

Dynamics of classical spins on a lattice: Spin diffusion

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The transport of magnetization and of spin-spin energy are calculated numerically for classical gyromagnets, coupled by truncated dipole-dipole and nearest-neighbor exchange interactions, on sc, bcc, and fcc cubic lattices. For the exchange case the results agree with previous theory, and also agree with experiments on ^3He for diffusion of magnetization (but not of spin-spin energy). For the case of dipole-dipole coupling, the results are in satisfactory agreement with previous theory for the angular average of the Zeeman diffusion, but disagree with experiment. Calculations are also presented for a lattice containing alternating moments of two different species: These show how the attenuation of diffusion by foreign spins ("diffusion barrier") depends independently on the moment and gyromagnetic ratio of the foreign species.

INTRODUCTION

Spin diffusion is the transport of some entity, usually magnetization, by means of flip-flops between neighboring magnetic spins which remain physically fixed in space. It is supposed that over long distances and times the transport obeys a diffusion equation. The existence of spin diffusion has not been in doubt since it was first invoked by Bloembergen¹ to explain nuclear spin-lattice relaxation in insulating crystals, which had been found to be much faster than predicted theoretically.² It was proposed that a few of the nuclear spins were rapidly relaxed by neighboring accidental electronically paramagnetic impurities, and that relaxation of the remainder depended on diffusion of magnetization to these sinks.

The order of magnitude of the effect is easily estimated, but accurate theories are difficult. Redfield³ showed that the diffusion constant D_Z for magnetization (equivalent to Zeeman energy in an external field) could be related to the moments of a line-shape function. Later, with Yu,⁴ he extended the theory to include diffusion of spin-spin energy (D_E) and presented numerical results for the case of nearest-neighbor exchange coupling. Cowan, Mullin, and Nelson have extended the moment theory to other lattices.⁵ For the longer-range dipole-dipole coupling the theory is more difficult and requires heavier approximations: By a perturbation procedure Redfield and Yu obtained a result for D_Z on a simple cubic (SC) lattice equivalent to one of Lowe and Gade,⁶ who had made similar approximations in the context of a different theoretical approach. Borckmans and Walgraef,⁷ using more elaborate methods of irreversible statistical mechanics, also obtained results for the sc lattice which are essentially equivalent to lowest order. Morita⁸ has given a memory-function theory with applications to exchange coupling and the XY model.

Experimental comparisons are available for bcc ^3He , where very strong exchange coupling leads to diffusion sufficiently rapid to be studied by spin-echo techniques.⁹ The experimental values for D_Z (Ref. 10) are in good

agreement with Redfield and Yu, but values for D_E (Ref. 11) apparently disagree by a factor of 2. For all other cases—in particular the classic case of dipole-dipole coupling in CaF_2 —the diffusion is much slower and has had to be studied indirectly through the analysis of spin-lattice relaxation in paramagnetically doped crystals. The interpretation of such experiments is complicated by the presence of a "diffusion barrier" near the impurities: Two neighboring nuclear spins at different distances from the impurity suffer different hyperfine shifts and are thus inhibited from making energy-conserving mutual spin flips. It has been shown^{12,13} that the early part of the relaxation exhibits a $t^{1/2}$ behavior which can be related to D_Z . Leppelmeier and Jeener¹⁴ took advantage of this situation to determine D_Z in U^{3+} -doped CaF_2 . However, the large anisotropy they found for D_Z disagrees with theory, and the anisotropy in T_1 they observed is not seen in numerous other relaxation measurements.

One attempt has been made¹⁵ to measure D_Z in CaF_2 by a direct tracer method, taking advantage of the large equilibrium magnetizations and very long T_1 available at millikelvin temperatures. The experiment failed as a result of T_1 being much shorter than had been expected, but could probably be successfully repeated with suitable modifications.

