Spin relaxation in disordered media

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Abstract. A review is given on theoretical grounds and typical experimental appearances
of spin dynamics and relaxation in solids containing randomly distributed nuclear and/or
electronic spins. Brief content is as follows.
Disordered and magnetically diluted systems. General outlines of the spin transport
theory. Random walks in disordered systems (RWDS). Observable values in phase spin
relaxation, free induction decay (FID). Interrelation of longitudinal and transversal
relaxation related to dynamics of occupancies and phases.
Occupation number representation for equations of motion.
Continuum media approximation and inapplicability of moment expansions. Long-range
transitions vs percolation theory.
Concentration expansion as a general constructive basis for analytical methods. Scaling
properties of propagators. Singular point. Dynamical and kinematical memory in RWDS.
Ways of regrouping of concentration expansions. CTRW and semi-phenomenology.
Coherent medium approximation for nuclear relaxation via paramagnetic impurities.
Combining of memory functions and cumulant expansions for calculation of FID.
Path integral representations for RWDS.
Numerical simulations of RWDS.
Spin dynamics in magnetically diluted systems with low Zeeman and medium low dipole
temperatures.
Cluster expansions, regularization of dipole interactions and spectral dynamics.

1. Introduction

We will discuss statically disordered media, this means that constituent particles (atoms, ions and so
on) do not participate in significant translational motions during the relaxation time under discussion,
and therefore their positions are fixed (frozen) in the main approximation. The relaxation is assumed
as forced by magnetic dipole interactions typical for nuclear magnetic (NMR) and electron
paramagnetic (EPR) resonance studies. Considered systems are suggested as translational invariant in
average.

In the simplest situation, when considered system consists of spins, placed in static fluctuating
fields, we have inhomogeneous broadening of resonance line – very old and simple one-particle
problem that nevertheless produces new results up to now [1]. Other systems of interest are much
more complex for analytical studies and we will try to clarify the nature of this complexity here. Our
consideration will be concentrated on peculiarities of theoretical methods which are necessary for
adequate description of observable phenomena. More extended description of theoretical and
experimental results can be found in [2, 3].

We will distinguish below disordered and magnetically diluted spin systems.
Typical nuclear disordered spin system is formed by impurity spins randomly placed in sites of diamagnetic (in electronic degrees of freedom) matrix crystal when host nuclei have faster phase relaxation and flip-flop transitions than impurities. Important example of such systems is formed by nuclei $^6\text{Li}$ in the single crystal $^7\text{Li}^{19}\text{F}$. The system is accessible for direct experimental $\beta$-NMR study due to unique coincidence of $g$-factors of stable nuclei $^6\text{Li}$ and $\beta$-active nuclei $^8\text{Li}$ ($^E\text{Li}$-nuclei) [4, 5]. Therefore, we will consider below a model system (consisting of $^6\text{Li}$ nuclei in a LiF single crystal with addition of one $^E\text{Li}$-nucleus $^8\text{Li}$) where nuclear polarization transfers from initially polarized $^8\text{Li}$ nucleus to the nearest nonpolarized $^6\text{Li}$ nuclei and then migrates over other $^6\text{Li}$ nuclei and might return back to the $^8\text{Li}$. Difference in speeds of processes in disordered subsystem $^8\text{Li}^-^6\text{Li}$ and matrix spins $^7\text{Li}^{19}\text{F}$ produce a possibility to describe the polarization transfer within the subsystem by master equations [6]

$$\frac{\partial p_{i0}}{\partial t} = -\sum_j \left( v_{ij} p_{i0} - v_{ji} p_{0j} \right) = - \left( Ap \right)_{i0} \cdot p_{i0} \left( t = 0 \right) = \delta_{i0}, \tag{1}$$

where $p_{i0} = \langle I_i^z \rangle$ is the quantum statistical average value of the $z$-component (polarization) of the $i$-th nucleus of the system $^6\text{Li}^-^6\text{Li}$, placed at $r_i$ ($i=0$ corresponds to $^8\text{Li}$ and $i\neq0$ to $^6\text{Li}$). Polarization of $^E$-nucleus $p_{00}(t)$ is a measurable value, and $t=0$ corresponds to creation of the nucleus in the sample via $(n,\gamma)$-reaction. Here $\langle ... \rangle$ means averaging over random positions of $\beta$-nuclei in the sample, that can by replaced by ensemble averaging over random positions of all impurity nuclei $^6\text{Li}^-^6\text{Li}$. For qualitative discussion we use simplified polarization transfer rates of the form $v_{i\neq j} = v_{ij}^o / r_{ij}^6$, where $v_0$ is transfer rate at nearest distance $r_0$, $r_{ij} = r_i - r_j$, and $v_{ij} = 0$. Dipole transport corresponds to $s=6$. In this case discussed problem has a name “random walks in disordered system (RWDS) with dipole transitions”. Other details, necessary for description of real experiments can be found in cited references. It should be noted, that from more general point of view our problem presents “site problem” in RWDS in difference from “bond problem”, where positions $r_i$ fill full crystal lattice, but transition rates $v_{ij}$ are independent for different $i$ and $j$.

Typical magnetically diluted nuclear systems are formed by spins $^{29}\text{Si}$ and $^{13}\text{C}$ in insulator silicon and diamond crystals correspondingly. Electron paramagnet systems formed by impurity spins in diamagnetic media have the same nature of spin evolution, but corresponding Hamiltonians are more complicated due to anisotropy of $g$-factors and influence of fine and hyperfine interactions. Therefore, for the sake of brevity, we will discuss below homonuclear magnetically disordered systems mainly. Examples of corresponding experimental studies can be found in [7-10]. Observable values for these systems are free induction decay (FID)

$$F(t) = \langle I_+^z(t) I_-^z \rangle_0 / \langle I_+ I_- \rangle_0 \tag{2}$$

or magnetic resonance form function

$$g(\omega) = \int \frac{dt}{2\pi} \exp(i\omega t) F(t), \tag{3}$$

where $I_+ = \sum_{j=1}^N I_j^+, \ I_-(I-) = (I_-)^+, \ I_+^z(t) = \exp(iH_d t) I_+ \exp(-iH_d t)$, $\langle A \rangle_0 = \text{Tr}(A) = \text{Tr}(A) / \text{Tr}(1)$ for any operator $A$, and high temperature approximation for density matrix $\rho$ was applied. We use traditional notations for other parameters [11], for example, $I_x^y = I_x^y + iI_y^x$ and $I_x^z$ is $\alpha$–component of spin operator for $j$-th nucleus placed at $r_j$, $N$ is total number of spins in the sample and the Hamiltonian is of the form

$$H_d = \frac{1}{2} \sum_{j} b_{\alpha j} \left( I_x^2 I_x^0 - \frac{1}{3} I_x I_x^0 \right), \ b_{\alpha j} = \frac{3\hbar^2}{2\alpha_j^3} \left( 1 - 3\cos^2 \vartheta_j \right), \ b_{\alpha 0} = 0, \tag{4}$$
where \( \theta \) is an angle between \( r \) and external static magnetic field \( H_0 = (0, 0, H_0) \). Here and below we use \( \hbar = 1 \), but sometimes it is written explicitly.

Eqs. (1) and (2) present examples of longitudinal and transversal (phase) relaxation. Processes of these types are not independent. For example, transfer rate \( v \) is connected with phase relaxation of impurity nuclei, because it is proportional to \( H(t)H(t)\exp(\Delta_0 t) \), and in main approximation \( H(t)H(t)\exp(\Delta_0 t) \). Here \( \Delta_0 \) is corresponding difference of Larmor frequencies, and time dependence is produced by interaction of impurities with \( z \)-components of host spins, which should be presented by longitudinal correlation functions and so on. Important simplification consists in neglecting of influence of impurities on host spins evolution that is correct, if impurity concentration is small. Similar interrelation of longitudinal and phase relaxation exist in magnetically diluted systems as well, but both processes are realized in disordered system here.

2. Occupation numbers

Effective formalism for work with random spin systems is occupation number representation [12]. Occupation number \( n_x = 1(0) \) if the crystal site \( x \) is (is not) filled by a spin of disordered system. When all \( n_x = 1 \) the spins fill out all crystal lattice. In disordered spin systems \( n_x \) is random variable with

\[
\langle n_x n_y \rangle = c \delta_{xy} + c^2 \left(1 - \delta_{xy}\right), \quad \left\langle \prod_{j=1}^m n_{x_j} \right\rangle = c^m.
\]

Last relation implies that all \( x_j \) are different, and coinciding positions can be excluded applying the identity \( \langle n_x \rangle^2 = \langle n_x \rangle \). Other important relation: \( \varphi(n_x) = \varphi(0) + n_x \left( \varphi(1) - \varphi(0) \right) \). It is valid, if \( \varphi(0) \) and \( \varphi(1) \) exist.

