Effects of dilution and disorder on magnetism in diluted spin systems

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The influence of configurational disorder on the magnetic properties of diluted Heisenberg spin systems is studied with regard to the ferromagnetic stability of diluted magnetic semiconductors. The equation of motion of the magnon Green’s function is decoupled by Tyablikov approximation. With supercell approach, the concentrations of magnetic ions are determined by the size of the supercell in which there is only one magnetic ion per supercell in our method. In order to distinguish the influence of dilution and disorder, there are two kinds of supercells being used: the \textit{diluted and ordered} case and the \textit{diluted and disordered} case. The configurational averaging of magnon Green function due to disorder is treated in the augmented space formalism. The random exchange integrals between two supercells are treated as a matrix. The obtained magnon spectral densities are used to calculate the temperature dependence of magnetization and Curie temperature. The results are shown as following: (i) dilution leads to increasing the averaged distance of two magnetic ions, further decreases the effective exchange integrals and is main reason to reduce Curie temperature; (ii) spatial position disorder of magnetic ions results in the dispersions of the exchange integrals between two supercells and slightly changes ferromagnetic transition temperature; (iii) the exponential damping of distance dependence obviously reduces Curie temperature and should be set carefully in any phenomenological model.

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I. INTRODUCTION

Following the discovery of a ferromagnetic transition at temperatures in excess of 100K, the diluted III-V magnetic semiconductors (DMS), which are realized by doping a semiconducting host material with low concentrations of magnetic impurities (typically manganese), have attracted a great deal of attention from both the experimental and theoretical point of view due to their potential in spintronics applications\textsuperscript{1,2,3,4,5,6,7,8,9}.

In DMS, low concentrations of magnetic impurities carrying localized magnetic moments (spins) form a diluted spin system. The random spatial distribution of the magnetic impurities breaks the translational symmetry of the crystal and thus greatly complicates the theoretical description of the material\textsuperscript{10,11,12,13}. Several different theoretical methods\textsuperscript{14,15,16,17} have been performed to get the transition temperatures. In some stages of these calculations, disorder effects have been completely neglected or treated within the mean-field approximation (MFA). Monte Carlo (MC) simulations\textsuperscript{18,19,20,21,22,23,24,25} seem to provide a better way to include the positional disorder, but these theories usually assume classical spins. A proper treatment of the positional disorder of the localized moments and their quantum nature is necessary\textsuperscript{12,26}. Recently, \textit{ab initio} calculations with supercell approach\textsuperscript{27,28,29,30} are used to investigate the effects of disorder. Alternatively, the stochastic series expansion (SSE) quantum MC (QMC) method with $L \times L$ supercell is used to investigate the order-disorder transition in the diluted two-dimensional Heisenberg model with random site dilution\textsuperscript{31}. But there are still doubts concerning the effect of disorder on magnetism.

In fact, the effect of disorder on magnetism is an old and important problem in diluted spin systems\textsuperscript{32,33}, although only during the last few years have disorder effects in DMS been considered. In these early papers, a typical method is using coherent potential approximation (CPA) that is initially developed by Soven and Taylor\textsuperscript{34,35} to treat the dynamics of a random Heisenberg-type Hamiltonian. However, most of these methods only include the short-range interaction and are hard to extend to the long-range exchange interaction. Up to now, lots of methods of investigating spin systems such as SSE-QMC\textsuperscript{36} and density matrix renormalization group theorem\textsuperscript{37} only take into account short-range or nearest-neighbor interactions. But the experimental fact of the transition temperature with low concentrations implies that the exchange interaction between the localized moments is long-ranged in DMS\textsuperscript{38}. It means the long-range exchange interactions should be also included in the theoretical calculation of the magnetization and the transition temperature.

