Magnetic properties of fcc Ni-based transition metal alloys

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Electronic properties and finite-temperature magnetism of Ni-based transition metal alloys with the face-centered cubic structure are studied theoretically by *ab initio* calculations. While the calculated total and local magnetic moments agree well with the experiment, the evaluation of the Curie temperature from first principles represents a much more delicate problem. The mean-field approximation and the random-phase approximation (RPA), as well as the renormalized RPA by Bruno that was extended to random alloys, were tested: the latter giving the most satisfactory agreement with the experiment for a broad class of Ni-based alloys of the type $Ni_{1-x}M_x$ (M=Cu, Pd, Co, Fe, and Mn) over the whole concentration range.

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I. INTRODUCTION

Electronic and magnetic properties of transition metal alloys were intensively studied in the past, both experimentally and theoretically. For example, the concentration trend of the magnetization in the $Ni_{1-x}Cu_x$ alloys is shown in any standard textbook of the solid-state physics.[1] In spite of this, a detailed understanding of the concentration dependence of magnetization in random magnetic alloys, particularly on the first-principle level, is more recent.[2–6] Also, the above first-principle studies had accurately described some of the conventional explanations. On the other hand, a reliable estimation of the alloy Curie temperature from the first-principles is less frequent and the problem itself still represents a challenge for the solid-state theory. We mention a recent paper (Ref. 7) in which the authors study some fcc- and bcc-magnetic transition alloys using the semiempirical approach on Mano[8] to estimate the Curie temperature.

The two-step approach, as suggested by Lichtenstein *et al.*,[9] became popular and rather successful in explaining thermodynamical properties of a broad class of magnetic materials.[10] In the first step, the total energies of the perturbed reference ferromagnetic (FM) (Refs. 9–12) or disordered-local moment (DLM) (Refs. 13 and 14) states are mapped onto the classical Heisenberg Hamiltonian. By the perturbation, we mean (classical) spin rotations at two different lattice sites. The corresponding total-energy change is related to the exchange integral, which appears in the Heisenberg Hamiltonian.[9] In the second step, the resulting random Heisenberg Hamiltonian is studied by methods of statistical physics. Typically, the mean-field approximation (MFA),[9] the Monte-Carlo (MC) simulation,[11,13] and the random-phase approximation (RPA) (Ref. 12) are used, the latter two approaches giving more reliable estimates of the critical temperature as compared to the MFA. Usually, the results of the MC and RPA estimates are similar.[10] While estimates of the Curie temperature of bcc Fe were rather successful when compared to the experiment, the Curie temperature of fcc Ni was underestimated.[11–13] It was clarified recently that the most probable reason for this disagreement between the theory and the experiment is the inadequacy of the adiabatic approximation for fcc Ni and a simple correction was suggested by various authors.[15–17] A good agreement for the calculated Curie temperature for fcc Ni was obtained while preserving previous good estimates for bcc Fe. Of the above approaches, the renormalized RPA method of Bruno[15] seems to be the most suitable for generalization to random substitutional Ni-rich magnetic alloys.

The problem of randomness is an additional new feature that appears in simulations. There are only few first-principles calculations of the finite-temperature magnetism of disordered alloys because of the subtlety of correctly treating the effect of disorder. A recent interest in diluted magnetic semiconductors seems to change this situation and progress in the understanding of the problem has been achieved independently by few groups.[18] It is therefore challenging for the theory to investigate the disordered Ni-based magnetic alloys such as fcc $Ni_{1-x}M_x$, where M is the other, either nonmagnetic or magnetic, transition metal. The reason is a combination of two effects; namely, the presence of randomness and the inadequacy of the adiabatic approximation expected in such alloys, particularly in the Ni-rich ones. It is the main purpose of this paper, to perform such a study for broad concentration ranges of both the nonmagnetic (Cu and Pd) and magnetic components (Co and Fe), where the face-centered cubic (fcc) phase exists, and to compare the results with the experiment. Finally, we consider also Ni-rich NiMn alloys for which the existence of two magnetic states in the ferromagnetic phase was found.[5] For completeness, we also show results for the concentration dependence of magnetic moments, but for this quantity, the first-principle approach is

quite successful, as it was demonstrated previously (see e.g., Refs. 2–4 and 6).

