# Simulation of spin diffusion in a disordered system

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We simulated the effect of dilution on the transport of magnetization between classical gyromagnets, coupled by a dipole-dipole interaction in a high external magnetic field, on a simple cubic lattice for a system with quenched disorder. We studied the dependence of the spin-diffusion coefficient on the concentration of dipoles on the lattice for various system sizes. For small concentrations we find that the diffusion coefficient depends *linearly* on concentration. We extrapolated the simulation results to the thermodynamic limit, i.e., to the limit of *large* system sizes and wavelengths. We also tested the consequences of a strong exponential cutoff of the dipole interactions. In this case our results show indications of a percolation transition at a finite value of the concentration. [S0163-1829(96)00725-4]

## I. INTRODUCTION

The concept of spin diffusion was introduced by Bloembergen<sup>1</sup> in 1949 to explain spinlattice relaxation measurements on insulating crystals. He proposed that impurities with paramagnetic electrons create inhomogeneities in the magnetization by transmitting Zeeman energy from the spin system to the lattice. This effect would induce a transport of magnetization between the spatially fixed nuclear spins coupled by dipolar interaction. Bloembergen derived a diffusion equation for this transport in first-order perturbation theory and estimated the diffusion coefficient. Further theoretical work was done by Redfield and Yu,<sup>2</sup> Lowe and Gade,<sup>3</sup> and Borckmans and Walgraef.<sup>4</sup> In all these papers a diffusion equation was derived under the assumptions of high external field, high temperature, and small magnetization gradient, but from different theoretical viewpoints. Nevertheless, the resulting expressions for the spin-diffusion coefficient were similar.

Experimental measurements of the spin-diffusion coefficient unfortunately appear to be rather difficult. The reason is that the diffusion is very slow, so that conventional methods cannot be used. In most of the experiments one therefore has to determine diffusion coefficients from measurements of spin-lattice relaxation times. This can only be achieved with the help of assumptions concerning the microscopic mechanism of magnetization transport in the neighborhood of the paramagnetic impurities and the coupling to the lattice vibrations. These assumptions generally cannot be verified independently.<sup>5</sup> An attempt to measure the diffusion coefficient in CaF<sub>2</sub> by a completely different method at temperatures below 1 K failed because of unexpectedly rapid spinlattice relaxation.<sup>6</sup>

In view of the difficulties in determining the microscopic transport mechanisms experimentally, it appears to be very much worthwhile to simulate the microscopic dynamics of coupled spin systems numerically. Numerical simulations, however, are also not unproblematic: since it is at present impossible to perform simulations for sufficiently large quantum-mechanical systems one has to resort to calculations for *classical gyromagnets.*<sup>7</sup> Fortunately this restriction

to classical spin systems can be justified by results from the theory of line shapes.  $^{8-10}$ 

Additional justification for the choice of classical gyromagnets comes from previous numerical work by Tang and Waugh<sup>7</sup> who showed that curves for the free-induction decay obtained for classical spins agree fairly well with experimental results. Tang and Waugh performed calculations for classical gyromagnets in a strong external magnetic field on fully occupied lattices. In their work the magnetization gradient, which is caused by the paramagnetic impurities in the experiments, is induced by an inhomogeneous initial condition for the average magnetization density with a periodic profile in one lattice direction. Tang and Waugh studied how the spin diffusion is influenced by different orientations of the lattice in the external field, by alloys of different nuclei on the same lattice, and by different lattice types. They also carried out calculations for systems with nearest-neighbor exchange couplings instead of dipole interactions between the spins. In general they found good agreement with theoretical results<sup>3,4</sup> and with experimental results by Leppelmeier and Jeener,<sup>11</sup> at least for one orientation of the lattice in the external field.

The work described in this paper was stimulated by the question how a system of nuclear spins would behave if it were diluted with randomly distributed vacancies or nonmagnetic impurities. This situation is quite common in NMR experiments on solids and biological material. It would be most interesting if experiments could be performed in the future, in which the amount of dilution (or defects) could be well controlled. We expect that such experiments, though difficult, could in principle be performed. The aim of our paper is to make predictions for the concentration dependence of the diffusion coefficients that can be tested in such future experiments. As we shall show below, one of our more important results is that the diffusion coefficient decreases relatively slowly (namely linearly) at small concentrations, suggesting that experimental determination of the diffusion coefficient at lower concentrations might still be feasible.