Another method for calculating the properties of disordered systems is the augmented space formalism (ASF)\textsuperscript{39,40}, which is introduced by Mookerjee\textsuperscript{41} and centers around averaging functions of independent random variables. Rather than expanding the Green function in some manner and then averaging an appropriate set of terms, the random problem is transformed into an ordered one which is defined in a larger Hilbert space. This new Hilbert space is referred to as the augmented space, which may be described as the direct product of the Hilbert space spanned by the original Hamiltonian with a “disorder” space that describes the random variables. On transforming to this augmented space, a new
nonrandom Hamiltonian can be defined such that configurational averages in real space for the random solid are equal to inner products in the augmented space.\textsuperscript{40, 41, 42}

In this article, we present a new approach, which combines the supercell approach and the ASF, to study theoretically the influence of position disorder of magnetic ions on magnetization and the transition temperature in diluted spin systems on a disordered Heisenberg model. Firstly, the size of the supercell determines the concentration of magnetic ions in the host materials. There is one magnetic ion per supercell which, however, can only occupy the same site in a supercell (the diluted and ordered case) or can occupy any site in a supercell (the diluted and disordered case). In the diluted and disordered case, the distance between two magnetic ions therefore becomes a random variable and then the effective Heisenberg exchange integrals, which are assumed to be a function of distance only, are random variables. In the framework of the ASF, the random exchange integrals are extended to matrices. Furthermore, the obtained spectral densities are used to calculate the temperature dependence of magnetization and Curie temperature of systems. Significantly, the spins are treated quantum mechanically in our approach although we use the supercell approach just like MC simulations. Moreover, the long-range exchange integrals are included. It should be mentioned here that the direct numerical diagonalization of the Green function is only applicable to the case of the finite size systems although it is a good method to include the long-range exchange integrals and to treat the spins quantum mechanically.

The article is organized as follows. The theoretical methods are described in section II. Section III is concerned with the numerical studies and discussion. In section IV we conclude the article with a summary.

II. THE MODEL

For a diluted spin material $A_{1-x}B_x$ (A: non-magnetic; B: magnetic), the concentration $x$ ($x \in (0,1]$) of magnetic ion B is the ratio of the number of magnetic ions to all ions. If we consider a supercell that is built by non-magnetic ions with only one substituted by magnetic ions, the size of the supercell decides the concentration $x$. For example, for simple cubic (sc) systems, the concentration $x$ equals to $1/(l \times m \times n)$ for the supercell with size $\{l \times m \times n\}$, where $l$, $m$ and $n$ refer to the numbers of ions in $X$, $Y$ and $Z$ axis, respectively. In Fig. 1 as an example, we give two kinds of possible realizations of a sc $\{2 \times 2 \times 1\}$ supercell that corresponds to the concentration $x = 25\%$: (a) diluted and ordered case, and (b) diluted and disordered case. Each magnetic ion is located in the same lattice site of the supercell in (a), but in any possible site of the supercell in (b).

To study the magnetic properties of diluted magnetic systems, we use the effective Heisenberg Hamiltonian

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

(1)

Here $i$ and $j$ label the sites occupied by the magnetic ions and $\mathbf{S}_i = \{S_i^x, S_i^y, S_i^z\}$ is the magnetic moment at lattice site $i$ with lattice vector $\mathbf{r}_i$. Moreover, in our supercell approximation, $i$ and $j$ can be also referred to the labels of the supercell because there is only one magnetic ion in each cell. The exchange parameter $J_{ij}$ is assumed to be a function of the distance $|\mathbf{r}_i - \mathbf{r}_j|$ only, independent of local environment. The Hamiltonian also contains a Zeeman coupling with external magnetic field $\mathbf{B} = (0,0,B)$. Introducing the retarded magnon Green’s function

$$G_{ij}(E) = \left\langle \left\langle S_i^+; S_j^- \right\rangle \right\rangle \frac{\gamma}{E}.$$

(2)

where $S_i^{\pm} = S_i^x \pm i S_i^y$, its equation of motion reads

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

(3)

To decouple the higher-order Green’s function, we make the Tyablikov approximation on the rhs. of (3). After rearranging, the equation of motion for $G_{ij}(E)$ is

$$\sum_{m} \left[ \left( \omega - \sum_{n} J_{in} \right) \delta_{im} + J_{im} \right] G_{mj} = \hbar \delta_{ij},$$

(4)

where $\omega = (E - g_{J} \mu_B B)/(2 \hbar \left\langle S_i^z \right\rangle)$. Thus, the matrix form of Green’s function can be expressed as:

$$G(\omega) = h (\omega I - H)^{-1}$$

(5)

where $I$ is the identity matrix and the matrix $H$ has the elements $H_{ij} = \delta_{ij} \sum_{n=1}^{N} J_{in} - J_{ij}$ which belong to the Hilbert space $\mathcal{H}$.

