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On-site Coulomb interaction and the magnetism of (GaMn)N and (GaMn)As

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We use the local-density approximation (LDA) and LDA+U schemes to study the magnetism of (GaMn)As and (GaMn)N for a number of Mn concentrations and varying number of holes. We show that for both systems and both calculational schemes the presence of holes is crucial for establishing ferromagnetism. For both systems, the introduction of U increases delocalization of the holes and, simultaneously, decreases the p-d interaction. Since these two trends exert opposite influences on the Mn-Mn exchange interaction the character of the variation of the Curie temperature T_C cannot be predicted without direct calculation. We show that the variation of T_C is different for two systems. For low Mn concentrations we obtain the tendency to increasing T_C in the case of (GaMn)N whereas an opposite tendency to decreasing T_C is obtained for (GaMn)As. We reveal the origin of this difference by inspecting the properties of the densities of states and holes for both systems. The main body of calculations is performed within a supercell approach. The Curie temperatures calculated within the coherent-potential approximation to atomic disorder are reported for comparison. Both approaches give similar qualitative behavior. The results of calculations are related to the experimental data.

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I. INTRODUCTION

After the discovery of the ferromagnetism of $Ga_{0.947}Mn_{0.053}As$ with the Curie temperature as high as 110 K (Ref. 1) the diluted magnetic semiconductors (DMS) became the subject of intensive experimental and theoretical studies being considered as promising materials for semiconductor spintronics. Although much new understanding of the physics of DMS has been won by the investigation of (GaMn)As, the Curie temperature of this prototype system could not be raised above 150 K.[2] The design of technologically useful DMS materials with the Curie temperature exceeding the room temperature remains a challenge.

The theoretical prediction by Dietl *et al.*[3] of the high-T_C ferromagnetism in (GaMn)N played an important role in the formulation of the directions of the search for new ferromagnetic DMS's. A number of experimental studies indeed detected the ferromagnetism of (GaMn)N samples with the Curie temperature higher than the room temperature.[4–6] However, the experimental data on the magnetic state of (GaMn)N are strongly scattered and range from a paramagnetic ground state to the ferromagnetic state with a very high Curie temperature of 940 K.

New proof of the complexity of the magnetism of (GaMn)N was given by the recent magnetic circular dichroism measurements by Ando.[7] The measurements were performed on a high-T_C sample and led Ando to the conclusion that the (GaMn)N phase in this sample is paramagnetic. The ferromagnetism of the sample was claimed to come from an unidentified phase. On the other hand, in a recent preprint by Giraud *et al.*[8] a high-temperature ferromagnetism was detected in the samples with a low Mn concentration of about 2%. The authors rule out the presence of precipitates in the system and argue that the room-temperature ferromagnetism is an intrinsic property of (GaMn)N. Thus the situation is controversial and calls for further research efforts. The purpose of the present work is a theoretical comparative study of (GaMn)As and (GaMn)N with a focus on the influence of the on-site Coulomb interaction on the Curie temperature of both systems. The calculations are performed on the basis of the density-functional theory (DFT) within both local-density approximation (LDA) and LDA+U (Ref. 9) approach.

Numerous LDA calculations of the electronic structure of (GaMn)As have been reported (see, e.g., reviews Refs. 10 and 11 and more recent calculations Refs. 12–15). For (GaMn)N the number of such calculations is still small (see Refs. 16–19 and recent preprint[20]). The study of the properties of both systems which goes beyond the LDA is at the very beginning. There are a short report on the LDA+U calculation for (GaMn)As (Ref. 21) and a very recent article[19] on LDA+U calculation for both systems. We mention also the preprint on the self-interaction corrected LDA (SIC-LDA) calculation[20] for these systems.

The present work is important in a number of aspects. First, we investigate the characteristics of the holes and discuss them from the viewpoint of the mediation of the ferromagnetic exchange interaction between the Mn impurities. Second, we calculate interatomic exchange interactions and the Curie temperatures. Third, we separate the contributions of the antiferromagnetic superexchange and the ferromagnetic kinetic exchange to the Curie temperatures. In all studies we focus on the influence of the on-site Coulomb interaction.

