic moment μ (crosses) in bulk fcc Ni. (b) Spin fluctuation energy $\Delta F(\xi) = F'(\xi) - F'(\xi^0)$, where $\xi^0 = \xi^0(T)$ refers to the minimum of $F'(\xi)$. The considered reduced temperatures $t = T/T_C$ are indicated in the inset.

B. Ni monolayer

The effects of reduced dimensionality and local atomic environment on the temperature-dependent magnetic properties are investigated by considering an fcc (001) Ni monolayer and by varying the nearest-neighbor distances d. In Fig. 2 results for the d magnetization $M^{(d)}(T)$ are shown for several values of $r = d/d_b$, where d_b refers to the bulk value. One observes that the zero-temperature magnetization $M^{(d)}(T = 0)$ increases monotonically with increasing r reaching the saturation value $10 - n_d$ at approximately $r \simeq 1.1$. This is a well-known consequence of the d band narrowing, which follows the reduction of the hopping integrals and the associated enhancement of the density of states at the Fermi energy. It is important to remark that the present finite-temperature theory takes into account spin fluctuations described by Ising-like scalar fields on top

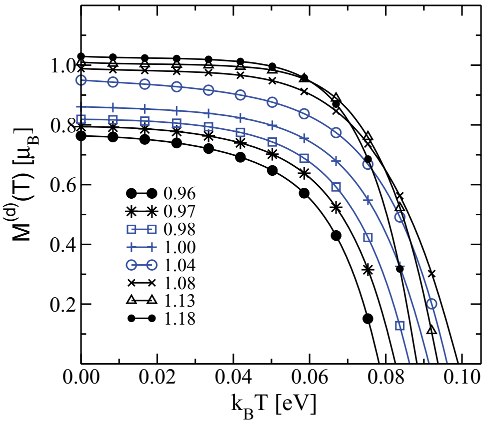

FIG. 2. (Color online) Temperature dependence of the d magnetization of an fcc (001) Ni monolayer calculated by using the CPA. Different representative values of the monolayer nearest-neighbor distance d are considered. The numbers in the inset indicate the ratio $r = d/d_b$ between d and the bulk NN distance d_b.

of a symmetry-broken ground state. Therefore, the Mermin-Wagner theorem, which actually holds for Heisenberg-like vector spins having a continuous symmetry, does not apply to our case. Moreover, our approach takes explicitly into account the itinerant character of the d electrons responsible for magnetism, while in Heisenberg-like models the spins are localized. As a result the effective interactions between exchange fields in itinerant-electron magnetism are not strictly short ranged and therefore we obtain finite values of the Curie temperature for the monolayer.

At finite temperatures one finds very different and more complex behaviors depending on the value of r. For example, first T_C increases slowly with increasing d up to about $r \simeq 1.08$ and then drops rather suddenly for $r > 1.08$. Let us first focus on distances where the ground-state moments are essentially saturated. For $1.1 < r < 1.18$, $M^{(d)}(T)$ decreases faster with increasing T when the NN distances are larger. This implies that for large NN distances, magnetism is less stable when the electrons are more localized, that is, when the kinetic or band energy is smaller. This is consistent with the fact that in this range of r, T_C decreases with increasing r. As we discuss below, this corresponds to a localized-like magnetic behavior. Concerning T_C, one observes a nonmonotonous behavior with a maximum value at approximately $r \simeq 1.08$. Notice that T_C decreases for $r > 1.08$ even if all the curves start at approximately the same saturated magnetization per atom. Therefore, the changes in T_C for large values of r cannot be ascribed to an environmental dependence of the local moments but rather to changes in the effective couplings J_H between them. A similar distance dependence of T_C has been found in thin Fe films.

