Investigation of the External-Magnetic-Field Dependence of Polarization Transfer in a ^{8}Li–^{6}Li Disordered Nuclear System

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The process under study is random walk in disordered meduim. It is managed by magnetic dipole-dipole interactions only.

$g(^{19}F)/g(^{6}Li) \approx 6.5,\ g(^{19}F)/g(^{107}Ag) \approx 45$

Examples:

1) nuclei ^{6}Li in the single crystal ^{7}Li^{19}F,

2) nuclei ^{107}Ag in the single crystal ^{109}Ag^{19}F.

Experiment: ^{8}Li-^{6}Li in the single crystal ^{7}Li^{19}F,

$(g(^{8}Li) - g(^{6}Li))/g(^{6}Li) = 0.00563$

β-NMR.

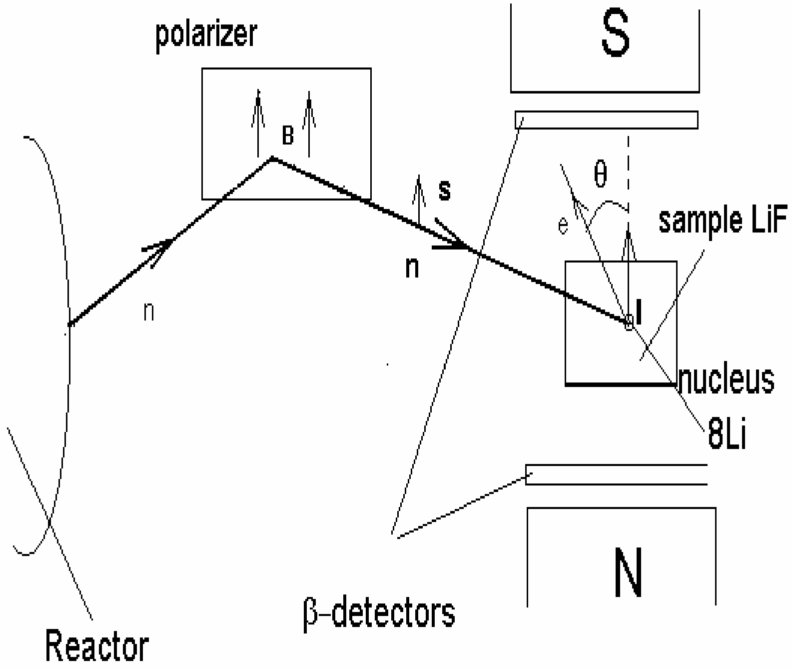
Polarized neutrons → polarized β-active nuclei (β-nuclei):

$^{7}Li(\vec{n},\gamma)^{8}\vec{Li} \leftrightarrow t = 0, \quad p(t=0) \approx 1 \sim 10^{6} \cdot \mu H_0/T.$