It was in this context that we decided to explore spin transport phenomena by numerical computation, i.e., by what would be called molecular dynamics if the dynamical entities were atoms or molecules rather than spins. At first sight this might appear unpromising in view of the fact that nuclei of spin $\frac{1}{2}$ —at least when considered individually—would seem to display essentially quantum mechanical behavior. A genuinely quantum approach to the behavior of N coupled spins would require diagonalization of matrices of dimension 2^N : this would create a hopeless situation for N sufficiently large to bring out the "many-body" behavior that distinguishes irreversible transport from local coherence.

However, a case can be made for treating the spins as

classical gyromagnets. Van Vleck's theory of line shapes¹⁶ yields explicit formulas for the second and fourth central moments M_2 and M_4 of the line in terms of nuclear properties and lattice sums. Jensen and Hansen¹⁷ have extended this theory to M_6 and M_8 . In all these formulas, the dependences on spin quantum number I as distinct from magnetic moment m and magnetogyric ratio γ can be seen by replacing $\gamma^2 \hbar^2 I(I+1) \rightarrow m^2$ throughout. Table I shows the differences between spin $\frac{1}{2}$ and the classical limit, i.e.,

$$I \rightarrow \infty, \hbar \rightarrow 0, \sqrt{I(I+1)}\hbar = m/\gamma = \text{const} \quad (1)$$

for an sc lattice (e.g., CaF_2) with the external field in the various directions. For the [001] direction Fig. 1 shows the free-induction decay (FID) calculated numerically for classical spins by the methods to be described below, together with a representation of the experimental FID by Abragam's¹⁸ empirical fitting function

$$F(t) = e^{-(at^2/2)} \frac{\sin bt}{bt} \quad (2)$$

in which a and b are determined by the second and fourth moments. The curves agree fairly well to beyond the first zero crossing, i.e., to what might be called the characteristic time T_2 of the transverse relaxation.

Similar conclusions can be drawn from the approximate line-shape theory of Gade and Lowe,¹⁹ who found some change between $I = \frac{1}{2}$ and $I = 1$, but almost none as I was increased further.

On this basis we conclude that classical calculations of spin dynamics should yield results which, while not exact, are likely of semiquantitative accuracy. The unconvinced may regard the remainder of this paper as a possibly instructive study of the way in which spin diffusion in a *hypothetical* system of many classical gyromagnets depends on coupling Hamiltonian, field direction, and lattice structure.

Briefly put, the computational approach is as follows: a finite crystal lattice is defined in which each spin j is labeled by three array indices (l, m, n) in terms of which its crystal coordinates $\mathbf{r} = (x, y, z)$ are expressed. The Hamiltonian is expressed in terms of the interaction of each spin j with its local field \mathbf{B}_j arising from all the neighbors k

$$\mathcal{H} = -\frac{1}{2} \sum_j \mathbf{m}_j \cdot \mathbf{B}_j; \quad \mathbf{B}_j = \sum_{k,j} \mathbf{B}_j^k \quad (3)$$

The truncated dipole-dipole interaction among like spins

TABLE I. Ratios of moments for classical spins to those for spin $\frac{1}{2}$ for various field directions. Values are a simple cubic lattice, including 9^3 spins.

Orientation	M_2	M_4	M_6	M_8
[001]	1.000	1.038	1.134	1.336
[110]	1.000	1.019	1.068	1.170
[111]	1.000	1.008	1.028	1.073

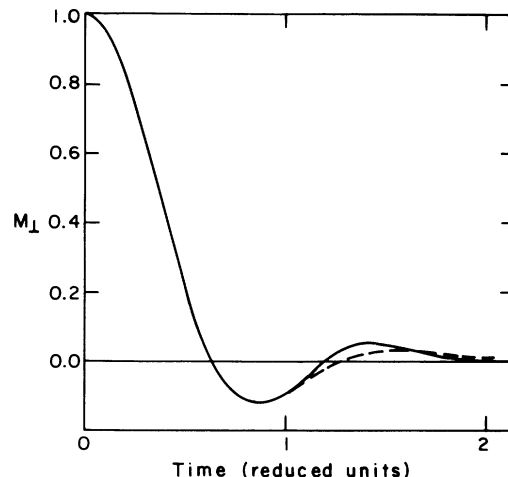


FIG. 1. Free-induction decays for a simple cubic lattice with \mathbf{B}_0 on the [001] direction. The dashed curve is the result of numerical simulations using the methods of this paper. The solid curve is a representation of the experimental FID. Similar agreement is obtained for other orientations.