Evolution (1) for disordered system can be rewritten now as

\[
\frac{d}{dt} \bar{P}_{xy} = -\sum_z \left( n_z v_{zx} \bar{P}_{xy} - n_x v_{xz} \bar{P}_{zy} \right) = -\left( A P \right)_{xy}, \quad \bar{P}_{xy}(t = 0) = \delta_{xy} \frac{n_x}{c}.
\]

Here propagator \( \bar{P}_{xy}(t) = n_x \bar{P}_{xy}(t) \) gives polarization of the lattice site \( x \) at time point \( t \) when initially the site \( y \) was polarized, and \( v_{zx} = v_{x}(r_i = z, r_j = x) \). Equation (6) is a direct consequence of Eq. (1), that is evident, if we omit all empty sites for which \( n_x = 0 \). Average propagator \( \bar{P}_{xy}(t) = \left\langle \bar{P}_{xy}(t) \right\rangle \) produces information about observable values. For example,

\[
\left\langle p_{00}(t) \right\rangle = \bar{P}_{xy}(t) = \bar{P}_{00}(t).
\]

The Hamiltonian (4) of magnetically diluted system became a form

\[
H_d = \frac{1}{2} \sum_{x,y} n_x n_y b_{xy} \left( I_x^2 I_y^2 - \frac{1}{3} I_x I_y \right), \quad (7)
\]

(where \( I_x \) is \( z \)-component of spin, placed at lattice site \( r \), and \( b_{xy} = b_{y}(r_i = z, r_j = x) \)) and FID (2) can be rewritten as

\[
F(t) = \frac{\left\langle I_x^0 I_x \right\rangle}{\left\langle I_x I_x \right\rangle} \left/ \left\langle I_x I_x \right\rangle \right\rangle, \quad I_x = \sum_x n_x I_x^x \cdot \quad (8)
\]

Here self-averaging of \( F(t) \) is applied, that is correct for macroscopic samples.

3. Continuum media approximation and inapplicability of moments expansions

Both types of spin systems have simple and very instructive exactly solvable examples with experimental realization.
If nondiagonal part of operator $\tilde{A}$ in (6) is neglected, then the equation has evident solution, and exact ensemble averaging can be fulfilled:

$$P_\gamma(t) = \delta_\gamma Q(t), \quad Q(t) = \left\{ \exp\left(-\sum_r n_r V_{\gamma r} t\right) \right\}_c = \exp\left(\sum_r \ln\left(1 - c (1 - \exp(V_{\gamma r} t))\right)\right). \quad (9)$$

If we calculate FID in magnetically diluted system applying Anderson’s Hamiltonian

$$H_A = \frac{1}{2} \sum_{x,y} n_x n_y I_x^i I_y^i,$$

then

$$n_x I_x^i(t) = \exp(iH_A t) n_x I_x^i \exp(-iH_A t) = n_x I_x^i \exp\left(i \sum_r n_r b_{\gamma r} t\right),$$

and for spins $I_j = \frac{1}{2}$ we receive

$$F(t) = F_A(t) = \prod_r \left[1 - c (1 - \cos(\frac{1}{2} b_{\gamma r} t))\right] = \exp\left(\sum_r \ln\left(1 - c (1 - \cos(\frac{1}{2} b_{\gamma r} t))\right)\right). \quad (10)$$

Simple derivation of relations (9) and (10) can be found in [2, 3].

Very important continuum media approximation (CMA) corresponds to limit $c \to 0$ and prime cell volume $\Omega \to 0$ at a fixed value of impurity density $n = c / \Omega$. Calculation of $Q(t)$ and $F_A(t)$ in CMA corresponds to retaining of leading terms in expansion of logarithms in powers of $c$ together with substitution

$$\sum_r \rightarrow \int \frac{d^d r}{\Omega},$$

where $d$ is a space dimension of the lattice. As a result, in this limit

$$Q^{\text{CMA}}(t) = \exp\left(-\left(\beta t\right)^{d/s}\right), \quad \beta = v_0 r_0^d \left\{ n \frac{\sigma_d}{d} \frac{1 - \frac{d}{s}}{\Gamma}\right\}^{1/d}, \quad (11)$$

$$F_A^{\text{CMA}}(t) = \exp\left(-\left|D_A t\right|^{d/3}\right), \quad D_A(d = 3) = \frac{2\pi^2}{3\sqrt{3}} \gamma^2 h n. \quad (12)$$

For other dimension $d \leq 2$ Anderson’s constant $D_A \sim c^{3/d}$ and it depends on orientation of the sample relative external field $\mathbf{H}_0$.

Relations (11) were first derived for $d = 3$ and $s = 6$ by Foerster [13] in studies of luminescence quenching, and relations (12) were obtained by Anderson and Abragam for $d = 3$ [11, 14].

Four main consequences should be extracted from these results:

1) Dependence on time is nonanalytical at $t = 0$ contrary to evident analytical dependence, realized before configurational averaging.

2) All dependence of effective rates of the processes on concentration consists in proportionality of these rates to transfer rates at average distance $r_\infty = \left(\Omega / c\right)^{1/d}$. 

3) If $c \ll 1$, but $v_0 t \gg 1$ (or $b_0 t \gg 1$), then deviation $1 - Q(t)$ (or $1 - F_A(t)$) is not small, and $Q(t) \approx Q^{\text{CMA}}(t)$ (or $F_A(t) \approx F_A^{\text{CMA}}(t)$). More precisely: $\left(\ln Q(t) - \ln Q^{\text{CMA}}(t)\right) / \ln Q(t) \sim c \ll 1$ and $\left(\ln F_A(t) - \ln F_A^{\text{CMA}}(t)\right) / \ln F_A(t) \sim c \ll 1$.

4) Decay of $Q(t)$ and $F_A(t)$ is fast enough to indicate, that application of a percolation theory is hopeless for such problems. For comparison we can indicate, that the percolation theory works [15] for other type of transition rates of the form $V_{\gamma r} = v_0 \exp\left(-|\mathbf{k} - \mathbf{y}| / \rho\right)$, when for large $v_0 t$ [16]

$$\ln \left\{ \exp\left(-\sum_r n_r V_{\gamma r} t\right) \right\}_c \sim -n(\rho \ln(v_0 t))^\delta$$

that is much more slow decay, than $Q(t)$ and $F_A(t)$. Main achievement of percolation theory consists in determining of a constant $\kappa_{\text{perc}}$ in exponential dependence of conductivity (which is
proportional to diffusion coefficient $D$ on concentration: $D \sim \exp(-\kappa_{\text{per}} n^{1/d} / \rho)$. This accuracy is insufficient for problems of dipole transport.

First property reflects existence of infinite transfer rates at small distances i.e. $v_y(r_y \to 0) \to \infty$ and $b_y(r_y \to 0) \to \infty$. Second statement means that $\beta \sim v_y(r_y = r_m)$ and $D_d \sim b_y(r_y = r_m)$. Third consequence indicates, that if $c \ll 1$, then CMA produces satisfactory approximation to exact results in most important time region, where deviations of observables from their initial values are not very small.

If $c = 1$ (ordered system), then spin dynamics is based on calculation of first terms of expansion of observables in time series and in consequent rearrangement of these series in physically adopted forms. Corresponding coefficients for FID have name “moments” [11], because they can be connected with moments of form function $M_n = \int d\omega \omega^n g(\omega)$. Examples of effective realization of this program can be found in [17, 18]. Consequences, listed above, indicate that this strategy is impossible for systems with strong disorder, where $c \ll 1$, because main constructive approximation for this case (CMA) produces infinite moments. Other equivalent point of view states, that finite number of moments can describe only small part ($\sim c$) of the total variation of observables during the evolution that is clearly seen in Eqs. (9) and (10).

4. Concentration expansions

Constructive way to treat disordered spin systems with small impurity concentration $c$ consists in application of expansions of observables in terms of powers of concentration instead of time Taylor’s series [19, 20]. Simple derivation of the expansion was invented in [19]. It is based on linearity in occupation numbers of the operator $\tilde{A}$ in (6)

$$\tilde{A}_{xy} = \sum_z A_{xy}^z n_z^x, \quad A_{xy}^z = \delta_{xy} V_{xy} - \delta_z V_{xy}, \quad (13)$$

and on application of linear symmetrization superoperator $S^z$ [21] which acts on operators $A^x$ and gives a possibility to remember the order of noncommuting operators $A^x$ in solution of master equation (6)

$$\tilde{P}_{xy}(t) = \left(\exp\left(-\sum_z n_z A^z t\right)\right)_{xy} \frac{n_y}{c} \equiv \left(S^x \exp\left(-\sum_z n_z A^z t\right)\right)_{xy} \frac{n_y}{c}. \quad (14)$$

Now operators $A^x$ commute under the symbol $S^z$ and averaging on occupation numbers is possible with the result

$$P_{xy}(t) = \left(S^x e^{-A^z t} \prod_{z \neq y} \left(1 + c(e^{-A^z t} - 1)\right)\right)_{xy}. \quad (15)$$

Expansion of last relation in powers of $c$ is simple, and it contains terms of the form

$$\left(S^x e^{-A^z t} \prod_{j=1}^m e^{-A^z t}\right)_{xy} = \left(\exp\left(-A^z t - \sum_{j=1}^m A^z t\right)\right)_{xy},$$

which are easily connected with solution of initial Eq. (6) for the case, when only $m + 1$ sites with positions $y, z_1, \ldots, z_m$ are filled by impurity spins. As a result

$$P_{00}(t) = \sum_{m=0}^{\infty} \frac{c^m}{m!} \sum_{x_1, \ldots, x_m} \sum_{k=0}^m (-1)^m k^m C_m^k P_{00}^{(k+1)}(0, z_1, \ldots, z_k; t), \quad (15)$$

$$P_{x0}(t) = c \sum_{m=0}^{\infty} \frac{c^m}{m!} \sum_{x_1, \ldots, x_m} \sum_{k=0}^m (-1)^m k^m C_m^k P_{10}^{(k+2)}(0, x, z_1, \ldots, z_k; t). \quad (16)$$
Here \( p_{00}^{(k+1)}(0, z_1, \ldots, z_k; t) \) is solution \( p_{00} \) of Eq. (1) for the problem of \( k+1 \) spins, placed at sites
\( r_0 = 0, z_1, \ldots, z_k \), \( p_{10}^{(k+2)}(0, x, z_1, \ldots, z_k; t) \) is solution \( p_{10} \) for the problem of \( k+2 \) spins, placed at \( r_0 = 0, r_1 = x, z_1, \ldots, z_k \), \( C_m^k \) is binomial coefficient. It is supposed here that for \( k=0 \) variables \( z_1, \ldots, z_k \) are absent, and for \( \mu=0 \) the variables and corresponding summations are absent. The prime in coordinate summation indicates that coincidences between spin positions are forbidden and summations are limited by common big cubic volume \( V \). Therefore symmetrisation with respect to all \( z_1, \ldots, z_m \) should be fulfilled before calculation of the limit \( V \to \infty \).

