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Nuclear spin diffusion in a rare spin species

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Résumé. — Nous développons une théorie approchée du tenseur de diffusion de spin pour un système de spins rares inclus dans un système de spins abondants, avec référence spéciale à 43 Ca dans CaF₂. L'utilité de la théorie est illustrée par la description d'une étude du ferromagnétisme à domaine des spins de 19 F, où la connaissance de la constante de diffusion de 43 Ca permet de déterminer l'épaisseur des domaines ferromagnétiques.

Abstract. — We derive an approximate theory for the spin diffusion tensor in a rare nuclear spin species imbedded into an abundant one, with special reference to ${}^{43}Ca$ in CaF₂. The usefulness of the theory is examplified by the description of an investigation of ferromagnetism with domains of the ${}^{19}F$ spins, where the knowledge of the ${}^{43}Ca$ spin diffusion constant allows a determination of the ferromagnetic domain thickness.

1. Introduction. — In a system of nuclear spins at normal concentration in a solid, it is well known that flip-flop processes between the spins tend to smear out the inhomogeneities of polarization or dipolar temperature. The evolution of these inhomogeneities is tentatively described by a diffusion equation. The whole process is known as « spin diffusion ». It plays a central role for nuclear spin-lattice relaxation by fixed paramagnetic centres randomly distributed at low concentration in the solid, and accounts reasonably well for the experimental observations (see e.g. Ref. [1], p. 378).

In an experimental investigation of nuclear ferromagnetism with domains in CaF_2 [2], which will be recalled at the end of this article, we have observed phenomena attributed to the spin diffusion of the rare isotope ⁴³Ca, which could be used to determine the domain thickness if the spin diffusion constant of ⁴³Ca were known. This was the incentive for obtaining a theoretical estimate for this diffusion constant.

In the system that we consider, the spins S are randomly located on a fraction $c \ll 1$ of the sites of a crystalline lattice, and are imbedded into a regular lattice of different spins I much more abundant than the spins S. We *assume* that there is a spin diffusion among the spins S, and we use a simple argument to calculate their diffusion tensor.

The article is arranged as follows. In section 2 we calculate the flip-flop rate between two spins S. In section 3 we recall briefly the theory of diffusion within

a regular lattice, and we show why its naïve extension to a diluted lattice is incorrect. In section 4 we develop the approximate theory of the spin diffusion tensor for a diluted lattice. In section 5 we compare this theory with a different approach [3]. Finally, section 6 describes the use of the diffusion constant in the investigation of nuclear ferromagnetism with domains.

2. Flip-flop rate between two spins S. — We use a frame which is rotating with respect to each spin species at its respective Larmor frequency. In this frame, the effective Hamiltonian reduces to the secular dipole-dipole Hamiltonian

$$\mathcal{K}'_{\mathbf{D}} = \mathcal{K}'_{II} + \mathcal{K}'_{IS} + \mathcal{K}'_{SS} \tag{1}$$

with

$$\mathcal{H}_{II} = \frac{1}{2} \sum_{\mu,\nu} A_{\mu\nu} [2 I_z^{\mu} I_z^{\nu} - I_x^{\mu} I_x^{\nu} - I_y^{\mu} I_y^{\nu}] \quad (2a)$$

$$\mathcal{K}_{IS}' = \sum_{i\,\mu} C_{i\mu} \, 2 \, I_z^{\mu} \, S_z^i \tag{2b}$$

$$\mathcal{H}_{SS} = \frac{1}{2} \sum_{i,j} B_{ij} [2 S_z^i S_z^j - S_x^i S_x^j - S_y^i S_y^i]. \quad (2c)$$

The coefficients are :

$$A_{\mu\nu} = \frac{1}{2} \gamma_I^2 \,\hbar (1 - 3\cos^2\theta_{\mu\nu}) \, r_{\mu\nu}^{-3} \tag{3a}$$

$$C_{i\mu} = \frac{1}{2} \gamma_I \gamma_S \hbar (1 - 3 \cos^2 \theta_{i\mu}) r_{i\mu}^{-3} \qquad (3b)$$

$$B_{ij} = \frac{1}{2} \gamma_s^2 \hbar (1 - 3 \cos^2 \theta_{ij}) r_{ij}^{-3}$$
(3c)

Since the spins S are rare, one has

$$\mathcal{K}'_{SS} \ll \mathcal{K}'_{II}, \mathcal{K}'_{IS}$$

and the only observable effect of the small coupling \mathcal{H}'_{SS} is to induce flip-flops between spins S.

We suppose that the density matrix σ is of the form :

$$\sigma = 1 - \sum \alpha_j S_z^j \tag{4}$$

and we look for the time evolution of

$$\langle S_z^i \rangle = -\frac{1}{3}S(S+1)\alpha_i$$
.

 S_z^i commutes with \mathcal{K}'_{II} and \mathcal{K}'_{IS} , and its evolution is entirely due to \mathcal{K}'_{SS} . By a standard second-order expansion of the density matrix with respect to the perturbation \mathcal{K}'_{SS} (see e.g. ref. [1], p. 276), one obtains :

$$\frac{\mathrm{d}\sigma}{\mathrm{d}t} = -\int_0^\infty \left[\mathscr{K}_{SS}', \left[\widetilde{\mathscr{K}}_{SS}'(t'), \sigma \right] \right] \mathrm{d}t' \tag{5}$$

where :

$$\widetilde{\mathscr{H}}_{SS}(t) = \exp\left[-i(\mathscr{H}_{II}' + \mathscr{H}_{IS}')t\right] \mathscr{H}_{SS}' \times \exp\left[i(\mathscr{H}_{II}' + \mathscr{H}_{IS}')t\right]$$
(6)

whence

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle S_{z}^{i} \rangle = \mathrm{Tr} \left\{ S_{z}^{i} \frac{\mathrm{d}\sigma}{\mathrm{d}t} \right\}$$
$$= \int_{0}^{\infty} \mathrm{Tr} \left\{ \left[[S_{z}^{i}, \mathcal{K}_{SS}^{\prime}], \sigma \right] \tilde{\mathcal{K}}_{SS}^{\prime}(t^{\prime}) \right\} \mathrm{d}t^{\prime} .$$
(7)

We use reduced traces, such that Tr 1 = 1.