in a strong external field, for example, would have

$$\begin{aligned} \mathbf{B}_j^k &= b_{jk} (m_{Xk} \mathbf{i} + m_{Yk} \mathbf{j} - 2m_{Zk} \mathbf{k}), \\ b_{jk} &= -\mathbf{r}_{jk}^{-3} P_2(\cos \vartheta_{jk}). \end{aligned} \quad (4)$$

Here (XYZ) and (ijk) refer to a laboratory coordinate system with \mathbf{k} parallel to the external field, and \mathbf{j}, \mathbf{k} rotating about the field at the Larmor frequency. ϑ_{jk} is the angle between \mathbf{r}_{jk} and \mathbf{k} . An initial condition on \mathbf{m}_i is established in which all spins are random in direction except for certain constraints which are required by the desired initial departure from a uniform infinite spin temperature. The local fields at all sites are then calculated, using cyclic boundary conditions. Each spin is now allowed to precess for a suitably short time under the torque exerted by its local field:

$$d\mathbf{m}_j/dt = \gamma \mathbf{m}_j \times \mathbf{B}_j \quad (5)$$

The calculation of local fields and precessions is repeated *ad libitum* and at suitable intervals the desired macroscopic average quantities are computed. Typically such a run is repeated a number of times, each time rerandomizing the unconstrained part of the initial conditions, to suppress fluctuations arising from the finite size of the system.

The calculations turned out to be heavier than we had anticipated, and required many hours of CPU time on the IBM 3090 system at the Cornell Supercomputer Center. In this paper we concentrate on the physically interesting of the methods used and the results. A detailed discussion of computational matters having to do with algorithmic stability, vectorization and parallel programming, dynamic error control, etc., will be published elsewhere.²⁰

DIFFUSION ON AN SC LATTICE: DIPOLAR INTERACTION

Zeeman energy

We now describe the calculation of spin diffusion in somewhat more detail. For concreteness we will focus on the diffusion of Zeeman energy in the classic case of CaF_2 , where the only nonzero nuclear moments are those of ^{19}F , occupying 100% of the points of a simple cubic lattice. Our object is to calculate the diffusion of the magnetization density M parallel to a strong external field \mathbf{B}_0 .

The general form of the diffusion equation is

$$\frac{\partial M(\mathbf{r}, t)}{\partial t} = \sum_{\mu, \nu} D_{\mu\nu} \frac{\partial^2 M(\mathbf{r}, t)}{\partial \mu \partial \nu}, \quad (6)$$

where μ, ν are any Cartesian coordinates. The anisotropy in D is of two kinds: The first is its familiar property of transforming in the laboratory as a second-rank tensor. Because of the high field, the diffusion with respect to rotating coordinates has axial symmetry about Z : In the cases we will be interested in, crystal symmetry will guarantee axial symmetry with respect to laboratory coordinates as well. The second is that the Hamiltonian itself depends on the orientation of the field with respect to crystal coordinates, as evident in the $P_2(\cos\theta)$ factor in (4). For any such direction, writing D in its principal axis system, the Fourier transform of (6) is

$$\frac{\partial A(\mathbf{k}, t)}{\partial t} = \sum_{\mu} -D_{\mu\mu} k_{\mu}^2 A(\mathbf{k}, t), \quad (7)$$

where A denotes the amplitude of the magnetization component with wave vector \mathbf{k} . Defining

$$D_{\mathbf{k}} = k^{-2} \sum_{\mu} k_{\mu}^2 D_{\mu\mu}, \quad (8)$$

(7) becomes

$$\frac{\partial A(\mathbf{k}, t)}{\partial t} = -k^2 D_{\mathbf{k}} A(\mathbf{k}, t). \quad (9)$$

This expresses the expectation that diffusion depends on direction but not on wavelength. Of course this, as well as satisfaction of a diffusion equation in the first place, is an assumption to be tested experimentally or in microscopic calculations.