II. FORMALISM

The classical Heisenberg Hamiltonian, as used in the framework of the two-step model, has the form

$$H_{\mathrm{eff}} = -\sum_{i,j} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j, \tag{1}$$

where i,j is the site index, $\mathbf{e}_i$ is the unit vector pointing along the direction of the local magnetic moment at the site i, and J_{ij} is the exchange integral between magnetic atoms at sites i and j, which by construction contains the values of atom magnetic moments and its positive (negative) value indicates the ferromagnetic (antiferromagnetic) coupling. There is just one kind of exchange integral, $J_{i,j}^{\mathrm{Ni,Ni}}$, in the case of Ni-based alloys with nonmagnetic atoms (Cu or Pd). More precisely, the exchange integrals between Ni moments, and small induced moments on Cu and Pd atoms, are very small and can be safely neglected. This is even more justified for exchange integrals between small induced moments on nonmagnetic atoms. On the other hand, there are three kinds of exchange integrals in the Ni-based alloys containing magnetic impurities, namely, $J_{i,j}^{Q,Q'}$ (Q,Q'=Ni, Co; Ni, Fe; and Ni, Mn). In both cases it is assumed that atoms are distributed randomly on the host fcc lattice. An extension of the two-step model to random alloys, particularly the evaluation of corresponding exchange integrals $J_{i,j}^{Q,Q'}$, is discussed in Ref. 19 and we refer the reader for details there.

The reference electronic structure calculations were performed using the tight-binding linear muffin-tin orbital (TB-LMTO) approach and the effect of disorder was described by the coherent-potential approximation (CPA).[20] The same radii for constituent atoms were used in the TB-LMTO-CPA calculations, but in the case of alloys with very different atom sizes (NiPd), we have included local Madelung corrections[21] to constituent LDA potentials. All calculations were performed for experimental lattice constants measured at low temperatures.[22–24] With the exception of Fe-rich NiFe alloys, all studied systems obey reasonably well Vegard's law.

It should be noted that exchange integrals for random alloy can be also determined in the framework of the supercell approach. In this case, one obtains naturally exchange interactions, which fluctuate in their size due to varying local environment. It was shown in Ref. 25 that if such fluctuating exchange interactions are properly averaged, their values are close to those obtained from the CPA approach.

Some comments are needed on the application of the MFA, the RPA, and the renormalized RPA to the case of disordered magnetic alloys. The simplest approach to this problem is to use the averaged-lattice model or the virtual-crystal approximation. In the framework of the averaged-lattice model, the disordered Heisenberg Hamiltonian corresponding to random $A_{1-x}B_x$ alloy is treated as a crystal with the nonrandom effective exchange interactions

$$J_{ij}^{\mathrm{eff}} = (1-x)^2 J_{ij}^{AA} + x(1-x)(J_{ij}^{AB} + J_{ij}^{BA}) + x^2 J_{ij}^{BB}. \tag{2}$$

By using the above effective exchange interactions, we can directly employ the MFA, RPA, or renormalized RPA estimates of the Curie temperatures for crystals and also for random magnetic alloys. The validity of the averaged-lattice approximation was questioned in recent studies[18] of diluted magnetic semiconductors because of its neglect of the magnetic percolation effect. This effect is, however, relevant for alloys with low concentration of magnetic atoms in a nonmagnetic host, which have, in addition, spatially well-localized exchange interactions. In concentrated magnetic alloys, the effect of magnetic percolation is much less important (see Fig. 1 in Ref. 26 and discussion there). It should be noted that ferromagnetism disappears for low concentration of Ni impurities in Cu or Pd hosts. The situation is even more favorable in the case of the alloys of Ni with Co, Fe, or Mn atoms because now all sites are occupied by magnetic atoms like in a crystal (although with different local moments). We have recently demonstrated the success of the averaged-lattice model in the case disordered $Ni_{2-x}MnSb$ Heusler alloys.[27]

Once the effective exchange integrals are determined, the MFA estimate of the Curie temperature is

$$k_B T_c^{\mathrm{MFA}} = \frac{2}{3} \sum_{i \neq 0} J_{0i}^{\mathrm{eff}}, \tag{3}$$

where the sum extends over many nearest-neighbor (NN) shells. An improved description of finite-temperature magnetism is provided by the RPA, which is given by

$$(k_B T_c^{\mathrm{RPA}})^{-1} = \frac{3}{2} \frac{1}{N} \sum_{\mathbf{q}} [J^{\mathrm{eff}}(0) - J^{\mathrm{eff}}(\mathbf{q})]^{-1}. \tag{4}$$

Here N denotes the number of $\mathbf{q}$ vectors used in the sum over the Brillouin zone and $J^{\mathrm{eff}}(\mathbf{q})$ is the lattice Fourier transform of the real-space exchange integrals J_{ij}^{eff}. It can be shown that T_c^{RPA} is always smaller than T_c^{MFA}.[12] We have used up to 231 shells in the MFA [Eq. (3)] and in the lattice Fourier transform of $J^{\mathrm{eff}}(\mathbf{q})$ in Eq. (4), and tested the convergence with respect to the number of shells included in the statistical study. The estimated computational error corresponding to a limited number of shells used in calculations is below ± 5 K. We refer the reader to Refs. 10 and 12 for more details. Finally, the Curie temperature estimate $\tilde{T}_c^{\mathrm{RPA}}$ in the framework of the renormalized RPA approach is[15]

$$(k_B \tilde{T}_c^{\mathrm{RPA}})^{-1} = (k_B T_c^{\mathrm{RPA}})^{-1} - \frac{6}{M^{\mathrm{eff}} \Delta^{\mathrm{eff}}}. \tag{5}$$