II. CALCULATIONAL SCHEME

The calculational scheme is discussed in Refs. 12 and 22 to which the reader is referred for more details. The scheme is based on the DFT calculations for supercells of semiconductor crystals with one Ga atom replaced by a Mn atom. The size of the supercell determines the Mn concentration. To calculate the interatomic exchange interactions we use the frozen-magnon technique and map the results of the total energy of the helical magnetic configurations onto a classical Heisenberg Hamiltonian

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

where J_{ij} is an exchange interaction between two Mn sites (i,j) and $\mathbf{e}_i$ is the unit vector pointing in the direction of the magnetic moment at site i.

To estimate the parameters of the Mn-Mn exchange interaction we perform calculation for the frozen-magnon configurations:

$$\theta_i=\text{const},\qquad \phi_i=\mathbf{q}\cdot\mathbf{R}_i\,, \tag{2}$$

where θ_i and ϕ_i are the polar and azimuthal angles of vector $\mathbf{e}_i$, $\mathbf{R}_i$ is the position of the ith Mn atom. The directions of the induced moments in the atomic spheres of the atoms of the semiconductor matrix are kept parallel to the z axis.

It can be shown that within the Heisenberg model (1) the energy of such configurations can be represented in the form

$$E(\theta,\mathbf{q})=E_0(\theta)-\sin^2\theta J(\mathbf{q}), \tag{3}$$

where E_0 does not depend on $\mathbf{q}$ and $J(\mathbf{q})$ is the Fourier transform of the parameters of the exchange interaction between pairs of Mn atoms:

$$J(\mathbf{q})=\sum_{j\neq 0} J_{0j}\exp(i\mathbf{q}\cdot\mathbf{R}_{0j}). \tag{4}$$

Performing back Fourier transformation we obtain the parameters of the exchange interaction between Mn atoms:

$$J_{0j}=\frac{1}{N}\sum_{\mathbf{q}}\ \exp(-i\mathbf{q}\cdot\mathbf{R_{0j}})J(\mathbf{q}). \tag{5}$$

The Curie temperature is estimated in the mean-field (MF) approximation

$$k_BT_C^{MF}=\frac{2}{3}\sum_{j\neq 0} J_{0j}\,. \tag{6}$$

We use a rigid-band approach to calculate the exchange parameters and Curie temperature for different electron occupations. We assume that the electron structure calculated for a DMS with a given concentration of the $3d$ impurity is basically preserved in the presence of defects. The main difference is in the occupation of the bands and, respectively, in the position of the Fermi level.

In the main body of the LDA$+U$ calculations we use $U=0.3$ Ry that corresponds to the value determined experimentally[23] and is used in some previous calculations.[19,21,24] The dependence of the Curie temperature on U is illustrated at the end of the paper. The results of the supercell calculations are compared with the results of the calculations within the coherent-potential approximation (CPA).[25]