It is interesting to analyze or interpret these results in the framework of an Ising or Heisenberg model. In mean-field approximation, the effective Heisenberg coupling J_H between NN spins is given by $J_H \simeq 3k_BT_C/[zM(0)^2]$, where z is the coordination number and $M(0)$ is the $T = 0$ moment. On the basis of this expression for J_H, one concludes that the effective exchange coupling between local moments has reached its maximum value at $r \simeq 1.1$ and that the decrease of T_C for $r > 1.1$ is a consequence of a reduction of J_H as the atoms are pulled apart. Still, the reduced value of J_H cannot explain the temperature dependence completely since also the form of $M^{(d)}(T)$ changes as r increases. Indeed, for larger r the drop in $M^{(d)}(T)$ is faster. This implies that for more localized electrons, strong spin fluctuations are only allowed close to T_C. The changes in the form of $M^{(d)}(T)$ can be illustrated more clearly by comparing, for example, the results for $r = 1.18$ with the case of smaller values of d (e.g., $r = 0.96$). For smaller values of r ($r < 1$) we observe a reduction of T_C, which is consistent with the trends found in compact cluster structures, where higher coordination number (reduction of the kinetic energy) implies a reduction in T_C.[36]

It is interesting to compare our results with experimental findings. Previous experimental studies in Ni$_n$ thin films of n layers have shown a reduction of T_C with respect to the bulk value.[5,54,55] In general, T_C approaches monotonically to the bulk value as a function of n. For instance, Li *et al.*[5] measured the T_C of Ni$_n$/W(110) as a function of $n \lesssim 20$. Ney *et al.*[55] reported the local moments at each layer in Ni$_n$/Cu(001) for $n \simeq 4$. Huang *et al.*[54] found a qualitative difference between the T_C of Ni$_n$/Cu(111) and that of Ni$_n$/Cu(001) for $n \lesssim 20$. In the first case, T_C is larger than that of the second case for a given value of n. Moreover, as n increases, T_C approaches faster to the bulk value in Ni$_n$/Cu(111) than in Ni$_n$/Cu(001). Our results for the single Ni monolayer show an enhancement of T_C with respect to the bulk value, which is opposite to the trend found in deposited Ni thin films. One should notice, however, that the model system studied in the present paper is a free monolayer. Therefore, a direct comparison with experiment is not straightforward, since hybridization effects between the monolayer, which most likely decrease T_C, and substrate are absent. Thus, free-standing monolayers should, in principle, overestimate T_C as compared to the deposited ones. Nevertheless, it is important to recall that in itinerant-electron magnetism the changes in the Curie temperature associated to a reduction of dimensionality or coordination number are far less obvious *a priori*. As already discussed, besides the reduction of coordination number, which in the monolayer tends to reduce T_C, one also finds that T_C is modified by the enhancement of the local moments, which is also a direct consequence of the reduction of local coordination and d-band width. The latter implies an enhancement of the local exchange splittings and is likely to yield an enhancement of the effective exchange interactions between the local moments. In fact, the interplay between these three factors (reduced number of pairs of interactions, enhancement of the local moments, and changes in the effective exchange couplings) is what defines T_C. Indeed, in the case of Fe monolayers[35] we have found, in agreement with experiments of deposited ultrathin films, that T_C is smaller in the free monolayer than in the bulk. Ni shows the opposite trend, which implies the admittedly nonobvious behavior that the enhancement of the local moments and effective exchange interactions dominate. This may be a consequence of the specific electronic structure of the idealized free monolayer. The trends could change for small finite film thicknesses (bilayer, trilayer, etc.). In addition, to our knowledge, experiments of a deposited single Ni monolayer have not been reported. Moreover, a recent experiment suggests that interface mixing between the Ni film and the substrate should be considered.[56]