In the concentrated case, after Fourier transforming, one can evaluate the Green’s function from its momentum representation:

$$G(q, \omega) = \frac{\hbar}{\omega - \left[ J(0) - J(q) \right]}.$$  

(6)
Here $J(q) = \sum_{I} J_{RI} z_{I} \gamma_{I}(q)$ where $\sum_{I}$ corresponds to the summation over the $I$-th shell with a distance $R_{I}$ from a given site 0, $\gamma_{I}(q)$ is the number of sites in the $I$-th shell and $\gamma_{I}(q) = \frac{1}{\pi z_{I}} \sum_{n} e^{-ir_{I}^{2} q}$ where the summation over $r_{I}^{2}$ runs over each site located in the $I$-th shell.

The equation (5) can also be applied to the calculation of the dilated and ordered case if one considers all the distances $r_{ij}$ between two magnetic ions as equal to the $R_{ij}$ between the respective supercells. Due to the translational symmetry of the supercell, it is chosen as the Wigner-Seitz unit cell. The shell structure is determined by the spatial distribution of the supercells. For example, in Fig. 1(i) if the supercell $i$ is set as the central supercell that refers to the 0-th shell, the supercells $j$ and $k$ belong to the first shell and the supercell $l$ belongs to the second shell.

But for the diluted spin system, equation (5) is not appropriate to be used directly because each lattice has different neighboring environment and the exchange integral between two supercells is different. The translational symmetry on lattice and supercell is absent. According to (b), the dilute and disordered case of the Fig. 1 the distance between two magnetic ions, which can occupy any site in their own supercell, is a random variable.

Then all possible distances between two magnetic ions in different supercells are described as a set of independent random variables. For example, in the framework of the shell structure, the set of all possible distances between two magnetic ions, which belong to the $n$-th supercell of the $I$-th shell and the central supercell, is $\{r_{1}, r_{2}, \ldots, r_{k}\}$ with the probability $\{c_{r_{1}}, c_{r_{2}}, \ldots, c_{r_{k}}\}$ where $\sum_{i} c_{r_{i}} = 1$, respectively. It leads to the exchange integrals $J_{RI_{n}}$ represented as a random variables set $\{J_{R_{1}}, J_{R_{2}}, \ldots, J_{R_{k}}\}$ with the probability density

$$p^{k}(J_{R_{in}}) = \prod_{i=1}^{k} c_{r_{i}} \delta(J_{R_{in}} - J_{R_{i}}).$$

It should be mentioned that, according to the shell structure, the set of exchange integrals $J_{RI_{n}}$, and the probability density $p^{k}(J_{R_{in}}) \prod_{i=1}^{k} c_{r_{i}} \delta(J_{R_{in}} - J_{R_{i}})$ in the $I$-th shell are the same when $n$ runs over each supercell in the $I$-th shell. So, one simplifies them as $J_{R_{i}}$ and $p^{k}(J_{R_{i}})$.

For each random variable $J_{R_{i}}$, according to the augmented space theorem, one can introduce a new Hilbert space $\phi^{k}$ such that $p^{k}(J_{R_{i}})$ corresponds to a suitably chosen operator $M_{ij}^{k}$ on $\phi^{k}$ of rank $k$, spanned by $k$ possible values of $J_{R_{i}}$. If $\{f_{0}^{k}\}$ (usually $\{1, 0, \ldots, 0\}$) belongs to an orthonormal basis in $\phi^{k}$, then

$$p^{k}(J_{R_{i}}) = -\frac{1}{\pi} \lim_{J \to J_{RI_{n}}^{0}} \text{Im} \left( f_{0}^{k} \langle J H_{k} - M_{ij}^{k} \rangle^{-1} f_{0}^{k} \right)$$

where $I_{k}$ is the identity operator on the space $\phi^{k}$. In other words, $\{f_{0}^{k}\}$ and $M_{ij}^{k}$ are chosen such that the spectral density of the operator $M_{ij}^{k}$ with respect to $\{f_{0}^{k}\}$ is the given probability distribution. With the suitable choice of the basis, the tridiagonal matrix representation of $M_{ij}^{k}$ has $a_{l}$ ($l = 1, \ldots, k$) down the diagonal and $b_{m}$ ($m = 1, \ldots, k - 1$) down the off-diagonal positions, which can be obtained by the continued-fraction expansion:

$$p^{k}(J_{R_{i}}) = -\frac{1}{\pi} \lim_{J \to J_{RI_{n}}^{0}} \text{Im} \left( J - a_{l} - \frac{1}{\frac{b_{l}}{J - a_{l} - \frac{1}{\frac{b_{l}}{J - a_{l} - \frac{1}{\ddots}}}} \right).$$

Thus the random exchange integral $J_{RI_{n}}$ is transformed into the matrix representation $M_{ij}^{k}$ in the Hilbert space $\phi^{k}$ where the randomness of the exchange parameters is completely included.