Through the use of equations (3c) and (4) one obtains after a little algebra :

$$\frac{1}{3}S(S+1)\frac{\mathrm{d}}{\mathrm{d}t}\alpha_{i} = -\frac{1}{2}\sum_{j}\left(\alpha_{i}-\alpha_{j}\right)B_{ij}\sum_{k,l}B_{kl}\times$$
$$\times \int_{0}^{\infty}\mathrm{Tr}\left\{\left(S_{x}^{i}S_{x}^{j}+S_{y}^{i}S_{y}^{j}\right)\left(\tilde{S}_{x}^{k}\tilde{S}_{x}^{l}+\tilde{S}_{y}^{k}\tilde{S}_{y}^{l}\right)\right\}\mathrm{d}t'.$$
(8)

The evolution of $\tilde{S}_{x}^{k}(t)$ is determined by the longitudinal dipolar field

$$2\sum_{\mu}C_{k\mu}I_{z}^{\mu}$$

which is more or less randomly modulated by the flip-flops between the spins I in the vicinity of the spin S_k . The evolutions of two spins S whose distance is much larger than the interatomic spacing between spins I will therefore be uncorrelated. If the spins S_i and S_j are well apart, a trace such as :

$$\operatorname{Tr}\left\{S_{x}^{i}S_{x}^{j}\widetilde{S}_{x}^{k}(t)\widetilde{S}_{x}^{l}(t)\right\}$$

will be non-negligible only if, say, S_k is close to S_i or is the spin S_i itself, and S_l is close to S_i or is S_i itself.

This trace is then approximately equal to :

$$\operatorname{\Gammar}\left\{S_{x}^{i}\widetilde{S}_{x}^{k}(t)\right\} \times \operatorname{Tr}\left\{S_{x}^{j}\widetilde{S}_{x}^{l}(t)\right\}.$$

The concentration c of the spins S being very small, very few spin S will have another spin S in their vicinity and, to within a negligible correction of order c, the only traces to be retained in the right-hand side of equation (8) are of the form :

$$\operatorname{Tr} \left\{ S_{x}^{i} S_{x}^{j} \tilde{S}_{x}^{i}(t) \tilde{S}_{x}^{j}(t) \right\} \simeq$$
$$\simeq \operatorname{Tr} \left\{ S_{x}^{i} \tilde{S}_{x}^{i}(t) \right\} \times \operatorname{Tr} \left\{ S_{x}^{j} \tilde{S}_{x}^{j}(t) \right\} \quad (9)$$

and similar terms for the y components. Since \mathcal{K}'_{D} is invariant by rotation around Oz, it is easily shown that :

$$\operatorname{\Gammar}\left\{S_{\mathbf{x}}^{i}\,\widetilde{S}_{\mathbf{y}}^{i}(t)\right\}=0$$

The approximation (9) is incorrect for spins S_i and S_j which are at short distance. This is of no consequence since, as will be seen in section 4, flip-flops between spins S at short distance play a negligible role in the spin diffusion.

With the approximation (9), and using the fact that :

$$\operatorname{Tr}\left\{S_{x}^{i}\widetilde{S}_{x}^{i}(t)\right\}=\operatorname{Tr}\left\{S_{y}^{i}\widetilde{S}_{y}^{i}(t)\right\}$$

is the same for all spins S_i , equation (8) becomes :

$$\frac{\mathrm{d}}{\mathrm{d}t} \alpha_i = -\sum_j W_{ij}(\alpha_i - \alpha_j) \qquad (10)$$

where :

$$V_{ij} = \frac{2}{3} S(S+1) B_{ij}^2 \int_0^\infty G^2(t) dt \qquad (11)$$

and :

with

$$G(t) = \operatorname{Tr} \left\{ S_x^i \, \widetilde{S}_x^i(t) \right\} / \operatorname{Tr} \left\{ S_x^{i^2} \right\}$$
(12)

is the free-induction decay function of the spins S, independent of \mathcal{K}'_{SS} for $c \ll 1$.

In the case when $\gamma_S \ll \gamma_I$, one has :

$$M_4/(M_2)^2 \sim (\gamma_I/\gamma_S)^2 \gg 1$$
,

where $-M_2$ and M_4 are the 2nd and 4th derivatives of G(t) at t = 0, and it can be shown (Ref. [1], p. 122) that G(t) is approximately exponential :

$$G(t) = \exp(-t/T_2) \tag{13}$$

$$\frac{1}{T_2} = \xi (M_2)^{1/2} (M_2^2/M_4)^{1/2}$$
$$= \xi M_2 (M_2/M_4)^{1/2}$$
(14)

where ξ is a numerical factor of order unity.

In CaF₂, where $\gamma_I/\gamma_S \simeq 14$ (S = ⁴³Ca; I = ¹⁹F) and $c = N({}^{43}Ca)/N({}^{40}Ca) \simeq 1.3 \times 10^{-3}$, a nearly exponential f.i.d. is indeed observed for ⁴³Ca [4].

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When G(t) is of the form (13), equation (11) yields :

$$W_{ij} = \frac{1}{3} S(S+1) T_2 B_{ij}^2, \qquad (15)$$

or else, according to equation (3c):

$$W_{ij} = K(3\cos^2\theta_{ij} - 1)^2 r_{ij}^{-6}$$
(16)

$$K = \frac{1}{12} S(S+1) T_2 \gamma_S^4 \hbar^2.$$
 (17)

The remarkable simplicity of the rate equation (10), where W_{ij} is independent of the various α_k , is a direct consequence of the presence of the abundant spins *I*. In a system containing a single spin species, either at normal or low concentration, rate equations of the form (10) are but a crude approximation.

Equations (10) are valid only in the limit when the polarizations of the spins S are small ($\alpha_i \ll 1$). We assume in the following that this is the case.