Throughout we express all physical quantities in reduced units. The nearest-neighbor distance r_0 and magnetic moment of one spin are taken to be unity. One unit of time is then $r_0^3/|\gamma m|$ and one unit of D is $|\gamma m|/r_0$. For CaF_2 these amount to 35.1×10^{-6} s and 2.12×10^{-15} $\text{m}^2 \text{s}^{-1}$, respectively. By definition the magnetization \mathbf{M} , in units of m/r_0^3 , is the average magnetic moment in volume ΔV :

$$\mathbf{M} = \sum_{\Delta V} \frac{\mathbf{m}_i}{\Delta V}. \quad (10)$$

In our calculations the initial condition is generally one in which the magnetization is uniform in two of the crystal directions and follows a single cycle of a cosinusoid in

the third direction: If this is the z direction,

$$M(\mathbf{r}, 0) = A(0) \cos(kz) \quad (11)$$

with $k = 2\pi/N$, where N is the number of layers of spins in the z direction of the (finite) crystal. This is done by setting $m_{z_i}(0)$ between -1 and 1 with a biased pseudorandom number generator arranged, for each crystal plane along the z axis, to return values from a distribution appropriate to (11). m_{x_i} and m_{y_i} are then chosen to be totally random except for the requirement that \mathbf{m}_i have unit length. (A further adjustment has to be made in certain cases; see below.) N is chosen by experience to be large enough that the transport of magnetization obeys a diffusion equation and is independent of N (see above): typical values are 32 or 64. $A(0)$ is taken to be as large as possible, sometimes even approaching unity, to minimize the statistical variations of magnetization. However, care must be taken to make $A(0)$ small enough that the calculated D is independent of $A(0)$. This corresponds to the usual high-temperature (small polarization) approximation that characterizes most experimental situations. Moreover if $A(0)$ is too large, the generation of harmonic magnetization waves occurs during subsequent time development.

In practice the orientation of the field in the crystal system is always taken to be the z [001] or x [100] direction, in which case diffusion occurs parallel or perpendicular to the field, or the [111] direction, in which case the diffusion front moves at an angle of 54.7° to the field. In the last case we do not calculate the principal elements of D parallel and perpendicular to the field. However, because of the cubic symmetry we know that the result calculated is

$$\bar{D}[111] = \frac{1}{3} \text{Tr}(D) = \frac{1}{3} D_{\parallel} + \frac{2}{3} D_{\perp}.$$

An initial set of local fields at all spins is now computed. At each point only those neighbors within an "interaction region" centered on that point were included. The number of neighbors required to give stable results is typically found to be $7^3 - 1$ to $63^3 - 1$, depending on circumstances.

The spins are now allowed to precess for a short interval Δt in the local fields just computed. The precessions could easily be treated as explicit finite rotations arising from the explicit trigonometric integration of (5). Since this assumes that the local fields are constant over Δt it is tantamount to approximating by a Taylor series to first order. If Δt is small enough to give accurate results the computation becomes inefficient. Moreover this method does not assure conservation of the total Zeeman energy, which in fact is a constant of the motion. For example, consider an isolated pair of spins, with one parallel to Z and the other parallel to X . Their local fields will be along X and Z , respectively, so that during the precession m_{z1} decreases while m_{z2} remains zero. It was found that accuracy and efficiency could both be well served by an explicit use of Taylor series. From (3)–(5), denoting the n th time derivative by a superscripted (n) ,

$$\mathbf{B}_j^{(n)} = \sum_k b_{jk} (m_{Xk}^{(n)} \mathbf{i} + m_{Yk}^{(n)} \mathbf{j} - 2m_{Zk}^{(n)} \mathbf{k}) \quad (12)$$

and

$$\mathbf{m}_j^{(n+1)} = \gamma (\mathbf{m}_j \times \mathbf{B}_j)^{(n)} = \gamma \sum_{m=0}^n \binom{n}{m} \mathbf{m}_j^{(n-m)} \times \mathbf{B}_j^{(m)}. \quad (13)$$

Given the initial conditions, derivatives up to $\mathbf{m}_j^{(n)}$ can be calculated by recursion and new spin components calculated by Taylor series up to the n th order.