It can be shown that relations (15) and (16) are very general and they can be derived [19, 20, 22] from the fact, that any reasonable symmetric function of occupation numbers \( Z(n_r \{n_r\}) \) can be expanded into series of productions of occupation numbers using the identity
\( Z(n_r \{n_r\}) = \phi(0) + n_x (\phi(1) - \phi(0)) \).

Therefore
\[
Z(n_r \{n_r\}) = Z(0) + \sum_{m=1}^{\infty} \frac{1}{m!} \sum_{r_1, \ldots, r_m} \left( \prod_{j=1}^{m} n_{r_j} \right) \sum_{k=0}^{m} C_m^k (-1)^{m-k} Z^{(k)}(r_1, \ldots, r_k)
\tag{17}
\]

Averaging of this relation consists in substitution \( \prod_{j=1}^{m} n_{r_j} \to \left( \prod_{j=1}^{m} n_{r_j} \right)_c = c^m \) that (in application to the propagator \( P_{xy}(t) \)) produces relations (15) and (16).

Calculation of the FID can be carried out in similar way. Indeed, if we introduce a correlation function
\[
F_{xy}(t) = \left\langle n_x n_y \left\langle I_x^+ (t) I_y^- \right\rangle_0 \right\rangle_c : \left( \left\langle I_x^+ I_y^- \right\rangle_0 \right)_c^{1/2},
\tag{18}
\]
then we can receive for it relations, similar to (15) and (16), and than calculate
\[
F(t) = \sum_{x} F_{xy}(t).
\tag{19}
\]

The correlator \( F_{xy}(t) \) presents orthogonal circular polarization of the site \( x \), if initially the polarization existed at site \( y \) only.

Free induction decay \( \tilde{F}(t) \) can be calculated as well directly using Eq. (17).

If \( \nu x \gg 1 \), then \( p_{00}^{(k+1)}(0, z_1, \ldots, z_k; t) \) and \( p_{10}^{(k+2)}(0, x, z_1, \ldots, z_k; t) \) in concentration expansions (15) and (16) became smooth functions of summation variables \( z_i \) and sums can be replaced by integrals in exact analogy with derivation of Eq. (11). This calculation produces correct result for \( P_{xy}(t) \) if \( c \ll 1 \) only, because we omit forbidding on coincidence of integration variables, which is not negligible if concentration \( c \) is not small. Now, replacing the variables \( z_i \to (c/\Omega)^{-1/d} z_i \) we arrive to relations
\[
P_{00}(t) = f_0(\beta t), \quad P_{10}(t) = cf_1(\beta t, x / r_{aw}),
\tag{20}
\]
which were derived in [12] and named as theorem on singularity point.

All dependence on time \( t \), distance \( x \), and concentration \( c \) is written here explicitly, while unknown functions \( f_0(\tau) \) and \( f_1(\tau, \xi) \) do not contain dependence on \( c \). It will be recalled that \( \beta \sim c^{1/d} \) and \( r_{aw} \sim c^{-1/d} \). For example, in application to Foerster’s problem (9), (11) we have \( f_0(\tau) = \exp(-\tau^{d/x}) \).

Similar relations for magnetically diluted systems are of the form
\[
F_{00}(t) = \phi_0(D, t), \quad F_{x0}(t) = c\phi(x / r_{aw}).
\tag{21}
\]
Here \( D, c \sim c^{3/d} \).
It is clear from (20) and (21) that expansions (15) and (16) in terms of $c^m$ is in reality expansion in powers $(\beta t)^{m/d}$ and therefore $c$ is not a small parameter if $\beta t \geq 1$. Similarly, concentration expansion of FID is in reality expansion in powers $(D t)^{m/3}$.

Physical meaning of Eqs. (20) and (21) is simple. Natural scales for time and distance are presented by $\beta$ (or $D$) and average distance $r_{av}$. The singularity is direct consequence of the fact, that $P_{xy}$ and $F_{xy}$ are the polarization of lattice site and the processes take place if initial site $y$ is occupied by the impurity spin certainly, while any other point $x \neq y$ is occupied with probability $c$ only. Polarization on nearly placed spins becomes equal with increasing of the time, therefore [12]

$$\lim_{t \to 0} P_{x00}(t) \sim c P_{x00}(t), \quad \lim_{t \to 0} F_{x00}(t) \sim c F_{x00}(t).$$

(22)

It means, that both $P_{x00}(t)$ and $F_{x00}(t)$ are not smooth functions of $x$ in the point $x=0$. To recognize a peculiarity of this fact we should compare it with property of $P_{x0}(t \to \infty)$ for $c = 1$, when migration of polarization takes place on regular lattice. Then operator $\sim A$ is invariant relative lattice translations and solution of Eq. 6 can be received by applying lattice Fourier transformation. At large $t$ the solution go to propagator of diffusive equation and is of the form

$$P_{x0}(t \to \infty, c = 1) = \frac{\Omega}{(4\pi D t)^{d/2}} \exp\left(-\frac{x^2}{4\pi D t}\right),$$

having no singularity similar to (22). As a result we should expect, that if long time behavior of $P_{x0}(t \to \infty)$ is diffusion like, then

$$P_{x0}(t \to \infty) = \left(\frac{1}{c} \delta_{x0} + (1 - \delta_{x0})\right) \frac{\Omega}{(4\pi D t)^{d/2}} \exp\left(-\frac{x^2}{4\pi D t}\right),$$

(23)

and $c (4\pi D t)^{d/2}/\Omega \sim (\beta t)^{d/2}$, it means, that

$$D = \frac{K}{2d} \beta r_{av}^2,$$

(24)

where $\kappa$ is numerical coefficient, its modern values are obtained via numerical simulations, discussed below.

Calculation of terms of concentration expansions is more complex than calculation of moments, therefore very limited number of these terms is calculated up to now. Terms of order $c$ and $c^2$ for $P_{00}(t)$ in real dipole RWDS problem were calculated in [6, 19] (with taking into account possible influence of external static field). Term of first order for FID $F(t)$ for correlated distribution of spin positions was considered in [20]. Next term for uncorrelated positions was determined in [23]. Possible variation of coefficient before $S_i S_j$ in the Hamiltonian (4) was considered there.

5. Ways of rearrangement of concentration expansions

Observables, averaged over impurity distribution, contain less information then they contain before averaging. Standard formalism for excluding of unnecessary details of description of physical evolution is projection technique of Nakajima-Zwanzig (see for example [17] and [24]). As a rule it produces convolution master equation, if we start from local in time differential equation of motion.

5.1. Projection operator technique and memory kernels for RWDS

Simplest form of the resulting master equation can be received, if projection (super)operator $\tilde{E}$ (main function of which is calculation of average value (mathematical expectation) of observable $\langle \tilde{P}_{x0}(t) \rangle$ for
example) and initial condition $\tilde{P}_{xy}(t = 0)$ obey the relation $\tilde{E} \tilde{P}_{xy}(t = 0) = \tilde{P}_{xy}(t = 0)$. Therefore the projector can be chosen as

$$\left(\tilde{E} \chi\right)_s = \frac{n_x}{c} \langle \chi_s \rangle.$$

(25)

It is supposed here, that $n_x \chi_x = \chi_x$. In other way, using Dirac’s notations,

$$\langle x | \tilde{E} | \chi \rangle = \frac{n_x}{c} \langle \langle x | \chi \rangle \rangle = \frac{n_x}{c} \langle \chi_s \rangle, \quad \tilde{E} = \sum_s |x\rangle \frac{n_x}{c} E_n_s \langle x|.$$

(26)

Here operator $E$ produces simple averaging on occupation numbers: $Ef = \langle f \rangle$. It is evident, that $\tilde{E}$ and $\tilde{E} = 1 - \tilde{E}$ are projectors, that means $\left(\tilde{E}\right)^2 = \tilde{E}$, and $\left(\tilde{E}\right)^2 = \tilde{E}$. Therefore, starting from Eqs. (6) and treating them in standard manner using introduced projection operators we arrive to convolution master equation

$$\frac{\partial}{\partial t} P = -\int_0^t d\tau M(\tau) P(t - \tau), \quad P_{xy}(t = 0) = \delta_{xy},$$

(27)

where

$$M_{xy}(t) = \left\langle \tilde{A}_s \frac{n_x}{c} \right\rangle \delta(t - \eta) - \left\langle \left(\tilde{A} E e^{-\eta \tilde{H}} \tilde{E} \tilde{A}\right) \frac{n_x}{c} \right\rangle e^\eta.$$

(28)

Here $\eta \to +0$ in order to receive $\int_0^t d\tau \delta(t - \eta) = 1$ for $t > 0$. Remembering that the system has a conservation low $\sum_s P_{xy} = 1$, we can separate diagonal and nondiagonal parts:

$$M_{xy}(t) = D(t) \delta_{xy} - N_{xy}(t), \quad D(t) = \sum_s N_{sx}(t).$$

(29)

The memory matrix $M_{xy}(t)$ is invariant to lattice translations, therefore it can be diagonalized by lattice Fourier transformation that gives important simplification in solution of the equation.