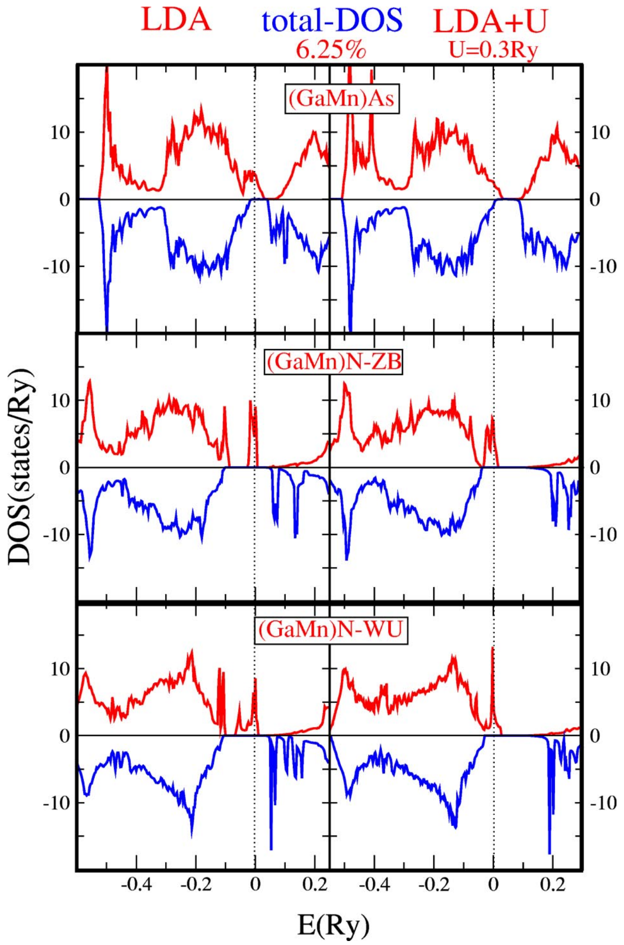

FIG. 1. (Color online) The spin-resolved total DOS of (GaMn)As and (GaMn)N for Mn concentration of 6.25%. Calculations are performed within LDA and LDA$+U$ approaches. For (GaMn)N, both zinc-blende (ZB) and wurzite (WU) structures are presented. The spin-up/spin-down DOS is shown above/below the abscissa axis. The total DOS is given per chemical unit cell of the semiconductor.

III. CALCULATIONAL RESULTS AND DISCUSSION

A. Density of states

We begin the discussion with the properties of the density of states (DOS). In Figs. 1 and 2 we present the DOS for $x=6.25\%$. The main features of the DOS discussed below are common for all Mn concentrations studied. The replacement of one Ga atom by a Mn atom adds to the energy structure five valence bands related to the Mn $3d$ spin-up states. On the other hand, there are only four valence electrons added. As a result, both (GaMn)As and (GaMn)N have one hole per Mn atom. (GaMn)N is half metallic in both LDA and LDA$+U$ calculations with only majority-spin states present at the Fermi level. (GaMn)As is half metallic in LDA calculation and possesses small contribution of the minority-spin states in LDA$+U$ calculation. In all cases the systems are characterized by high spin polarization of the electron states at the Fermi energy. This property is in good agreement with a recent measurement of high carrier spin polarization in (GaMn)As.[26]

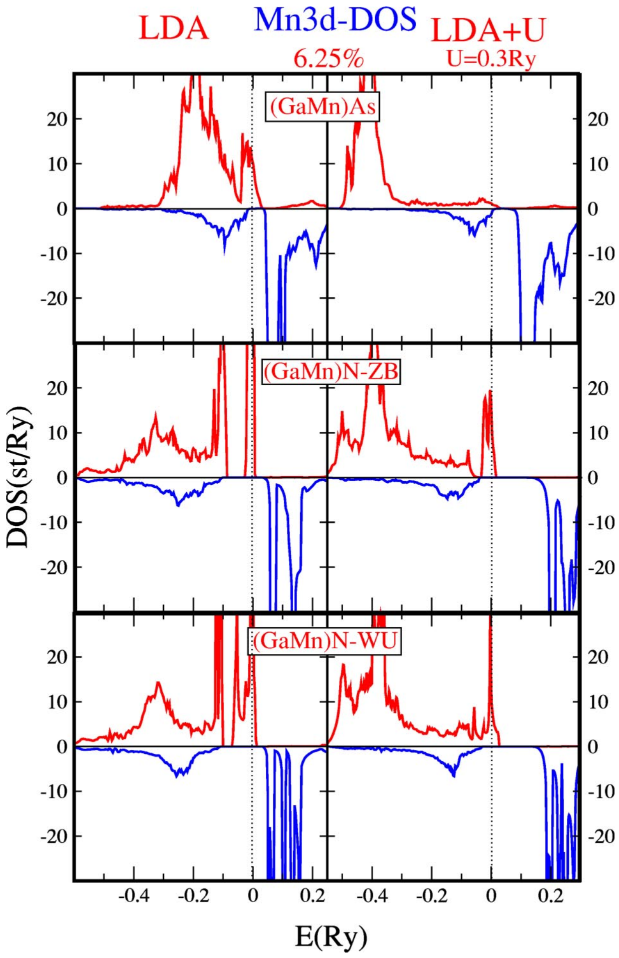

FIG. 2. (Color online) The same as in Fig. 1 but for partial Mn 3d DOS.

Despite this similarity, important differences between (GaMn)As and (GaMn)N are obtained in both LDA and LDA+U calculations. In LDA, the spin-up impurity band of (GaMn)As merges with the valence band. On the other hand, in (GaMn)N the impurity band lies in the semiconducting gap. This is valid for both wurzite and zinc-blende crystal structures of (GaMn)N. These features are in agreement with the results of previous calculations.[10–12,16–20] In all LDA calculations the impurity bands have large Mn 3d contribution (Fig. 2).