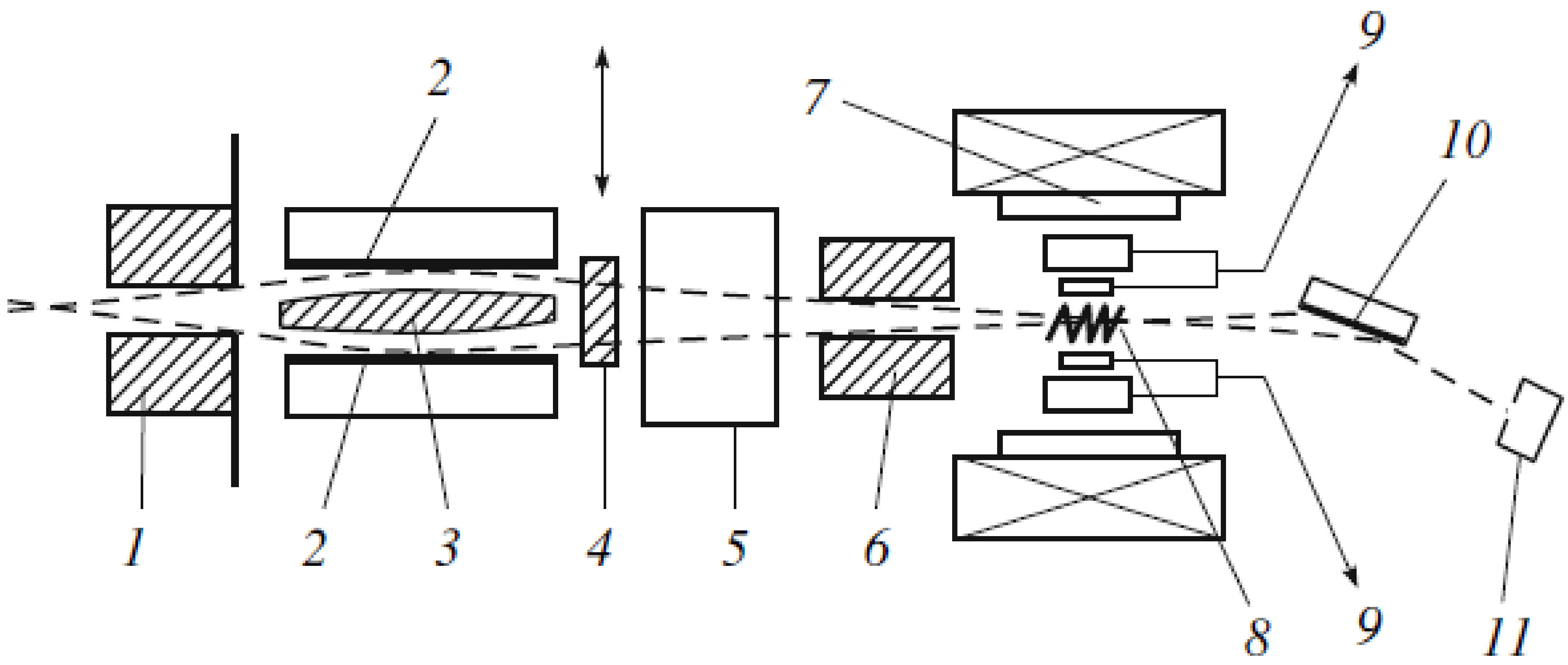
Angular distribution of β-radiation

$W(\vartheta,t) \propto 1 + a \cdot p(t) \cdot \cos\vartheta.$ Measurable value:

$$\varepsilon(t) = \frac{N(\vartheta=0,t) - N(\vartheta=\pi,t)}{N(\vartheta=0,t) + N(\vartheta=\pi,t)} \Leftrightarrow a \cdot p(t).$$



$|a| \approx 0.1$, $T_{1/2} = 0.84$ s.



Layout of the neutron beta-NMR spectrometer (ITEP, Moscow). It includes a unit for obtaining polarized neutrons (1, 3, 6 - collimators for neutrons, 2 - neutron-polarizer mirror, 4 - polarized-neutron-beam chopper, 5 - spin flipper for reorienting the polarization of the neutron beam incident on the sample, 10 - analyzer mirror, and 11 - neutron detector) and a unit for measuring the asymmetry of the emission of electrons from the decay of beta-nuclei (7 - electromagnet, 8 sample under study, and 9 scintillation telescope counters for detecting electrons from the decay process)

^8Li-^6Li system, characteristics: random distribution of nuclei ^6Li in the crystal, impurity concentration $c \leq 10\%$.

Naïve expectation for ^8Li depolarization kinetics is $p(t)=exp(-wt)$.

Foerster decay: if the polarization leaves the β-nuclei only, then

$$p(t)=\langle exp(-wt) \rangle = exp(-(\beta t)^{1/2}).$$

Actual evolution.

Very short time: $p(t)=1-cz(b_0 t)^2/2$, $t<T_2(^6Li)$, $b_0 t<<1$,

b_0 – dipole interaction between nearest Li nuclei,

z – effective coordination number.

Short time: $p(t)=1-cz\nu_0 t$, $1-p(t)<<1$,

ν_0 – polarization transfer rate between nearest Li nuclei,

Intermediate time: $p(t)=exp(-(\beta t)^{1/2}/2)$, $\beta t \leq 5$, . $\beta \approx 40c^2 \nu_0$,

$c^2 \nu_0$ – polarization transfer rate at average distance $r_c \sim c^{-1/3}$.

Long time asymptotics: $p(t) \sim 1/(\beta t)^{3/2}$, $\beta t \geq 30$, diffusion behavior.

Spin dynamics.