Main strategy of application of master equations consists in approximation of the memory kernel with consequent calculation of propagator, solving obtained master equation. For example, if $c = 1$, then first step was fulfilled during derivation of Eq. (6), and at second step we arrive to diffusion process, while the memory at first step had no direct indication on this diffusion. Situation in strong disorder is opposite [6].

Indeed, relations (20) will be fulfilled for $c \ll 1$ if equation on diagonal part of the propagator is separated and has a form

$$\frac{\partial}{\partial t} P_{00} = -\int_0^t d\tau D(\tau) P_{00}(t - \tau).$$

(30)

It means that long time asymptotics $P_{00}(t \to \infty) \sim t^{-d/2}$ is one to one related with $D(t \to \infty)$. For example if $d=3$, then $D(t \to \infty) \sim t^{-3/2}$ as well. Similar relations can be derived for other dimensions using Laplace transformation to solve the Eq. (30) and Tauberian theorems. Therefore, if we want to receive the long time asymptotics of $P_{00}(t)$ solving the Eqs. (27) and (28), we should know long time asymptotics of $D(t)$ that is impossible, basing on first terms of concentration expansion. Nevertheless we will obtain reasonable diffusion asymptotics for $P_{x_0 y_0}(t)$ applying approximation for $N_{xy}(t)$, which is correct in initial region of time and if $\sum_x x^3 w_{x_0} < \infty$, where $w_{x_0} = \int_0^\infty dt N_{x_0}(t)$. By other words we have no possibility to calculate directly measurable $P_{00}(t)$ basing on master equation (27), (28) and concentration expansions.
We can introduce other kind of memory (geometrical, contrary to dynamical one in (27), (28)) if we will take into account, that
\[ P_{x y}(t) = \tilde{P}_{x y}(t) n_y = \tilde{P}_{x y}(t; n_y = 1) n_y. \]
Therefore if we introduce auxiliary propagator \( \tilde{K}_{x y}(t) = c \tilde{P}_{x y}(t; n_y = 1) \), then \( \tilde{K}_{x y}(t) = P_{x y}(t) \), and \( \tilde{K}_{x y}(t) \) obey the equation
\[
\frac{d}{dt} \tilde{K}_{x y} = -\left( \tilde{A}^x \tilde{K} \right)_{x y}, \quad \tilde{K}_{x y}(t = 0) = \delta_{x y}
\]
with much more simple initial condition, then in (6). Here \( \tilde{A}^x = \tilde{A}(n_y = 1) \). It is sufficient now to chose the projectors of Nakajima-Zwanzig as simple averaging \( E \) and \( 1 - E \). As a result the master equation for \( P_{x y}(t) \) receive a form
\[
\frac{\delta}{\delta t} P_{x y} = -\int_0^t d\tau \sum_x M_{x x}^x (\tau) P_{x y} (t - \tau),
\]
where
\[
M_{x x}^x (\tau) = \left( \tilde{A}_{x x}^x \right) \delta(t - \eta) - \left( \tilde{A}_{x x}^x E e^{-\tilde{E}_{x x}^x E} \tilde{A}_{x x}^x \right)_{x x}.
\]
Memory of this type gives [6] reasonable diffusive asymptotics (23), but the matrix \( M_{x x}^x (t) \) can not be diagonalized by Fourier transformation, and Eq. (32) has no solution in quadratures.

5.2. Similarities with the theory of continuum time random walks. Semi-phenomenology
The theory of Scher and Lax [16] belongs to so called continuum time random walks (CRTW) theories. It was among the first, directed on analytical treatment of RWDS problem. The theory was formulated in the form, which is equivalent to master equation (27) with memory kernel of dynamical type (29) for transition rates of the form \( \nu_{x y} = \nu_0 \exp(-|x - y|/\rho) \), and without exact correspondence to concentration expansions. The property (30) was not recognized at that time, and asymptotics \( P_{x y}(t \to \infty) \) was not of diffusion type, while increasing of \( \left< x^2 \right> = \sum_x x^2 P_{x 0}(t \to \infty) \sim t \) was diffusion like. In Ref. [6] the Scher-Lax theory was updated to produce correct first term of concentration expansion and qualitatively correct diffusion asymptotics (23) for dipole transport on the basis of application of geometrical memory (32), (33). This variant received a name “semi-phenomenological theory”. The memory kernel was presented as
\[
M_{x x}^x (t) = \delta_{x y} D_x^x(t) - N_{x y}^x(t), \quad D_x^x(t) = \sum_y N_{x y}^x(t),
\]
and explicit form was received in Laplace representation:
\[
N_{x y}^x (\lambda) = \left( 1 - \delta_{x y} \right) w_{x x}^x (\lambda) + \frac{\delta_{x y}}{c} w_{x y}^x (\lambda), \quad w_{x y}^x (\lambda) = cv_{x y} Q(\lambda + v_{x y}) / Q(\lambda).
\]
Here \( f(\lambda) = \int_0^\infty dt \exp(-\lambda t) f(t) \) for any function \( f(t) \), and \( Q(t) = Q_{\text{CM}}(t) \) (see (9) and (11)). Such definition of \( w_{x y}^x (\lambda) \) is specific for Scher-Lax theory, but application of geometrical memory produce correct terms of the propagator in the first order in concentration in difference from primary version [16].

Important consequence of this analysis consists in formulation of statement, that in calculation of long time asymptotics of \( P_{x y}(t) \) the condition \( n_y = 1 \) can be omitted, and then the result of Eq. (32) is of the form
\[
\begin{align*}
P_{x_0}(t \to \infty) &= \frac{\Omega}{(2\pi)^3} \int d^3k e^{i\mathbf{k} \cdot \mathbf{x}} \exp(-\overline{M}(\mathbf{k})t), \quad \overline{M}(\mathbf{k}) = \sum_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} \int_0^\infty dt M_{x_0}(t). \tag{34}
\end{align*}
\]

For small \(k\) we have
\[
\overline{M}(\mathbf{k}) = Dk^2 - \sigma k^3 + O(k^4).
\tag{35}
\]

Here \(D\) is diffusion coefficient, which depends on details of approximation, used for construction of \(M\), while second term is produced by dipole long range nature of primary transition rates \(v_{xx} \sim |\mathbf{z} - \mathbf{x}|^{-6}\), and it should not depend on the model treatment of \(M\). As a result, prognosis for dipole transport in impurity system was written as
\[
P_{00}(t) \approx P(t) = Q(t) + \frac{\xi(1 - Q(t))}{(\mu\beta(t + \tau))^{3/2}} \left(1 + \frac{\phi}{(\mu\beta(t + \tau))^{1/2}}\right)^{\frac{1}{2}}. \tag{36}
\]

Here coefficient \(\xi\) reflects the asymmetry of transition rates existing in real \({}^8\text{Li-}^6\text{Li}\) system (\(v_{0j} = \xi v_{j0}, \xi = 3\), that was omitted above), \(\mu\) is determined by the relation \(\mu\beta = 4\pi Dc^{2/3}\), coefficient \(\phi\) is produced by term \(\sigma k^1 > 0\) from (35) and it is model independent (as well as \(\sigma\)), and \(\tau\) produces coincidence of first terms of expansion of \(P(t)\) in powers of \((\beta t)^{1/2}\) with exact
\[
P_{00}(t) = 1 - \left(\frac{\beta t}{\xi + 1}\right)^{1/2} + O(\beta t).
\]

Relation (36) interpolates exact asymptotical behaviour of short and long times. It should be valid for other values of diffusion coefficient as well with new \(\mu\) and \(\tau\), and with the same \(\xi\) and \(\phi\).

Primary experimental studies [4, 25, 26] confirmed this prediction at \(\beta t \leq 5\), with taking into account 1) necessary orientation anisotropy of transition rates, 2) new values of diffusion tensor, obtained in numerical simulation [27-29, 31], and 3) influence of correlation of local fields on transition rates [25, 30]. Consequent studies [5, 31-33] revealed, that for \(c < 10\%\) and \(\beta t \leq 30\) more correct relation is of the form \(P_{00}(t) = P(t)(1 + \chi(t))\), where (for static external fields \(H_0 \approx 200\) G) variation \(\chi(t)\) is relatively small (-0.1 < \(\chi(t) < 0.7\)) and it increases the “reoscillation”, presented by term in (36), which is proportional to \(\phi\).

