Disorder in Diluted Spin Systems

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(Dated: June 19, 2018)

The influence of substitutional disorder on the magnetic properties of diluted Heisenberg spin systems is studied with regard to the magnetic stability of ferromagnetic diluted semiconductors (DMS). The equation of motion for the magnon Green's function within Tyablikov approximation is solved numerically for finite systems. The resulting spectral density is then used to estimate the magnetization and Curie temperature of an infinite system. This method is suitable for any form of a ferromagnetic exchange interaction. Besides different lattices and spin magnitude, exchange interactions of different range are examined. The results show that, for short-range interaction, no magnetic order exists below the critical percolation concentration, whereas a linear dependence of the Curie temperature on the concentration of spins is found for ferromagnetic long-range interaction.

PACS numbers: 75.10.Nr, 75.10.Jm, 75.50.Pp, 02.60.-x

I. INTRODUCTION

The discovery of ferromagnetism in (III,Mn)-V diluted magnetic semiconductors (DMS) has attracted considerable attention among scientists during the past years. The great interest in these materials is motivated by the idea of using their spins degrees of freedom in conjunction with their electronic degrees to build new electronic devices ranging from fast nonvolatile memories to quantum computers. However, progress has been impeded by the fact that most of the DMS studied so far have a Curie temperature $T_C$ below room temperature.

For the development of ferromagnetic DMS with desired properties such as high Curie temperatures, the theoretical understanding of the magnetism in these materials plays an important role. In DMS, a small fraction of the non-magnetic host-semiconductor ions is substituted by ions, which carry a localized magnetic moment (spins). These magnetic ions are mainly randomly distributed over the lattice sites of one of the host-semiconductor species. This positional disorder breaks the translational symmetry of the crystal and thus greatly complicates the theoretical description of the material.

One of the first to consider the substitutional disorder present in diluted spin systems was Brout in the late 1950s. Since then, various attempts have been made to tackle the problem of including the disorder into the theoretical description of ferromagnetism in diluted spin systems. Many approaches combine mean-field or spin-wave theories with configurational-average methods, which not only rely on uncontrolled approximations with respect to the disorder, but also encounter difficulties when applied to spin systems. Some use Monte-Carlo simulations, which make the simplification of classical spins instead of quantum spins. Other approaches are based on percolation theory or replica method, but these treat the magnetism itself only on a mean-field level. However, one must properly take into account the positional disorder of the spins and their quantum nature to obtain reliable predictions about the magnetic properties of diluted spin systems.

In this article, we will present an approach, which is based on the Tyablikov approximation to the Heisenberg model and uses numerical studies of finite systems to estimate spontaneous magnetizations and Curie temperatures of the corresponding systems in the thermodynamic limit. It should be mentioned here that the idea of using numerical studies of finite spin systems in spin-wave-type approximation is not new. According to our knowledge, however, the obtained spectral densities have never been used before to calculate spontaneous magnetizations and Curie temperatures. The presented approach is suitable for any form of ferromagnetic exchange interaction. Within this approach, we are able to treat the quantum fluctuations of the spins within random-phase approximation. It means that our approach goes beyond the classical-spin approximation and the mean-field theory, which notoriously overestimates ferromagnetic stability. Furthermore, we are able to treat the positional disorder of the spins numerically exact in contrast to approaches employing configurational-average methods.