Single crystal LiF, $\frac{g(^8Li)-g(^6Li)}{g(^8Li)} = 0.00563.$

$H_0 = 200\text{G} \leftrightarrow$ flip-flop ^{8}Li-^{6}Li has the same speed as flip-flop ^{6}Li-^{6}Li, other cross-relaxation transitions are forbidden.

$$\frac{\partial p_{i0}}{\partial t} = -\sum_j (\nu_{ji} p_{i0} - \nu_{ij} p_{j0}), \qquad p_{i0}(t=0) = \delta_{i0},$$

$p_{i0}(t) = \langle I_i^z(t) \rangle$ – quantum statistical average value of the z-component (polarization) of the i-th nucleus, placed at $\mathbf{r}_i$ ($i = 0 \leftrightarrow$ ^{8}Li, and $i \neq 0 \leftrightarrow$ ^{6}Li). The rates of polarization transfer:

$$\nu_{ji} = \xi_j \nu_{ji}^0 \left(\frac{1-3\cos^2\theta_{ji}}{(r_{ij}/d)^3} \right)^2, \;\; \nu_{ji}^0 = \frac{\pi S(S+1)}{6} \left(\frac{g_i g_j \beta_n^2}{\hbar d^3} \right)^2 g_{ij}(\omega_{ij}),$$

$\xi_j = \xi\delta_{j0} + 1 - \delta_{j0}, \;\; \xi = I(I+1)/[S(S+1))] = 3,$

$I = I(^8Li) = 2,\ S = I(^6Li) = 1,\ g_i$ – g-factor, β_n – nuclear magneton,

θ_{ji} is the angle between $\mathbf{H}_0$ and $\mathbf{r}_{ji} = \mathbf{r}_j - \mathbf{r}_i$,

$d = 2.0136 \cdot \sqrt{2}$ Å is minimal Li-Li distance,

ω_{ij} is difference of the Larmor frequencies.

Basic approximation:

$g_{ij}(\omega) \approx \exp(-\omega^2/(2M))/(2\pi M)^{1/2}, \quad M = 2M_2,$

M_2 – second moment of the ^{8}Li NMR line. This approximation is correct qualitatively at least for $\omega^2 \leq 2M_2$. As a result ν_{ij}^0 have two values only:

$\nu_{ij}^0 = \nu_0 \leftrightarrow \omega_{ij} = 0 \leftrightarrow$ transfer between ^{6}Li spins, and

$\nu_{i0}^0 = \nu_1 \leftrightarrow \omega_{ij} = \Delta \leftrightarrow$ transfer between ^{8}Li and ^{6}Li.

The approximation will be improved later.

The way to $\frac{\partial p_{i0}}{\partial t} = -\sum_j(\nu_{ji}p_{i0} - \nu_{ij}p_{j0})$ is long and complex, but disorder is not important here and standard small parameters were utilized: $zc^2\nu_0 T_2 \ll 1$; $zc^2\nu_0\tau_c \ll 1$, T_2 is phase relaxation time for ^{8}Li, τ_c is flip-flop time for surrounding spins ^{7}Li and ^{19}F, and $zc^2\nu_0$ is estimation of the main process rate.

Nontrivial condition $z \gg 1$ should be fulfilled as well, if spins $I_j \geq 1$. Expected accuracy of the Eq.(1) is about 1%.

Roughly speaking the way is similar to derivation of hydrodynamics from quantum mechanics via Boltzmann equation.

At next step we should solve the equations

$$\frac{\partial p_{i0}}{\partial t} = -\sum\nolimits_j (\nu_{ji} p_{i0} - \nu_{ij} p_{j0}), \qquad p_{i0}(t=0) = \delta_{i0}, \tag{1}$$

and average the solution $p_{00}(t)$ over random distribution of spin positions $\{\mathbf{r}_j\}$.

The situation is similar to derivation of the hydrodynamics equations. We consider Eq.(1) as microdynamical one and we go to corresponding macrodynamics for polarization $P_{\mathbf{x}0}(t)$ of arbitrary lattice site $\mathbf{x}$, if the site 0 was polarized initially. Observable polarization of ^8Li will be described by $P_{00}(t) = \langle p_{00}(t) \rangle$.

The problem has no analytical solution now, but the propagator $P_{\mathbf{x}0}(t)$ can be expanded in terms of c^m. For $P_{00}(t)$ real parameter of the expansion is $(\beta t)^{1/2}$ instead of c. In order to calculate the term $\propto c^m$ the solutions of Eq.(1) for $k \leq m$ spins are necessary, while initial system (1) is written for infinite number of spins.