5.3. Connections with effective (or coherent) medium theories. Nuclear relaxation via paramagnetic impurities

This class of theories is considered as mean field ones. Application of these theories to our problems with random site positions meet both all difficulties, which were listed for theories similar to CTRW, and new obstacles, connected with absence of ways to find analytical solutions of equations, which are typical for equations of main approximation. Therefore we will consider here only relatively simple problem of nuclear relaxation via paramagnetic impurities, where nuclear spin polarization can diffuse over crystal lattice and it can decay due to interaction with electron paramagnetic impurities (traps), randomly distributed on crystal sites. Examples of experimental realization of the process can be found in Refs. [11, 17, 34-38]. Basic equations of evolution are of the form
\[
\frac{\partial}{\partial t} p(\mathbf{x}, t) = D \Delta p(\mathbf{x}, t) - \sum_z n_z v_{\alpha z} p(\mathbf{x}, t), \quad v_{\alpha z} = \frac{v_{d0}^6}{|\mathbf{z} - \mathbf{x}|^6} = \frac{C}{|\mathbf{z} - \mathbf{x}|^6}, \quad p(\mathbf{x}, t = 0) = 1. \tag{37}
\]

Here \(p(\mathbf{x}, t)\) is polarization of the nucleus, placed at the crystal site \(\mathbf{x}\), \(\Delta\) is Laplacian, \(D\) is spin-diffusion coefficient, and the angular dependence of \(v_{\alpha z}\) is neglected together with the difference in eigenvalues of the diffusion tensor. The observable nuclear polarization (normalized to \(p(t = 0) = 1\) is
Here $d$ is the space dimensionality, $V$ is the crystal volume, the symbol $|0\rangle$ presents a vector having components $\langle x |0\rangle = 1$ and the propagator $G_{xy}(t) = \langle x | G(t) | y \rangle$ obeys the Eq. (37), but for initial condition $G_{xy}(t=0) = \delta(x - y)$.

Eq. (37) presents example of random walks in a system with disordered traps.

Expansion (17) of the observable $\bar{p}(t)$ in concentration powers gives in first terms

$$
\bar{p}(t) = \langle 0 | G^{(0)}(t) | 0 \rangle + n \int d^d x \left( \langle 0 | G^{(1)}(x,t) | 0 \rangle - G^{(0)}(t) \right) + O(n^2) = 1 - M_0(t) + O(n^2),
$$

$$(39)
$$

$M_0(t) = n \int d^d x \left( \langle 0 | G^{(1)}(t) | 0 \rangle - 1 \right).$$

$$(40)$$

Preceding studies [11, 17, 34-36] used (for $d=3$) the substitution $\bar{p}(t) = 1 - M_0(t) + O(n^2) \to \exp(-M_0(t))$ without justification. Analysis of two- and one-dimensional problems was absent whereas experiments were started to study fractal objects [38]. Therefore the work [39] was directed on founding of natural way for rearrangement of the expansion (39) and improving and extension of the results of Ref. 34 on systems with spatial dimension $d \leq 3$. Our attention will be directed on the rearrangement of concentration expansion. Other important results can be found in [39].

To construct a theory of mean field class we represent the propagator $G(t)$ in the form

$$
G(t) = \exp\left( -A + B(t) t \right) - \exp(\bar{p}(t)),$$

$$(41)
$$

$U(t) = \sum_{z} n_z U_z, \quad U_z = \delta_{zy} V_{xz}, \quad B(t) = A t + M(t),$$

$$(42)
$$

where $A= -DA$ and the operator $M(t)$ is as-yet undefined. We can write it in the form

$$
M(t) = \sum_{z} M_z(t),$$

$$(43)
$$

similar to the form of "potential" $U$ in (42). It can be said that the operators $M_z(t)$ must adequately describe the effect of traps in the so-called effective medium that appears upon averaging over the configurations of traps. It is therefore natural to assume that, on average, the propagator $G(t)$ undergoes no changes if one of the sites of the effective medium is replaced by an actual one and if the result is thereupon averaged over the distribution of traps; that is,

$$
G(t) = \exp\left( -A + M(t) + M_z U_z t \right) \right) f_c$$

$$(44)
$$

Relations (41)-(44) form a closed set of nonlinear operator equations. The solution coincides with (39) in main order in $c$. According to [39], it practically coincides with $\bar{p}(t) = \exp(-M_0(t))$, if $M_0(t)$ $\sim 1$ and $c \ll 1$, and corrections are important for longer time.

The derivation of Eq. (44) illustrates a way of application of “coherent medium approximation” to our problems in time representation. Other way can consists in application of similar ideas to Laplace representation of the propagator $G(\lambda)$. We should expect, that

$$
G(\lambda) = \left( \lambda + A + U \right)^{-1} f_c, \quad M_z(\lambda) = \sum_{z} M_z(t),$$

$$(45)
$$

and then equation of self-consistency is

$$
G(\lambda) = \left( \lambda + A + M(t) + M_z U_z \right)^{-1} f_c.$$}

$$(46)
$$

Probably, solution of Eqs. (41)-(44) and time version of solutions of Eqs. (45) and (46) are close in the same region ($M_0(t) \sim 1$ and $c \ll 1$) for three dimensional systems. But they are very different for $d<3$, and Eqs. (41)-(44) look as more preferable.
In order to complete the picture we should notice, that (1) to clarify the influence of corrections to this solution in more detail we can calculate the next ($\sim \hbar^2$) term of the concentration expansion of $p(t)$ or $G(t)$, and (2) there exist physical [40] and mathematical [41, 42] studies producing the law $\ln\left(\frac{1}{\overline{p}(t \to \infty)}\right) \sim t^{d+2}$, which is expected [44] to be valid at $\overline{p}(t) \sim 10^{-12}$. The asymptotics can be received by methods of superfield theory [45], but up to a preexponential multiplier only, as well as in Refs. 40–42. Rather artificial mathematical constructions [41, 42] receive natural simple clarifying via steepest descent method in superfield theory [45]. No way to calculate the pre-exponential factor was found. As a result, these methods were insufficient up to now for study of RWDS without traps.

5.4. Combining of memory functions and cumulant expansions for calculation of FID

Initial equations of motion (1), (6) and (37) in RWDS problem can be considered mathematically as one particle Schrödinger equation for imagine time. Therefore conceptually the problem of calculation of FID in magnetically diluted systems is more complex, because it starts (according to Eqs. (7), (18) and (19)) from many-particle Heisenberg equation of motion

$$\frac{\partial}{\partial t} n_s L_s^*(t) = i [H_s, n_s L_s^*(t)] = iH_s n_s L_s^*(t).$$

(47)

Here last equality defines Liouville superoperator $H_s^*$ acting in a Hilbert space, spanned on usual operators of quantum mechanics [17, 24] with scalar product $\langle A | B \rangle = \langle A^\dagger B \rangle_0$, where $A$ and $B$ are usual operators. Projection superoperator $P$ can be chosen as

$$PQ = \sum n_s I_s^*(n_s I_s^* Q)_{0,0}^{-1},$$

(48)

where $Q$ is arbitrary quantum mechanical operator. Introducing $\overline{P} = 1 - P$ and treating Eq. (47) in standard manner we arrive to convolution master equation

$$\frac{\partial}{\partial t} F_{rq}(t) = -\int_0^t d\tau \sum M_{rq}(\tau) F_{rq}(t - \tau),$$

(49)

$$M_{rq}(t) = \frac{1}{n_s \langle I_s^* I_s^* \rangle_0} \left\langle \left\{ [I_s^* H_s^*], H_d \right\} \exp \left( i\overline{P}H_s^* \overline{P}t \right) [H_d, n_s I_s^*] \right\rangle_{0,0}.$$  

(50)

We will discuss later three dimensional systems only.

Let us start from a dense system when all $n_q = 1$. Main property of the FID here consists in existence of oscillation, approximately presented by a relation [46]

$$F(t) = \frac{b \sin at}{a \sinh bt},$$

where constants $a$ and $b$ are of order $\sqrt{M_2}$.

We will formulate a simple approximation, which reproduce this oscillating behavior of the FID basing on known moments $M_2$ and $M_4$, and than we will generalize it to magnetically diluted systems, using first two terms of the concentration expansion, calculated in Ref. 23. Main approximation of the analysis is founded on findings of Refs. 45 and 25, were NMR form function $g(\omega)$ for impurity $^7\text{Li}$ in $\text{LiF}$ was measured in frequency range, where $g(\omega) \approx 10^{-4}g(0)$, and the results were satisfactory described [45, 25, 30] without adjusting parameters by an extension of Anderson-Weiss-Kubo (AWK) theory, developed primary for explaining of “narrowing by motion”.

5.4.1. Dense systems. Memory kernel $M_{rq}(t)$ is most complex object in the theory. To calculate it for dense system we use next simple and natural approximation:
\[
M_{rr}(t) = m_{rr} \chi(t), \tag{51}
\]
where \( m_{rr} = M_{rr}(t=0) \) is easily calculable, and \( \chi(t) \) is yet unknown function at that \( \chi(t=0) = 1 \).