In (12) the summation is over all $N-1$ neighbors in the system. Although the sum often can be restricted to an interaction region containing a smaller number L of spins, it still dominates the computing time. Fortunately a large saving is possible by using the properties of Fourier transforms, especially when a vector processor is available. For the X component, for example,

$$\begin{aligned} B_X^{(n)}(\mathbf{r}_j) &= \sum_{k=1}^N b(\mathbf{r}_j - \mathbf{r}_k) m_X^{(n)}(\mathbf{r}_k) \\ &= b(\mathbf{r}_j) \otimes m_X^{(n)}(\mathbf{r}_j) \end{aligned} \quad (14)$$

$$= \mathcal{F}^{-1}(\mathcal{F}[b(\mathbf{r}_j)] \mathcal{F}[m_X^{(n)}(\mathbf{r}_j)]) \quad (15)$$

where \mathcal{F} denotes Fourier transformation and \otimes denotes convolution. By using this procedure the computing time is reduced by a factor of about $L/\log_2 N$.

The calculation of local fields and updating of spin orientations form the basis for an iteration over long times. At suitable intervals the magnetization $M(z)$ is computed by summing over slices normal to the z axis and Fourier transformed to obtain $A(t)$. According to (9) this quantity should decay exponentially with time, so that D could be obtained by curve fitting. A much more direct procedure is possible because of our knowledge of the *microscopic* state of the system at all times. This permits us to calculate the time derivative of $A(t)$ directly by summing the derivative of \mathbf{m}_j over slices and Fourier transforming. Then from (9),

$$D_{\mathbf{k}} = - \frac{\partial A(\mathbf{k}, t) / \partial t}{k^2 A(\mathbf{k}, t)}. \quad (16)$$

Figure 2 shows results of a typical run for $A(0) = 0.5$

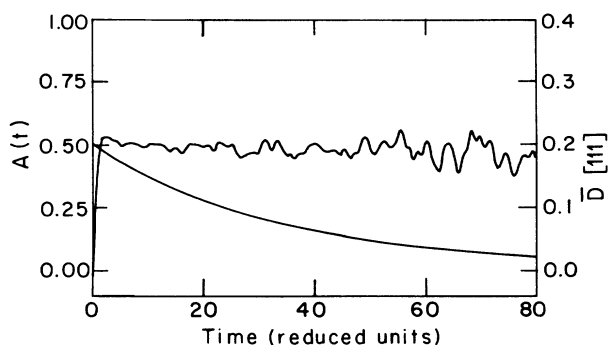


FIG. 2. Output of a representative run calculating D_z for the [111] orientation of an sc lattice. The lower curve shows the decay of the amplitude of a sinusoidal magnetization inhomogeneity with $l=16$. The upper curve traces the "instantaneous" diffusion coefficient (see text). The fluctuations in the upper curve grow with time (expressed in reduced units as defined earlier) as the amplitude of the inhomogeneity decreases.

and the field in the [111] direction. $A(t)$ shows the anticipated exponential decay. After an initial rise from zero in about one unit of time (T_2) D remains approximately constant apart from statistical fluctuations, which of course grow relatively larger as A decreases. The initial rise in D , and the corresponding but no so obvious zero initial slope of A , are consequences of the time-reversal symmetry of the dynamics and the initial condition chosen. During this transient period of unit duration the spin system approaches local thermodynamic equilibrium.