This article is organized as follows: In section II we will generalize the Tyablikov approximation to spin systems without translational symmetry. Section III is concerned with the numerical studies we performed on the basis of the generalized Tyablikov approximation. In subsection III A we will explain the algorithm we used for our numerical studies. In the following subsections, we will discuss the results we obtained for systems with ferromagnetic short-range, ferromagnetic long-range and oscillating long-range exchange interactions. In section IV, we will conclude the article with a summary and proposals for future work.
II. THE MODEL

To study the magnetic properties of a material with a concentration $c \in [0,1]$ of atoms carrying a localized magnetic moment, we consider a lattice with a fraction $c$ of the lattice sites occupied by a spin. We assume the dynamics of these spins to be described by the isotropic Heisenberg Hamiltonian

$$H = - \sum_{i,j=1}^{N} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{\hbar} g_{ij} \mu_B B \sum_{i=1}^{N} S_i^z .$$

(1)

Here, $i$ and $j$ label the occupied lattice sites and $S_i = (S_i^x, S_i^y, S_i^z)$ is the spin operator of the localized magnetic moment at lattice site $i$ with lattice vector $\mathbf{R}_i$. The spins – whose total number is denoted by $N$ – interact via a Heisenberg exchange coupling with exchange parameters $J_{ij}$ obeying $J_{ij} = J(|\mathbf{R}_i - \mathbf{R}_j|)$ and $J_{ii} = 0$. In addition, the Hamiltonian contains a Zeeman coupling of the spins to a homogeneous external magnetic field $\mathbf{B} = (0,0,B)$.

The Tyablikov approximation\textsuperscript{52} has been developed for and very successfully applied to concentrated spin systems with translational symmetry. In the following, we will generalize the Tyablikov approximation to derive expressions for the magnetization and the Curie temperature for diluted spin systems, where we cannot presuppose translational symmetry. Thereby, we will keep the form of the appearing expressions as close as possible to those known from the usual Tyablikov approximation. However, the absence of translational symmetry complicates the evaluation of the derived expressions, as will be seen later.

We will first consider spins with $S = 1/2$. In this case, the magnetic properties of the system can be studied by use of the retarded magnon Green’s function

$$G_{ij}(E) = \langle \langle S_i^+; S_j^- \rangle \rangle \delta_{ij} E .$$

(2)

containing the step operators $S_i^\pm = S_i^x \pm i S_i^y$. The equation of motion for $G_{ij}(E)$ reads

$$\left( E - g_{ij} \mu_B B \right) G_{ij}(E) = 2\hbar^2 \delta_{ij} \langle S_i^z \rangle + 2\hbar \sum_{m} J_{im} \times \left( \langle \langle S_i^z S_m^z; S_j^- \rangle \rangle E - \langle \langle S_i^z S_m^z; S_j^- \rangle \rangle E \right) .$$

(3)

Making the Tyablikov approximation, which consists in decoupling the higher Green’s function on the rhs. of (3), and assuming a uniform magnetization $\langle S_i \rangle \equiv (0,0,\langle S^z \rangle)$, one obtains after rearrangement:

$$\sum_{m} \left( \frac{E - g_{ij} \mu_B B}{2\hbar \langle S^z \rangle} - \sum_{n} J_{in} \right) \delta_{im} + J_{im} \right] G_{mj} = \hbar \delta_{ij} .$$

(4)

In the following, it is convenient to interpret this equation as a matrix equation:

$$\left( \frac{E - g_{ij} \mu_B B}{2\hbar \langle S^z \rangle} \mathbf{1} - \mathbf{H} \right) \mathbf{G}(E) = \hbar \mathbf{1} .$$

(5)

Here, the Hamilton matrix $\mathbf{H}$, which is defined by its matrix elements $H_{ij} = \delta_{ij} \sum_{n=1}^{N} J_{in} - J_{ij}$, has been introduced.

In order to solve equation (5) for $\mathbf{G}$, one has to diagonalize $\mathbf{H}$. In the concentrated case, this can be done by a Fourier transformation because of the translational symmetry of the system. For a diluted system, translational symmetry is absent, and hence, no general method exists for this task. However, for a finite system the Hamilton matrix can be diagonalized numerically. Since $\mathbf{H}$ is Hermitian, this diagonalization yields $N$ real eigenvalues $E_r$ with eigenvectors, which can be chosen to form an orthonormal basis. For notational convenience, we will use indices $r$ and $r'$ to refer to matrix elements in this $\mathbf{H}$-diagonal” basis, whereas we retain $i$ and $j$ to refer to matrix elements in the ‘site-diagonal’ basis, where matrix indices are identified with site labels.