Apart from the experimental relevance of our paper, our results are also of interest from a theoretical point of view. At present theoretical results for disordered models with long-range interactions, with coupling constants of variable

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sign (such as dipole interactions), are to the best of our knowledge lacking entirely. Therefore we chose to resort to numerical simulations of the microscopic dynamics. Our results can be viewed as direct generalization and extension of the numerical methods used previously by Tang and Waugh.<sup>7,12</sup>

Some aspects of the research reported here are also treated in a recent paper by Sodickson and Waugh,<sup>13</sup> albeit for relatively small systems (up to  $16^3$  for the concentration dependence of diffusion coefficients). In our extrapolation to large system sizes (see below) we show that the results for  $16^3$  systems are still relatively far from the thermodynamic limit.

This paper is organized as follows. In Sec. II we give an overview over the model used for the simulation. Next (in Sec. III) we discuss the details of the simulation for the diluted spin system. In Sec. IV we present the results of our numerical simulations. Finally, in Sec. V, we discuss and summarize our findings.

### **II. THEORETICAL BACKGROUND**

We consider a system of classical spins occupying the sites of a simple cubic lattice with a probability determined by the prescribed spin concentration. The system is in a state of quenched disorder, since the spins remain on their initial sites. The spins interact with each other through the usual dipole-dipole coupling. We assume that there is a strong external magnetic field which makes transitions between states of the system highly unfavorable if the Zeeman energy is not conserved. If the dipolar interaction is modified in such a way that these transitions cannot occur at all,<sup>14</sup> the Hamiltonian of the spin system can be written as follows:

$$H = H_Z + H_1 + H_2,$$

where the Zeeman term  $H_Z$ ,  $H_1$ , and the flip-flop term  $H_2$  are given by

$$H_{Z} = -\gamma \hbar B_{0} \sum_{j} I_{jZ},$$

$$H_{1} = \sum_{i \neq j} A_{ij} I_{iZ} I_{jZ},$$

$$H_{2} = -\frac{1}{4} \sum_{i \neq j} A_{ij} (I_{i}^{+} I_{j}^{-} + I_{i}^{-} I_{j}^{+}),$$

if the external field  $B_0$  is chosen in the Z direction. Here  $\mathbf{I}_i$  is related to  $\mathbf{m}_i$ , the magnetic moment of spin *i*, in the usual way ( $\mathbf{m}_i = \gamma \hbar \mathbf{I}_i$ ). The coefficients  $A_{ij}$  are given by

$$A_{ij} = \gamma^2 \hbar^2 r_{ij}^{-3} (1 - 3\cos^2 \theta_{ij})$$
(2.1)

with  $r_{ij}$  the distance between spins *i* and *j* and  $\theta_{ij}$  the angle between the distance vector  $\mathbf{r}_{ij}$  and the direction of the external field  $B_0$ . In the frame rotating about  $\mathbf{B}_0$  at an angular velocity  $-\gamma B_0$  the term  $H_Z$  is compensated and the remaining Hamiltonian can be written in the form

$$H = -\mathbf{m}_i \cdot \mathbf{B}_i \,, \tag{2.2}$$

(2.5)

where  $\mathbf{B}_i$  is the local field for spin *i* generated by all the other spins in the system:

$$\mathbf{B}_i = \sum_{j \neq i} \mathbf{B}_{ij} \tag{2.3}$$

with

and

$$\mathbf{B}_{ij} = b_{ij} (m_X^j \hat{\mathbf{e}}_{\mathbf{X}} + m_Y^j \hat{\mathbf{e}}_{\mathbf{Y}} - 2m_Z^j \hat{\mathbf{e}}_Z)$$
(2.4)

 $b_{ii} = -\gamma^{-2} \hbar^{-2} A_{ii}.$ 

Coefficients  $A_{ij}$  are defined in Eq. (2.1). Here (X, Y, Z) refer to the coordinate system with  $\hat{\mathbf{e}}_{\mathbf{Z}}$  parallel to the external field and  $\hat{\mathbf{e}}_{\mathbf{X}}, \hat{\mathbf{e}}_{\mathbf{Y}}$  rotating about the field at the Larmor frequency  $\gamma B_0$ . The form (2.2) of the Hamiltonian suggests that the spins can be treated as classical gyromagnets precessing around their local fields  $\mathbf{B}_i$  given in Eq. (2.3). In this picture the microscopic equations of motion for the spins are<sup>14</sup>

$$\frac{d}{dt}\mathbf{m}_i = \gamma \mathbf{m}_i \times \mathbf{B}_i \quad (i = 1, \dots, N_{\text{spin}} \le N).$$
(2.6)