Effects of spin-spin energy on Zeeman diffusion

As already stated, the calculated diffusion constant may depend on the initial amplitude $A(0)$ if this is too large. Then one might expect a plot such as Fig. 2 to show a secular change in D , whose value at any time $D_1(t)$ would correspond to the *initial* value $D_2(0)$ for a second run with $A_2(0) = A_1(t)$. However, it is found that D sometimes depends on $A(0)$, but *not* dynamically on $A(t)$. There must therefore be some conserved quantity on which D depends. This quantity is the total spin-spin energy. When $M(\mathbf{r})$ is a function of z only, the average per spin of this quantity over the whole crystal is

$$\bar{E}_{\text{int}} = - \frac{1}{2l} \sum_{z=1}^l \overline{\mathbf{m}_j(z) \cdot \mathbf{B}_j(z)} = - \frac{1}{2l} \sum_{z=1}^l \overline{\mathbf{m}_j(z) \cdot \mathbf{B}_j(z)}, \quad (17)$$

where, for example, $\mathbf{m}_j(z)$ means the moment of a single spin in the layer at z . (Recall that z is an integer.) The average is over j (on the layer at z) and can be broken because $\mathbf{m}_j(z)$ is initialized in a way that does not depend on the moments of the neighbors k , and therefore not on the local field \mathbf{B}_j which they exert on j . Remembering that the initial averages of m_{Xj} and m_{Yj} are zero and using the fact that the spin density is unity to replace the average of m_z by M , we have

$$\bar{E}_{\text{int}} = - \frac{1}{2l} \sum_z M(z) \bar{B}_{zj}(z) = \frac{1}{l} \sum_z M(z) \sum_v b(\mathbf{r}_{jk}) \bar{m}_{zjk}, \quad (18)$$

where \mathbf{r}_{jk} refers to an interspin vector within the interaction region v centered on any j of layer z . Rewriting \bar{m}_{zjk} as $M(z+z_{jk})$, where z_{jk} is the z component of \mathbf{r}_{jk} ,

$$\bar{E}_{\text{int}} = \sum_v b(\mathbf{r}_{jk}) \langle M(z) M(z+z_{jk}) \rangle. \quad (19)$$

If the interaction region has tetragonal symmetry it can be shown that

$$\bar{E}_{\text{int}} = -P_2(\cos\alpha) \sum_v r_{jk}^{-3} P_2(\cos\beta_{jk}) \langle M(z) M(z+z_{jk}) \rangle, \quad (20)$$

where α is the angle between the z axis and the field and β is the angle between an interspin vector and z . (It can also be shown that scaling the interaction region and the wavelength of the magnetization profile by the same fac-

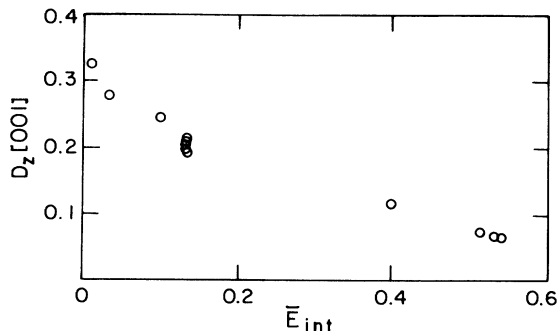


FIG. 3. Dependence of $D_Z[001]$ on spin-spin energy \bar{E}_{int} . \bar{E}_{int} is a constant of the motion, determined by the initial condition for the moments of the system.

tor does not change the spin-spin energy.)

Equation (20) shows that for the [111] orientation $\bar{E}_{\text{int}}=0$, so the effect of $A(0)$ on D does not appear. For the other orientations \bar{E}_{int} is proportional to $[A(0)]^2$ and can be adjusted, for given $A(0)$, by introducing into the initial condition a suitable correlation between the X components of the spins. Figure 3 indicates the sensitivity of $D_Z[001]$ to \bar{E}_{int} , varied by choosing $A(0)$ between 0.0625 and 0.5 and the size of the interaction region between 7^3 and 63^3 . $\bar{E}_{\text{int}} \rightarrow 0$ is appropriate to the usual experimental high-temperature approximation.

The results for diffusion of Zeeman energy are summarized and compared with previous results in Table II.