After going to the $\mathbf{H}$-diagonal basis, one obtains for the magnon Green’s function:

$$G_{rr'}(E) = \frac{2\hbar^2 \langle S^z \rangle}{E - g_{ij} \mu_B B - 2\hbar \langle S^z \rangle E_r + i0^+} \delta_{rr'} .$$

(6)

From this expression, one can read off the magnon spectral density $S_{rr'}(E) = \frac{1}{\pi} \Im (G_{rr'}(E))$:

$$S_{rr'}(E) = -\frac{1}{\pi} \Im (G_{rr'}(E))$$

$$= 2\hbar^2 \langle S^z \rangle \delta (E - g_{ij} \mu_B B - 2\hbar \langle S^z \rangle E_r) \delta_{rr'} .$$

(7)

Thus, the energies $E_r = g_{ij} \mu_B B - 2\hbar \langle S^z \rangle E_r$ are the excitation energies of the elementary excitations of the systems, which we call magnons.

To obtain the local magnon spectral density $S_{ii}(E)$, one has to transform $S_{rr'}(E)$ back to the site-diagonal basis. However, this may yield different $S_{ii}(E)$ for different lattice sites $i$. This contradicts the assumption of a uniform magnetization, since $S_{ii}(E)$ and $\langle S_i^z \rangle$ are connected by the spectral theorem:

$$\langle S_i^z \rangle = \hbar S \frac{1}{\hbar} \langle S_i^z S_i^- \rangle = \hbar S \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \frac{S_{ii}(E)}{E^2 - 1} dE .$$

(8)

where $\beta = 1/k_B T$ with $k_B$ as the Boltzmann constant and $T$ as the temperature. One can circumvent the problem by abandoning the simplification of a uniform magnetization at the cost of having to solve a system of $N$ non-algebraic equations in an iterative procedure, where one has to diagonalize an $N \times N$ matrix similar to $\mathbf{H}$ in each step. Here, a numerically less demanding approach is taken, which we believe to capture the important physics of the system: We consider $\langle S^z \rangle$ an approximation for $\langle S_i^z \rangle$ and use the site-averaged spectral density

$$\tilde{S}_{ii}(E) = \frac{1}{N} \sum_{i=1}^{N} S_{ii}(E) = \frac{1}{N} \sum_{r=1}^{N} S_{rr}(E)$$

(9)

as an approximation for $S_{ii}(E)$ and then use the spectral
theorem:
\[
\langle S^z \rangle \approx \langle S^z_i \rangle = \hbar S - \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \frac{S_{ii}(E)}{e^{\beta E} - 1} \, dE \\
\approx \hbar S - \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \frac{\tilde{S}_{ii}(E)}{e^{\beta E} - 1} \, dE . \tag{10}
\]

In short, we have to solve:
\[
\langle S^z \rangle = \hbar S - \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \frac{\tilde{S}_{ii}(E)}{e^{\beta E} - 1} \, dE . \tag{11}
\]

After inserting (9) and doing some rearrangements, this yields:
\[
\langle S^z \rangle = \frac{\hbar S}{1 + \frac{2}{N} \sum_{r=1}^{N} \left[ e^{\beta g \mu B + 2\hbar (S^z)_r E_r} - 1 \right]^{-1}} . \tag{12}
\]

From this equation, one obtains an expression for the Curie temperature by linearization with respect to \( \langle S^z \rangle \):
\[
T_C = \frac{\hbar^2}{2k_B} \left( \frac{1}{N} \sum_{r=1}^{N} \frac{1}{E_r} \right)^{-1} . \tag{13}
\]

Note that for translationally invariant systems, the indices \( r \) can be identified with the wave vectors of the first Brillouin zone, and thus the expressions (12) and (13) reduce to the standard Tyablikov formulae for concentrated spin systems.