All existing analytical information was obtained by this method.

More quantitative results were received by numerical simulations.

Wy the problem is very interesting?

1) It has very simple form and challenge to an intellectual duel.

2) It has many applications in physics, chemistry and biology.

3) Being written in universal theoretical language - path integrals form - it produces very complex field and superfield theories. Any progress in solution of the problem should be transformed into progress in solutions of these universal problems. All story of theoretical physics of 20 century can be considered as search of ways to solve the problems of field theories.

The path integrals:

$$\mathsf{P}_{\mathbf{xy}}(t) = (e^{-At})_{\mathbf{xy}} = \int_{\mathbf{q}(0)=\mathbf{x}}^{\mathbf{q}(1)=\mathbf{y}} D\mathbf{p}(\tau) D\mathbf{q}(\tau) \exp(I[p,q]),$$

$$I[p,q] = i\int_{\mathbf{x}}^{\mathbf{y}} \mathbf{p}d\mathbf{q} + n\int d^3z\left(e^{-t\int_0^1 d\tau A^z(\mathbf{q}(\tau),\mathbf{p}(\tau))} - 1 \right),$$

$$A^z(\mathbf{q},\mathbf{p}) = \nu_{\mathbf{zq}}(1 - e^{-i\mathbf{p}(\mathbf{z}-\mathbf{q})}), \quad \nu_{\mathbf{zq}} \propto |\mathbf{z}-\mathbf{q}|^{-6}.$$

The representation is similar to, but more complex than path integrals in famous polaron problems, where the action (for partition function at temperature T) is

$$S[x] = \frac{1}{2}\int_0^{1/T} dt\left(\frac{d\mathbf{x}}{dt}\right)^2 - \alpha\int_0^{1/T} dt du \frac{\exp(-|t-u|)}{|\mathbf{x}(t) - \mathbf{x}(u)|}$$

Superfield path integral representations for $P_{\mathbf{xy}}(t) = \langle \tilde{P}_{\mathbf{xy}} \rangle_c$ exist as well.

Example of superfield representation

$$P_{\mathbf{xy}}(\lambda)=\int_0^{\infty} dt\exp(-\lambda t)P_{\mathbf{xy}}(t)=\left(\lambda+A\right)_{\mathbf{xy}}.$$

Bose-fields $a_{\mathbf{x}}$ and $a_{\mathbf{x}}^{+}$, Fermi-fields $\alpha_{\mathbf{x}}$ and $\alpha_{\mathbf{x}}^{+}$,

superfields $\phi_{\mathbf{x}}=\{a_{\mathbf{x}},\alpha_{\mathbf{x}}\}$ and $\phi_{\mathbf{x}}^{+}=\{a_{\mathbf{x}}^{+},\alpha_{\mathbf{x}}^{+}\}$,

$$\phi^{+}O\phi=\sum\nolimits_{xy}\left(a_{\mathbf{x}}^{+}O_{\mathbf{xy}}a_{\mathbf{y}}+\alpha_{\mathbf{x}}^{+}O_{\mathbf{xy}}\alpha_{\mathbf{y}}\right),$$

$$P_{\mathbf{xy}}(\lambda)=\int\delta a\delta a^{+}\delta\alpha\delta\alpha^{+}\alpha_{\mathbf{x}}\alpha_{\mathbf{y}}^{+}\exp\left(-I(\phi^{+},\phi)\right),$$

$$I(\phi^{+},\phi)=\lambda\phi^{+}\phi+c\sum\nolimits_{\mathbf{z}}\left(1-\exp(-\phi^{+}A^{\mathbf{z}}\phi)\right),$$

$$A_{\mathbf{xy}}^{\mathbf{z}}=\nu_{\mathbf{xy}}\delta_{\mathbf{xz}}-\nu_{\mathbf{xz}}\delta_{\mathbf{zy}},\quad \nu_{\mathbf{xy}}\sim\left|\mathbf{x}-\mathbf{y}\right|^{-6}.$$

Divergencies are seen clearly, but the theory exists as expansion in powers of c^{m} at least.