Free induction decay of impurity nucleus is defined by a Hamiltonian having no flip-flop terms in dipole interaction of the nucleus with surrounding host nuclei. Therefore, in order to calculate \( \chi(t) \) using experience of Refs. 45 and 25, we introduce auxiliary system which has modified interaction for spin \( I_0 \) only. Its Hamiltonian is
\[
H_d^n = \frac{1}{2} \sum_{r} \sum_{q \neq 0} b_{rq} \left( I_r^z I_q^z - \frac{1}{3} I_r I_q \right) + \frac{2}{3} \sum_r b_{rr} I_r^z I_0^z. \tag{52}
\]

Repeating the derivation (47)-(51) with Hamiltonian \( H_d^n \), we receive, that master equation for \( F(t) = F_{00}(t) = \langle I_0^z I_0^z(t) \rangle \) is decoupled from other \( F_{rq}(t) \), and we can write it as
\[
\frac{\partial}{\partial t} F(t) = -\int_0^t d\tau M_f(\tau) F(t-\tau), \tag{53a}
\]

\[
M_f(t) = m_f \chi_f(t), \tag{53b}
\]
where unknown \( \chi(t=0) = 1 \), and \( m_f \) is easily calculable coefficient (second moment for \( F(t) \)).

Difference between the functions \( \chi(t) \) and \( \chi_f(t) \) should be small, of order \( 1/z_e \), where \( z_e \) is effective number of neighbors, therefore next main approximation is
\[
\chi(t) = \chi_f(t). \tag{54}
\]

Methods for calculation of FID for impurity nuclei [45, 25, 30] can be applied for independent calculation of \( F(t) = F_{00}(t) \) here directly. They use two main approximations. The first one consists in hypothesis that we can neglect of influence of impurity spin on evolution of the local field that is similar to approximation (54). The second approximation considers the local field on impurity spin
\[
\omega_{00}(t) = \frac{2}{3} \sum_q b_{q0} I_q^z(t),
\]
(where time dependence is defined by Heisenberg evolution) as smooth classical normal stochastic process with correlator
\[
\langle \omega_{00} \omega_{00}(t) \rangle = \frac{4}{9} \sum_{qr} b_{q0} b_{r0} \langle I_q^z I_r^z(t) \rangle_0 = m_i \kappa(t), \tag{55}
\]
\[
m_i = \frac{4 I(I+1)}{9 \cdot 3} \sum_q b_{q0}^2 = \langle \omega_{00}^2 \rangle_0,
\]
that produces
\[
F_f(t) = \exp \left( -m_i \int_0^t d\tau (t-\tau) \kappa(\tau) \right). \tag{56}
\]

Last relation is main in the AWK-theory.

The time dependence of operator \( I_q^z(t) \) in (55) is Hamiltonian again, and corresponding ways to take in into account are described in [45, 25, 30]. The relation (56) can be considered as well as first term of cumulant expansion of the correlation function \( F_f(t) \) in powers of local frequency \( \omega_0(t) \) [21]:
\[
F_f(t) = \left( T \exp \left( \int_0^t d\tau \omega_0(\tau) \right) \right)_0 \rightarrow \exp \left( -\int_0^t d\tau (t-\tau) \langle \omega_0(\tau) \omega_0 \rangle_0 \right).
\]

As a result, if correlation function (56) is calculated, then we can
a) define the memory kernel \( M_f(\tau) \) solving the Eq. (53) with known (from (56)) \( F_f(t) \);

b) define from (53) the functions \( \chi(\tau) \) and \( \chi_f(\tau) = \chi(\tau) \), and
c) calculate $F_{rq}(t)$ using Eqs. (49)-(51).

If the aim consists in calculation of FID $F(t) = \sum_{q} F_{rq}(t)$, then at the step “c” we can solve more simple master equation

$$\frac{\partial}{\partial t} F(t) = -\int_{0}^{t} d\tau M(\tau) F(t-\tau),$$

(57a)

$$M(t) = \sum_{r} M_{rx}(t) = \sum_{r} m_{rx} \chi(t) = \frac{9}{4} M_{f}(t),$$

(57b)

which follows directly from Eq. (49), (50) and (51). The translation invariance of the crystal was taken into account as well.

Realization of this program was fulfilled for $M_{d}/M_{z}^{2} = 19/9$, that can be considered as reasonable universal approximation for any simple lattice. Three different forms for function $\kappa(t)$ were applied:

1) $\kappa(t) = \exp(-\frac{1}{2} M_{z} t^{2})$,

2) $\kappa(t) = 1/\cosh^{2}\left(-\frac{1}{2} (M_{z})^{1/2} M_{f} t^{2}\right)$,

3) $\kappa(t) = 1/(1 + \frac{1}{2} M_{z} t^{2})^{3/4}$.

Existence of oscillation of the FID was stable, and relative variations of FID due to application of these $\kappa(t)$ were reasonably small. Agreement of the results with famous measurement [46] of FID in CaF$_{2}$ is better, than solution for simple Gaussian memory function represented by Fig. 1.1 in [17] for NMR form function.

In conclusion of the section we see, that existence of oscillations of $F(t)$ and absence of oscillation in $F_{f}(t)$ is connected directly with coefficient 9/4 in (57), which reflects transfer of transversal polarization in Eqs. (49) and (50). From mathematical point of view we have the same effect, as in equation of motion for oscillator coordinate $x(t)$ with frequency $\Omega_{0}$ and friction coefficient $\mu$, written in integral form

$$\frac{\partial}{\partial t} x(t) = -\Omega_{0}^{2} \int_{0}^{t} d\tau \exp(-\mu \tau) x(t-\tau).$$

It is evident, that with increasing of $\Omega_{0}$ we have here transition from simple decay to oscillation decay at $\Omega_{0}=\mu/2$.

5.4.2. Magnetically diluted systems. In order to generalize the construction to magnetically diluted systems we should invent substitutions for relations (51), (53) and (55), because all second moments became infinite, as it was discussed in section 3. For the sake of brevity we will use Laplace representation, and according to section 4 we will apply concentration expansion. The relations (51) and (53) for memory kernels can be substituted by

$$M_{rs}(\lambda) = m_{rs}(\lambda) \sigma(\lambda), \quad M_{f}(\lambda) = m_{0}(\lambda) \sigma_{0}(\lambda),$$

(58)

where functions $m_{rs}(\lambda)$ and $m_{0}(\lambda)$ contain terms of order $\epsilon^{1}$ only and they produce the terms of the same order in concentration expansions of $F_{rq}(t)$ and $F_{f}(t)$.

Following (54) we will apply the approximation

$$\sigma(\lambda) = \sigma_{0}(\lambda).$$

(59)
For independent determining of auxiliary FID $F_i(t)$ we use the fact [47], that if instead of exact calculation (10) of Anderson FID we suppose, that in any random configuration the FID is Gaussian with exact second moment, then

$$F_d(t) = \left\{ \exp \left[ i \sum_q n_q I_q^i \right] \right\}_{t=0} \rightarrow$$

$$\rightarrow F_g(t) = \exp \left[ -\frac{1}{2} \sum_q \left( (I_q^i)^2 \right) n_q b_q^2 t^2 \right] \right\}_{t=0} = \exp (-0.80 D_d t).$$

It means that this approximation produces surprisingly exact result. Therefore, if local fields are directed along the $z$-axis and depend on time, we obtain qualitatively correct representation of the process applying AWK-theory in any fixed configuration of impurities with result of the form

$$F_i(t) = \exp \left\{ -2B^2 \int_0^t d\tau (t-\tau) \exp (-\alpha B\tau) \right\}. \quad (60)$$

where coefficients $B$ and $\alpha$ can be received from concentration expansions and the function $\kappa(\tau) = \exp (-\alpha B\tau)$ here has the same sense as $\kappa(\tau)$ in Eqs. (55) and (56). It is simplest function, which represents influence of flip-flop transitions in surrounding spins and is compatible with analytical structure of concentration expansion for $d=3$. Therefore choice of this simple exponential form corresponds to Gaussian variant of $\kappa(\tau)$ for dense systems. The radical in (60) represents static fluctuations of interactions due to random distribution of positions of spins. If $\alpha = 0$, then (60) coincides with exact Anderson’s solution for impurity nucleus in absence of flip-flop transitions for surrounding spins, that defines $B = 2D_d / 3$.

For calculation of the FID it is sufficient to solve the Eq. (57a) with memory kernel

$$M(t) = \frac{3}{2} M(t), \quad (61)$$

where $M(t)$ is solution of Eqs. (53a) and (60), in particular

$$M(t) = (\lambda + \frac{1}{2} M(t))^{-1}, \quad F_i(t) = (\lambda + \frac{1}{2} M(t))^{-1}, \quad (62)$$

and the coefficient 3/2 in (61) again reflects the relation between exact first terms of the concentration expansions for $F_i(t)$ and $F(t)$.

First two terms of concentration expansion for $F(t)$, calculated in Ref. 23 are of the form

$$F(t) = 1 - D_d t + \frac{5}{9} (D_d t)^2 + O((D_d t)^3), \quad (62)$$

that produces $\alpha = 1 / 2$ and, instead of (60), we have

$$F_i(t) = \exp \left\{ -2 \left( B t + 2 \exp (-Bt/2) - 2 \right)^{1/2} \right\}. \quad (60a)$$

Numerical analysis indicates, that in the time region where $Bt=2D_d t/3>3.2$ and $F_d(t)>1/30$ the solution $F(t)$ can be approximated by a relation

$$F_{\text{app}}(t) = \frac{2}{5} F_i(t) \left( 1 + \frac{3}{2} F_i(t) \right)^{5/6} - \frac{1}{12} F_i(t) (1 - F_i(t))^2, \quad (63)$$

and relative error does not exceed 1%. The error monotonically increases later with increasing of the time and it comes up to 10% for $Bt=5$, and 25% for $Bt=10$.