For general \( S \)-values, an analogous derivation using a set of Green’s functions instead of the Green’s function alone leads to the implicit equation
\[
\langle S^z \rangle = \hbar \left( \frac{1 + S + \Phi}{2S + 1} + \frac{(S - \Phi)}{2S + 1} \right) \Phi \tag{14}
\]
for the magnetization. Here, the average magnon number
\[
\Phi = \frac{1}{N} \sum_{r} \frac{1}{e^{\beta(g \mu B + 2\hbar (S^z)_r E_r)} - 1} \tag{15}
\]
has been used as a convenient abbreviation. The Curie temperature is then given by
\[
k_B T_C = \frac{2}{3} \frac{\hbar^2 S (S + 1)}{k_B} \left( \frac{1}{N} \sum_{r} \frac{1}{E_r} \right)^{-1} \tag{16}
\]
for general \( S \)-values.

In the following we will only consider systems in an infinitesimal external magnetic field \( B = 0 \)° just to break rotational invariance and therefore, usually drop terms including the \( B \)-field explicitly.

### III. NUMERICAL STUDIES OF FINITE SYSTEMS

#### A. Algorithm

The equations (12) to (16) are of no practical use for the calculations of the magnetic properties of the system unless one can compute the eigenvalues \( E_r \) of the Hamilton matrix \( H \). As already mentioned above, the Fourier transformation, which is the weapon of choice for concentrated spin systems, does not work for diluted systems, because translational symmetry is absent. However, for a finite system, we can diagonalize the Hamilton matrix numerically to obtain its eigenvalues. Compared to configurational average methods, the numerical diagonalization has the advantage that no approximations with respect to the disorder are needed. Furthermore, the spins can be treated quantum mechanically and the simplification to classical spins as in usual Monte Carlo simulations is not necessary.

The algorithm we used in our numerical studies of finite systems is as follows: First of all, the system parameters must be fixed. These are the concentration \( c \) of spins on the lattice, the exchange parameter \( J(\langle R \rangle) \) as a function of the distance \( R \) between two spins and the size and geometry of the system. The next step is to decide by the help of a pseudo-random-number generator which lattice sites of the system are occupied by a spin. Now that the positions of all occupied lattice sites in the system are known, the exchange parameters \( J_{ij} = J(\langle|\vec{R}_i - \vec{R}_j|\rangle) \) between occupied lattice sites \( i \) and \( j \) can be calculated, and hence, the matrix elements \( H_{ij} \). Now, the matrix \( H \) can be diagonalized numerically using standard diagonalization routines for large matrices.

For the shape of the simulated samples, we chose a cuboid lattice section, which we closed to a torus to eliminate surface effects. To avoid multiple- and self-interaction due to the periodic boundary conditions, we cut off any long-range interaction at a distance half the system size. We chose the size of the samples such that for each concentration we obtained approximately the maximal number of spins in the system that we could handle (ca. 14,000 spins on an AMD Athlon PC, 2GHz CPU, 1 GB RAM). For simplicity, we only considered the case that each lattice site is occupied by a spin with probability \( c \) independent of the occupation of neighboring sites. However, if one is interested in clustering effects, a different routine to distribute the spins on the lattice could be used.

To decrease statistical errors, we averaged the eigenvalue distribution, i.e. \( \tilde{S}_{ii}(E) \), over several simulated systems with equal system parameters, but different pseudo-random distribution of the spins. Test calculations showed that this averaging seems hardly necessary for the large samples we used, which is due to the self-averaging property of the site-averaged local magnon spectral density \( \tilde{S}_{ii}(E) \). As a trick to smoothen the eigenvalue spectra, we sometimes used slightly differ-
ent sample geometries for different simulation runs and weighted the obtained eigenvalue distributions with their number of lattice sites. Further weighting was not necessary, since for a fixed system size the probability of generating a certain distribution of spins with the used algorithm coincides with the statistical weight of this distribution among those having an equal number of lattice sites.