These representations demonstrate the relation of the RWDM to general problems of the modern field theory, but they are too complex, and real calculations now are based on concentration expansion (real parameter is $\left(\beta t\right)^{1/2}\sim ct^{1/2}$) for $\beta t\lesssim 1$, and numerical simulation for $\beta t\gtrsim 1$.

Main problem of numerical simulation -

infinite disordered sample in finite computer program.

We use infinite **crystal** with large **disordered unite cell,** containing $100<N_d<4000$ spins of ^6Li. As a result we can apply Bloch's theorem and receive Bloch's eigenvalues and eigenfunctions for matrix with dimension $N_d \bullet N_d$ and than we can calculate observable values, applying integration over Brillouin's zone.

Results are stable within 2% for N_d>400 spins.

Main problem in experiment.

$\beta \leq 10\ s^{-1}$, and measurements at $t \geq 3$ s are required, while $T_{1/2}$=0.84 s. ⇔ small statistics – long measurements.

Related problem: Concentration c=10% (corresponding to $\beta \approx 10\ s^{-1}$) is not small enough to neglect the correlations of local fields – dependence of ν_{ij}^0 on $\mathbf{r}_{ij}$ is important =

= **correlation of local fields on impurity spins**.

$$\nu_{ji} = \xi_j \nu_{ji}^0 \left(\frac{1 - 3\cos^2\theta_{ji}}{(r_j/d)^3} \right)^2, \quad \nu_{ji}^0 = \frac{\pi S(S+1)}{6} \left(\frac{g_i g_j \beta_n^2}{\hbar d^3} \right)^2 g_{ij}(\omega_{ij}),$$

$$g_{\mathbf{rq}}(\Delta) = \int_{-\infty}^{\infty} \frac{dt}{2\pi} e^{i\Delta t} \langle I_{\mathbf{r}}^{+}(t) I_{\mathbf{q}}^{-}(t) I_{\mathbf{q}}^{+} I_{\mathbf{r}}^{-} \rangle_0 / \langle I_{\mathbf{r}}^{+} I_{\mathbf{q}}^{-} I_{\mathbf{q}}^{+} I_{\mathbf{r}}^{-} \rangle_0$$

$$\langle \bullet \bullet \bullet \rangle_0 = Tr(\bullet \bullet \bullet)/Tr(1)$$

$$H = H_0 + H_I + H_{IF} + H_{IL}, \qquad H_0 = H_F + H_L + H_{FL}$$

$$H_A = \frac{1}{4} \sum_{\mathbf{xq}} b_{\mathbf{xq}}^{A} (3A_{\mathbf{x}}^{z} A_{\mathbf{q}}^{z} - \mathbf{A}_{\mathbf{x}} \mathbf{A}_{\mathbf{q}}), \qquad H_{AB} = \sum_{\mathbf{xq}} b_{\mathbf{xq}}^{AB} A_{\mathbf{x}}^{z} B_{\mathbf{q}}^{z}$$

$$b_{\mathbf{xq}}^{AB} = \frac{g_A g_B \beta_n^2}{\hbar |\mathbf{x}-\mathbf{q}|^3} (1 - 3\cos^2 \vartheta_{\mathbf{xq}}).$$

$$\langle I_{\mathbf{r}}^{+}(t) I_{\mathbf{q}}^{-}(t) I_{\mathbf{q}}^{+} I_{\mathbf{r}}^{-} \rangle_0 / \langle I_{\mathbf{r}}^{+} I_{\mathbf{q}}^{-} I_{\mathbf{q}}^{+} I_{\mathbf{r}}^{-} \rangle_0 \Rightarrow \left\langle \exp\left(i \int_0^t d\tau (\omega_{\mathbf{r}}^{l}(\tau) - \omega_{\mathbf{q}}^{l}(\tau)) \right) \right\rangle_0 \Rightarrow$$

$$\Rightarrow \exp\left(-\frac{1}{2} \left\langle \left(\int_0^t d\tau (\omega_{\mathbf{r}}^{l}(\tau) - \omega_{\mathbf{q}}^{l}(\tau)) \right)^2 \right\rangle_0 \right) =$$