2.1 INFLUENCE OF THE POLARIZATION OF THE SPINS I. — In the above calculation, we have assumed through equation (4) that the spins I were not polarized. The existence of a non vanishing polarization of the spins I will modify the transition rate W_{ij} through its influence on the free-decay shape of the spins S, which will be of the form :

$$G(t) \propto \operatorname{Tr} \left\{ \sigma_{I} S_{x}^{i} \widetilde{S}_{x}^{i}(t) \right\}$$
(18)

where the density matrix σ_I , of the form :

$$\sigma_{I} = \exp(-\beta\omega I_{z})/\operatorname{Tr}\left\{\exp(-\beta\omega I_{z})\right\}$$

corresponds to a polarization p of the spins I.

We limit ourselves to the case when $\gamma_s \ll \gamma_I$ and to spins I = 1/2, a case pertaining to CaF₂.

A straightforward calculation not given here yields the result that both M_2 and M_4 are proportional to $(1 - p^2)$ so that, according to equations (14) and (15):

 $\frac{1}{T_2} \propto (1-p^2)$

and

$$W_{ij}(p) = W_{ij}(0)/(1 - p^2).$$
 (19)

3. Summary of diffusion theory. — Consider a particle which can be located on each one of a set of fixed sites, and jump between sites *i* and *j* with a probability per unit time W_{ij} . The rate equations for the probabilities α_i of occupation of the various sites are identical with equation (10). In the following we discuss diffusion in terms of this model of a particle jumping between sites.

Let us first consider the case when the N_0 available sites form a Bravais lattice. By using the space Fourier transforms :

$$\alpha(\mathbf{q}) = \frac{1}{\sqrt{N_o}} \sum_{i} \alpha_i \exp(i\mathbf{q} \cdot \mathbf{r}_i)$$
(20)

$$W(\mathbf{q}) = \sum_{j} W_{ij} \exp[i\mathbf{q} \cdot (\mathbf{r}_{i} - \mathbf{r}_{j})]$$
(21)

equation (10) can be written :

$$\frac{\mathrm{d}}{\mathrm{d}t} \alpha(\mathbf{q}) = - \left[W(0) - W(\mathbf{q}) \right] \alpha(\mathbf{q}) \,. \tag{22}$$

We consider the limit when $q = |\mathbf{q}|$ is small : $qa \ll 1$, where a is the lattice parameter. Since $W_{ij} = W_{ji}$, we have :

$$\begin{bmatrix} W(0) - W(q) \end{bmatrix} = \sum_{j} W_{ij} \begin{bmatrix} 1 - \exp(i\mathbf{q} \cdot \mathbf{r}_{ij}) \end{bmatrix}$$
$$\simeq \frac{1}{2} q^{2} \sum_{j} (\mathbf{n} \cdot \mathbf{r}_{ij})^{2} W_{ij}$$
$$= q^{2} D(\mathbf{n})$$
(23)

where $\mathbf{n} = \mathbf{q}/q$. $D(\mathbf{n})$ is the diffusion coefficient in the direction **n**. With respect to **n** it is a tensor with three principal values and orthogonal principal axes, say D_x , D_y , and D_z .

When the vectors q characterizing the distribution of the α_i are small, i.e. when the variation of the α_i takes place over distances much larger than the interatomic spacing, one may replace this discrete variation by a continuous one : $\alpha(\mathbf{r})$. By performing a space-Fourier transform of equation (22) one obtains, according to equation (23) :

$$\frac{\partial}{\partial t} \alpha(\mathbf{r}) = \left(D_X \frac{\partial^2}{\partial X^2} + D_Y \frac{\partial^2}{\partial Y^2} + D_Z \frac{\partial^2}{\partial Z^2} \right) \alpha(\mathbf{r}),$$
(22')

which is the usual form of a diffusion equation. The preceding treatment of diffusion is standard.

When W_{ij} is of the form (16), equation (23) yields :

$$D(\mathbf{n}) = \frac{1}{2} \sum_{j} (\mathbf{n} \cdot \mathbf{r}_{ij})^2 W_{ij}$$

= $\frac{1}{2} K \sum_{j} \cos^2 \beta_{ij} (3 \cos^2 \theta_{ij} - 1)^2 r_{ij}^{-4}$ (24)

where β_{ij} is the angle between **n** and **r**_{ij}.

Consider for instance a f.c.c. lattice of parameter a, with $\mathbf{H}_0 // \mathbf{q} // [111]$. A computation of equation (24) yields :

$$D = 18.4 K a^{-4}$$

= 2.9 K N^{4/3} (25)

where $N = 4 a^{-3}$ is the number of sites per unit volume.

The proportionality of D to $N^{4/3}$ is general, and not restricted to a particular lattice.

3.1 REMARK. — We cite without proof a classical result of diffusion theory (easily derived from equation (22')):

When the particle is at a given site at t = 0, the average of the square of its displacement in a direction **n** at time t is :

$$\langle (\mathbf{R}.\mathbf{n})^2 (t) \rangle = 2 D(\mathbf{n}) t$$

= $\sum_j (\mathbf{n}.\mathbf{r}_{ij})^2 W_{ij} t$ (26)

i.e. it is obtained by summing the squares of the displacements along \mathbf{n} of the various jumps. This result is typical of independent random processes.

Equation (26) is valid only when $\langle (\mathbf{R}.\mathbf{n})^2 \rangle^{1/2}$ is much larger than the inter-site distance, that is when the time is sufficiently long. This results from the fact that equation (23) holds only for small vectors q.

When W_{ij} is of the form (16) we can estimate the value of $D \propto \langle (\mathbf{R}.\mathbf{n})^2 \rangle$ by neglecting in equation (24) the angular factors and replacing the discrete summation by an integral :

$$D \propto KN \int_{r_0}^{\infty} r^{-4} 4 \pi r^2 dr = 4 \pi KN \int_{r_0}^{\infty} r^{-2} dr \quad (27)$$

where r_0 is of the order of *a*, which yields the qualitative conclusion that in this case diffusion proceeds essentially through frequent jumps to short distances rather than unfrequent jumps to large distances.