The disadvantage of the numerical diagonalization is that only finite systems can be treated, but there is no spontaneous magnetization in the isotropic Heisenberg model for any finite system. The Tyablikov approximation preserves this property, which is seen in our calculations by the fact that, due to its structure, \( H \) has at least one zero-eigenvalue, which has a finite spectral weight for finite systems. More precisely, the number \( N_{\text{zeros}} \) of zero-eigenvalues of \( H \) equals the number \( N_{\text{clusters}} \) of clusters of spins connected by a non-zero exchange interaction for systems with only ferromagnetic interaction and \( N_{\text{zeros}} \geq N_{\text{clusters}} \) for systems containing antiferromagnetic interaction. Hence, and always yield zero for the spontaneous magnetization and the Curie temperature, if the computed eigenvalues of the simulated samples are used directly as input. In order to obtain a finite spontaneous magnetization and Curie temperature, one has to go to infinite systems. However, the eigenvalue distribution of a sufficiently large finite system represents a good approximation to the eigenvalue distribution of the corresponding infinite system except for energies close to zero, where the discreteness of the eigenvalue spectrum of the finite system comes into play. We solved this problem by shifting the zero-eigenvalues of \( H \) slightly to higher energies, which will be explained in more detail in the following subsection.

### B. Short-Range Interaction

As first application we investigate systems of spins on a simple cubic (sc) lattice with nearest-neighbor interaction only:

\[
J(R) = \begin{cases} 
J^0 & \text{for } R = a, \\
0 & \text{otherwise.}
\end{cases}
\tag{17}
\]

Here, \( a \) denotes the nearest neighbor distance and \( J^0 \) the nearest neighbor interaction strength.

In figure 1 we plotted the spectral density for different concentrations \( c \) of spins. The spectral density is zero for all \( E < 0 \) for all concentrations, which is due to the fact that \( H \) is positive semi-definite for systems with purely ferromagnetic interactions. The calculations for the concentrated system show the well-known symmetric shape of a simple cubic density of states. Dilution increases the spectral density for lower energies at the cost of the spectral density at higher energies. For concentration above \( c \approx 0.5 \), the spectral density stays smooth, whereas for lower concentrations, peaks at certain energies \( (E/2\hbar (S^z))^0 = 0, 1, 2, 2 - \sqrt{2}, \ldots) \) become apparent, which constitute the whole spectral density for \( c < 0.3 \). The smooth part of the spectral density can be attributed to one large percolating cluster of spins, which has a quasi-continuous spectrum. This percolating cluster only exists above the critical percolation concentration \( c_P \), which is \( c_P^0 \approx 0.3 \) for the simple cubic lattice with nearest-neighbor interaction. The peaks stem from smaller clusters, which contain a considerable fraction of the spins only for concentrations around and below \( c_P \) and which have a discrete spectrum.

To obtain non-zero Curie temperatures from the obtained \( H \)-eigenvalues of finite systems, we used a small energy \( \delta \) to shift the eigenvalues close to zero slightly to higher energies. To be more specific, we used

\[
E_r \mapsto \begin{cases} 
\delta & \text{for } E_r < \delta, \\
E_r & \text{otherwise, } \forall r \in \{1, \ldots, N\}
\end{cases}
\tag{18}
\]

for practical reasons. The effect of this shift \( \delta \) on the Curie temperature \( T_C \) is illustrated in figure 2. Let us first discuss the curves for \( c = 1 \), where the numerically obtained data for finite systems can be compared with the analytical results for the infinite system. For \( N = \infty \), the Curie temperature has a finite value at \( \delta = 0 \) and is quite insensitive to small shifts \( \delta \ll J^0 \). For large enough but finite \( N \), the calculated Curie temperature rapidly rises from \( T_C = 0 \) at \( \delta = 0 \) to a value comparable to the infinite system at \( \delta \approx J^0/N \). In this steep region, \( T_C(\delta) \) is dominated by the shifted zero-eigenvalue, which has a spectral weight \( \nu_{\text{zeros}} = N^{-1} \), and \( T_C(\delta) \approx \frac{2}{\hbar^2} S (S + 1) N \delta \). For \( \delta \gg J^0/N \), however, the shifted zero-eigenvalue becomes less important, and