AWK-theory

$$= \exp\left[-2 \int_0^t d\tau \left(t - \tau \right) \left(\Lambda_{\mathbf{rr}}(\tau) - \Lambda_{\mathbf{rq}}(\tau) \right) \right]$$

$$\Lambda_{\mathbf{rq}}(\tau) = \langle \omega_{\mathbf{r}}^{l}(\tau) \omega_{\mathbf{q}}^{l} \rangle_0 = \sum_{A=F,L} \sum_{\mathbf{xy}} b_{\mathbf{xr}}^{AI} b_{\mathbf{qy}}^{AI} \langle A_{\mathbf{x}}^{z}(\tau) A_{\mathbf{y}}^{z} \rangle_0$$

$\langle A_{\mathbf{x}}^{z}(\tau) A_{\mathbf{y}}^{z} \rangle_0$ - spin-diffusion propagator for ordered spins

$$\langle I_{\mathbf{r}}^{+}(t) I_{\mathbf{q}}^{-}(t) I_{\mathbf{q}}^{+} I_{\mathbf{r}}^{-} \rangle_0 / \langle I_{\mathbf{r}}^{+} I_{\mathbf{q}}^{-} I_{\mathbf{q}}^{+} I_{\mathbf{r}}^{-} \rangle_0 =$$

$$= \exp\left[-2 \int_0^t d\tau \, (t-\tau) \left(\Lambda_{\mathbf{rr}}(\tau) - \Lambda_{\mathbf{rq}}(\tau) \right) \right]$$

Static correlation: $\Lambda_{\mathbf{rq}}(t=0) \neq 0$

The effect exists in absence of flip-flop motion of surrounding spins and is near 50% for nearest neighbors.

Dynamical correlation: $|\Lambda_{\mathbf{rq}}(t)|$ starts to increase with time, then decays, that increases total influence of correlations of local fields on cross-relaxation form-functions.

As a result the form function $g_{\mathbf{r}0}(\omega=0)$ has deviations about 20% even for **r** at 4-th coordination sphere. Deviation for form function at $\omega > \left\langle \left(\omega_{loc}\right)^2 \right\rangle^{1/2}$ can be much more significant.

Spin diffusion on ordered lattice

$$\frac{d}{dt_{eff}} G_{\mathbf{xy}}\left(t_{eff}\right) = -\sum_{\mathbf{r}} w_{\mathbf{rx}}\left(G_{\mathbf{xy}} - G_{\mathbf{ry}}\right), \quad G_{\mathbf{xy}}(0) = \delta_{\mathbf{xy}}$$

$$G_{\mathbf{xy}}^{A}\left(t_{eff}(t)\right) = \left\langle A_{\mathbf{x}}^{z}(t) A_{\mathbf{y}}^{z} \right\rangle_{0} / \left\langle \left(A_{\mathbf{y}}^{z}\right)^{2} \right\rangle_{0}$$

$$t_{eff}^{A}(t) = \int_0^t \frac{d\tau}{T_{2A}}(t-\tau) g_{cA}(\tau), \qquad T_{2A} = \int_0^\infty d\tau g_{cA}(\tau).$$