3.2 NAIVE EXTENSION TO A DILUTED LATTICE. — The sites available to the particle are now randomly distributed over the lattice points of a Bravais lattice at a concentration $c \ll 1$. We can tentatively use the same kind of treatment as above, with the only difference that each lattice point would be weighted by the probability that is a site available to the particle : when the particle is at a site *i*, the probability per unit time that it jumps to another site *j* of the Bravais lattice is W_{ij} multiplied by the probability that the site *j* is available. The latter is equal to *c* for all sites of the Bravais lattice, since these sites are available at random at the relative concentration *c*. We thus obtain :

$$D(c) = cD(1)$$

that is, according to the first equation (25) :

$$D \propto ca^{-4}$$
, (28)

a form independent of the lattice. The number N of available sites per unit volume being proportional to ca^{-3} , we obtain from equation (28):

$$D = \xi N^{4/3} c^{-1/3} \tag{29}$$

which means that at constant N, D is larger the smaller c, that is the smaller the parameter of the Bravais lattice. D would tend to infinity if at constant concentration per unit volume, the available sites were randomly distributed in the continuous space.

This conclusion is erroneous. The more refined theory developed in the next section in the limit $c \ll 1$ will yield a value of *D* proportional to $N^{4/3}$, but independent of *c*.

The flaw in the present treatment is the following. Let us consider a pair of sites 1 and 2 at close distance, however rare such a case may be. The concentration cbeing very small, the probability is very small to find another site close to them. In most cases, the nearest sites to the pair will be at a distance comparable with the average inter-site distance. If at a given time the particle is, say, at site 1, its subsequent motion will consist on the average of many fast jumps back and forth between sites 1 and 2 before escaping to another site. These jumps between 1 and 2 do not contribute to the diffusion, whereas in the present treatment each one is included into the book-keeping for computing the square of the average displacement of the particle. According to equation (27), these short jumps yield a large contribution to the computed value of D, which is therefore grossly overestimated.

4. Diffusion in a diluted spin system. — In this section we develop an approximate theory for the correlation between successive jumps of the particle in the limit of vanishingly small concentration c. We derive for each pair of sites i, j an « efficiency » coefficient λ_{ij} , defined as the probability that a jump between i and j contributes to increasing $(\mathbf{R}.\mathbf{n})^2$. We obtain then for the diffusion coefficient D, in place of the first equation (24) :

$$D = \frac{1}{2} \sum_{i} (\mathbf{n} \cdot \mathbf{r}_{ij})^2 W_{ij} \lambda_{ij} .$$
 (30)

The limit $c \ll 1$ corresponds to the case when the available sites are randomly distributed in a continuous space. The probability of finding a site in a volume element dv around **r** is :

$$\pi(\mathbf{r}) \,\mathrm{d}v = N \,\mathrm{d}v \,, \tag{31}$$

where N is the average number of available sites per unit volume.

In this limit, equation (30) is replaced by :

$$D = \frac{N}{2} \int (\mathbf{n} \cdot \mathbf{r})^2 W(\mathbf{r}) \lambda(\mathbf{r}) d^3r . \qquad (32)$$

4.1 SCENARIO FOR CORRELATED JUMPS. — We take a large statistical set of systems where sites available to the particle are distributed at random at an average concentration of N per unit volume, and consider statistical averages over this set. Each system of the set has a site at the origin, called site 1.

We begin by selecting the subset of systems with a second site at a position 2, and where all other sites are subjected to the conditions :

$$\sum W_{2j} = A \tag{33a}$$

$$\int_{k \neq 2} W_{1k} = B \tag{33b}$$

We suppose that at t = 0 the particle has arrived at site 1, coming from elsewhere than site 2, and we make a partial book-keeping of the subsequent jumps of the particle, irrespective of the time at which they take place, as follows. Starting from site 1, the particle may jump either to site 2 or to another site k. In the subset, a fraction :

$$\rho = B/(W_{12} + B) \tag{34}$$

of particles go to $k \neq 2$, and a fraction $(1 - \rho)$ go to site 2.

Among those which have gone to site 2, the next jump will send a fraction :

$$\mu = A/(W_{12} + A) \tag{35}$$

to a site $j \neq 1$, and a fraction $(1 - \mu)$ will jump back to site 1.

For the latter, the two successive jumps cancel each other and have no net effect on the displacement of the particle.

In order to determine the effect of those which jumped to $j \neq 1$, it would be necessary to investigate the correlations between jumps involving 3 and more sites. On physical grounds, such correlations will be important only if the various sites are at distances smaller than the average inter-site distance. The probability for such configurations being very small, we will neglect multi-site correlations and assume that the jump $2 \rightarrow j$ has rendered the preceding jump $1 \rightarrow 2$ « efficient ». For sites distributed at random in the continuous space we have on the average :

$$\langle \mathbf{r}_{12} \cdot \mathbf{r}_{2i} \rangle = 0 \tag{36}$$

and the effect of the « efficient » jump $1 \rightarrow 2$ is to increase $\langle (\mathbf{R}.\mathbf{n})^2 \rangle$ by the amount $(\mathbf{r}_{12}.\mathbf{n})^2$.

We continue the book-keeping of the jumps starting from either 1 or 2. With respect to the number of particles at site 1 at t = 0 in the subset, the proportions of those involved in these successive jumps are as follows :

1st jump :

$$1 \to k : \rho$$

$$1 \to 2 : (1 - \rho)$$

2nd jump :

$$2 \to j : (1 - \rho) \mu$$

 $2 \to 1 : (1 - \rho) (1 - \mu).$

2n th jump :

$$\begin{aligned} 2 \to j &: (1 - \rho)^n \, (1 - \mu)^{n-1} \, \mu \\ 2 \to 1 &: (1 - \rho)^n \, (1 - \mu)^n \, . \end{aligned}$$

(2n + 1)th jump :

$$\begin{split} 1 \to k : (1 - \rho)^n \, (1 - \mu)^n \, \rho \\ 1 \to 2 : (1 - \rho)^{n+1} \, (1 - \mu)^n \end{split}$$