Diffusion of spin-spin energy

The spin-spin energy, quite apart from its influence on D_Z , has importance as an entity which undergoes diffusion (D_E) on its own right. In the case of exchange coupling (see below) an attempt has been made to derive a value from experiments.¹¹ This phenomenon can be calculated by essentially the same means described above. It was found that D_E depends on the distribution of magnetization even when $\bar{E}_{\text{int}}=0$. To calculate D_E for uniform magnetization and vanishing \bar{E}_{int} the initial state was assigned in such a way that alternate xy planes received equal and opposite magnetizations, but the contributions of the m_{zj} to \bar{E}_{int} are compensated by the contri-

TABLE II. Diffusion of Zeeman energy on a simple cubic lattice. The diffusion always occurs along a fourfold axis of the crystal. This is either parallel (D_{\parallel}) or perpendicular (D_{\perp}) to the external field, or else the field is in the crystal [111] direction. In the last case symmetry requires that the diffusion coefficient along any fourfold axis is the average over all directions with respect to the field.

D_{\parallel}	D_{\perp}	$\bar{D}[001]$	$\bar{D}[111]$	Ref.
0.35	0.14	0.21	0.22	This work
		0.24	0.26	6
0.33	0.11	0.19	0.18	7
		0.20	0.036	14

TABLE III. Diffusion of spin-spin energy under dipole-dipole interaction on an sc lattice.

$D_{E\perp}[001]$	$D_{E\parallel}[001]$	$\bar{D}_E[001]$
0.19	0.63	0.34

bution of the m_{xj} , needed to create a distribution of $E_{\text{int}}(z)$. It was found that D_E , by contrast with D_Z , is insensitive to A . Because of the difficulty of establishing a suitable initial distribution of $E_{\text{int}}(z)$ for the [111] orientation, $D[111]$ was not calculated for this case. Results for the [001] orientation are given in Table III.

DIFFUSION OF HEISENBERG MAGNETS

Calculations similar to the preceding have been carried out using an exchange Hamiltonian $H_{jk} = J\mathbf{I}_j \cdot \mathbf{I}_k$ with J nonzero only for nearest neighbors. For this case the local field corresponding to Eq. (4) is

$$\mathbf{B}_j^k = \frac{J}{\gamma^2 \hbar^2} \mathbf{m}_k. \quad (21)$$

Accordingly D is measured in units of $r_0^2 J \hbar^{-1} \sqrt{I(I+1)}$. Of course there is no dependence on field direction.

Inasmuch as the best known experimental case of exchange-mediated spin diffusion is in bcc ^3He , we calculated diffusion for bcc as well as sc lattices. Of course the three array indices used to label a particular spin bear an extremely simple relationship to Cartesian spin coordinates on an sc lattice. While a similar but slightly more complicated relationship can be written for other lattices, such as bcc, it is easier to make use of the fact that an sc lattice can be represented as four interpenetrating bcc lattices. By suitable logic determining which combinations of indices are to be omitted in calculating local fields, exactly the same methods already described can be used to calculate the behavior of four independent bcc crystals simultaneously.

It was found that D_Z is sensitive to A , so that quite small values of $A(0)$ (0.08) had to be used and considerable computing time incurred in order to get the desired high-temperature limit. It is interesting that the convolution method for calculating the local fields is still faster than direct summation, even though only six neighbors contribute.

Our results for D_Z and D_E on sc and bcc lattices are given in Table IV.

TABLE IV. Diffusion of Zeeman energy D_Z and internal energy D_E under nearest-neighbor exchange interactions for sc and bcc lattices.