In the LDA+U calculations, the impurity band of (GaMn)As disappears from the energy region at the top of the valence band. Almost all spin-up Mn 3d states lie at about -0.4 Ry below the Fermi level. In contrast, (GaMn)N still possesses impurity band which lies now closer to the valence band. The Mn 3d contribution to the impurity band decreases compared with the LDA-DOS but is still large (Fig. 1).

B. Properties of the holes

Now we turn to the discussion of the properties of the holes. We again present the results for one Mn concentration (Fig. 3) and begin the discussion with the spatial distribution of the hole in the LDA calculations. In (GaMn)As 29% of the hole is located on the Mn atom. The first and second coordination spheres of As include four atoms each and contain 35% and 14% of the hole, respectively. The rest is distributed between Ga and empty spheres.

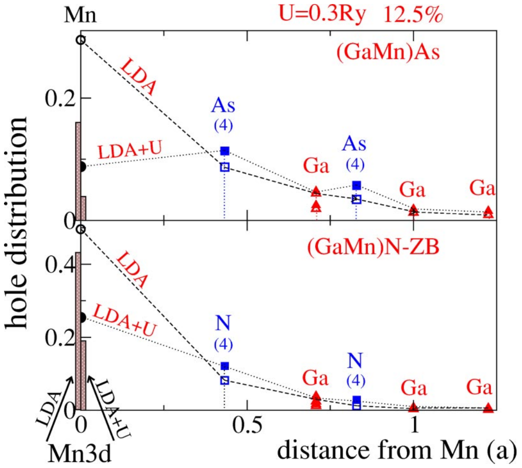

FIG. 3. (Color online) Distribution of the hole in LDA and LDA+U calculations. Numbers in parentheses give the number of atoms in the coordination sphere. The hole part is given for one atom. The distance from Mn is given in units of the lattice parameter. The narrow rectangulars to the left and right of the ordinate axis present the Mn 3d contribution into the hole for LDA and LDA+U calculations.

In (GaMn)N the holes are much stronger localized about the Mn atom. For the ZB structure we find 50% of the hole in the Mn sphere. First and second coordination spheres of N contain, respectively, 33% and 5% of the hole. Although the part of the hole located at the distant atoms decreases compared with (GaMn)As it is still sizable. This property allows the mediation of the exchange interaction between the Mn atoms. Similar result we obtain for the wurzite (GaMn)N (not shown).

In the LDA+U calculations, the holes become more delocalized (Fig. 3). In (GaMn)As, the Mn contribution decreases to 9%. On the other hand, the contribution on the first and second coordination spheres of As increased to 45% and 23%. The contribution of the Ga atoms changes much less. For (GaMn)N in ZB structure the corresponding numbers are 25%, 48%, and 12%. Again similar behavior is obtained for (GaMn)N in the wurzite structure.

The increased delocalization of the holes is a factor favorable for the mediation of the exchange interaction between the Mn atoms and therefore for a higher Curie temperature. There is, however, another factor that is equally important for establishing the long-range ferromagnetic order. This factor is the strength of the p-d exchange interaction which is the physical origin of the interaction mediated by the holes. This second factor is less easy to characterize quantitatively. A convenient quantity for such a characterization is the p-d exchange parameter usually denoted as $N\beta$. In the mean-field approximation $N\beta$ describes the spin-splitting of the semiconductor valence-band states which appears as a consequence of the interaction of these states with Mn 3d elec-