Here N is the number of lattice sites,  $N_{spin}$  the number of spins. If site *i* is in fact unoccupied by a spin the corresponding magnetic moment  $\mathbf{m}_i$  is understood to be identically zero.

As in the case of the ordered system,<sup>7</sup> the equations of motion (2.6) are used for the simulation: starting from a certain initial orientation  $\mathbf{m}_i(0)$ , the spins are allowed to precess around their local fields during a short time interval  $\Delta t$ . The interaction of the new magnetic moments  $\mathbf{m}_i(\Delta t)$ leads to new local fields  $\mathbf{B}_i(\Delta t)$  that have to be calculated according to Eqs. (2.3) and (2.4). These new local fields are then inserted into the equations of motion for  $t = \Delta t$ . This procedure must be repeated at each time step. For higher accuracy the magnetic moments at time  $t + \Delta t$  are approximated by a Taylor series

$$\mathbf{m}_{i}(t+\Delta t) = \sum_{n=0}^{m} \frac{\Delta t^{n}}{n!} \mathbf{m}_{i}^{(n)}(t), \qquad (2.7)$$

where the superscript (n) denotes the *n*th time derivative. The time derivatives of the magnetic moments can be calculated recursively by differentiation of Eq. (2.6)

$$\mathbf{m}_{i}^{(n+1)}(t) = \gamma [\mathbf{m}_{i}(t) \times \mathbf{B}_{i}(t)]^{(n)}$$
$$= \gamma \sum_{m=0}^{n} {n \choose m} \mathbf{m}_{i}^{(n-m)}(t) \times \mathbf{B}_{i}^{(m)}(t). \quad (2.8)$$

Here  $\mathbf{B}_{i}^{(m)}(t)(m \leq n)$  is obtained from the derivatives of Eqs. (2.3) and (2.4), that in turn depend on lower-order derivatives of the magnetic moments.

Obviously the calculation of the local fields and their derivatives costs the largest amount of computing time since it requires N sums over N-1 lattice sites for each derivative at each time step. One can however take advantage of the convolution structure of Eq. (2.3): note that the coefficients  $b_{ij}$ depend only on the distance  $r_{ij}$  between spins *i* and *j*, while the magnetic moments depend only on  $\mathbf{r}_j$ . It is therefore possible to calculate the Fourier transforms of  $b_{ij}$  and  $\mathbf{m}_j$  separately and to perform the inverse transform on their product to obtain  $\mathbf{B}_{ij}^{(n)}$ . By using this procedure one can reduce the computing time<sup>7</sup> by a factor of about *N*/ln*N*.

From the microscopic magnetic moments the magnetization density parallel to the strong external field  $\mathbf{B}_0$  can be calculated for each time step. We assume that a *diffusive* transport of magnetization occurs as a result of a spatially inhomogeneous initial condition of the magnetization. The general form of the diffusion equation is

$$\frac{\partial M(\mathbf{r},t)}{\partial t} = \sum_{\mu,\nu} D_{\mu\nu} \frac{\partial^2 M(\mathbf{r},t)}{\partial \mu \partial \nu},$$

where  $\mu, \nu$  refer to Cartesian coordinates and  $D_{\mu\nu}$  to the matrix elements of the diffusion tensor. After a transformation to principal axes the Fourier transform of the diffusion equation is

$$\frac{\partial A_{\mathbf{k}}(t)}{\partial t} = -k^2 D_{\mathbf{k}} A_{\mathbf{k}}(t), \qquad (2.9)$$

where  $A_{\mathbf{k}}(t)$  denotes the amplitude of the magnetization with wave vector  $\mathbf{k}$  and  $D_{\mathbf{k}}$  is defined by

$$D_{\mathbf{k}} = k^{-2} \sum_{\mu=1}^{3} k_{\mu}^{2} D_{\mu\mu}. \qquad (2.10)$$

The initial condition for the magnetization in the simulation is chosen in the same manner as for the fully occupied lattice:<sup>7</sup> it is uniform in the y, z plane of the crystal lattice and follows one or more cycles of a cosine profile in the x direction

$$M(\mathbf{r},0) = A_0 \cos(kx), \qquad (2.11)$$

where  $\mathbf{k} = k \hat{\mathbf{e}}_{\mathbf{x}}$  is the wave vector of this inhomogeneity. Note that the lattice axes *x*,*y*,*z* are not necessarily along the axes *X*,*Y*,*Z* of the frame rotating about  $\mathbf{B}_0$ .