Here $X^{\mathrm{eff}} = M^{\mathrm{eff}}$ and Δ^{eff} are the calculated averaged magnetic moment and the corresponding exchange splitting. The latter is estimated from the difference $\Delta^Q = C_d^{Q,\uparrow} - C_d^{Q,\downarrow}$ of the potential parameter $C_d^{Q,\sigma}$ of the LMTO theory for d states ($Q = A,B$). We thus have $X^{\mathrm{eff}} = (1-x)X^A + xX^B$, where $X^Q = M^Q$, Δ^Q, and A=Ni while B=Cu, Pd, Co, Fe, Mn. While such a choice seems to be the natural one for the averaged-lattice model and it is consistent with Eq. (2), we are not able, at present, to give a better theoretical justification. It is also

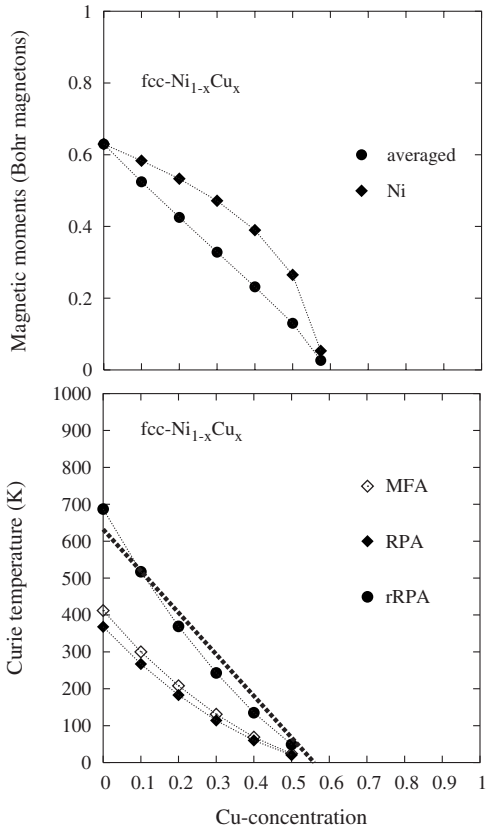

FIG. 1. fcc-$Ni_{1-x}Cu_x$ disordered alloy. Upper frame: Averaged and component-resolved magnetic moments as a function of the alloy composition. Lower frame: Curie temperatures as a function of the alloy composition for various approximations (MFA, RPA, and renormalized RPA). The dashed line is the experiment.

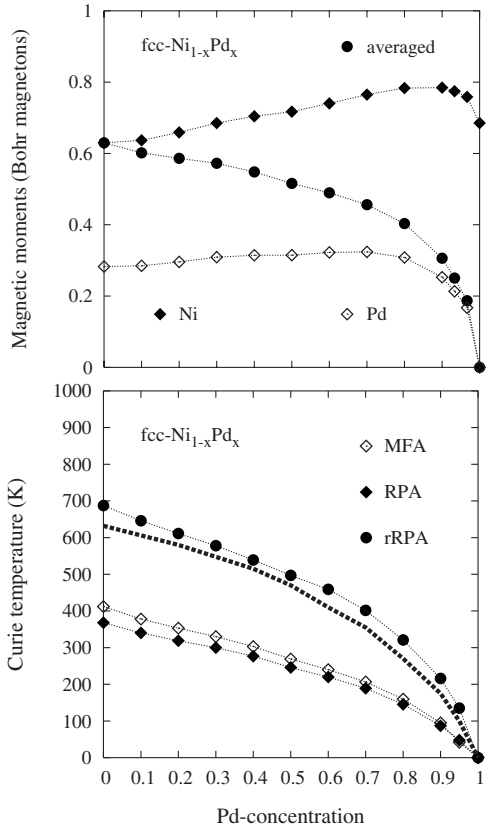

FIG. 2. The same as in Fig. 1 but for fcc-$Ni_{1-x}Pd_x$ disordered alloy.

obvious from Eq. (5) that the renormalized Curie temperature is enhanced, as compared to the unrenormalized one.

III. RESULTS AND DISCUSSION

Results for magnetic moments and Curie temperatures of $Ni_{1-x}Cu_x$, $Ni_{1-x}Pd_x$, $Ni_{1-x}Co_x$, $Ni_{1-x}Fe_x$, and $Ni_{1-x}Mn_x$ alloys are summarized in Figs. 1–4 and 6, respectively.