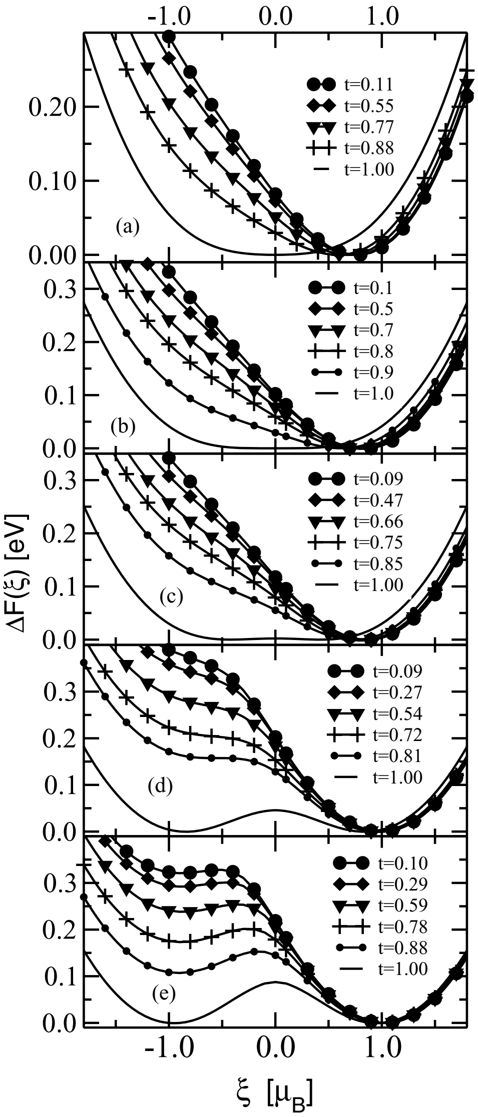

FIG. 3. Local spin-fluctuation energy $\Delta F(\xi) = F'(\xi) - F'(\xi^{0+})$ in fcc (001) Ni monolayers. Different NN distances d are considered as given by $r = d/d_b$, where d_b refers to the bulk value: (a) $r = 0.96$, (b) $r = 0.98$, (c) $r = 1.00$, (d) $r = 1.10$, and (e) $r = 1.18$. $F'(\xi^{0+})$ refers to the minimum of the free energy $\xi > 0$. The considered reduced temperatures $t = T/T_C$ are indicated in the inset.

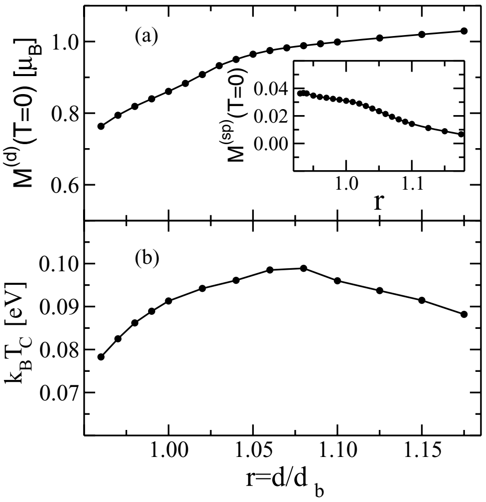

FIG. 4. (a) The ground-state d-electron magnetization $M^{(d)}(T = 0)$ and (b) the Curie temperature T_C of an fcc (001) Ni monolayer as a function of the NN distance d. The contribution of the sp electrons to the total magnetization at $T = 0$ is shown (in μ_B) in the inset.