![FIG. 1: Site-averaged local magnon spectral density \( \tilde{S}_c(E) \) for nearest-neighbor interaction on an sc lattice for various concentrations \( c \) of spins (shown as histogram of the eigenvalue distribution of the Hamilton matrix, each curve is an average of 5 to 10 configurations of 8000 to 14000 spins).](image)
the curves of the finite system and the infinite system are almost identical. Hence, the Curie temperature for the finite system with \( c = 1 \), large \( N \) and \( J^0/N \ll \delta \ll J^0 \) approximates the Curie temperature for the infinite system with \( c = 1 \) very well. Furthermore, for large \( N \) and \( J^0/N \ll \delta \ll J^0 \), the results are quite insensitive to the actual choice of \( \delta \).

For \( c < 1 \), the situation is more complicated. Firstly, there are no exact analytical results for \( T_C \) we could compare our numerical results with. Furthermore, for an exchange interaction with finite range, there is always a finite fraction of spins in smaller clusters. Hence, always a finite fraction \( w_{\text{zeros}} = N_{\text{zeros}}/N \) of eigenvalues is zero. Therefore, \( T_C(\delta) \) maintains a finite slope \( \propto w_{\text{zeros}} \) for \( \delta \ll w_{\text{zeros}}J^0 \) even for very large \( N \) as is seen in figure 2(b). This problem could be avoided if only the percolating cluster, which is the only cluster that can support long-range order in the system, were considered in the calculation of \( T_C \). However, identifying and removing all the eigenvalues due to smaller clusters is numerically quite demanding. Moreover, for concentrations well above the critical percolation concentration, the number of smaller clusters is very small and thus \( w_{\text{zeros}} \ll 1 \). Hence, the behavior of \( T_C(\delta) \) is dominated by the eigenvalues of the percolating cluster for \( w_{\text{zeros}}J^0 \ll \delta \) and \( c > c_p \) and is not very sensitive to changes in \( \delta \) for \( \delta \ll J^0 \) as seen in figure 2(b).

For \( c \lesssim c_p \), most or all eigenvalues are due to smaller clusters and therefore, the applicability of the presented method may be questionable. However, since a large fraction of the eigenvalues is zero, \( T_C(\delta) \approx \frac{2}{3}\hbar^2 S(S+1)\delta \), which simply means that \( T_C \approx 0 \) for small \( \delta \). This agrees with percolation theory, from which follows that \( T_C = 0 \) for \( c < c_p \), since there can not exist any long-range ordering in the system. Therefore, we believe that the Curie temperature calculated using the eigenvalues of a large finite system and a shift \( \delta \) chosen such that \( J^0/N \ll \delta \ll J^0 \) gives a good estimate for the Curie temperature of the corresponding infinite system for finite dilution, too. In the following, all given values for the Curie temperature and spontaneous magnetization refer to \( \delta = 0.01J^0 \), which corresponds for systems with \( N \approx 14000 \) to a Curie temperature \( T_C(\delta) \) just outside the steep region at small \( \delta \).

The Curie temperature \( T_C \) we calculated for various concentrations is shown in figure 3. Below \( c = 0.3 \), the Curie temperature is practically zero in accordance with percolation theory. Above \( c = 0.3 \), the Curie temperature increases with increasing concentration and reaches its maximum value \( k_B T_C = 2.61S(S+1)\hbar^2J^0 \) at \( c = 1 \) in good agreement with the analytical result of \( k_B T_C = 2.64S(S+1)\hbar^2J^0 \).