A. $Ni_{1-x}Cu_x$ alloys, Fig. 1

We shall start with explanation of magnetic behavior of NiCu alloys. In spite of its prototypical character, the first-principles study of the alloy thermodynamics, particularly of the concentration dependence of the alloy Curie temperature, is missing in the literature. The averaged magnetization decreases linearly with Cu contents, as seen from the figure. A simple explanation of this effect is a gradual filling of Ni band (0.6 holes per atom) due to the Cu atoms so that for $x_{Cu} \approx 0.6$, we have a nonmagnetic state.[1] The calculated averaged and Ni-local moments agree well with the experiment,[23] as well as with previous theoretical studies.[3,4] In agreement with the experiment, we find an almost linear decrease of the averaged magnetic moment with Cu concentration. There is a small induced moment on Cu sites (smaller than $0.01\mu_B$, not shown). While qualitatively correct, the above simple explanation of the concentration dependence of the magnetization misses some details. The interatomic *sp-d* charge-transfer effects on Ni atoms and the change of the local Ni density of states with alloying leads to an almost perfect charge neutrality, which underlines the textbook explanation.[2,4] We find the extinction of magnetization for about $x_{Cu}=0.58$.

The linear decrease of the Curie temperature and its magnitude as a function of the Cu concentration, as observed in the experiment, is reasonably well reproduced only by the renormalized RPA. On the other hand, a significant bending of the concentration dependence of the Curie temperature is found for both the MFA and RPA approaches. For each alloy concentration, the MFA value of the Curie temperature is larger than the RPA value, as expected. In both cases, however, calculated Curie temperatures are underestimated, particularly on the Ni-rich end. This is a direct consequence of the violation of the adiabatic approximation in Ni-rich NiCu alloys. On the other hand, the renormalized RPA takes this effect approximately into account, which leads to a good agreement with the experiment.

B. $Ni_{1-x}Pd_x$ alloys, Fig. 2

This system is similar to the above studied NiCu alloy in the sense that only one of the alloy components (Ni) is magnetic as a pure crystal. In contrast to Cu atoms, Pd atoms can be easily polarized and, thus, carry non-negligible induced local magnetic moments. The calculated averaged magnetic

moments in fcc NiPd alloys agree well with the experimental results in Ref. 28 while slightly smaller values, particularly in Pd-rich region, are reported in Ref. 29 ($m_{aver}=0.3\mu_B$ and $0.47\mu_B$ for $x_{Pd}=0.7$ in Refs. 28 and 29, respectively). The component-resolved magnetic moments were also estimated in the experiment.[29] The general trend is well reproduced in our calculations: an increase of local magnetic moments on Ni atoms with a maximum in Pd-rich region and a much weaker concentration variation of Pd moments with Pd concentration. The calculated moments on Ni sites are slightly smaller than those estimated in Ref. 29, e.g., the largest calculated and measured moments for Ni atoms are $0.8\mu_B$ and $1.1\mu_B$, and for Pd atoms $0.3\mu_B$ and $0.25\mu_B$, respectively.

Calculated Curie temperatures in the framework of the renormalized RPA agree again very well with the experimental data.[28] On the other hand, both the MFA and RPA approximations underestimate the Curie temperature. The differences are largest for Ni-rich alloy, as expected. The reason is the same as for CuNi alloys: the violation of the adiabatic theorem, as explained in Ref. 15. There is a good agreement between the present MFA calculations and an early study of NiPd alloys[9] in the framework of the MFA. Only the first three shells of exchange integrals were used in Ref. 9, as contrasted with many shells employed here. The explanation is a rather localized character of exchange integrals in Ni (Ref. 12) and Ni-rich alloys.

Curie temperatures of both NiCu and NiPd alloys decrease monotonically with concentration of the nonmagnetic component. This is due to the fact that exchange interactions $J_{ij}^{\mathrm{Ni,Ni}}$ between Ni moments depend only weakly on composition: they slightly decrease in NiCu and slightly increase in NiPd due to the high polarizability of Pd atoms. Because other exchange interactions in NiCu and NiPd alloys are negligible, the magnitude of effective interactions J_{ij}^{eff} [Eq. (2)] and, thus, also Curie temperatures decrease with Cu (Pd) concentrations monotonically in NiCu (NiPd) alloys.

C. $Ni_{1-x}Co_x$ alloys, Fig. 3

This alloy exists in fcc phase up to about 80% of Co, and over this concentration range, the averaged magnetic moment increases linearly with Co content.[22,23] This concentration trend, as well as magnitudes of the averaged moments, are well reproduced by present calculations. The largest averaged moment is close to $1.5\mu_B$ for $x_{Co}=0.8$ in agreement with the experiment. The concentration dependence of the local Co- and Ni-magnetic moments is very weak in accordance with the experiment.[23] This fact also explains the observed linear dependence of the averaged magnetic moment on the alloy composition.