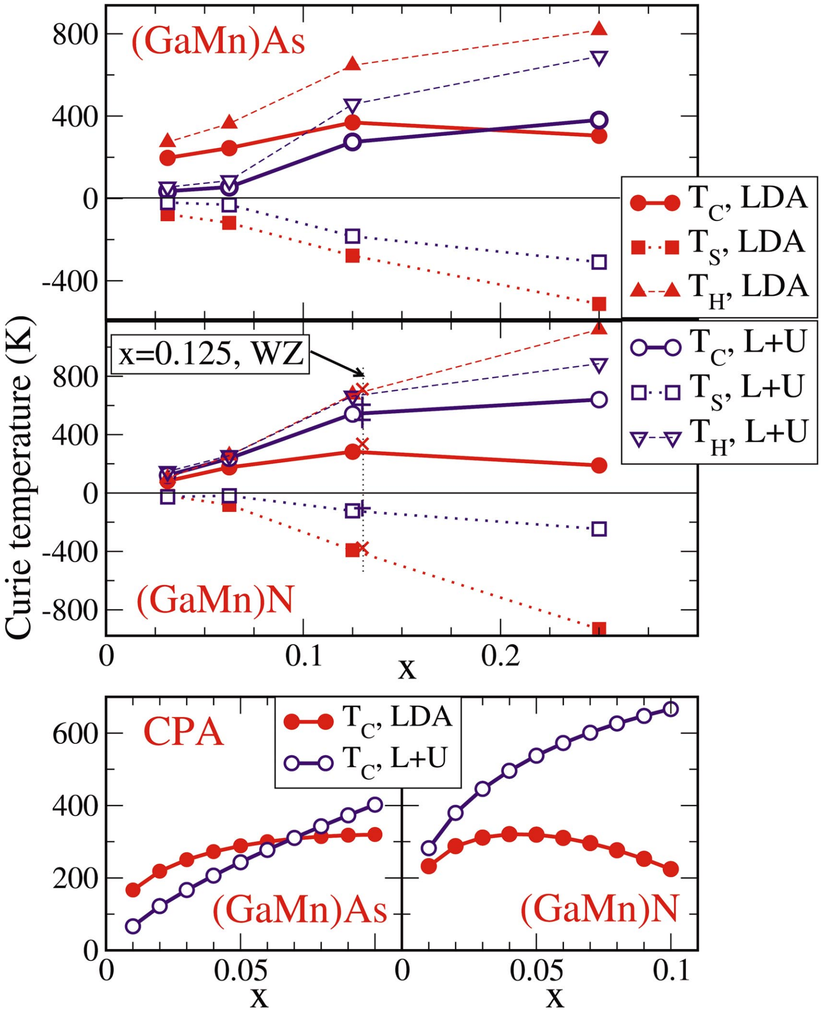

FIG. 4. (Color) T_C as a function of the Mn concentration. The superexchange contribution T_S and hole contribution T_H are shown (see the text for definitions of these contributions). The calculations are performed for ZB structure. For (GaMn)N, the results for WU structure for $x=12.5\%$ are presented for comparison. The lower part of the figure shows the Curie temperature calculated within CPA. L+U is abbreviation for LDA+U.

trons. However, our calculations (Fig. 3) show that the spatial form of the hole states can differ strongly from the unperturbed semiconductor states making the mean-field definition of the p-d interaction poorly founded.

The experimental estimations of $N\beta$ made for (GaMn)As on the basis of different experimental techniques vary strongly from large values of $|N\beta|=3.3$ eV (Ref. 27) and $N\beta=2.5$ eV (Ref. 28) to much smaller values $N\beta=-1.2$ eV (Ref. 23) and $|N\beta|=0.6$ eV.[29] (We use the traditional sign convention: negative $N\beta$ corresponds to antiparallel directions of the spins of the d and p states.) A possible reason for this variation is strong p-d hybridization in III-V DMS. Note that for many II-VI systems $N\beta$ is well defined experimentally[30] and allows good DFT description within the LDA+U approach.[22] In the present paper we do not consider the problem of the estimation of the $N\beta$ parameter in systems with strong p-d hybridization. For characterization of the strength of the p-d interaction we use the value of the Mn 3d contribution to the hole (Fig. 3).

For both systems the LDA+U calculations give a strong decrease of the Mn 3d contribution to the hole (Fig. 3). For (GaMn)As, the Mn 3d contribution drops from 16% to a small value of 4%. For (GaMn)N in ZB structure we get 43% and 19% and for (GaMn)N in WU structure 41% and 16% (the results for WU structure are not shown in Fig. 3). Important that in (GaMn)N the LDA+U the hole still contains large Mn 3d contribution.

The presence of two competing trends in the properties of holes—increasing delocalization, on the one hand, and decreasing p-d interaction, on the other hand—makes direct calculation of T_C necessary to predict the influence of the Hubbard U on the Curie temperature.