The question of the effect of the substitutional disorder at finite dilution on the Curie temperature is connected to the question, how one defines the ordered system for finite dilution. Placing the remaining spins of the diluted system on an artificial lattice with an appropriate lattice constant to regain translational invariance would give a Curie temperature merely depending on how one alters the nearest-neighbor exchange interaction strength with increasing lattice constant. The virtual-crystal approximation, which is widely used to incorporate dilution effects because of its simplicity, would give \( T_C(c) = c T_C(1) \), which is certainly wrong for low \( c \) in the case of an exchange interaction with finite range.

The reduced magnetization as a function of tempera-
than 100 K in DMS such as Ga
pronounced for higher spins. Any anomalous curvature
where, the magnetization curves show a
S = 1/2 concentration values as low as
0.5 of magnetic atoms
for different ranges n of the exchange interaction. We performed calculations for the sc and fcc lattice structure, but no qualitative differences could be found between them. As expected, a decreasing value of the threshold concentration, above one finds a finite Curie temperature, is found with increasing range n of the interaction. Moreover, the observed values of the threshold concentration agree quite well with the corresponding critical percolation concentration, which is a lattice-structure- and interaction-range-dependent quantity. Furthermore, the results show that the exchange interaction must have a range beyond the fourth shell to obtain a finite Curie temperature for concentration values of c ≈ 0.05, which are typical of the recently studied DMS. This is consistent with the values n ≥ 5 for the fcc and n ≥ 6 for the sc lattice obtained by the formula

\[ n \gtrsim \left( \frac{3}{4\pi f_{\text{geom}}} \right)^{2/3} \]  

with f_{\text{geom}}^{\text{sc}} = 2.4 and f_{\text{geom}}^{\text{fcc}} = 2.4/\sqrt{2}, which is based on percolation arguments.

For n = ∞, the Curie temperature is essentially linear in the concentration for both lattice structures. Hence, a small, but finite value is found for the Curie temperature even for very low concentrations. Thus, it seems that the applicability of the virtual-crystal approximation is restored in the limit n → ∞. However, calculation for other forms of ferromagnetic long-range exchange interactions show that the perfect linear dependence \( T_C \propto c \) is a peculiarity of the exchange interaction, although the proportionality seems to be roughly fulfilled for all ferromagnetic long-range exchange interactions. Note that placing the remaining spins of the diluted system on an artificial lattice with an appropriate lattice constant \( a(c) \propto c^{1/3} \) and changing the exchange parameter according to eq. gives a Curie temperature \( T_C \propto c^{4/3} \) in contrast to the observed \( T_C \propto c \).

We also calculated magnetization curves for different concentrations and exchange-interaction ranges for the sc and fcc lattice structure. However, since any unusual features of the magnetization curves have not been observed, we will refrain from an extensive discussion of these.
D. Oscillating Long-Range Interaction

It is believed that the ferromagnetism in DMS is carrier-induced, with holes in the valence band of the host semiconductor mediating the interaction between the localized moments of the impurity atoms. One feature of such an indirect exchange mechanism is that it may give rise to a long-ranged effective exchange interaction \( J(R) \) which is non-monotonous as function of the spin-spin distance \( R \) and even may be ferromagnetic for some, but antiferromagnetic for other values of \( R \). To study such an oscillatory exchange interaction, we choose

\[
J(R) = \begin{cases} 
J^0 \left( \frac{R}{a} \right)^{-3} \cos \left( \pi \left( \frac{R}{a} - 1 \right) \right) & \text{for } a \leq R, \\
0 & \text{otherwise.} \end{cases} \tag{21}
\]