Calculation of the form function $g(\omega)$ can be fulfilled in terms of Fourier-Laplace transformation of $F_d(t)$. Indeed, if we introduce the functions

$$g_x(\omega) = \int_0^\infty dt \cos \omega t F_i(t), \quad g_y(\omega) = \int_0^\infty dt \sin \omega t F_i(t),$$

then
\begin{align}
g(\omega) = \frac{1}{2\pi} \left( F(\lambda = \varepsilon - i\omega) + F(\lambda = \varepsilon + i\omega) \right) = \frac{6}{\pi} \left( \frac{g(\omega)}{(a g(\omega) - 3)^2 + (b g(\omega))^2} \right). \tag{64}
\end{align}

Obtained FID (63) has no oscillations on the scale \(T_2 = 1/\Delta a\) and it is monotonically decreasing function of time, because it is a solution in continuum media approximation. But oscillations on the scale \(t_0 = 1/b_0\) can be significant in real crystals even for concentration \(c\) of order several percents. Here \(b_0\) is a constant of dipole interaction between nearest spins in the sample. To recover them it is sufficient to replace \(D a t\) in the solution by initial sum, wherefrom \(D a t\) was produced:

\begin{align}
D a t \to c \sum_{x} \left( 1 - \cos \frac{1}{2} b_0 t \right). \tag{65}
\end{align}

As a consequence of this substitution monotonic \(g(\omega > 0)\) can receive satellite maxima at \(\omega = b_0\). Both this properties are similar to relations between FIDs (10) and (12) and between corresponding NMR form-functions.

There exist no experimental results which can be compared directly with relations (63), (64) or with their modification for real crystals. Probably it is connected with difficulties in obtaining of homogeneous polarization for magnetically diluted systems. Best results were received in measuring of spin echo for nuclei \(^{29}\text{Si}\) in silicon in Ref. 10 and in studies of NMR form functions in Refs. 7-9. But these results are contradictory.

The FID presented on Fig.1 from [10] has decay rate \(D_{10} = 2\Delta a/3\) for \(t < 1/\Delta a\) instead of expected \(\Delta a\). The result indicates that the sample did not correspond to the model under consideration, because theoretical predictions for this time range are reliable. Probably the value \(D_{10}\) is a consequence of existence of inhomogeneous Knight shift. Estimations indicate that for conditions, corresponding to this measurement, scale of the shift is of order \(2\pi \cdot 3 \cdot 10^3\) Hz, while \(\Delta a = 2\pi \cdot 42\) Hz, and typical variation of the shift at interspin distance exceeds \(\Delta a\). In these conditions flip-flop term of the dipole Hamiltonian is not secular, and it should be omitted, producing decay rate \(2\Delta a/3\).

NMR form functions, received in Refs. 7-9, have line widths substantially larger than the theoretical expectation (contrary to [10]), that indicates on additional source of broadening and on impossibility of direct application of the Hamiltonian (4) to the samples. But authors of Ref. 9 found a way to separate additional line broadening. After this correction line width, corresponding to FID (63), is in better agreement with results [9] than line width for simple exponential Anderson FID (12).

6. Path integral representations for RWDS
Path integrals serve as common general language of modern theoretical physics. Path integral representations of propagators of RWDM did not produced up to now the results, which we can not receive by operator methods. Nevertheless this way is very useful for understanding of the place of the problem in physical theory as a hole, and to recognize its complexity. Discussion of the problem requires special technique, therefore we will make only some general remarks here.

Many different path integrals representations for RWDS are possible. We expect, that most interesting are those, where configurational averaging over random impurities positions is fulfilled analytically. Equations (6) and (13) produce general basis for this action in the way, similar to derivation of relation (9). Path integral representations of this type were introduced in Ref. 48. For the sake of brevity we will consider only simplified version of the theory, where Eq. 6 has reduced initial condition \(\hat{P}_y(t = 0) = \delta_{\alpha y}\) that is sufficient for studies of long time asymptotics.

“Quantum mechanical” type of corresponding reduced propagator in continuum media approximation is of the form
\[
P_{\gamma}(t) = \left\langle \exp\left( -\sum_{\gamma} n_{\gamma}(t) \right) \right\rangle_{\gamma} = \int Dp(\tau) Dq(\tau) \exp \left( i \int_{\gamma} p dq - L[p, q, t] \right),
\]
\[
L[p, q, t] = n \int d^d z \left( 1 - \exp \left( -\int_0^t d\tau A^\gamma(p(\tau), q(\tau)) \right) \right),
\]
\[
A^\gamma(p, q) = \nu \left( 1 - \exp\left( -p(z - q) \right) \right).
\]

Exact definition of the integral, which is very important for real calculations, can be found in [48].

The representation (66) is similar to, but more complex than path integrals in the famous polaron problems (PP) [49]. The main difference consists of multi-time action \( L \) in (66) instead of two-time action in PP, in strong singularity of \( \nu \) instead of the less singular kernel \( |z-q|^{-1} \) in PP, in additional path integral over all \( p(\tau) \) and in strong dependence of the infinite-fold integral (66) on the exact form of approximations by the integrals with finite multiplicity.

Representation of superfield type is of the form [48]
\[
P^\gamma(\lambda) = \left\langle \exp\left( \lambda + \sum_{\gamma} n_{\gamma} A^\gamma \right) \right\rangle_{\gamma} = \int D\Phi D\Phi^* \alpha_\gamma \alpha^*_\gamma \exp \left( -\lambda \Phi^* \Phi - L[\Phi^*, \Phi] \right),
\]
\[
L[\Phi^*, \Phi] = c \sum_{\gamma} \left( 1 - \exp\left( -\Phi^* A^\gamma \Phi \right) \right).
\]

Here superfield \( \Phi_\gamma \) has two components: \( \Phi_\gamma = (a_\gamma, a_\gamma^*), \) where \( a_\gamma \) is usual Bose field, while \( a_\gamma^* \) is anticommuting Fermi field, and the product \( \Phi^* \tilde{\Phi} = \sum_{\gamma} (a_\gamma^* \tilde{Q}_\gamma a_\gamma + \alpha_\gamma^* \tilde{Q}_\gamma \alpha_\gamma) \) for any operator \( \tilde{Q} \).

It is evident, that relation (67) defines strongly nonlinear superfield theory, which has strong divergences in continuum media approximation.

7. Numerical methods in RWDS

Main problem for numerical simulation of RWDS, and especially in determining of long time asymptotics, consists in description of infinite system in a finite computer. Effective solution was constructed in Refs. 27-29 and 31.

The method consists in substitution of infinite disordered media by a crystal with a large disordered elementary cell (supercell), containing \( N_d \sim 1000 \) impurity spins, and on checking of stability of the results with regard to variation of \( N_d \). It starts from Eq. (1) and \( N_d \) impurities randomly placed in sites of a supercell, having \( N = N_d / c = N_g^3 \) lattice sites, and edges \( R_{\beta} = N_g b_{\beta} \), where \( b_{\beta} \) form the basis of the matrix crystal (LiF for the system \(^6\)Li-\(^7\)Li). Then the supercell is continued periodically to cover all space. If \( N_d \to \infty \), then we go back to random media. It is very important that the initial condition remains of the correct form and that it is not continued periodically. The eigenvalues problem
\[
\sum_{j} A_{ij}(m) = \varepsilon_{\mu} \phi_j(m)
\]
has Bloch’s solution
\[
\phi_j(m) = \exp\left( ikr \right) \chi_j(k, \mu), \quad m = (k, \mu),
\]
where \( \chi_j(k, \mu) \) has periodical dependence on impurity position \( r \) and \( k \) belongs to Brillouin zone \( V_B \) formed by all \( k \) satisfying the condition \( k B_{\alpha} \leq \pi \). Therefore, we have a finite eigenproblem for \( \chi_j(k, \mu) \):
\[
\sum_{\gamma=0}^{N_d-1} B_{j\gamma}(k) \chi_j(k, \mu) = \varepsilon_{\mu}(k) \chi_j(k, \mu),
\]
\[
B_{j\gamma}(k) = \sum_{\alpha=0}^{N_d-1} \exp\left( -i k (r_j - R(n)) \right) A_j(r_j + R(n)), \quad R(n) = \sum_{\alpha=1}^{N_d} n_{\alpha} R_{\alpha}.
\]

Here \( A_j(r_j + R(n)) \) is \( A_j \) with substitution \( r \to r_j + R(n) \).

As a result the propagator in fixed configuration of impurities
\[ p_{\gamma_0}(t) = V_B^{-1} \int_{V_B} d^3k \exp \left( ik r_j \right) \left( \exp \left( -B(k)t \right) \right)_{\gamma_0}, \]

and averaged propagator
\[ P_{\gamma_0}(t) = V_B^{-1} \int_{V_B} d^3k \left( \exp \left( -B(k)t \right) \right)_{\gamma_0} \]
\[ P_{\gamma_0}(t) = V_B^{-1} \int_{V_B} d^3k \exp \left( ik x \right) \left( \exp \left( -B(k)t \right) \right)_{\gamma_0}^{(r=\infty)}, \]

where subscript \((r=\infty)\) indicates that averaging is fulfilled under condition that site \(x\) is occupied by an impurity spin having number 1 (the site 0 is always occupied by spin “0”).