D_Z (sc)	D_Z (bcc)	D_E (bcc)	Ref.
0.307	0.34	0.32	This work
0.296	0.328	0.34	4
	0.35	0.75	11
0.397	0.32		8

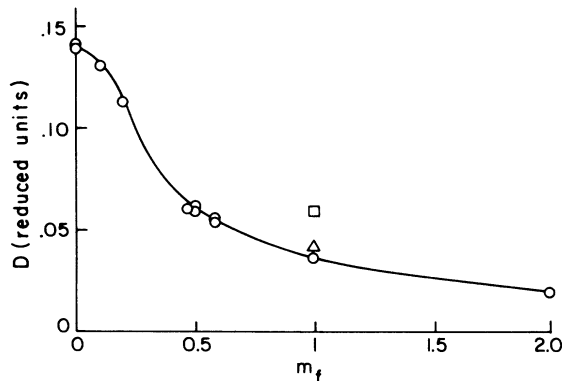


FIG. 4. Attenuation of D_Z (in the reduced units discussed in the text) for spins j on an sc lattice when alternate spins f are of a foreign species having a different magnetic moment m_f but the same magnetogyric ratio: $\gamma_f = \gamma_j$. In the limit $m_f \rightarrow 0$ the result corresponds to an fcc lattice of a single species. The extra points at $m_f = 1$ correspond to $\gamma_f = 6.7 \gamma_j$ and $67 \gamma_j$: The faster precession of the foreign spin results in partial averaging of the "diffusion barrier" which they produce for the spins j .

CRYSTAL CONTAINING TWO SPIN SPECIES

Spin diffusion depends on energy-conserving flip-flops between neighboring spins, which can occur in a strong external field only when both spins have the same Larmor frequency. A foreign moment \mathbf{m}_f can influence the diffusion among spins \mathbf{m}_j of the original species by shifting the local field at \mathbf{m}_j and thus "detuning" it to a degree from potential flip-flop partners \mathbf{m}_k . We have calculated this effect for a simple cubic lattice in which alternating spins are of different species. The local field at j was computed in the usual way except, for those neighbors of the opposite species, Eq. (4) was replaced by

$$\mathbf{B}_j^f = b_{jf}(-2m_{Zf}\mathbf{k}). \quad (22)$$

The initial condition was chosen so that the total spin-spin energy vanished. While γ_j and m_j were set equal to unity as before, the two new adjustable parameters γ_f and m_f were introduced and played appropriate roles in Eqs. (22) and (5). Note that the angular momentum of either species is m/γ . D_Z as a function of m_f is summarized in Fig. 4 for $\gamma_f = 1$. The diffusion of the normal species is inhibited, as indicated. [Note that even for $m_f = 1$ the species are distinct since (22) rather than (4) was used to calculate the local field.] Also D_Z shows a strong dependence on field direction *except* when $m_f \rightarrow 0$, when it becomes quite small, comparable to our results for an sc lattice. These results are consistent with a study by Andrew, Swanson, and Williams,²¹ who found nearly

TABLE V. Diffusion of Zeeman energy of spins j in an sc crystal in which alternate spins are of a foreign species f . The interactions are dipolar.

m_f	$\bar{D}[001]$	$\bar{D}[111]$
$=m_j$	0.050	0.14
$=0$	0.176	0.166

isotropic diffusion for single-species sc and fcc lattices, but a strong anisotropy for alkali halide lattices. The results are summarized in Table V. Note that the limit $m_f \rightarrow 0$ corresponds to a face-centered lattice of a single species.

The figure also shows two points at $m_f = 1$ for $\gamma_f = 6.7$ and $\gamma_f = 67$. The partial restoration of diffusion is a result of the increased rapidity of precession of the foreign species, which partly averages out the contribution of the local field caused by f at j . This is related to the "self-decoupling" phenomenon which has been observed in the form of line narrowing for low- γ species in solids where they have high- γ neighbors.¹⁸

CONCLUSIONS

The computed numerical values of diffusion coefficients agree quite well for the most part with previous results from approximate analytical theories based on quantum spin dynamics and from experiment. This circumstance can be taken as independent corroboration of the earlier results or as a vindication of the use of classical mechanics for the calculation of macroscopic averages of spin behavior in many-body systems. The one conspicuous disagreement involves a particularly difficult and indirect experiment on diffusion of spin-spin energy in ³He: We suspect it is the experiment or its interpretation which is at fault there.

The relatively satisfactory comparisons with previous work give us some confidence in the results for which no previous theory or experiments exist.

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