Now, in the framework of the ASF, each supercell has the same spin term and the exchange integral from the central supercell to one special shell has the same matrix form. No supercell in the configurational distribution is distinguishable from any other. In other words, in the diluted spin system, the translational invariance that is destroyed on lattice is recovered on the supercell while the randomness is masked in the exchange integral matrix $M_{ij}^{k}$. Note, the exchange integral matrix $M_{ij}^{k}$ is the same within one and the same shell, but different among the different shells. Here, we want to mention an interesting technique: the combinatorial method which reduces initial eigenproblem to a less-dimensional one. In the case of finite systems, considering symmetry properties including methods of algebraic combinatorics, the Hamiltonian matrix is transformed to a block (quasidiagonal) form and the numerical solutions of eigenproblem are high precision and small resultant. In our approaches, due to recovering in the translational invariances, one can use the equation (5) to study an infinite system.

In the ASF, the configurational averages in real space are replaced by inner products in the augmented space $\Sigma = \mathcal{H} \otimes \Phi$, where the "disorder" space $\Phi = \phi^{1} \otimes \phi^{2} \otimes \cdots \otimes \phi^{k}$ with $\phi^{i} = \prod_{I} f_{0}^{i} \otimes \phi^{I}$ being $z_{I}$ times the direct product of $\phi^{I}$ and $z_{I}$ is the total number of the supercells in the $I$-th shell. Moreover, $|F_{0}\rangle = |F_{0}^{1}\rangle \otimes |F_{0}^{2}\rangle \cdots \otimes |F_{0}^{k}\rangle \cdots$ is an orthonormal basis in the "disorder" space $\Phi$, where $|F_{i}\rangle = \prod |f_{0}^{i} \rangle \otimes |f_{0}^{k} \rangle$. Note, in the expression of $|f_{0}^{k}\rangle$, we add the index $I$ to represent $|f_{0}^{k}\rangle$ in the I-shell. About the details of constructing the augmented space for a randomly disordered system with independent site-occupation variables, we refer the reader to a series of papers.39-40,41-42 The elements of Green function can be expressed as

$$G_{0n}(\omega) = \langle R_{0} \otimes F_{0}|\hbar \left( \omega I - \tilde{H} \right)^{-1}|R_{n} \otimes F_{0}\rangle \quad (10a)$$

$$\tilde{H} = \sum_{ij} |R_{i}\rangle\langle R_{j}| \otimes \delta_{ij} \sum_{m} J_{imi} - J_{ij} \rangle . \quad (10b)$$

Here, if $i$ refers to the central supercell and $j$ refers to the $n$-th supercell that is located in the $I$-th shell, then

$$J_{ij} = I^{1} \otimes \cdots \otimes I^{l-1} \otimes M^{I} \otimes I^{l+1} \cdots , \quad (11a)$$

$$M^{I} = \left( \prod_{1}^{n-1} \otimes I_{k} \right) \otimes M^{k}_{ij} \otimes \left( \prod_{n+1}^{+} \otimes I_{k} \right) , \quad (11b)$$
where $\prod_{m}^{m'} \otimes I_{kL}$ means $(m - m' + 1)$ times the direct product of $I_{kL}$ and $I^{L} = \prod_{i}^{+1} I_{kL}$ is the direct product of all $z_{L}$ identity matrices of rank $k_{L}$ in the $L$-th shell.