Figure 3 shows the corresponding spin fluctuations energies $\Delta F(\xi) = F'(\xi) - F'(\xi^{0+})$, where ξ^{0+} indicates the position of the minimum of the free energy for $\xi > 0$. Results are given for several representative temperatures T and NN distances. It is interesting to identify three different behaviors. First, for the smallest NN distance, the curves are similar to those obtained for the bulk, particularly well bellow T_C ($t \leqslant 0.9$). $\Delta F(\xi)$ shows a single minimum at $\xi = \xi^{0+} > 0$, which approaches to $\xi^{0+} = 0$ for $t \to 1$. Consequently, the dominant spin excitations involve mainly amplitude fluctuations of the local exchange fields around the average exchange splitting or d-electron magnetization $M^{(d)}(T) = \langle \xi \rangle$. This is probably due to the fact that the magnetic moment at this interatomic distance is relatively small and similar to the bulk value [$M_{\text{bulk}}(T = 0) \simeq 0.6\mu_B$]. However, note that close to T_C $\Delta F(\xi)$ is very flat, much flatter than in the solid [$\Delta F(\xi) \sim \xi^4$ for the monolayer, while $\Delta F(\xi) \sim \xi^2$ in the bulk; see Fig. 1(b)]. This means that much larger amplitude fluctuations of ξ are present in the monolayer for $T \simeq T_C$. This can be interpreted as a consequence of the reduced coordination number. Second, for $r = 0.98$–1.00 one observes that the curves for $\Delta F(\xi)$ get wider, showing that the negative spin fluctuations start to become energetically more favorable even well below T_C [e.g., $t = 0.85$ for $r = 1.0$; see Fig. 3]. These negative ξ fluctuations accelerate the decrease of the magnetization and are the precursor of the development of a second minimum in $\Delta F(\xi)$ for $\xi < 0$. Notice, moreover, that the curves are extremely flat for $T \simeq T_C$. Finally, for $r \geqslant 1.1$ a second minimum develops at $\xi^{0-} \simeq -\xi^{0+}$, first close to T_C and then, as r increases, also at lower temperatures [see Figs. 3(d) and 3(e)]. For low temperatures only an inflexion point is found for $\xi \simeq \xi^{0-}$. These trends can be interpreted as an enhancement of the local character of the spin fluctuations due to the reduction of the kinetic energy of the d electrons.

Ground-state and finite-temperature properties can be correlated by comparing the results for the d-electron magnetization $M^{(d)}(T = 0)$ and the Curie temperature T_C as a function of the nearest-neighbor distance d. In this way the interplay between kinetic and Coulomb energies can be explored, since shorter NN distances yield larger d-band widths, which correspond qualitatively to larger coordination numbers. In addition, in the inset, we show the sp contribution to $M(T = 0)$. In Fig. 4 one observes that the ground-state d moment grows monotonically with increasing r as the density of states at the Fermi energy increases (Stoner criterion). Nearly saturated values $M^{(d)}_{\text{sat}} \simeq 10 - n_d$ are reached for the region $r \simeq 1.05$–1.10, beyond which $M^{(d)}(T = 0)$ increases more slowly. Notice that the sp-electron spin polarization, shown in the inset of Fig. 4, is parallel to the d-electron moment ($M^{(sp)} > 0$), in contrast to the solid. Since $M^{(sp)} > 0$ is a consequence of the sp-d hybridization, it decreases with increasing d as the sp-d hopping integrals decrease despite the small enhancement of the d-electron exchange splitting.

The Curie temperature, which measures the stability of ferromagnetic order, shows a more interesting nonmonotonous behavior. As in the case of $\Delta F(\xi)$, different regimes can be distinguished. For $0.96 \lesssim r \lesssim 1.04$ one observes that T_C increases due to the increase of the local moments and the associated enhancement of the exchange splitting $\Delta\varepsilon_x^d$ [see Fig. 4]. In this case, the significant increase of $M^{(d)}(T=0)$ and $\Delta\varepsilon_x^d$ dominates over possible reductions of the NN spin coupling in the increasing distance. Subtle effects of itinerant magnetism are present in almost all Ni monolayers. For example, for $1.04 \lesssim r \lesssim 1.1$, $M^{(d)}(T=0)$ increases but T_C is nearly constant. As discussed above, this behavior can only be ascribed to the specific electronic structure, which yields very little changes in the corresponding spin fluctuation energies [see Figs. 3(b) and 3(c)]. In fact, the increase in the ground-state local moments is compensated by a decrease of the effective coupling J_H as r increases. The behavior of T_C for $r \gtrsim 1.1$ can be qualitatively interpreted in terms of a mean-field Heisenberg model. As already discussed, J_H decreases with increasing r for these values of r. In this case, the electronic hoppings and the kinetic energy of the electrons are relatively small and the spin fluctuations have a more localized character [see Figs. 3(d) and 3(e)]. Consequently, the spin-fluctuation energies decrease, since the perturbations introduced by spin disorder are less significant as r increases. One may conclude that for $r \simeq 1.1$ a transition or crossover from itinerant- to localized-like character of the dominant d-electrons occurs. In summary, the distance dependence of the finite-temperature properties are the result of a subtle competition between localized and itinerant aspects of magnetism. Interesting magnetic phenomena can be expected as a function other variables, for example, the local coordination number, which also affect the relative importance of kinetic and Coulomb contributions.