We shall discuss first the Curie temperature of fcc phase of Co.[10,12] The MFA and RPA values are, respectively, 1645 and 1311 K, and are to be compared with the experimental value of 1395 K. It should be noted that the experimental value is for the hcp phase but it is close to the fcc phase. The renormalized RPA enhances the RPA value of the Curie temperature to 1770 K. While the RPA value underestimates the experimental one only by about 6%, the renormalized RPA overestimates it by about 25% despite the fact that it has

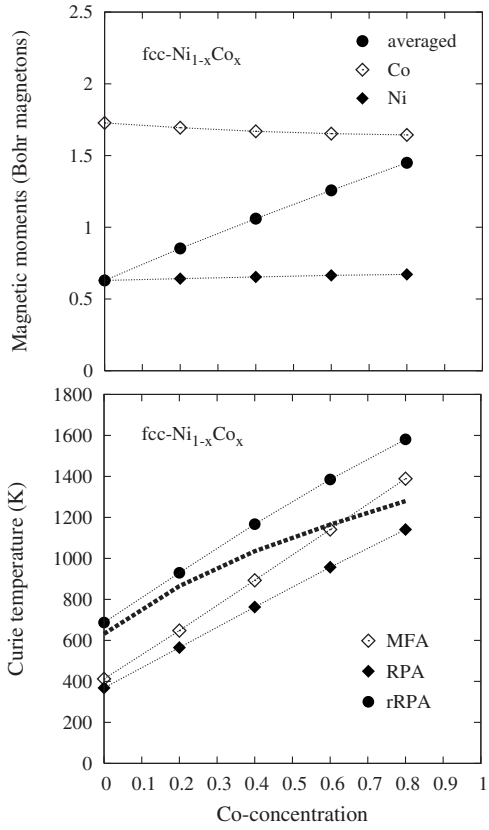

FIG. 3. The same as in Fig. 1 but for fcc-$Ni_{1-x}Co_x$ disordered alloy.

improved results for both Ni and Fe.[15] The origin of this failure of the renormalized RPA for the case of pure Co metal is not clear yet. The renormalized RPA is physically better justified than the RPA for systems with less developed local magnetic moment, which is also the case of cobalt. The success of the RPA for Co-rich NiCo alloys can be, thus, fortuitous. One of the possible mechanisms (in addition to the inclusion of longitudinal fluctuations) that could influence the calculated Curie temperature is the electronic entropy, namely, the fact that exchange integrals should be evaluated at the Curie temperature rather than at $T=0$ K, as is usually done. Modifications due to the electronic entropy could lead to a non-negligible reduction of high Curie temperatures, which is the case of the cobalt.

In agreement with the above facts, we find a good agreement of calculated Curie temperature with the experiment in the framework of the renormalized RPA in the Ni-rich region. On the other hand, the RPA agrees better with experimental data for Co-rich alloys ($x_{Co}>0.4$). Incidentally, the MFA agrees in this concentration range with the experiment even slightly better than the RPA.

The monotonic increase of the Curie temperature is easy to understand from Eq. (2): exchange interactions $J_{ij}^{Q,Q'}$ $(Q,Q'=A,B)$ are all weakly depending on the composition while $J_{ij}^{\mathrm{Co,Co}}$ are a few times larger as the remaining two interactions. As a result, effective interactions J_{ij}^{eff} increase monotonically and, thus, also increases the Curie temperature.

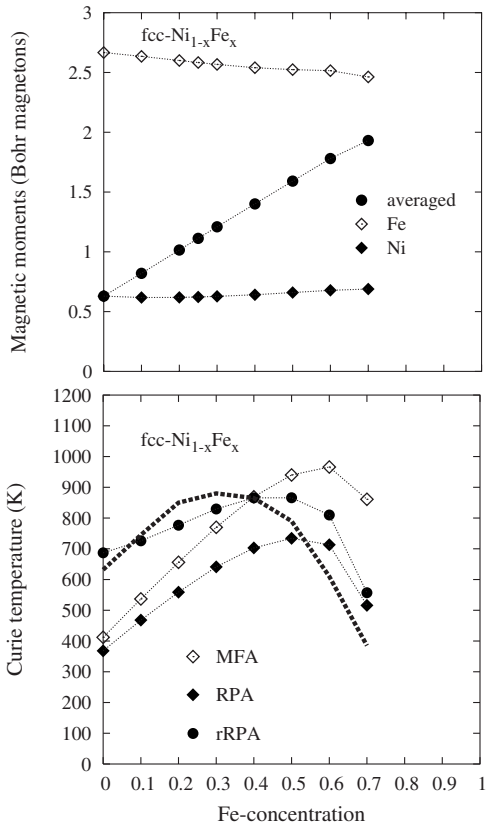

FIG. 4. The same as in Fig. 1 but for fcc-$Ni_{1-x}Fe_x$ disordered alloy.