Primarily [27-29] calculations were directed on determination of the diffusion tensor via calculation of smallest eigenvalue of the operator \(B\) solving the equation of motion with special initial condition and using modification of Adams algorithm for ordinary differential equations. After checking of stability of the results to values of \(N_d \geq 500\) full diagonalization of \(B\) and relation (71) were applied in [31]. The results are in satisfactory agreement with experimental studies of the model system \(^{6}\)Li-\(^{6}\)Li in LiF [5, 33].

8. Spin dynamics in magnetically diluted systems with low Zeeman and medium low dipole temperatures
Development of EPR repeated in many problems findings of NMR. Particularly two temperature NMR theory of Provotorov was successfully generalized to EPR that produced a possibility to study many new effects [50]. Standard two temperature theory was developed in high temperature approximation (HTA), when \(\omega_b / T_Z \ll 1\) and \(\omega_{loc} / T_d \ll 1\). Here \(\omega_b\) and \(\omega_{loc}\) are Zeeman and local frequencies, and \(T_Z\) and \(T_d\) are temperatures of Zeeman and dipole reservoirs. Conditions with \(\omega_b / T_Z \geq 1\) are not seldom in EPR, therefore the theory received corresponding extension in Refs. [51-54]. Local frequency in NMR has two different definitions – from heat capacity and from resonance line width [17]. In HTA both definitions produce close results for NMR and are related with second moment \(M_2 \sim \omega_{loc}^2\). In EPR the line width is scaled on \(D_\perp\) at any temperatures, while heat capacity in HTA is scaled on \(M_2\) again, and \(M_2 \sim D_\perp^2 / c\). Therefore we come to natural problem of extension of the theory to temperature region where dipole temperature is medium low, that is \(D_\perp / T_d \ll 1\), but \(M_2^{1/2} / T_d \geq 1\). This step was fulfilled in Refs. 55-57, where two temperature fluctuation-dissipation theorem was derived, Provotorov’s equations were generalized to arbitrary Zeeman and medium low dipole temperatures, dipole and Zeeman heat capacities and resonance form functions were constructed on the basis of concentration expansions and fluctuation shift was revealed. More detailed information can be found in original articles and the review [58]. Applications of these results to studies of EPR in magnetic fluids [1, 59] can be indicated among more modern investigations. These references contain generalization of the theory to samples where local fields are produced by elongated aggregates of nanoparticles instead of point dipoles and corresponding experimental studies.

9. Cluster expansions and spectral dynamics
Preceding review of magnetically diluted systems was directed in main on studies of spin evolution with simple well defined initial condition (FID) or with small external perturbation (resonance form function), when resonance line can be considered as homogeneous. If resonance perturbation is not small the dynamics became more complex [58]. Important example was investigated in [60], where dipole broadened EPR line, which as a norm should be homogeneous, revealed inhomogeneous properties. In particular, in reaction on strong resonance field burning of the hole in resonance line was observed. It is important that wing of the hole was nearly exponential contrary to standard expectation.
of Lorentzian line wings for homogeneous dipole lines. One of important conclusion of the work consisted in understanding of importance of close spin pairs with interspin distances much smaller than average distance between spins $r_{av}$. Special influence of similar spin pairs was studied as well in spin disordered systems \[61, 4\] among investigations \[62, 63, 4\] of other processes beyond the Eqs. 1. Later a theory of two-spin resonance in magnetically diluted systems was constructed, where influence of close pairs determined unusual dependence of the process rate on magnitude of RF field $Z_1$ \[64\]. Importance of influence of close pairs on lack of homogeneity of dipole line was supported as well by Ref. 47.

Regular construction for consideration of similar effects is cluster analysis (or cluster expansions) developed in Ref. 65 and 66.

First of all we should underline, that concentration expansions and cluster expansions are very different. Both methods use occupation numbers but have different aims. Destination of concentration expansions consists in expansion of observable in powers of impurity concentration $c$, while cluster analysis is directed on decomposition of the system itself in such constituents as spins of mass, and two-spin, three-spin and so on clusters. The decomposition admits then to separate processes with different time scale and spectra of different nature. Very important guiding line consists in elimination of divergences, existing in naive high temperature spin dynamics in continuum media approximation. First of them is dipole heat capacity and (proportional to it) second moment of usual resonance line form function.

By definition $k$-cluster is a group of $k$ spins in which every spin has more strong intragroup interaction than all its out-of-group interactions. As a measure of interaction between spins $i$ and $j$ we use the value of $b_{ij}$ from dipole Hamiltonian (4). With this definition a spin can belong to several clusters simultaneously, for example, to a 2-cluster, and to 3-cluster. In applications it is convenient to have a division in which each spin belongs to no more than one cluster. This is obtained if we fix the rank $k_{max}$ of the largest cluster, and then exclude from the $k$-clusters with $k < k_{max}$ all those clusters that also belong to clusters of higher rank $q (k < q < k_{max})$. It is natural to designate the clusters thus obtained as orthogonal.

It was found \[65\] that if from the entire system we separate out 2- and 3-particle clusters as carriers of the discrete spectrum, the remaining spins (the totality of which we call the mass) have finite dipole heat capacity and second moment, which are determined by interactions at average distance. Therefore, in analogy with standard NMR of dense spin systems, we can expect, that spectrum of the mass of spins can be considered as continuous. As a result spins of mass produce homogeneous broadening of cluster spectra. Transitions between states of the cluster take place due to interactions with other clusters and with mass spins, and they are slow. Numerical analysis \[65\] shows, that orthogonal 2- and 3-clusters contain 51(1)% and 11(1)% of all spins correspondingly if $k_{max}$=3, but 2-clusters define all Lorentz wings of the EPR line.

The regularization of the spin-spin interactions in the mass, consisting in finiteness of the second moment, makes it possible to hope that for the mass and for the system as a whole the fastest process will be the establishment of quasiequilibrium in the Zeeman and dipole reservoirs of the mass, for which the two-temperature description is thereby preserved, while the kinetics of the clusters should be constructed with allowance for the discreteness of their spectra. Spins of the mass are still subject to inhomogeneous broadening from the interaction with the spins of the clusters, and the two-temperature description for them will be valid only over times greater than the time of establishment of equilibrium in this inhomogeneously broadened spectrum.

As a whole, the cluster expansion gives a constructive alternative to spin packets theory of Portis \[67\], used in \[60\] for interpretation of the data. The spin packet is a global concept, characterizing the entire spin system as a whole, and its shape does not depend on the intensity of the field $\omega_A$. But the absorption of the external field occurs locally, and has an essential dependence on the real configuration of the spins that participate in each elementary act. Unlike the shape of a spin packet, the line-shape function describing the homogeneous broadening of these spins is not a self-averaging
quantity, and only directly observable quantities are subject to configurational averaging. In accordance with this, we shall examine the results of Ref. 60 starting from the ideas developed above.

Transitions between states of the cluster take place due to interactions with other clusters and with mass spins, and they are slow. As a result saturation at the EPR line wing induces transitions between states of the 2-clusters, and the line shape of the hole is defined by the interaction of the cluster with mass spins. If this interaction is estimated within the AWK model, then any mass configuration produces exponential wings of the hole \( g(\Delta) \sim \exp(-\Delta / \mu) \), where \( \Delta \) is detuning of the saturating field from the cluster transition frequency, and \( \mu \) is defined by magnitude of the fields produced by other spins on the cluster, and by the rate of their fluctuations. After configurational averaging the wings become \( g(\Delta) \sim \Delta^{-4} \), but averaged hole shape describes the transitions for small magnitude \( \omega_l \) of the saturating field only. In a general case the observable area of the hole \( \sigma \) should be averaged. For large \( \omega_l \) we have \( \sigma \sim \ln(t_p \omega_l^2 / \mu) \), where \( t_p \) is the duration of the saturating pulse. Logarithm is a slow function, therefore \( \langle \sigma \rangle_c \sim \ln(t_p \omega_l^2 / \langle \mu \rangle_c) \), which was observed in [60] and was interpreted as the exponential wing of Portis’s packets. The described theory produces a microscopic picture of the phenomenon and indicates that Portis’s packets have limited relation to the problem, because they are homogeneous by definition, i.e. \( g(\Delta) = \langle g(\Delta) \rangle_c \), while in more microscopic cluster theory this relation isn’t fulfilled.

10. Conclusion
Author believes that spin dynamics and magnetic resonance are most developed branches of nonequilibrium statistical physics. This position is based on such brilliant studies as investigation of phase transitions in nuclear magnetism [17] and inversion of evolution of spin systems [68]. Existence of ergodic theorem for realistic spin systems with nontrivial interspin interaction [69] is, probably, unique as well.

Preceding discussion indicates that spin dynamics of disordered systems exists now as developed section of the physics. It incorporates achievements of spin dynamics of dense systems and has its own problems, conceptions, methods and perspectives.

Modern state of understanding of the processes in disordered spin systems is satisfactory, because experimental studies in model spin system \(^7\text{Li}-^6\text{Li}\) and successful numerical simulation of the process gave enough information for construction relatively full theoretical description. Nevertheless new measurements will be useful for better understanding of the nature of primary transition rates and their dependence on interspin distance. Of course, analytical solutions for long time and intermediate asymptotics of the processes still very desirable.

We have much less progress in description of magnetically diluted systems. New experimental investigations are necessary. First of all we need in precise measurement of free induction decay in diluted nuclear systems, which can be compared with classical work [46], fulfilled for dense nuclear paramagnets.

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