IV. CONCLUSION

The finite-temperature magnetic properties of bulk Ni and of Ni monolayers have been determined in the framework of a functional-integral itinerant-electron theory. The effects of disorder in the electronic structure due to spin fluctuations have been taken into account by means of the single-site coherent potential approximation. The stability of ferromagnetism has been studied as a function of the local environment of the atoms. A remarkable nonmonotonous dependence of the magnetization curves has been obtained by varying the interatomic distances (strain effects). Moreover, it has been shown that the reduction of the system dimensions, from three to two, enhances the localized character of the spin fluctuations and yields a clear transition from itinerant to localized behavior. These trends have been correlated with the environmental dependence of the electronic properties and with the resulting changes in the ground-state magnetic moments and spin-fluctuation energies. The electronic structure contributions and the itinerant character of the s, p, and d valence electrons have been found to be crucial for determining the magnetic behavior of low-dimensional systems at finite temperatures.

The present electronic model and the local approach to the electronic structure are well suited to investigate more complex systems with reduced symmetry, such as clusters and nanostructures on surfaces, or substrate and interlayer effects on thin films and monolayers. Besides the applications, a number of methodological improvements are worthwhile. For instance, short-range magnetic order, noncollinear magnetic order, and possible fluctuations of the direction of vector exchange fields $\vec{\xi}_l$ are likely to affect the magnetization curves and value of T_C. It would be therefore very interesting to quantify the role of these spin fluctuations in the framework of the present electronic theory. Moreover, the effects of interfaces with nonmagnetic substrates could be incorporated in order to achieve a more realistic comparison with experiment. In addition, the model and CPA method could be extended by including spin-orbit[2,57] and dipole-dipole interactions, which are responsible for the magnetic anisotropy and for the spin reorientation transitions as a function of temperature and structure.[17,18,58,59] Finally, from a more fundamental perspective, it remains a major challenge to improve on the treatment of many-body effects beyond the static approximation, since the electronic correlations are expected to become increasingly important as the system dimensions decrease.

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[1] G. M. Pastor, J. Dorantes-Dávila, and K. H. Bennemann, Phys. Rev. B **40**, 7642 (1989).

[2] G. M. Pastor, J. Dorantes-Dávila, S. Pick, and H. Dreyssé, Phys. Rev. Lett. **75**, 326 (1995); J. Dorantes-Dávila and G. M. Pastor, *ibid.* **81**, 208 (1998); R. A. Guirado-López, J. Dorantes-Dávila, and G. M. Pastor, *ibid.* **90**, 226402 (2003).

[3] See, for instance, G. M. Pastor in *Atomic Clusters and Nanoparticles*, Lectures Notes of the Les Houches Summer School of Theoretical Physics, edited by C. Guet, P. Hobza, F. Spiegelman, and F. David (EDP Sciences, Les Ulis/Springer, Berlin, 2001), p. 335 ff.