D. $Ni_{1-x}Fe_x$ alloys, Figs. 4 and 5

The NiFe alloys were intensively studied in the past.[23] The fcc phase exists in the region $x_{Fe} \in (0, 0.7)$ while for higher Fe concentrations the bcc phase is stable. There are two specific concentrations to be mentioned, namely, the Ni-rich permalloy alloy (20%–25% of Fe) and the Fe-rich invar-type alloys (65% of Fe).

Measured and calculated magnetic moments agree again reasonably well.[23] Similarly to NiCo alloys, the weak concentration dependence of local Fe- and Ni-magnetic moments results in an almost linear increase of the average magnetic moment with Fe content. The local Ni moment is close to its bulk value of $0.6–0.7\mu_B$, the local Fe moment on the fcc lattice is about $2.5–2.7\mu_B$.

The experimental Curie temperature exhibits a dramatic change with the Fe concentration having the maximum for about 30% of Fe and then decreases. This trend is qualitatively reproduced by all approximations used. The maximum of the Curie temperature is, however, shifted to higher Fe concentrations: 60% for the MFA, 50% for the RPA, and it is between 40%–50% for the renormalized RPA. Both the MFA and RPA underestimate the experimental Curie temperature in the Ni-rich concentration range. The renormalized RPA gives the most satisfactory overall agreement with the experiment also concerning the calculated Curie temperatures. It should be noted, however, that the slope of the concentration dependence of the Curie temperature in the Ni-rich region is larger in the experiment, as compared to that obtained from the renormalized RPA.

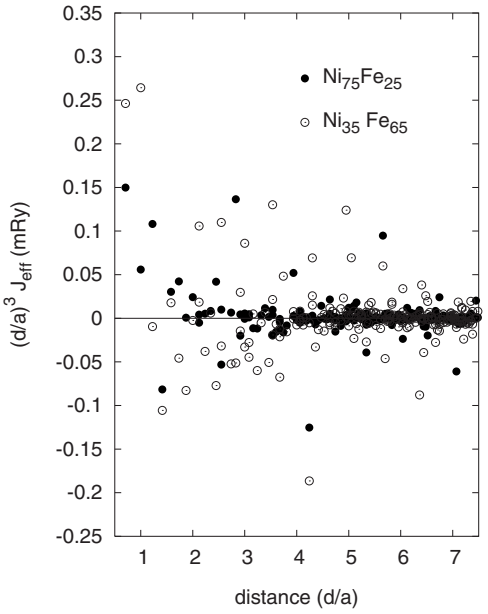

FIG. 5. Effective exchange interactions [Eq. (2)] as a function of the distance d (in units of the lattice constant a) for two fcc-$Ni_{1-x}Fe_x$ alloys: $x=0.25$ and $x=0.65$. Exchange interactions are multiplied by the factor $(d/a)^3$.

We also wish to address here the origin of the concentration maximum of the Curie temperature in fcc-NiFe alloys, which is in a striking contrast to the monotonic composition variation found in fcc-NiCu, NiPd, and NiCo alloys. The maximum is an interplay of the two trends: (i) the increase of effective exchange integrals J_{ij}^{eff} due to increasing Fe content, which also increases the Curie temperature, and (ii) the increasing amount of the frustration in the Fe-rich sample, which decreases the Curie temperature. By the frustration, we mean an increasing amount of the antiferromagnetic couplings in the Fe-rich alloy. The long-range oscillating character of exchange interactions in fcc Fe was discussed recently[25] and, as it is illustrated in Fig. 5, it is also present in Fe-rich alloys. We demonstrate this fact by comparing exchange interactions for fcc-$Ni_{75}Fe_{25}$ and fcc-$Ni_{35}Fe_{65}$ alloys in Fig. 5. It is also obvious from Fig. 5 that the magnitude of the dominating first interactions are larger in fcc $Ni_{35}Fe_{65}$ but the observed decrease of the Curie temperature is really due to the large amount of antiferromagnetic couplings. It is also seen that the real-space extent of exchange interactions is significantly larger in fcc $Ni_{35}Fe_{65}$, as compared to fcc $Ni_{75}Fe_{25}$. The short-range ferromagnetic character of interactions in fcc Ni is well documented in the literature[12] and it should be contrasted with the long-range oscillating behavior of interactions in fcc Fe.[25] The Ni-rich and Fe-rich NiFe alloys, thus, share relevant features of corresponding pure crystal counterparts.