The total proportions of jumps $1 \rightarrow 2, 2 \rightarrow 1$, and $2 \rightarrow j$ are then :

$$\begin{aligned} \Im(1 \to 2) &= (1-\rho) \sum_{0}^{\infty} \left[(1-\rho) (1-\mu) \right]^{p} \\ &= (1-\rho) / \left[1 - (1-\rho) (1-\mu) \right] \end{aligned} \tag{37a}$$

$$\mathfrak{T}(2 \to 1) = (1 - \rho) (1 - \mu) / [1 - (1 - \rho) (1 - \mu)] \quad (37b)$$

$$\mathcal{F}(2 \to j) = \mu(1-\rho)/[1-(1-\rho)(1-\mu)]. \quad (37c)$$

Since the efficient jumps between 1 and 2 are those followed by a jump $2 \rightarrow j$, the partial efficiency coefficient λ_{12} for this subset is :

$$\lambda_{12}(A, B) = \mathfrak{f}(2 \to j) / [\mathfrak{f}(1 \to 2) + \mathfrak{f}(2 \to 1)] = \mu / (2 - \mu)$$
(38)

which is independent of *B*. According to equation (35) we have :

$$\lambda_{12}(A, B) = \lambda_{12}(A) = \frac{A}{2 W_{12} + A}.$$
 (39)

The efficiency coefficient λ_{12} for the whole set is the average of (39) over all values of A :

$$\lambda_{12} = \int_0^\infty \frac{A}{2 W_{12} + A} \mathcal{F}(A) \, \mathrm{d}A \tag{40}$$

where $\mathfrak{T}(A)$ is the measure of the subsets where the probability per unit time that the particle performs a jump from site 2 is equal to A. The sites being distributed at random in a continuous space, this measure $\mathfrak{T}(A)$ is independent of the fact that, according to condition (33*a*), one should exclude the jumps to site 1. It is therefore also independent of the distance between sites 1 and 2.

4.2 SPIN DIFFUSION THROUGH DIPOLE-DIPOLE INTER-ACTIONS. — The distribution function $\mathcal{T}(A)$ is calculated in the Appendix for jump probabilities W_{ij} of the form (16). The result is :

$$\Im(A) = \frac{k}{2\sqrt{\pi}} A^{-3/2} \exp(-k^2/4A)$$
(41)

with :

$$k = \frac{16}{3} \left(\frac{\pi}{3}\right)^{3/2} N \sqrt{K} \tag{42}$$

where K is given by equation (17). Equation (40) yields then :

$$\lambda = \frac{k}{2\sqrt{\pi}} \int_0^\infty \frac{1}{(2 W + A)\sqrt{A}} \exp(-\frac{k^2}{4}A) \, dA$$

or else, by using the new variable $z = (2 W/A)^{1/2}$:

$$\lambda = \frac{k}{\sqrt{2 \pi W}} \int_0^\infty \frac{1}{1 + z^2} \exp(-z^2 k^2/8 W) \, \mathrm{d}z \,. \quad (43)$$

In the limit of large and small jump probabilities W, the efficiency coefficient λ takes the following values :

i) $k^2/8 W \ll 1$, i.e. $W \gg k^2/8$.

According to equation (42), this corresponds to :

$$W \gg \frac{32}{9} \left(\frac{\pi}{3}\right)^3 N^2 K = \frac{32 \times 16}{9} \left(\frac{\pi}{3}\right)^3 \frac{K}{d^6} \sim \frac{K}{(0.5 d)^6}$$

where d is the parameter of a f.c.c. lattice with N sites per unit volume. This case corresponds on the average to an inter-site distance $r \ll d$.

In the right-hand side of equation (43) we can replace the exponential by unity and we obtain :

$$\lambda \simeq \frac{k}{\sqrt{2 \pi W}} \int_0^\infty dz / (1 + z^2)$$
$$= \frac{\sqrt{\pi}}{2\sqrt{2}} \frac{k}{\sqrt{W}}.$$
(44)

According to equations (16) and (42) the probability for an efficient jump between two sites i and j at short distance is

$$\lambda_{ij} W_{ij} = \frac{\sqrt{\pi}}{2\sqrt{2}} k \sqrt{W_{ij}} \sim K d^{-3} r_{ij}^{-3}$$

and the contribution of these jumps to the diffusion constant, of the order of :

$$N\int_{r_0}^{r_1 \leqslant d} \lambda(r) W(r) r^4 dr \sim \frac{KN}{d^3} \int_{r_0}^{r_1 \leqslant d} r dr$$

is negligible, by contrast with the naïve model of section 3.

ii) $k^2/8 W \gg 1$, i.e. $W \ll k^2/8$.

Under the integral on the right-hand side of equation (43), the exponential decays much faster than $(1 + z^2)^{-1}$, which can be replaced by unity, and we obtain :

$$\lambda \simeq \frac{k}{\sqrt{2 \pi W}} \int_0^\infty \exp(-z^2 k^2/8 W) dz$$

= 1, (45)

so that each jump between distant sites i and j contributes to the diffusion.

The diffusion constant, defined by equation (32) depends on the direction **n**. Since according to equation (16) $W(\mathbf{r})$ depends on the orientation of **r** only through its angle θ with the external field **H**, the tensor *D* will be axially symmetric around the direction of **H**, with principal values D_{\parallel} and D_{\perp} . For the parallel case, one has in equation (24) : $\cos^2 \beta_{ij} = \cos^2 \theta_{ij}$, and for the perpendicular case :

$$\cos^2\beta_{ij}=\sin^2\theta_{ij}\cos^2\varphi_{ij}.$$

According to equations (16), (32) and (44) the expression for, say, D_{\parallel} is, writing $\cos \theta = u$:

$$D_{\parallel} = \frac{N}{2} \times 4\pi \int_{0}^{\infty} r^{4} dr \int_{0}^{1} u^{2} W(r, u) \lambda(r, u) du$$

= $\sqrt{2\pi}Nk \sqrt{K} \int_{0}^{\infty} dz \int_{0}^{1} u^{2} | 3 u^{2} - 1 | du \int_{0}^{\infty} r dr$
 $\times \left\{ \frac{1}{1 + z^{2}} \exp[-k^{2} z^{2} r^{6} / (8K(3u^{2} - 1)^{2})] \right\}.$ (46)