C. Curie temperature

In Fig. 4, we show the Curie temperature for a number of concentrations of Mn impurities.[31]

The calculated Curie temperatures (Fig. 4) show again a strong difference between (GaMn)As and (GaMn)N.[32] In (GaMn)N, the Curie temperature increases with account for on-site Coulomb interaction for all Mn concentrations considered. On the other hand, in (GaMn)As the value of T_C decreases for concentration 3.125%, 6.25%, and 12.5%.

For comparison we present (Fig. 4) the Curie temperature calculated within the coherent-potential approximation. The supercell approach employed in the present paper and the CPA provide two opposite limits in the treatment of the atomic disorder: The supercell calculation deals with an ordered atomic pattern. On the other hand, the CPA assumes complete atomic disorder with the effects of the short-range

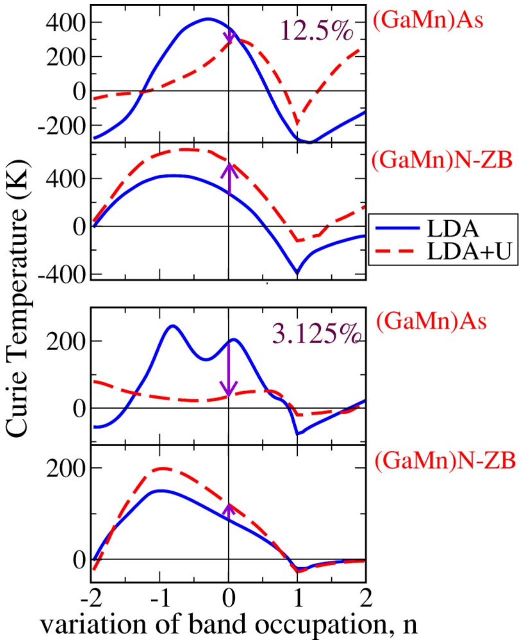

FIG. 5. (Color online) The Curie temperature as a function of the band occupation for Mn concentration of 3.125% and 12.5%. The Curie temperature is calculated according to mean-field formula [Eq. (6)] and negative values of T_C reflect prevailing antiferromagnetic exchange interactions.

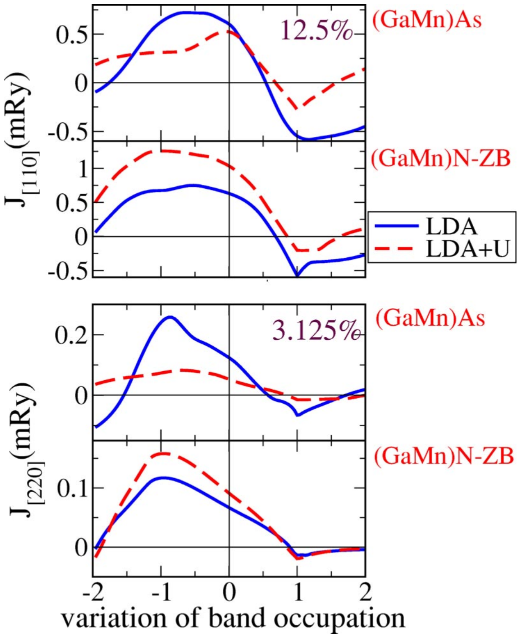

FIG. 6. (Color online) The exchange parameter between Mn atoms separated by the vectors $a(1,1,0)$ and $a(2,2,0)$ for, respectively, $x=3.125\%$ and $x=12.5\%$ as a function of the band occupation. a is the lattice parameter of the zinc-blende structure.

order neglected. Important that the influence of Hubbard U on the Curie temperature obtained within CPA is qualitatively similar to that obtained within the supercell calculations: In (GaMn)As, U gives a lower T_C for small x and a higher T_C for larger x. In (GaMn)N, account for Hubbard U leads to an increase of T_C for the whole Mn concentration range studied.