E. $Ni_{1-x}Mn_x$ alloys, Figs. 6–8

The last system we consider are Ni-rich NiMn alloys, which are ferromagnetic over the limited range of Mn-impurity concentration $x_{Mn} \lesssim 0.25$. The magnetic properties of this alloy were analyzed in detail in Ref. 5. The main

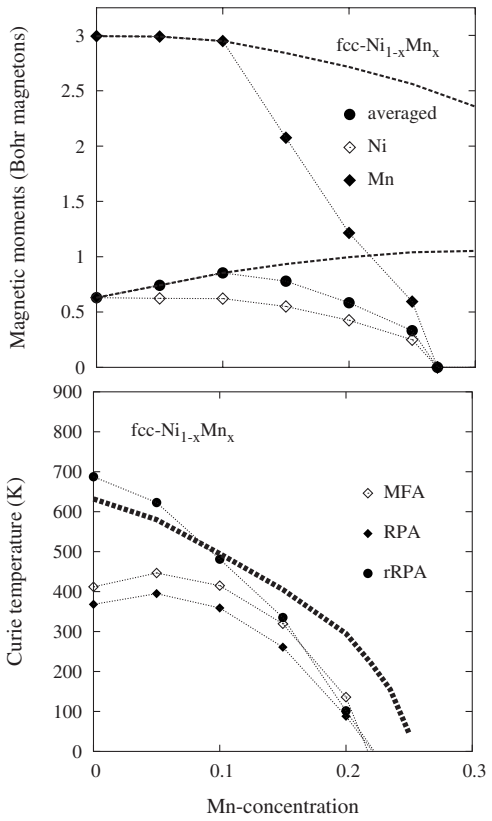

FIG. 6. The same as in Fig. 1 but for fcc-$Ni_{1-x}Mn_x$ disordered alloy. The thick dashed lines in the upper frame indicate averaged and local Mn moments, assuming the ferromagnetic state for all concentrations.

conclusion was that the system is ferromagnetic but the transition to the magnetic state with local moments parallel and antiparallel to the average magnetization (uncompensated DLM state) starts for $x_{\mathrm{Mn}} \in 0.1, 0.15$. Measured and calculated magnetic moments (Fig. 6) agree again reasonably well with those of Ref. 5 and experiment[24] giving a pronounced maximum for $x_{\mathrm{Mn}} \approx 0.1$. Our calculations also show that the pure ferromagnetic state (dashed thick line) fails to reproduce the experiment if the experimental lattice constant is used. The agreement with the experiment for purely ferromagnetic state, as obtained in Ref. 6, is the result of a wrong concentration dependence of the alloy lattice constant, which strongly deviates from the experimental one (see Figs. 18–20 of Ref. 6). The concentration dependence of the local magnetic moments on Ni and Mn atoms also agree well with the neutron-diffraction experiment:[24] both local moments decrease with increasing x_{Mn} and vanish for $x_{\mathrm{Mn}} \approx 0.25$. Again, the breakdown for purely ferromagnetic description of the local Mn moment is evident (thick dashed line).

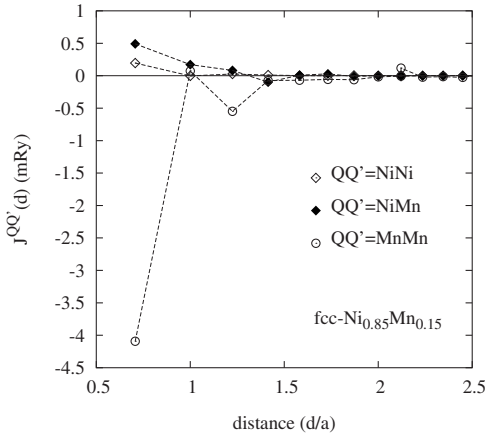

FIG. 7. Exchange interactions between Ni-Ni, Ni-Mn, and Mn-Mn pairs as a function of the distance d (in units of the lattice constant for fcc-$Ni_{0.85}Mn_{0.15}$ alloys.

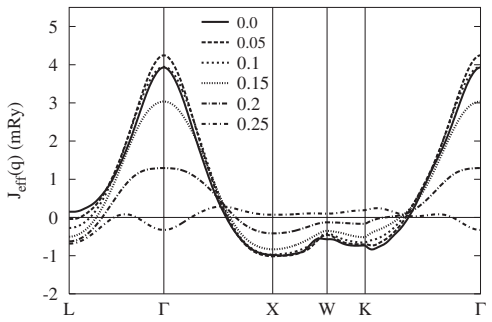

FIG. 8. Lattice Fourier transformation $J^{\mathrm{eff}}(\mathbf{q})$ of the real-space exchange integrals J_{ij}^{eff} plotted along the high-symmetry lines in the fcc-Brillouin zone.