Considering the translational symmetry of the supercell, after Fourier transforming, one can evaluate the averaged Green’s function:

$$G_{00}(\mathbf{q}, \omega) = \langle F_{0} | h \left[ \omega \mathbf{I} - \mathbf{\tilde{H}}(\mathbf{q}) \right]^{-1} | F_{0} \rangle, \quad (12a)$$

$$\mathbf{\tilde{H}}(\mathbf{q}) = \mathbf{Q}(0) - \mathbf{Q}(\mathbf{q}), \quad (12b)$$

where, in the shell structure form,

$$Q(q) = \sum_{l} Q_{l}^{I}(q), \quad (13a)$$

$$Q^{I}(q) = I_{l}^{1} \otimes \cdots \otimes I_{l}^{I-1} \otimes M_{l}^{I}(q) \otimes I_{l}^{I+1} \cdots, \quad (13b)$$

$$M_{l}^{I}(q) = \sum_{n=1}^{z_{l}} M_{l}^{I}(q), \quad (13c)$$

$$M_{l}^{I}(q) = \left( \prod_{n=1}^{z_{l}} \otimes I_{l_{n}} \right) \otimes M_{l}^{I_{l}(q)} \otimes \left( \prod_{n=1}^{z_{l}} \otimes I_{l_{n}} \right), \quad (13d)$$

where $M_{l}^{I_{l}(q)} = M_{l}^{I_{l}}(q)$ (the proof being given in Appendix).

Furthermore, the averaged Green function $\langle \mathbf{G}(\omega) \rangle$ can be evaluated from its momentum representation

$$\langle \mathbf{G}(\omega) \rangle = \sum_{q} G_{00}(\mathbf{q}, \omega), \quad (14)$$

where the Lambin-Vigneron algorithm is used to do summation in $q$-space over the Brillouin zone.

Then, the magnon spectral function can be expressed as

$$A(\omega) = -\frac{1}{\pi} \text{Im} \langle \mathbf{G}(\omega) \rangle. \quad (15)$$

Using the Callen equation, magnetization reads

$$\langle S^{z} \rangle = \frac{\hbar (1 + S + \Psi) \Psi^{2S+1} + (S - \Psi) (1 + \Psi)^{2S+1}}{(1 + \Psi)^{2S+1} - \Psi^{2S+1}} \quad (16)$$

where the average magnon number $\Psi$ can be calculated by

$$\Psi = \int_{-\infty}^{+\infty} d\omega \frac{A(\omega)}{e^{\hbar(\omega)/k_{B}T} - 1}. \quad (17)$$

Considering $\langle S^{z} \rangle \to 0$ in the limit $T \to T_{C}$, one can get the transition temperature

$$k_{B}T_{C} = \frac{2}{3} \hbar S(S + 1) \left( \int_{-\infty}^{+\infty} d\omega \frac{A(\omega)}{\omega} \right)^{-1}. \quad (18)$$

### III. NUMERICAL STUDIES

In this section, the influence of dilution and disorder on magnetization and Curie temperature is investigated in diluted spin systems. For simplicity, we only consider the case of a three-dimensional system on a simple cubic lattice with the lattice constant $a = 1$ and an infinitesimal external magnetic field. In addition, the shrink of the $q$-space is included because of the Fourier transforming based on the supercells.

To study the influence of the range of a ferromagnetic exchange interaction on magnetization, respectively, power-law, RKKY and damped-RKKY type exchange interactions are used:

$$J_{ij}(R) = \begin{cases} \frac{J_{0}}{(R/k_{0})^{4}}, & (R/k_{0})^{4} \\ J_{0} \left[ \sin \left( \frac{R}{a} \right) - \left( \frac{R}{a} \right) \cos \left( \frac{R}{a} \right) \right] \left( \frac{R}{k_{0}} \right)^{4}, & \end{cases} \quad (19)$$

where $a$ is the lattice constant, $J_{0}$ is the effective nearest-neighbor interaction strength and $R$ is the distance between two magnetic ions. If the damping factor $R_{0} \to \infty$, the damped-RKKY exchange interaction is equal to the RKKY exchange interaction. In order to calculate practicably, one needs to do cutting about the exchange interaction at a distance $R_{\text{cut-off}}$. Considering the Fourier transformation based on the supercells and comparing the calculations of the diluted and ordered case and of the diluted and disordered case, the diluted and ordered case is used to decide the shell structure of systems. If the distance between one supercell and the central supercell is larger than $R_{\text{cut-off}}$, the exchange interactions will be considered as zero. It should be mentioned that the equation (19) is used just for illustration of the present approach, although the neglected effect of disorder on the value of exchange integral should be included. In addition, superexchange effects and the influence of virtual bound states on the value of exchange integral are also neglected.