The integral over r is of the form :

$$J_{r} = \int_{0}^{\infty} r \exp(-\alpha r^{6}) dr = \frac{1}{2} \int_{0}^{\infty} \exp(-\alpha y^{3}) dy$$
$$= \frac{1}{2} \Gamma\left(\frac{4}{3}\right) \alpha^{-1/3}, \qquad (47)$$

 $\Gamma\left(\frac{4}{3}\right) \simeq 0.893$,

where the Γ function

and

$$\alpha^{-1/3} = \left(\frac{8 K}{k^2}\right)^{1/3} |3 u^2 - 1|^{2/3} z^{-2/3}.$$
 (48)

Then the integral over u is of the form :

$$\mathfrak{I}_{u} = \int_{0}^{1} u^{2} |3 u^{2} - 1|^{5/3} du \simeq 0.375$$

The integral over z is of the form :

$$J_z = \int_0^\infty \frac{1}{1+z^2} \, \frac{\mathrm{d}z}{z^{2/3}} = 3 \int_0^\infty \frac{\mathrm{d}y}{1+y^6} \simeq 3.11$$

and we obtain, according to equations (42), (47) and (48) and the numerical values of the integrals :

$$D_{\parallel} = 4.67 \ N^{4/3} \ K \ . \tag{49}$$

The calculation of D_{\perp} differs from that of D_{\parallel} by the form of the integral over u:

$$J_{u} = \frac{1}{2} \int_{0}^{1} (1 - u^{2}) | 3 u^{2} - 1 |^{5/3} du \simeq 0.199$$

and we obtain

$$D_{\perp} = 2.48 \ N^{4/3} \ K \ . \tag{50}$$

By comparison with equation (25), the fact of letting the sites be randomly distributed over the continuous space rather than regularly distributed over an f.c.c. lattice increases the diffusion coefficient D_{\parallel} merely by 1.6. The spin diffusion coefficient depends therefore essentially on the concentration N of spins per unit volume and is rather insensitive to the positions of these spins.

4.3 NUMERICAL ESTIMATE FOR ${}^{43}Ca$ IN CaF_2 . — The calcium spins in CaF_2 form a f.c.c. lattice whose parameter at low temperature is [5]

$$a = 5.446 \text{ Å}$$
.

The spins of ⁴³Ca occupy a fraction $c \simeq 1.3 \times 10^{-3}$ of the calcium sites. Their spin is 7/2 and their gyromagnetic ratio :

$$\gamma/2 \pi \simeq 286.5 \text{ Hz per G}$$
.

We limit ourselves to the field direction H/[111]. The experimental f.i.d. of ⁴³Ca is nearly exponential, with a time constant [4] :

 $T_2 = 217 \pm 18 \ \mu s$.

We have then :

$$N \simeq 3.2 \times 10^{19} \,\mathrm{cm}^{-3}$$

and, according to equation (17):

of errors that are listed below.

$$K \simeq 3.32 \times 10^{-45} \,\mathrm{cm}^6 \,\mathrm{s}^{-1}$$
.

Let p be the fluorine polarization. Equations (19), (49) and (50) yield :

$$D_{\parallel} \simeq 1.6 \times 10^{-18} (1 - p^2)^{-1} \,\mathrm{cm}^2 \,\mathrm{s}^{-1}$$
 (51)

$$D_{\perp} \simeq 8.4 \times 10^{-19} (1 - p^2)^{-1} \,\mathrm{cm}^2 \,\mathrm{s}^{-1}$$
. (52)

5. Discussion. Comparison with a different approach. — The rare spins are located on a Bravais lattice, and the fact of treating them as if they were distributed in the continuous space introduces a number

i) The jump from site 2 to a site other than 1 is not exactly uncorrelated with the jump $1 \rightarrow 2$, i.e. equation (36) is not strictly valid. However the correction is noticeable only for those sites 1,2 whose distance is comparable with the lattice parameter, that is much smaller than the average inter-site distance. These anomalously close pairs yield a negligible contribution to the diffusion.

ii) As stated earlier, correlations between jumps involving more than two sites were ignored, because they are important only for groups of spins at anomalously short distances.

iii) The distribution function $\mathcal{F}(A)$ is different from equation (41). On physical grounds, the difference is likely to be small except for large values of A. For spins on a Bravais lattice, there is in fact a maximum possible value for A. This will significantly affect the efficiency coefficient λ only for large W, that is for pairs of sites at close distance which contribute little to the diffusion.

iv) Finally, as stated in section 2, the flip-flop rate between close spins is not given by equations (16) and (17).

The error resulting from these approximations is likely to be of the order of c, the fraction of occupied sites of the Bravais lattice. A strong argument in favour of this estimate is that the value obtained for the diffusion tensor differs little from that for a regular f.c.c. lattice with the same spin concentration. One may indeed reasonably expect that by keeping the spin concentration constant and varying the lattice parameter, the diffusion constants will vary smoothly between c = 1 and $c \rightarrow 0$.

It is instructive to compare the present theory with a different approach to spin diffusion through dipoledipole interactions in a diluted system, developed by Vugmeister [3]. We sketch briefly the principle of this theory.

The rate equation (10) for the probability α_i can be formally integrated and yields :

$$\alpha_i(t) = \alpha_i(0) \exp\left(-\sum_j W_{ij} t\right) + \int_0^t \sum_j W_{ij} \exp\left(-\sum_k W_{ik}(t-t')\right) \alpha_j(t') dt'.$$
 (53)

One then takes the average of both sides of this equation over a statistical set of systems. The first term on the right hand side yields :

$$\langle \alpha_i(0) \rangle \left\langle \exp\left(-\sum_j W_{ij} t\right) \right\rangle = \langle \alpha_i(0) \rangle F(t)$$

where the function F(t) is computed in the Appendix (Eq. (A.9)).