For the occupied sites the Z components (i.e., the components parallel to  $\mathbf{B}_0$ ) of the initial microscopic magnetic moments are fixed between -1 and +1 with the help of a random number generator that is constructed to achieve a distribution appropriate to the magnetization profile (2.11) in each crystal plane along the x direction.<sup>7</sup> The X and Y components are then randomly selected from a uniform distribution, under the condition that  $\mathbf{m}_i$  has unit length. Magnetic moments equal to zero are assigned to the unoccupied lattice sites.

Our simulation was restricted to the situation where the external magnetic field is oriented along the [111] direction in the crystal coordinate system. For this case  $D_{\mathbf{k}}$  defined by Eq. (2.10) is proportional to the trace of the diffusion tensor:  $D_{\mathbf{k}}[111] = \frac{1}{3} \text{Tr}(D)$ . This choice is of course not a fundamental restriction. Different orientations could also be studied.

To determine  $D_{\mathbf{k}}$ , rather than fitting  $A_{\mathbf{k}}(t)$  to a decaying exponential function, one can take advantage of the fact that the microscopic state of the system is exactly known: the diffusion coefficient can be calculated directly as a function of time from Eq. (2.9)

$$D_{\mathbf{k}} = -\frac{\partial A_{\mathbf{k}}(t)/\partial t}{k^2 A_{\mathbf{k}}(t)}.$$
(2.12)

To this purpose one has to compute the magnetization and its first time derivative by summing the Z components of the magnetic moments or their time derivatives, respectively,

over slices of the lattice perpendicular to **k**, i.e., the x direction. Fourier transformation of both quantities yields  $A_{\mathbf{k}}(t)$  and  $\partial A_{\mathbf{k}}(t)/\partial t$ .

From hydrodynamics one knows<sup>15</sup> that the diffusion tensor  $D_{\mathbf{k}}$  has to be taken in the limit  $k \rightarrow 0$  or  $\lambda \rightarrow \infty$ . Simulations can of course only be performed for finite  $\lambda$ . In order to determine the diffusion tensor in the limit  $k \rightarrow 0$ , it will therefore be necessary to extrapolate the results from numerical simulations to infinitely large wavelengths and system sizes.

As the difference of two constants of the motion, namely the total energy and the Zeeman energy, the spin-spin interaction energy

$$E_{\text{int}} = -\frac{1}{2} (N_{\text{spin}})^{-1} \sum_{i=1}^{N_{\text{spin}}} \mathbf{m}_i \cdot \mathbf{B}_i$$
(2.13)

is a third constant of the motion. As remarked also by Tang in his thesis,<sup>12</sup> this quantity is always small if  $\mathbf{B}_0$  is chosen in the [111] direction. It can be used as a control parameter for the simulation.

Following Tang and Waugh,<sup>7</sup> we express all physical quantities in "reduced units." The nearest-neighbor distance  $r_0$  and the magnetic moment of one spin *m* are taken to be unity. These definitions imply that the time unit is  $r_0^3/|\gamma m|$  and the unit for the diffusion coefficient  $|\gamma m|/r_0$ . For CaF<sub>2</sub> these units are  $35.1 \times 10^6$  s and  $2.12 \times 10^{15}$ m<sup>2</sup> s<sup>-1</sup>, respectively.

# III. DETAILS OF THE SIMULATION FOR THE DISORDERED SYSTEM

Simulations were carried out for lattice sizes of  $N = l*m*n = 16^3, 32^3$  and  $64^3$ . The lower limit is due to accuracy requirements, the upper to restrictions on memory and CPU time. The routines that were used to perform the fast-Fourier transforms require that lattice dimensions are powers of 2. Periodic boundary conditions were chosen in all three spatial directions. This implies that the dipole coupling coefficients  $b_{ij}$  have to be set equal to zero on the boundaries of the interaction cell (of size N).