Fig. 2 shows the magnon spectral density of the diluted and ordered case and the diluted and disordered case for several concentrations $x$ that correspond to the different sizes of the supercells. Because of the cut-off distance $R_{\text{cut-off}} = 2.0a$, the exchange parameters between the nearest-neighbor supercells are only included for the $\{2 \times 2 \times 2\}$ supercells’ structure that corresponds to the concentration $x = 12.5\%$. That is the reason why the calculation of the $\{2 \times 2 \times 2\}$ structure in the diluted and ordered case shows the well-known symmetric shape of a simple cubic density of states except for the change of the bandwidth. For the $\{1 \times 1 \times 2\}$ and $\{2 \times 2 \times 1\}$ supercells’ structures, in the diluted and ordered case, there are more than one peak because of including the exchange interactions belonging to next nearest-neighbor(hnn) or other longer-range supercells. In addition, there is one peak in the $\{1 \times 1 \times 2\}$ and $\{2 \times 2 \times 1\}$ structures being in the same position as that in the $\{2 \times 2 \times 2\}$ structure. It means all the magnetic ions and the exchange interactions in the
FIG. 2: The magnon spectral density for $R_{cut-off}/a = 2.0$ on an sc lattice for various concentrations $x$ (that correspond to the different sizes of the supercells) for the diluted and ordered case and the diluted and disordered case with the effective nearest-neighbor integral $J_0 = 1.0$, $S = 2.5$ and the damping factor $R_0 = 2.0a$.

$\{2 \times 2 \times 2\}$ structure are also included in the $\{1 \times 1 \times 2\}$ and $\{2 \times 2 \times 1\}$ structures. It should be noted here that the long-range exchange interactions between magnetic ions are included in the nearest-neighbor supercells because the exchange interactions are based on the positions of magnetic ions. Fig. 2 also shows that the peaks of the magnon spectral density move toward lower energies for decreasing concentrations of the magnetic ions. It shows that dilution increases the magnon spectral density for lower energies at the cost of the magnon spectral density at higher energies.

Disorder also enhances the low energy part of the magnon spectral density, especially for the $\{1 \times 1 \times 2\}$ and $\{2 \times 2 \times 1\}$ structures, if one compares the diluted and ordered case and the diluted and disordered case in Fig. 2. However, it is clearly shown in the figures that the influence of dilution is more important than that of disorder. Furthermore, the obviously separated peaks in the diluted and ordered case disappear in the diluted and disordered case because the random position distribution of the magnetic ions leads to a dispersion of the exchange integrals, while the exchange integral between two supercells is a single value in the diluted and ordered case. At the same time, it is clear that the dispersion of the exchange integrals leads to the long tails that extend to high energy magnons’ area, especially for the $\{2 \times 2 \times 2\}$ structure in the panel of power-law interactions.

It is quite interesting to note that the damping factor also increases the magnon spectral density for lower energies. From the expression of the damped-RKKY exchange integrals, one finds that the exponential damping will decrease the strength of the short-range interaction less than that of the long-range one. Compared with the RKKY exchange interaction, it leads to the clearer separation of the exchange integral distribution for the damped-RKKY exchange interaction in the diluted and ordered case. It is especially clear for the $\{2 \times 2 \times 2\}$ structure. But in the diluted and disordered case, the changing of the separation is not so obvious due to the dispersions of the exchange integrals.

The temperature dependence of magnetization, which is calculated from the magnon spectral density in Fig. 2, is shown in Fig. 3. The figures show that all curves of magnetization decrease to zero monotonically and smoothly with increasing temperature from $T = 0$ to $T = T_c$. From the equation (18), one knows that increasing magnon spectral density at lower energies will lead to decrease the Curie temperature. It is clearly shown by the influence of dilution on Curie temperature because dilution strongly shifts the magnon spectral density to lower energies. Dilution increases the distance between two magnetic ions and further decreases the strength of the exchange integrals. In other words, the influence of dilution on Curie temperature mainly comes from decreasing of the effective exchange interactions.