For the second term the author uses the decoupling approximation :

$$\left\langle \begin{array}{l} W_{ij} \exp\left(-\sum_{k} W_{ik}(t-t')\right) \alpha_{j}(t') \end{array} \right\rangle$$
$$= \left\langle W_{ij} \exp\left(-W_{ij}(t-t')\right) \right\rangle \times$$
$$\times \left\langle \exp\left(-\sum_{k \neq j} W_{ik}(t-t')\right) \right\rangle \left\langle \alpha_{j}(t') \right\rangle$$
$$= F(t-t') c W_{ij} \exp\left(-W_{ij}(t-t')\right) \left\langle \alpha_{j}(t') \right\rangle \quad (54)$$

where, in the last line, the index j runs over all sites of the lattice of the spins S.

To proceed, one replaces the discrete summation in equation (53) by an integral over space variables, introduces the Laplace-Fourier transform :

$$\alpha(\mathbf{k}, z) = \int_0^\infty \exp(-zt) \,\mathrm{d}t \int \alpha(\mathbf{r}, t) \exp(i\mathbf{k} \cdot \mathbf{r}) \,\mathrm{d}^3 r,$$
 (55)

and obtains, in the limit $\mathbf{k}, z \to 0$ an expression of the form :

$$\alpha(\mathbf{k}, z) = \xi / [z + k^2 D]$$
(56)

whence, returning to the time variable :

$$\alpha(\mathbf{k}, t) = \alpha(\mathbf{k}, 0) \exp(-Dk^2 t)$$
 (57)

which is a diffusion equation. The diffusion constant $D(\mathbf{n})$ is of the same form as equation (32), with :

$$\lambda(W) = \int_0^\infty \frac{1}{W+A} \mathfrak{I}(A) / \left(\int_0^\infty \frac{1}{A} \mathfrak{I}(A) \, \mathrm{d}A \right). \quad (58)$$

The calculation of D_{\parallel} and D_{\perp} is analogous to that developed in the preceding section. One obtains the same anisotropy D_{\perp}/D_{\parallel} as for equations (49) and (50), and :

$$D_{\parallel} = 3.53 \ N^{4/3} \ K \tag{59}$$

which is a factor 0.76 lower than the value (49).

What is open to criticism in this theory is the decoupling approximation (54) which assumes that in the statistical set, the value of $\alpha_j(t')$ is not correlated with the existence of a site at the position *i* and with the distribution of the other sites.

The consideration of these correlations is the central point of the theory developed in section 4. The argument used for deriving these correlations is so simple and so transparent that it is our feeling that our approach is based on more convincing physical grounds than that of Vugmeister.

From a practical point of view the difference between the estimates obtained by these two methods for the diffusion tensor is insignificant.

6. Ferromagnetism of ¹⁹F and spin diffusion of ⁴³Ca in CaF₂. — We describe briefly the salient points of ferromagnetism with domains observed on the ¹⁹F spin system in CaF₂ [2, 6].

Nuclear magnetic ordering is produced in two steps : firstly a dynamic polarization of the nuclear spins by microwave irradiation close to the Larmor frequency of paramagnetic impurities $(Tm^{2+} in CaF_2)$, which decreases the nuclear spin entropy, and secondly a nuclear adiabatic demagnetization in high field by a fast passage stopped at resonance, which transforms the Zeeman order into dipolar order.

When the demagnetization is performed at negative spin temperature with the external d.c. field H_{\parallel} .[111], the theory predicts that the ordering of the ¹⁹F spins is ferromagnetic, with domains in the form of thin slices whose short axes are parallel to H, and whose magnetizations are parallel or antiparallel to H. This ordering shows up by the splitting of the ⁴³Ca resonance signal into two lines : ⁴³Ca spins located in different domains experience opposite dipolar fields from the ordered ¹⁹F spins.

It is possible to produce an imprint of the positions of the domains by saturating one of the 43 Ca lines : the 43 Ca polarization is then zero in the domains of one type and unaltered in those of the other type. It is observed that when the 19 F spins are remagnetized and then demagnetized again, the 43 Ca resonance line which had been saturated, and whose amplitude was zero, is now visible, but with an amplitude between 15 and 20 per cent of the total signal amplitude. This shows that the new ferromagnetic domains have nearly come back to the positions of the initial domains.

However, the « memory » of the 43 Ca signal, defined as the ratio $(J_1 - J_2)/(J_1 + J_2)$, where J_1 and J_2 are the intensities of the two lines, is observed to depend on the time spent in the remagnetized state prior to the second demagnetization. As a function of this time, it decays nearly exponentially with a time constant τ of the order of 20 h. This time is much longer than the dipolar spin-lattice relaxation time (of the order of 1 h), and much shorter than the Zeeman relaxation time (at least several hundred hours). The decay of the ⁴³Ca signal memory is attributed to the spin diffusion of the ⁴³Ca spins which smears out the imprint of the domains.

If we assume a uniform domain thickness d, and an initial ⁴³Ca polarization varying from p_0 to 0 along Oz in adjacent domains, we have :

$$p(z, 0) = \frac{1}{2} p_0 + \frac{2}{\pi} p_0 \sum_{n \text{ odd}} \frac{1}{n} \sin\left(\frac{\pi n z}{d}\right)$$
(60)

and at time t:

$$p(z,t) = \frac{1}{2} p_0 + \frac{2}{\pi} p_0 \sum_{n \text{ odd}} \frac{1}{n} \sin\left(\frac{\pi n z}{d}\right)$$
$$\times \exp\left(-\frac{D\pi^2 n^2}{d^2} t\right) \quad (61)$$

where D is the diffusion constant D_{\parallel} .

The term n = 1 becomes quickly dominant. The ⁴³Ca signal memory being proportional to the difference of average polarizations between different domains, is expected to decay exponentially with a time constant :

$$\tau = d^2 / (\pi^2 D) . \tag{62}$$

The ¹⁹F polarization in the domains was about $p \simeq 0.7$, and the diffusion coefficient is according to equation (51):

$$D \simeq 3.2 \times 10^{-18} \,\mathrm{cm}^2 \,\mathrm{s}^{-1}$$
. (63)

(A different value was used in reference [2], because of a numerical error). For $\tau \simeq 20$ h, equation (62) then yields a domain thickness :

$$d\simeq 150$$
 Å .