The configuration of occupied and unoccupied sites on the lattice was determined by the following method: for each lattice site a random number was generated from a uniform distribution on the interval [0,1]. The site was considered as unoccupied if that number was larger than the desired spin concentration  $\tilde{c} \leq 1.0$ , and occupied otherwise. The resulting spin configuration was stored in the form of occupation numbers  $p_i = 1$  or 0 for all lattice sites. The actual concentration is then given by  $c = N_{\text{spin}}/N$ .

Next we comment on the parameters  $A_0$  and  $k=2\pi/\lambda$  that determine the initial profile of the magnetization in Eq. (2.11).  $\lambda$  was chosen equal to l, l/2 and l/4 for each lattice size provided that  $l \ge \lambda \ge 16$ . The initial amplitude  $A_0$  of the inhomogeneity was fixed between 0.5 and 1.0 in such a way that the initial amplitude of the Fourier transform of the magnetization for  $k=2\pi/\lambda$ ,  $A_k(t=0)$  (that is approximately equal to  $cA_0$ ), was smaller than or equal to 0.5. We checked that different values for  $A_0$  (within sensible limits) do not

influence the results of the simulation. The number of derivatives for each time step was limited to 10 [see Eqs. (2.7), (2.8)]. Dynamic error control was used to adjust this number in the course of the simulation in order to reduce the calculating time: as explained in Ref. 12, one can assume that the cumulative error increases exponentially with time during the simulation. It is therefore most efficient to compute corrections in the form of higher-order derivatives only as long as they are larger than the estimated cumulative error at that time step. The minimum number of derivatives was chosen to be 4. Within these limits significant influence upon the results could not be observed.

A minor problem is that the decay of the amplitude  $A_{\mathbf{k}}(t)$  of the perturbation (2.9) slows down with increasing  $\lambda$  or with decreasing concentration. We therefore adjusted the length of the time interval  $\Delta t$  in order to obtain both a noticeable decline of the magnetization within 300–400 timesteps and a sufficiently large range over which  $D_{\mathbf{k}}$  could be accurately determined. On the other hand,  $\Delta t$  was not chosen larger than 1.5 reduced units in order to trace the microscopic motions of the spins with sufficient accuracy (see above, during 1 reduced unit of time a spin precesses through one radian of angle). The smallest value we used for  $\Delta t$  was 0.2.

The spin-spin interaction energy (2.13) was found to be small, namely of the order  $\pm 10^{-3}$  for all concentrations. The fact that  $E_{\rm int}$  should remain constant during the simulation was used to control the choice of the other parameters. In our simulation  $E_{\rm int}$  was found to be constant with an accuracy better than  $10^{-6}$  during one run of at least 300 time steps.

The memory required for system sizes of  $32^3$  and  $64^3$  lattice sites and 10-4 derivatives per time step is about 2.4 and 18 megawords, respectively. Computing times for 300 time steps with 10 derivatives each are about 12 min for  $32^3$  lattice sites and about 80 min for  $64^3$  lattice sites on a Cray X-MP. Because of the long calculation times only a few simulations could be carried out for each value of the concentration. Each of these was performed for a different configuration of the magnetic moments. We did not repeat simulations for the same spin configuration with varying initial conditions for the magnetization.

### **IV. RESULTS OF THE SIMULATION**

For given lattice size and wavelength of the initial perturbation we performed simulations for different spin concentrations in steps of  $\Delta c = 0.1$  or less. In the simulations we calculated the amplitude  $A_{\mathbf{k}}(t)$  of the magnetization with wave vector  $\mathbf{k}$  of the perturbation, the spin-spin interaction energy  $E_{int}(t)$  [from Eq. (2.13)] and the diffusion coefficient  $D_{\mathbf{k}}(t)$  according to Eq. (2.12). From the results for  $D_{\mathbf{k}}(t)$  we determined an appropriate time range to calculate an average value  $D_{\mathbf{k}}$  and its standard deviation: usually it took of the order of 10 time steps for the diffusion coefficient to rise from zero to a stationary value. This switch-on time was therefore neglected in the averaging procedure. We observed that the strength of the fluctuations in  $D_k$  increases when the amplitude  $A_{\mathbf{k}}(t)$  drops below 0.1 in reduced units. Furthermore the size of the fluctuations increases with decreasing lattice size or concentration, as one would expect, and with