To get a deeper insight into the formation of the Curie temperature we study the dependence of T_C on the band occupation. In Fig. 5 we present the results for two Mn concentrations of 3.125% and 12.5%. Here, $n=0$ corresponds to the nominal number of carriers (one hole per Mn atom). The kink at $n=1$ marks the point where the valence band is full and the conduction band is empty. In Fig. 6, we show the dependence of the leading interatomic exchange parameter[12] as a function of the band occupation. Indeed, the behavior of the Curie temperature (Fig. 5) repeats in gross features the behavior of the exchange parameter (Fig. 6). In particular, the decrease of the number of holes below the nominal value of one hole per Mn atom leads to the decrease and change of the sign of the exchange parameter. Thus our calculations show the crucial role of the valence-band holes for mediating ferromagnetism in both (GaMn)As and (GaMn)N. This result is obtained in both LDA and LDA+U approaches and agrees with the commonly accepted picture of the ferromagnetism in DMS used in the model-Hamiltonian studies (see, e.g., Refs. 33 and 34, and references therein).

It is useful to present the Curie temperature in the form $T_C=T_S+T_H$. Here T_S is the Curie temperature at $n=1$ and specifies the exchange interaction mediated by the completely filled valence band. This contribution can be related to the Anderson's superexchange.[35] T_H is the difference between the Curie temperatures at $n=1$ and $n=0$ and specifies the contribution of the hole states.

The value of T_S is negative for all concentrations in both LDA and LDA+U approaches reflecting the property that the exchange interaction through completely filled bands is antiferromagnetic. The hole contribution T_H is always positive and is responsible for the ferromagnetism of the system.

The analysis of Fig. 4 shows that the contribution of the antiferromagnetic superexchange is in general not small and must be taken into account in the estimation of the Curie temperature. The absolute value of the antiferromagnetic superexchange is always larger in the LDA case. The decrease of the superexchange for nonzero U can be explained by an increased energy distance between the occupied and empty Mn $3d$ states.

The influence of Hubbard U on the hole contribution T_H differs strongly for two systems. In (GaMn)N, LDA, and LDA+U give very similar results up to $x=12.5\%$. For larger x the LDA value of T_C is higher. This shows that for low Mn concentrations the increased delocalization of the holes is compensated by the decreased p-d interaction. For larger x the increase of the p-d interaction becomes more important since the distance between the Mn atoms decreases. For (GaMn)As, the strong decrease of the p-d interaction under the influence of Hubbard U is not compensated by the increased hole delocalization and the value of T_H drops strongly for small x.

Comparison of the results of the calculations with experiment shows that in the case of (GaMn)As the account for Hubbard U does not make the agreement with experiment better. Indeed, the calculated Curie temperature becomes too low. A possible reason for this can be an underestimation of the Mn $3d$ contribution into the hole states within the LDA

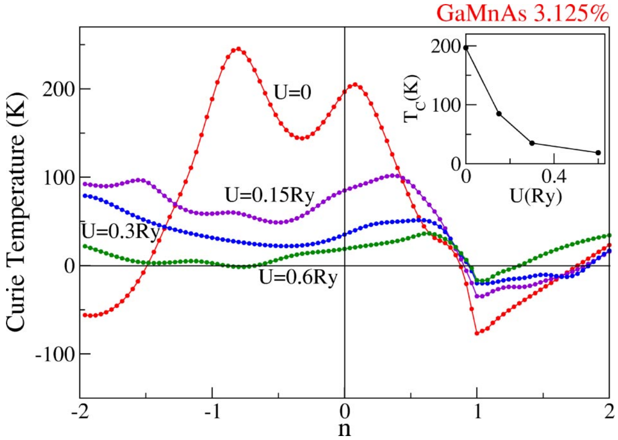

FIG. 7. (Color online) The Curie temperature of (GaMn)As with 3.125% of Mn as a function of the band occupation for a number of U values. In the inset, the T_C value as a function of U for nominal electron number $n=0$.

$+U$ calculation: these states disappear from the Fermi-level region almost completely (Fig. 1). The dynamical mean-field theory (DMFT) which treats the on-site correlations beyond the LDA$+U$ can improve the agreement with experiment since the DMFT provides an account for the quasiparticle states[36] that are neglected within the LDA$+U$. Before the LDA+DMFT study is performed the decision about more appropriate method for the study of the ferromagnetism in (GaMn)As should be made in favor of the LDA scheme which gives good correlation with experiment.[12]