Disorder also influences Curie temperature $T_c$, but not so important as dilution. The random distribution of the magnetic ions leads to the dispersions of the ex-
change integrals. That increases the lower energy part of magnon spectral density and is the reason why disorder lowers the Curie temperature. It is especially clear for the \( \{1 \times 1 \times 2\} \) and \( \{2 \times 2 \times 1\} \) structures. However, for the \( \{2 \times 2 \times 2\} \) structure of power-law interactions, Curie temperature \( T_c \) is slightly higher in the disordered case than in the ordered case because the dispersions also increases the high energy part of magnon spectral density. The final effect of disorder on \( T_c \) is the results of competition between the high energy magnons and the low energy magnons. If one uses an average value or a special value to substitute the dispersion of the exchange integrals, just like the mean-field approximation \( (k_B T_c \sim \frac{1}{3N} \sum_q J^\text{eff}(q)) \) or the diluted and ordered case in our calculation, the influence of the dispersions of the exchange integrals will be washed out.

The results show that the Curie temperature is mainly influenced by dilution or the effective exchange interactions. It may explain the following fact. For a long time, it was rather surprising that the models that neglect the disorder and treat the exchange interaction in MFA provide a Curie temperature in a good agreement with the experiment. Although the positions of magnetic ions is randomly distributed, one may get a reasonably calculated Curie temperature, which is near to the experimental Curie temperature, by adjusting suitably the effective exchange integrals. Moreover, given the level of dilution of the magnetic ions, the results suggest that the way to get high Curie temperature DMS materials is increasing the effective exchange integrals \( J_0 \).

Comparing the calculation of RKKY and damped-RKKY, we find there is a big influence of the exponential damping on Curie temperature, which is even a more important factor than the position disorder of magnetic ions for \( R_0 = 2.0a \) in our calculation. The reasons to introduce the exponential damping is due to substitutional disorder or to the half-metallic character of (Ga,Mn)As and (Ge,Mn) alloy, etc.\textsuperscript{47-49}. In our theory, only the latter is meant because the positional disorder is already explicitly taken into account in our calculation. The obvious influence of damping on Curie temperature suggests that the damping factor should be chosen carefully if one only wants to incorporate the damping exponential item to include the positional disorder influence, especially in model calculation.

IV. SUMMARY

The aim of this article is to study the influence of dilution and disorder on the ferromagnetic properties of diluted spin systems. By combining the supercell approach and the ASF, we study theoretically the influence of disorder on magnetization and Curie temperature in diluted spin systems on a disordered Heisenberg model. Firstly, the size of the supercell is used to determine the concentration of magnetic ions because there is only one magnetic ion per supercell in our calculations. In order to investigate the influence of dilution and disorder on Curie temperature, the positions of magnetic ions in the supercells can be arranged in two ways: (i) the diluted and ordered case in which the magnetic ion is only able to occupy the same lattice within each supercell and (ii) the diluted and disordered case in which the magnetic ion is able to occupy any lattice point in the supercell. For the diluted and ordered case and the concentrated case, the equations of motion of the magnon Green’s function, which is decoupled by making Tyablikov approximation, can be solved directly from Fourier transformation because of the translational symmetry of the supercells. For the diluted and disordered case, the effective Heisenberg exchange integrals, which are assumed to be a function of distance only, are random variables because the positions of two magnetic ions in the supercells is randomly distributed. By using ASF, the random exchange integrals between two supercells are extended to matrices, thereby resting translational symmetry, in order to calculate the averaged magnon spectral density. Then, the obtained averaged spectral densities have been used to calculate the temperature dependence of magnetization and Curie temperature. Significantly, the long-range exchange integrals are included and all the calculations correspond to an infinite system and there is no cutting in the real space.

The resulting theory is then solved numerically for a simple cubic system. The long-range ferromagnetic exchange integrals including power-law decaying, RKKY type and damped-RKKY type are used to calculate the temperature dependence of magnetization and Curie temperature. The results show that dilution and the damping exponential item increase the magnon spectral density for lower energies at the cost of the magnon spectral density at higher energies. That leads to a decrease of the Curie temperature of spin systems. The influence of dilution on Curie temperature mainly comes from decreasing the effective exchange interactions. The effect of disorder on Curie temperature is more complicated. The random distribution of the magnetic ions leads to dispersions of the exchange integrals, which mainly decrease Curie temperature but sometimes increase \( T_c \) in the concentration area of our calculations. The role of the exponential damping of distance is obvious and should be set carefully in any phenomenological model. From our calculations, to attack the problem of high temperature ferromagnetic DMS materials, the effective way is increasing the effective exchange integrals.