FIG. 1. Results of a simulation for a system of size  $N=64^3$ , wavelength  $\lambda = 16$  at the concentration c = 0.9496: amplitude of the perturbation  $A_k$ , diffusion coefficient  $D_k$ , and spin-spin energy  $E_{int}$  as functions of the time in reduced units. The time step is  $\Delta t = 0.2$ . The time average of  $D_k(t)$ ,  $D_k = 0.185$ , was calculated from the data for  $4 \le t \le 50$ ; the corresponding standard deviation was found to be 0.009.

increasing wavelength of the perturbation [as the decay in  $A_{\mathbf{k}}(t)$  slows down]. Two typical examples of the output of simulations at spin concentrations near c=1.0 and at low concentrations are shown in Figs. 1 and 2 for  $N=64^3$  and  $\lambda=16$ .

The concentration was lowered until the standard deviation of  $D_{\mathbf{k}}(t)$  became of the same order of magnitude as the average  $D_{\mathbf{k}}$  itself. This limit depends of course on the system size. For 64<sup>3</sup> sites concentrations as low as c = 0.01 could be reached. The standard deviations were of the order  $10^{-2}$  to  $10^{-3}$ . For c = 1.0 the standard deviations amounted to about 5% to 8% of the value of  $D_{\mathbf{k}}$  depending on the system size and the wavelength of the perturbation. The values  $D_{\mathbf{k}}$  of the diffusion coefficient at a given system size and wavelength were plotted as a function of the spin concentration as shown



FIG. 2. The same for  $N=64^3$  and  $\lambda = 16$  at the concentration c=0.0748,  $\Delta t=0.8$ ;  $D_{\mathbf{k}}=0.016$  with standard deviation 0.002 for  $32 \le t \le 208$ .



FIG. 3. Diffusion coefficient as a function of the spin concentration for  $\lambda = 16$  at system sizes  $N = 16^3, 32^3, 64^3$ .

in Figs. 3–5. Each data point represents the average value of  $D_{\mathbf{k}}$  from a single run over a suitable time interval that was determined according to the criteria described above. Standard deviations of the order  $10^{-2}$  are of the order of the diameter of the data points. Excellent fits of the data over the whole range of *c* can be obtained just by using third-order polynomials. These are the "guides to the eye" in Figs. 3–5.

For all studied system sizes and for all values of  $\lambda$  the diffusion coefficient as a function of the spin concentration shows *linear* behavior for small values of *c* as well as for small deviations from c = 1.0. In Figs. 3–5 there is no evidence whatsoever of a percolation threshold, at least for concentrations larger than 0.01.

Figures 3 and 4 show that, for a fixed value of  $\lambda$ , the diffusion coefficient  $D_{\mathbf{k}}(c)$  is insensitive to the lattice size. This shows that (for fixed  $\lambda \ge 16$ ) the limit of infinitely large systems is reached already at  $N = \lambda^3$ . On the other hand, from our results it is evident that, if the lattice size is kept fixed,  $D_{\mathbf{k}}(c)$  grows with increasing  $\lambda$  for all  $c \le 1$ . This occurs even for the largest studied system with  $64^3$  sites (see Fig. 5). For c = 1.0 these data are in agreement with Tang and Waugh.<sup>7</sup>



FIG. 4. Diffusion coefficient as a function of the spin concentration for  $\lambda = 32$  at system sizes  $N = 32^3, 64^3$ .



FIG. 5. Diffusion coefficient as a function of the concentration for system size  $N = 64^3$  at wavelengths  $\lambda = 16,32,64$ .