In (GaMn)N, the Mn $3d$ states are present at the Fermi level because of the strong hybridization with the $N2p$ states. We obtained an increase of T_C which is in correlation with the prediction by Dietl *et al.*[3] and with some of the experimental results. Our LDA$+U$ results for (GaMn)N correlate in many respects with the picture proposed recently by Dietl *et al.*[37] in the discussion of the Zhang-Rice limit for this system. They conclude that the high T_C can appear in (GaMn)N as a result of strong p-d interaction within the Zhang-Rice polaron which is efficiently transferred due to the relatively large extent of the p-wave functions.[38]

Figure 7 illustrates the dependence of the Curie temperature on the value of U for (GaMn)As with 3.125% of Mn. We obtain a systematic trend of decreasing scale of the variation of T_C with variation of the band occupation. The inset shows $T_C(U)$ for nominal electron number $n=0$. The dependence is monotonous though nonlinear.

Within our LDA$+U$ calculation we did not obtain the orbital ordering found in the SIC calculation.[20] We, however, do not exclude that such a state can be stabilized within the LDA$+U$ approach. Our numerical experiment with the impurity band split ‘‘by hand’’ to simulate the effect reported in Ref. 20 indeed resulted in a nonferromagnetic ground state.

IV. CONCLUSIONS

We use the local-density approximation (LDA) and LDA$+U$ schemes to study the magnetism of (GaMn)As and (GaMn)N. We show that for both systems and both calculational schemes the presence of holes is crucial for establishing ferromagnetism. The introduction of U increases delocalization of the holes and, simultaneously, decreases the p-d interaction. Since these two trends exert opposite influences on the Mn-Mn exchange interaction the character of the variation of the Curie temperature (T_C) cannot be predicted without direct calculation. We show that the variation of T_C is very different for two systems. For low Mn concentrations we obtain the tendency to increasing T_C in the case of (GaMn)N whereas an opposite tendency to decreasing T_C is obtained for (GaMn)As. We reveal the origin of this difference by inspecting the properties of the densities of states and holes for both systems. The main body of calculations is performed within a supercell approach. The Curie temperatures calculated within the coherent-potential approximation to atomic disorder are reported for comparison. Both approaches give similar qualitative behavior. We make a contact between calculational results and the strong scattering of the experimental data on T_C of the systems.

Note that the present experimental and theoretical situation does not allow to give a unique answer on the question which of two schemes, LDA or LDA$+U$, provides better description of the diluted magnetic semiconductors. This answer can be different for different systems and even samples. Our previous calculations for the II-VI DMS (ZnMn)Se (Ref. 22) gave strong arguments for the superiority of the LDA $+U$ approach in this case. In the present study of (GaMn)As, the LDA$+U$ method with a moderate U underestimates the strength of the p-d interaction and leads to too low Curie temperature for low Mn concentrations. For (GaMn)N, the experimental situation is too unclear to allow for a well-founded decision concerning the superiority of the theoretical scheme. The comparative study reported here aims to contribute to both the increase of the understanding of the systems studied and the formulation of the theoretical approaches most suitable for the investigation of concrete DMS’s. The efforts must, however, be continued and we hope that the results of this paper will be stimulating for further studies.

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[31] The calculation is performed for the case of one hole per Mn atom. Many experiments report the hole number smaller than the nominal Mn content. The reason for decreased hole number are different types of donor defects compensating the Mn acceptors. On the other hand, the number of the Mn atoms participating in the ferromagnetic ordering is also found experimentally to be smaller than the nominal Mn concentration. Therefore the ratio of one hole per one active Mn can still correspond to the factual relation in some samples. The scattering of the experimental data discussed in the Introduction reveals differences in nominally equivalent samples with equal Mn content. To address this problem we consider the variation of the number of carriers for a given Mn concentration.

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[38] At this point it is worth to remind the reader about the strong scattering of the experimental data discussed in the Introduction. Any theoretical model, also the models used in the given paper, does not take into account the whole diversity of the features characteristic for a given sample. The final effect of the trends revealed in our study in the case of a concrete sample depends on the combination of defects in the sample. As follows from our study, the hole number and the efficiency of the screening of the on-site Coulomb interaction are important characteristics. Another feature which can play an important role in the magnetism of DMS is clustering of the magnetic impurities.[16,39] Further experimental efforts for better characterization of the samples and further theoretical efforts for detailed study of different types of defects are needed to reach a quantitative agreement between experiment and calculations.

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