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V. APPENDIX

For $R_{ij}$, if the index $i$ refers to the $I$-th shell and $j$ refers to the $j$-th supercells in $I$-th shell, we write Fourier transformation of $J_{ij} = J(R_{ij})$ as:

$$Q(q) = \sum_{R_{ij}} e^{-iqR_{ij}} J(R_{ij})$$

$$= \left[e^{-iqR_{11}} J(R_{11}) + \cdots + e^{-iqR_{1z_{1}}} J(R_{1z_{1}})\right]$$

$$+ \left[e^{-iqR_{21}} J(R_{21}) + \cdots + e^{-iqR_{2z_{2}}} J(R_{2z_{2}})\right]$$

$$+ \cdots$$

$$+ \left[e^{-iqR_{I1}} J(R_{I1}) + \cdots + e^{-iqR_{Iz_{I}}} J(R_{Iz_{I}})\right]$$

$$= \sum_{I} Q^{I}(q)$$

where $z_{I}$ is the total number of the supercells in the $I$-th shell and

$$Q^{I}(q) = \sum_{n=1}^{z_{I}} e^{-iqR_{in}} J(R_{in})$$

$$= I^{1} \otimes \cdots \otimes I^{I-1} \otimes M^{I}(q) \otimes I^{I+1} \otimes \cdots$$

$$M^{I}(q) = \sum_{n=1}^{z_{I}} e^{-iqR_{in}} q^{I} M^{I}$$

If we refer the $n$-th supercells to that of the central position $R_{in}$, i.e.,

$$\{1,2, \cdots, (z_{I}-1), z_{I}\} \rightarrow \{R_{11}, R_{I2}, \cdots, R_{I(z_{I}-1)}, R_{Iz_{I}}\},$$

$M^{I}(q)$ can be expressed as

$$M^{I}(q) = e^{-iqR_{11}} (M^{k_{I}} \otimes I_{k_{I}} \otimes I_{k_{I}} \otimes \cdots \otimes I_{k_{I}})$$

$$+ e^{-iqR_{12}} (I_{k_{I}} \otimes M^{k_{I}} \otimes I_{k_{I}} \otimes \cdots \otimes I_{k_{I}})$$

$$+ \cdots$$

$$+ e^{-iqR_{Iz_{I}}} (I_{k_{I}} \otimes I_{k_{I}} \otimes I_{k_{I}} \otimes M^{k_{I}})$$

In addition, for

$$\{1,2, \cdots, z_{I}\} \rightarrow \{R_{I2}, R_{I3}, \cdots, R_{Iz_{I}}, R_{I}\},$$

$$\{1,2, \cdots, z_{I}\} \rightarrow \{R_{Iz_{I}}, R_{I1}, \cdots, R_{I(z_{I}-2)}, R_{I(z_{I}-1)}\},$$

correspondingly, one can write

$$M^{I}(q) = e^{-iqR_{I1}} (M^{k_{I}} \otimes I_{k_{I}} \otimes I_{k_{I}} \otimes \cdots \otimes I_{k_{I}})$$

$$+ e^{-iqR_{I2}} (I_{k_{I}} \otimes M^{k_{I}} \otimes I_{k_{I}} \otimes \cdots \otimes I_{k_{I}})$$

$$+ \cdots$$

$$+ e^{-iqR_{Iz_{I}}} (I_{k_{I}} \otimes I_{k_{I}} \otimes I_{k_{I}} \otimes M^{k_{I}})$$

To sum $z_{I}$ expressions of $M^{I}(q)$ and divided by $z_{I}$, one can get

$$M^{I}(q) = \frac{1}{z_{I}} \sum_{n=1}^{z_{I}} M^{I}_{n}(q),$$

$$M^{I}_{n}(q) = \left(\prod_{1}^{n-1} \otimes I_{k_{I}} \right) \otimes M^{k_{I}}_{n}(q) \otimes \left(\prod_{n+1}^{z_{I}} \otimes I_{k_{I}} \right),$$

where $M^{k_{I}}_{n}(q) = M^{k_{I}}_{I} \gamma_{I}(q)$ and $\gamma_{I}(q) = \frac{1}{z_{I}} \sum_{n=1}^{z_{I}} e^{-iqR_{in}} q^{I}$.
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