The fact that the distance between the curves in Fig. 5 decreases with increasing  $\lambda$  suggests that an extrapolation of the spin-diffusion coefficient to infinitely large wavelength (or  $k \rightarrow 0$ ) might be feasible with the available data. In order to do so, we assume that the diffusion coefficient  $D_c(\lambda)$  at fixed concentration c and for a finite value of  $\lambda$  approaches the extrapolated value  $D_c(\infty)$  in the following way:

$$D_c(\lambda) = D_c(\infty)(1 - \delta \lambda^{-\alpha}). \tag{4.1}$$

This form corresponds to a **k**-dependent diffusion coefficient, with algebraic corrections to the leading  $(k \rightarrow 0)$  behavior,  $\overline{D}_c(\mathbf{k}) = \overline{D}_c(0)[1 + \mathcal{O}(k)^{\alpha}]$ . Our assumption that the corrections are *algebraic* in **k** is based on an analogy with hydrodynamics, where algebraic corrections occur in conjunction with long-time tails in the correlation functions.<sup>16</sup> These long-time tails are caused by many-body or nonlocal effects in the dynamics of a physical system. In addition, we will see below that the data are fitted well by an ansatz of the form (4.1).

We assume the parameters  $\alpha$  and  $\delta$  in Eq. (4.1) to be independent of the concentration. In a first step we determine  $\alpha$  and  $\delta$  for  $1.0 \ge c \ge 0.5$  with the help of the third-order polynomials for  $\lambda = 16$ , 32, and 64 that have been used to fit the simulated data in Fig. 5. Our criterion for the restriction of the concentration interval is that the distance between the curves for  $D_{\lambda}(c)$  in Fig. 5 should be larger than the accuracy in the simulation data (i.e., the diameter of the data points, see above) in order to obtain meaningful results. By averaging over the chosen concentration interval we find the following values:  $\alpha = 0.45$  with a standard deviation  $\sigma_{\alpha} = 0.08$ and  $\delta = 1.14$  with a standard deviation  $\sigma_{\delta} = 0.16$ . Using these results we calculate  $D_c(\infty)$  from Eq. (4.1) for fixed values of c over the whole concentration range. In Fig. 6 the diffusion coefficient for  $\lambda \rightarrow \infty$  is plotted as a function of the concentration. The indicated error bars represent the influence of the errors  $\sigma_{\alpha}$  and  $\sigma_{\delta}$ . In this figure we also repeat the fitted curves from Fig. 5 in order to allow a comparison between the simulated data and the extrapolation. It is clear from Fig. 6 that the results for  $\lambda = 16$  are still far from the thermodynamic limit.



FIG. 6. Extrapolation of the diffusion coefficient for  $\lambda \rightarrow \infty$  based on the fitted curves for  $N = 64^3$  (see Fig. 5).

We also investigated how strongly the tendency towards percolation sets in if one crosses over from long-range (dipolar) interaction to nearest-neighbor interaction. Our results for the full range dipole coupling suggest, that the qualitative dependence of the diffusion coefficient on the concentration is not influenced by the size of the system or the use of wavelengths smaller than the lattice size in the *x* direction, we therefore restricted our studies to a system with  $32^3$  sites for  $\lambda = 16$ . The dipole coupling constants  $b_{ij}$  (2.5) were replaced by

$$\tilde{b}_{ij} = b_{ij} \exp(-r_{ij}/R_c)$$

for cutoff distances  $R_c = 1.0$  and 0.5. Obviously, the exponential cutoff leads to a strong reduction of the coupling constants at larger distances. For example,  $b_{ij}$  is reduced at  $r_{ij}=8$  by a factor of  $10^{-3}$  for  $R_c=1.0$  and by  $10^{-7}$  for  $R_c=0.5$ . The results for  $R_c=0.5$  are shown in Fig. 7. The most interesting effect of the exponential cutoff is the fact that in both cases  $D_{\mathbf{k}}$  depends no longer linearly on c for concentrations below 0.2. For  $R_c=0.5$  the diffusion coeffi-



FIG. 7. Exponential cutoff of the dipole interaction with  $R_c = 0.5$ . Diffusion coefficient as a function of the concentration for system size  $N = 32^3$  at wavelength  $\lambda = 16$ .

cient approaches zero for c=0.1 whereas for  $R_c=1.0$  the percolation threshold has to lie at a lower concentration. Both data sets can be fitted well by curves of the (purely empiric) form  $\exp(-\alpha/c)(\beta c^2 + \gamma c + \delta)$ , with  $\alpha$  of the order of  $10^{-1}$ .

The conclusion of our study is that one needs an extremely strong exponential cutoff to see any tendencies toward percolation for concentrations of order  $10^{-2}$  or larger. These results demonstrate that *if* percolation occurs at all for dipolar interaction, it must occur for concentrations so small that they are numerically unreachable and probably experimentally irrelevant.