Calculated and experimental Curie temperatures decrease monotonically with Mn content and we also mention a good quantitative agreement between experiment and the RPA model. The main difference is the onset of ferromagnetism that starts for slightly larger Mn concentration in the experiment. One can speculate that this could be partly due to the use of the averaged-lattice model that neglects the percolation effect. Because of Ni-rich character of the present alloy, both the MFA and RPA underestimate experimental Curie temperatures.

Exchange integrals in $Ni_{0.85}Mn_{0.15}$ are shown in Fig. 7. In agreement with phenomenological model based on the first NN interactions and measured Curie temperatures,[24] we observe a ferromagnetic character of Ni-Ni and Ni-Mn interactions but a large antiferromagnetic coupling for first NN Mn-Mn pairs. The large antiferromagnetic coupling between Mn atoms is an obvious reason for the extinction of the ferromagnetism at relatively small Mn-impurity concentration (about 25%). It should be noted that, in particular, Ni-Mn and Mn-Mn interactions extend beyond nearest-neighbor pairs in contrast with the phenomenological model.[24]

We also investigate the stability of the FM state as a function of the Mn content. To this end we show in Fig. 8 the concentration trend of the lattice Fourier transform $J^{\mathrm{eff}}(\mathbf{q})$ of the real-space exchange integrals J_{ij}^{eff}. The maximum (note the minus sign) in Eq. (1) corresponds to the magnetic ground state. A well-pronounced peak at $\mathbf{q}=\Gamma$ indicates the FM ground state. We observe a strong decrease of the stability of the FM state for $x_{\mathrm{Mn}} > 0.1$, in agreement with the onset of the uncompensated DLM, which becomes the ground state. Finally, a new complex magnetic ground state is formed for $x_{\mathrm{Mn}} \in (0.2, 0.25)$, in agreement with observed ex-

tinction of the ferromagnetism in this concentration range.[24]

In the present paper the ferromagnetic state was employed as a reference state for mapping to the Heisenberg Hamiltonian. It was suggested recently[13,14,30] that in some cases it can be more convenient to consider as a reference state the so-called disordered–local-moment state, combined possibly with the fixed-spin moment approach. In particular, it was demonstrated in Ref. 30 that such approach combined with an approximative treatment of the longitudinal fluctuations gives the Curie temperature of pure fcc Ni and bcc Fe in a better quantitative agreement with the experiment, as compared to the ferromagnetic reference state and the renormalized RPA approach adopted here. A generalization of such theory to the case of random alloys is still a challenge to the theory but it is definitely a step in a proper direction.

Recently, Takahashi *et al.*[7] studied the dependence of the Curie temperature in some transition metal alloys on the valence electron number (an analog of the Slater–Pauling curves) using the semiempirical approach of Mano.[8] They employed three leading exchange interactions found from first principles, similarly as done here, but their Curie temperature is not corrected for less developed moments such as, e.g., Ni. The present renormalized RPA estimates of the Curie temperature are in better quantitative agreement with the experiment for Ni-based alloys.

IV. CONCLUSION

We have presented a first-principles theory of electronic and thermodynamic properties for Ni-based magnetic transition metal alloys with both nonmagnetic (Cu and Ni) and magnetic (Co, Fe, and Mn) impurities over a broad concentration range. We have found a good agreement with the experiment and the previous theoretical studies for magnetic moments, both averaged and local ones. We have presented a systematic study of Curie temperatures of $Ni_{1-x}M_x$ compounds (M=Cu,Pd,Co,Fe,Mn), which was missing in the literature. We have used a simple generalization of the renormalized RPA theory on to random alloys based on the averaged-lattice model, which is justified for large component concentrations. While both the MFA and the RPA failed to give reasonable quantitative agreement with the experiment, particularly for Ni-rich alloys, the renormalized RPA approach, which accounts for violation of the adiabatic theorem, was successful over a broad range of concentrations. A generally weak concentration dependence of exchange interactions leads to a monotonic concentration dependence of the Curie temperature. Such behavior, however, is not observed in NiFe alloys with a pronounced concentration maximum of the Curie temperature. We have shown that this maximum is a result of interplay of two trends: the increase of the magnitude of effective exchange integrals with Fe concentration, and, at the same time, increasing frustration and long-range character of exchange integrals at the Fe-rich end. Ni-rich NiMn alloys are ferromagnetic only over a limited concentration range of Mn impurities. This is mainly due to large antiferromagnetic coupling between the nearest-neighbor Mn atoms. The concentration dependence of the Curie temperature is also reasonably well described by the present theory.

The theory should be improved in some respects, e.g., by removing the averaged-lattice model limitation, by investigating the effect of possible longitudinal fluctuations, by including the effect of electronic entropy relevant for systems with the high Curie temperature, or by considering a possible local environment effects that are observed in some alloys, e.g., in NiFe alloys, which all could bring the theory to a better quantitative agreement with the experiment.

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