[4] J. Bansmann, S. H. Baker, C. Binns, J. A. Blackman, J.-P. Bucher, J. Dorantes-Dávila, V. Dupuis, L. Favre, D. Kechrakos, A. Kleibert, K.-H. Meiwes-Broer, G. M. Pastor, A. Perez, O. Toulemonde, K. N. Trohidou, J. Tuaillon, and Y. Xie, Surf. Sci. Rep. **56**, 189 (2005).

[5] Y. Li, M. Farle, and K. Baberschke, Phys. Rev. B **41**, 9596 (1990); Y. Li and K. Baberschke, Phys. Rev. Lett. **68**, 1208 (1992).

[6] A. Hucht and K. D. Usadel, Phys. Rev. B **55**, 12309 (1997).

[7] N. Sandschneider and W. Nolting, Phys. Rev. B **79**, 184423 (2009).

[8] S. Schwieger, J. Kienert, K. Lenz, J. Lindner, K. Baberschke, and W. Nolting, Phys. Rev. Lett. **98**, 057205 (2007).

[9] N. Saratz, A. Lichtenberger, O. Portmann, U. Ramsperger, A. Vindigni, and D. Pescia, Phys. Rev. Lett. **104**, 077203 (2010).

[10] L. M. Falicov, D. T. Pierce, S. D. Bader, R. Gronsky, K. H. Hathaway, H. J. Hopster, D. N. Lambeth, S. S. P. Parkin, G. Prinz, M. Salamon, I. K. Schuller, and R. H. Victora, J. Mater. Res. **5**, 1299 (1990); M. T. Johnson, P. J. H. Bloemen, F. J. A. den Broeder, and J. J. de Vries, Rep. Prog. Phys. **59**, 15409 (1996); J. Shen and J. Kirschner, Surf. Sci. **500**, 300 (2002).

[11] C. A. F. Vaz, J. A. C. Bland, and G. Lauhoff, Rep. Prog. Phys. **71**, 056501 (2008).

[12] A. Lisfi, J. C. Lodder, H. Wormeester, and B. Poelsema, Phys. Rev. B **66**, 174420 (2002); J. Langer, J. H. Dunn, A. Hahlin, O. Karis, R. Sellmann, D. Arvanitis, and H. Maletta, *ibid.* **66**, 172401 (2002); H. J. Choi, W. L. Ling, A. Scholl, J. H. Wolfe, U. Bovensiepen, F. Toyama, and Z. Q. Qiu, *ibid.* **66**, 014409 (2002).

[13] S. Förster, G. Baum, M. Müller, and H. Steidl, Phys. Rev. B **66**, 134427 (2002); G. Gubbiotti, G. Carlotti, M. G. Pini, P. Politi, A. Rettori, P. Vavassori, M. Ciria, and R. C. O'Handley, *ibid.* **65**, 214420 (2002); R. Vollmer, S. van Dijken, M. Schleberger, and J. Kirschner, *ibid.* **61**, 1303 (2000).

[14] S.-K. Lee, J.-S. Kim, B. Kim, Y. Cha, W. K. Han, H. G. Min, J. Seo, and S. C. Hong, Phys. Rev. B **65**, 014423 (2001); W. Wulfhekel, T. Gutjahr-Löser, F. Zavaliche, D. Sander, and J. Kirschner, *ibid.* **64**, 144422 (2001); H. L. Meyerheim, D. Sander, R. Popescu, J. Kirschner, P. Steadman, and S. Ferrer, *ibid.* **64**, 045414 (2001).

[15] M. Farle, W. Platow, A. N. Anisimov, P. Poulopoulos, and K. Baberschke, Phys. Rev. B **56**, 5100 (1997).

[16] P. Poulopoulos, P. J. Jensen, A. Ney, J. Lindner, and K. Baberschke, Phys Rev. B **65**, 064431 (2002).