This value is sufficiently larger than the average distance between ${}^{43}Ca$ spins (~ 30 Å) to warrant the description of the evolution of the ${}^{43}Ca$ polarization by a diffusion equation. This thickness is comparable with the average distance between the paramagnetic impurities of Tm²⁺, which in this sample is about 130 Å, and suggests that the formation of the ferromagnetic domains is triggered by the paramagnetic impurities.

This result was later confirmed by a neutron diffraction study of nuclear ferromagnetism with domains in LiH [7]. The existence of domains which are thin in the direction of the (vertical) external field shows up by a vertical angular broadening of the diffracted neutron beam, as observed with a neutron multidetector. The average thickness of the domains can be obtained from this vertical elongation, and is found to be comparable with the average distance between paramagnetic impurities.

The consistency of these two experiments confirms the interpretation of the decay of the ${}^{43}Ca$ memory as being due to spin diffusion and shows that the theoretical estimate (56) for the parallel spin diffusion coefficient has the right order of magnitude.

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Appendix : distribution function $\mathfrak{I}(A)$. — The system considered in section 4 consists of a random distribution of sites with an average of N sites per unit volume. A particle can jump between sites with a probability per unit time of the form (16). We look for the proportion $\mathfrak{I}(A)$ of sites from which the probability per unit time of jumping elsewhere is equal to A.

This is done through the calculation of its Laplace transform :

$$F(t) = \int_0^\infty \mathfrak{I}(A) \exp(-At) \, \mathrm{d}A \,. \qquad (A.1)$$

This problem is formally identical with that of nuclear Zeeman spin-lattice relaxation through fixed paramagnetic impurities in the absence of spin diffusion [8], the only difference being in the angular variation of the transition probability : $\cos^2 \theta_{ij} \sin^2 \theta_{ij}$, rather than $(3 \cos^2 \theta_{ij} - 1)^2$ in the present case. As in section 4, we consider a statistical set of

As in section 4, we consider a statistical set of systems with a particle in the site 1 and calculate averages over this set.

The function F(t) is :

$$F(t) = \left\langle \exp\left(-\sum_{j} W_{1j} t\right) \right\rangle \qquad (A.2)$$

where the bracket means the average over this set.

Consider a subset $\Gamma(W)$ defined by the condition that all $W_{ij} < W$, i.e. that there is no site in a volume v(W) surrounding the site 1. The average over this subset of the decay function is called f(W, t):

$$f(W, t) = \left\langle \exp\left(-\sum_{j} W_{1j} t\right) \right\rangle_{\Gamma(W)}$$
 (A.3)

We have evidently :

$$F(t) = f(\infty, t). \qquad (A.4)$$

Consider now the subset $\Gamma(W + dW)$ with all $W_{1i} < W + dW$, that is with no site in a volume :

$$v(W + \mathrm{d}W) = v(W) - \mathrm{d}v$$

surrounding the site 1.

In this subset, there is a proportion 1 - N dvof systems with no site in the volume dv, for which the average decay function is equal to f(W, t), and a proportion N dv of systems with one site in the volume dv, for which the average decay function is $f(W, t) \times \exp(-Wt)$. We have then :

$$f(W + dW, t) = f(W, t) \{ 1 - N dv + N dv \exp(-Wt) \}$$
$$= f(W, t) \{ 1 - N dv [1 - \exp(-Wt)] \}$$
$$= f(W, t) + \frac{\partial}{\partial W} f(W, t) dW$$

whence :

$$\frac{\partial}{\partial W} \ln f(W,t) = -N[1 - \exp(-Wt)] \frac{\mathrm{d}v}{\mathrm{d}W}.$$
(A.5)

We calculate now the derivative dv/dW. An element of volume is of the form :

$$d'v = r^2 dr \sin \theta d\theta d\varphi$$
$$= \frac{1}{3} d(r^3) du d\varphi$$

where $u = \cos \theta$.

whence

The angle φ varies from 0 to 2 π and u varies from -1 to 1.

According to equation (16) we have

$$W = K(3 u^2 - 1)^2 r^{-6}$$

$$r^3 = \sqrt{\frac{K}{W}} |1 - 3 u^2|.$$
 (A.6)

The derivative dv/dW is :

$$\frac{\mathrm{d}v}{\mathrm{d}W} = \frac{1}{3} \int_0^{2\pi} \mathrm{d}\varphi \int_{-1}^1 \mathrm{d}u \times \frac{\partial |r^3|}{\partial W} \bigg|_u$$
$$= \frac{2\pi}{3} \int_{-1}^{+1} \mathrm{d}u \times \frac{\partial |r^3|}{\partial W} \bigg|_u$$

that is, according to equation (A.6) :

$$\frac{\mathrm{d}v}{\mathrm{d}W} = \frac{2\pi}{3} \frac{\sqrt{K}}{W^{3/2}} \int_0^1 |1 - 3u^2| \,\mathrm{d}u$$
$$= \frac{8\pi}{9\sqrt{3}} \frac{\sqrt{K}}{W^{3/2}}.$$
(A.7)

By inserting this value into equation (A.5) we obtain :

$$\frac{\partial}{\partial W} \ln f = -\frac{8 \pi}{9\sqrt{3}} N \sqrt{K} \qquad \frac{1 - \exp(-Wt)}{W^{3/2}}$$

whence :

and

$$\ln F(t) = \ln f(\infty, t) = -\frac{8\pi}{9\sqrt{3}} N\sqrt{K} \times \int_0^\infty \frac{1 - \exp(-Wt)}{W^{3/2}} dW$$

$$\frac{16\pi^{3/2}}{W} \sqrt{Kt} = (A, S)$$

$$= -\frac{16\pi^{3/2}}{9\sqrt{3}}N\sqrt{Kt}$$
 (A.8)

$$F(t) = \exp(-k\sqrt{t}) \qquad (A.9)$$

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with :

$$k = \frac{16 \pi^{3/2}}{9\sqrt{3}} N\sqrt{K} . \qquad (A.10)$$

The distribution function
$$\mathfrak{T}(A)$$
 whose Laplace transform is equal to $F(t)$ is [9]:

$$\Im(A) = \frac{k}{2\sqrt{\pi}} A^{-3/2} \exp\left(-\frac{k^2}{4A}\right).$$
 (A.11)

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