#### V. SUMMARY AND CONCLUSIONS

We studied the dependence of the spin-diffusion coefficient on the concentration of spins on a lattice by performing computer simulations of the microscopic dynamics of the system. For low concentrations we found linear behavior of the diffusion coefficient as a function of the spin concentration. We showed that the long range of the dipolar interaction is decisive for this behavior by examining how an increasingly strong exponential cutoff of the interaction leads to a percolation effect.

We investigated the concentration dependence of spindiffusion coefficients for increasing lattice size and wavelength  $\lambda$  of the perturbation. For  $\lambda$  fixed we found that the limit of infinitely large system size is already reached for  $N = \lambda^3$ .

On the other hand, the fact that the diffusion coefficients still grow with increasing  $\lambda$  for fixed system size, even for a system of  $64^3$  lattice sites (see Fig. 5), suggests that the limit  $\lambda \rightarrow \infty$  (or  $k \rightarrow 0$ ) is not yet reached. It is however possible to get an impression of what the limiting curve for the diffusion coefficient  $D_{\lambda}(c)$  would look like. As described in Sec. IV, we used the simulation results for a system size  $N = 64^3$  to determine Eq. (4.1) for the asymptotic behavior of the diffusion coefficient for  $\lambda \rightarrow \infty$  at a fixed concentration. The result of our extrapolation in Fig. 6 shows that, even at  $\lambda = 64$ , deviations from the limiting curve are rather large for all except the lowest concentrations. This is of course due to the fact that the extrapolation curve (4.1) approaches its limit very slowly, as can be illustrated by an example: if the extrapolation curve is to be taken seriously, the distance of the diffusion coefficient from the limiting value for c = 1 is reduced to 0.01 (corresponding to the error bar in Fig. 6) only for  $\lambda$  as large as 2000, which is obviously not reachable with present-day numerical methods. Accordingly, our prediction for the diffusion coefficient at  $N = 128^3$  and  $\lambda = 128$ , e.g., for c = 1, from Eq. (4.1),  $D_{128}(1.0) = 0.239 \pm 0.015$ , shows still a deviation from extrapolated large the value  $D_{\infty}(1.0) = 0.274 \pm 0.01$ . Hence, we cannot expect simulations for a system with 128<sup>3</sup> lattice sites to give a true picture of the long-wavelength limit either, but those simulations could be very helpful to improve the extrapolation curve. Such a project would no doubt require a large amount of CPU time on a computer with high memory capacity. However, the simulations could be restricted to  $\lambda = 128$ , since one can expect (see Figs. 3 and 4) that the diffusion coefficients for  $N = 128^3$  and  $\lambda = 64$ , 32, and 16 are the same as for the smaller systems.

0.3

For c = 1.0 there is generally good agreement between the simulation results<sup>7</sup> on the one hand and theoretical derivations<sup>3,4</sup> for the spin-diffusion coefficient as well as experimental results<sup>11</sup> on the other (at least for the [100] orientation of the external field relative to the crystal lattice).

In our simulation, where c < 1, it is not so easy to compare to these theories, since the theoretical work uses the translational invariance of the fully occupied lattice in an essential way. Hence, the theories of Refs. 3 and 4 cannot easily be adapted for the disordered system at low concentrations. An expansion for small deviations from c = 1.0might be possible. To our knowledge, a theory for a disordered system with dipole coupling is not available. Such a theory would necessarily be very complicated, not only because of the disorder. The additional difficulties arise from the long range of the dipole interaction and the fact that its character changes from attractive to repulsive as the orientation  $\theta_{ij}$  varies.

Comparison between our simulation results for the concentration dependence of the spin diffusion and experiments would be interesting. We are aware of the fact that an experimental investigation would be difficult. It would require preparation of a large number of samples with a specified concentration of atoms with a nonzero nuclear magnetic moment and of nonmagnetic impurities or vacancies. Furthermore, the diffusion for small concentrations of magnetic atoms is even slower than for c = 1.0, so that measurements of increased accuracy are necessary. On the other hand, for dipole coupling our simulations predict a relatively slow (linear) decrease of the diffusion coefficients for concentrations  $c \leq 0.2$ , suggesting that experiments at lower concentrations might still be feasible.

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