[17] R. K. Kawakami, Ernesto J. Escorcia-Aparicio, and Z. Q. Qiu, Phys. Rev. Lett. **77**, 2570 (1996); Hyuk J. Choi, Z. Q. Qiu, J. Pearson, J. S. Jiang, D. Li, and S. D. Bader, Phys. Rev. B **57**, R12713 (1998); R. K. Kawakami, M. O. Bowen, H. J. Choi, E. J. Escorcia-Aparicio, and Z. Q. Qiu, *ibid.* **58**, R5924 (1998).

[18] D. P. Pappas, K.-P. Kämper, B. P. Miller, H. Hopster, D. E. Fowler, A. C. Luntz, C. R. Brundle, and Z.-X. Shen, J. Appl. Phys. **69**, 5209 (1991); R. Allenspach and A. Bischof, Phys. Rev. Lett. **69**, 3385 (1992); R. Allenspach, J. Magn. Magn. Mater. **129**, 160 (1994).

[19] R. Pfandzelter and M. Potthoff, Phys. Rev. B **64**, 140405 (2001).

[20] E. Kisker, K. Schröder, W. Gudat, and M. Campagna, Phys. Rev. B **31**, 329 (1985); J. Kirschner, M. Globl, V. Dose, and H. Scheidt, Phys. Rev. Lett. **53**, 612 (1984); J. Kirschner, Phys. Rev. B **30**, 415 (1984).

[21] M. Pratzer and H. J. Elmers, Phys. Rev. B **69**, 134418 (2004).

[22] H. Hasegawa, J. Phys. F: Metal Phys. **16**, 347 (1986).

[23] H. Hasegawa, J. Phys. F: Metal Phys. **17**, 165 (1987).

[24] H. Hasegawa, Surf. Sci. **182**, 591 (1987).

[25] H. Hasegawa, J. Phys.: Condens. Matter **1**, 9325 (1989).

[26] J. B. Staunton and B. L. Gyorffy, Phys. Rev. Lett. **69**, 371 (1992); S. S. A. Razee, J. B. Staunton, L. Szunyogh, and B. L. Györffy, Phys. Rev. B **66**, 094415 (2002); J. B. Staunton, S. Ostanin, S. S. A. Razee, B. L. Györffy, L. Szunyogh, B. Ginatempo, and E. Bruno, Phys. Rev. Lett. **93**, 257204 (2004).

[27] M. Dantziger, B. Glinsmann, S. Scheffler, B. Zimmermann, and P. J. Jensen, Phys. Rev. B **66**, 094416 (2002).

[28] J. C. Neto, J. R. de Sousa, and J. A. Plascak, Phys. Rev. B **66**, 064417 (2002).

[29] M. Pajda, J. Kudrnovský, I. Turek, V. Drchal, and P. Bruno, Phys. Rev. Lett. **85**, 5424 (2000); Phys. Rev. B **64**, 174402 (2001).

[30] F. López-Urías, G. M. Pastor, and K. H. Bennemann, J. Appl. Phys. **87**, 4909 (2000).

[31] T. Herrmann, M. Potthoff, and W. Nolting, Phys. Rev. B **58**, 831 (1998); J. H. Wu, H. Y. Chen, and W. Nolting, *ibid.* **65**, 014424 (2001).

[32] P. Bruno, Phys. Rev. Lett. **87**, 137203 (2001).

[33] A. I. Lichtenstein, M. I. Katsnelson, and G. Kotliar, Phys. Rev. Lett. **87**, 067205 (2001).

[34] G. M. Pastor, J. Dorantes-Dávila, and K. H. Bennemann, Phys. Rev. B **70**, 064420 (2004).

[35] R. Garibay-Alonso, J. Dorantes-Dávila, and G. M. Pastor, Phys. Rev. B **73**, 224429 (2006).

[36] R. Garibay-Alonso, J. Dorantes-Dávila, and G. M. Pastor, Phys. Rev. B **79**, 134401 (2009).

[37] J. Dorantes-Dávila, G. M. Pastor, and K. H. Bennemann, Solid State Commun. **60**, 465 (1986).