In figure 5, we plotted the site-averaged local magnon spectral density \( \tilde{S}_n(E) \) for different concentrations. Although the exchange interaction \( J(R) \) contains antiferromagnetic interactions, the concentrated spin system has a saturated ferromagnetic ground state and a low-temperature ferromagnetic phase with a Curie temperature of \( k_B T_C \approx 1.6h^2 J^0 \). However, as soon as the concentration is decreased from \( c = 1 \), the spectral density becomes non-vanishing for \( E < 0 \). This effect is still small for high concentrations, and one could try to use the computed eigenvalues to calculate Curie temperatures by using a small shift of the lowest eigenvalues, but already at intermediate concentrations the negative \( H \)-eigenvalues substantially contribute to \( \tilde{S}_n \) and certainly can not be ignored any more. This means that we can not compute any spontaneous magnetization or finite Curie temperature for such systems with our method.

The results observed for the exchange interaction \( J(R) \) can be generalized. Eigenvalues \( E_n < 0 \) of the Hamilton matrix \( H \) occur for all systems with \( c < 1 \) and antiferromagnetic components in the exchange interaction. These negative eigenvalues stem from the fact that at finite dilution there are always some spins in the system which predominantly couple antiferromagnetically to the other spins of the system. As a result, the saturated ferromagnetic state is not the ground state of the diluted system and hence, is thermodynamically unstable against creation of magnons. However, the Tyablikov approximation with the assumption of a uniform magnetization is a reasonable approximation to the Heisenberg model only for a saturated ferromagnetic ground state, and therefore, the presented method fails for oscillatory exchange interactions. Note that these considerations rule out a highly oscillating exchange interaction for materials which are ferromagnetic despite a low concentration of magnetic atoms. For materials with highly oscillating exchange interaction, one rather expects a spin-glass-like phase at low concentrations instead of a ferromagnetic one.
IV. SUMMARY

The aim of this article was to study the magnetic properties of diluted spin systems. Our first step was to generalize the Tyablikov approximation to spin systems without translational symmetry to account for the substitutional disorder present in such systems. The resulting equation of motion for the magnon Green’s function was then solved numerically for finite spin systems. The obtained spectral densities were then used to estimate the Curie temperature and the spontaneous magnetization of the corresponding infinite system.

Compared with other methods, our approach has the advantage that the spins are treated quantum-mechanically within the Tyablikov approximation, which goes beyond mean-field and classical-spin approximation, and – apart from the simplification of a uniform magnetization – no approximations are needed with respect to the substitutional disorder. Furthermore, the numerical effort as well as the analytical is fairly low in our approach.

Our calculations for short-range exchange interactions show no magnetic order below the critical percolation concentration in accordance with percolation theory. For ferromagnetic long-range exchange interactions, the calculation show a linear dependence of the Curie temperature on the concentration of spins. Hence, a finite Curie temperature is found even for very low concentrations. For systems with oscillating long-range exchange interaction, the presented method fails to give answers due to the lack of a ferromagnetic ground state at finite dilution. For ferromagnetic materials with concentrations of magnetic atoms as low as $c \approx 0.05$, these results imply that the effective exchange interaction must be very long-ranged, but can not oscillate strongly with the inter-spin distance.

In this article, we have shown that our approach gives quite reasonable results for a variety of model systems with ferromagnetic ground state. The next step should be an application to real substances. To attack the problem of ferromagnetic DMS, for example, the presented method may be combined with a theory for the electronic degrees of freedom, which provides the values for the effective exchange interaction between the localized moments. Furthermore, the method can be extended to account for a site-dependent magnetization of the localized moments. However, this will highly increase the numerical effort due to the distribution of the magnetic atoms by choosing certain preparation parameters and hence, obtain certain desired magnetic properties. Furthermore, it should be investigated whether the presented method, in particular the treatment of the zero-eigenvalues, could be put on a firm theoretical basis.

Acknowledgments

This work benefited from the support of the SFB290 of the Deutsche Forschungsgemeinschaft. One of us (S. Hilbert) gratefully acknowledges financial support by the Friedrich-Naumann-Stiftung. Thanks to C. Santos for helpful discussions.

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