[38] S. Polesya, O. Šipr, S. Bornemann, J. Minár, and H. Ebert, Europhys. Lett. **74**, 1074 (2006).

[39] Y. Kakehashi, J. Phys. Soc. Jpn. **50**, 2251 (1981).

[40] Peter Fulde, *Electron Correlations in Molecules and Solids*, Springer Series in Solid State Sciences, Vol. 100 (Springer-Verlag, Berlin, Heidelberg, New York, 1991).

[41] J. Hubbard, Phys. Rev. B **19**, 2626 (1979); **20**, 4584 (1979); H. Hasegawa, J. Phys. Soc. Jpn. **49**, 178 (1980); **49**, 963 (1980).

[42] The contribution of atom l to E_K is approximately proportional to $\sqrt{z_l}$ in the second moment approximation.

[43] J. A. Morkowski and P. Wosicki, J. Magn. Magn. Mater. **71**, 285 (1988).

[44] For the sake of clarity, a "hat" (ˆ) is used to distinguish operators from numbers.

[45] In fact, a term of the form $\Delta\hat{H}_0 = \frac{1}{2}\sum_l\{(U_{dd} - J_{dd}/2)\hat{N}_l^d + U_{ss}\hat{N}_l^s\}$ should be subtracted from the single-particle Hamiltonian $\hat{H}_0$.

[46] A. Vega, J. Dorantes-Dávila, L. C. Balbás, and G. M. Pastor, Phys. Rev. B **47**, 4742 (1993); P. Alvarado, J. Dorantes-Dávila, and H. Dreyssé, *ibid.* **50**, 1039 (1994).

[47] E. N. Economou, *Green's Functions in Quantum Physics*, Springer Series in Solid State Sciences, Vol. 7 (Springer, Heidelberg, 1983).

[48] R. J. Elliot, J. A. Krumhansl, and P. L. Leath, Rev. Mod. Phys. **46**, 465 (1974).

[49] D. A. Papaconstantopoulos, *Handbook of the Band Structure of Elemental Solids* (Plenum, New York, 1996).

[50] R. Haydock, in *Solid State Physics*, edited by H. Ehreinreich, F. Seitz, and D. Turnbull (Academic, New York, 1980), Vol. 35, p. 215. $\rho_{i\alpha\sigma}(E) = -1/\pi\,\mathrm{Im}\{G_{i\alpha\sigma}\}$, where $G_{i\alpha\sigma}$ refers to the local Green's function.

[51] W. Nolting, W. Borgiel, V. Dose, and T. Fauster, Phys. Rev. B **40**, 5015 (1989).

[52] F. J. Himpsel, J. A. Knapp, and D. E. Eastman, Phys. Rev. B **19**, 2919 (1979).

[53] P. LLoyd, J. Phys. C **2**, 1717 (1969).

[54] F. Huang, M. T. Kief, G. J. Mankey, and R. F. Willis, Phys Rev. B **49**, 3962 (1994).

[55] A. Ney, A. Scherz, P. Poulopoulos, K. Lenz, H. Wende, K. Baberschke, F. Wilhelm, and N. B. Brookes, Phys Rev. B **65**, 024411 (2001).

[56] H. L. Meyerheim, D. Sander, N. N. Negulyaev, V. S. Stepanyuk, R. Popescu, I. Popa, and J. Kirschner, Phys. Rev. Lett. **100**, 146101 (2008).

[57] R. Garibay-Alonso, M. Reyes-Reyes, E. Urrutia-Bañuelos, and R. López-Sandoval, J. Phys. Condens. Matter. **22**, 056001 (2010).

[58] A. Enders, D. Peterka, D. Repetto, N. Lin, A. Dmitriev, and K. Kern, Phys. Rev. Lett. **90**, 217203 (2003).

[59] J. Dorantes-Dávila, H. Dreyssé, and G. M. Pastor, Phys. Rev. Lett. **91**, 197206 (2003).