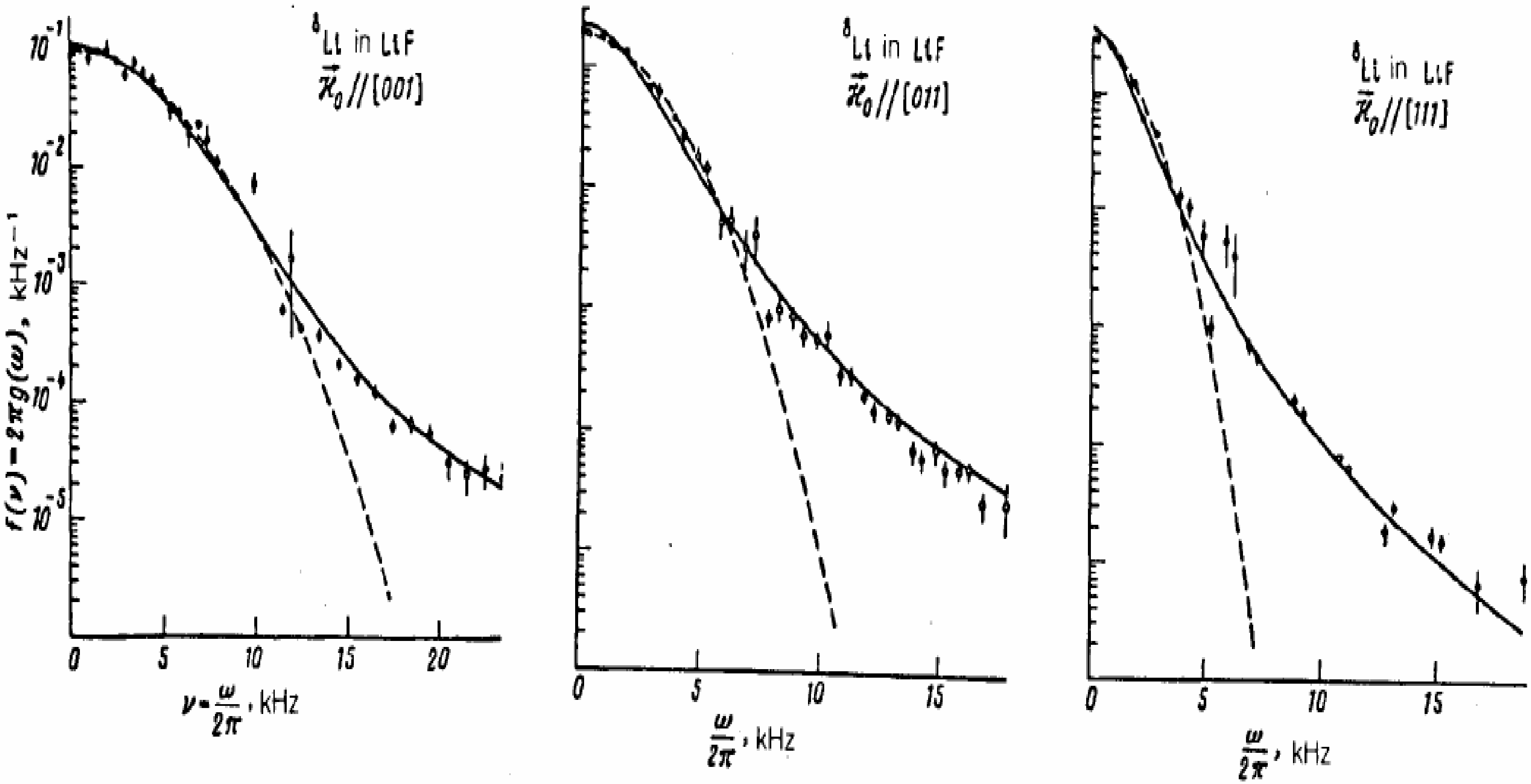
$$g_{cA}(t) = \left(\left\langle A_{\mathbf{r}}^{+}(t) A_{\mathbf{r}}^{-} \right\rangle_{0}\right)^{2}$$

Why this way is correct?

It produce NMR-form-function in satisfactory agreement with experimental data in range g(ω) ≥10^{-5}g(0)

$$g(\omega) = \int_{-\infty}^{\infty} dt e^{-i\omega t} \left\langle \exp\left(\int_0^t d\tau \omega_{\mathbf{r}}^{l}(\tau)\right) \right\rangle =$$

$$= \int_{-\infty}^{\infty} dt e^{-i\omega t} \exp\left(\int_0^t d\tau \Lambda_{\mathbf{rr}}(\tau)\right).$$



NMR form-function: theory vs experiment.
Gulko et al. JETP Lett. 1993, PPN 1996

Effect of correlation of local fields on cross relaxation form-function $g_{ij}(\Delta=0)$.

$g_G = g_G(\Delta = 0) = \left(2\pi \bullet 2M_2\right)^{1/2}$ – Gaussian approximation,

M_2 - second moment of NMR line.

r_{ij}/(2.0136 Å)	$1\text{-}3\cos^2\theta_{ij}$	$g_{ij}(\Delta=0)/g_G$	r_{ij}/(2.0136 Å)	$1\text{-}3\cos^2\theta_{ij}$	$g_{ij}(\Delta=0)/g_G$
1.414	-1	1.119	3.742	-0.1429	1.082
1.414	1	1.620	3.742	0.7143	1.139
2	0	1.055	3.742	1	1.180
2.449	1/3	1.081	3.742	1.571	1.135
2.449	1	1.300	4	0	1.092
2.449	-5/3	1.140	4.243	1/9	1.099
2.828	-1	1.081	4.243	-1	1.100
2.828	1	1.247	4.243	1	1.163
3.162	-3/5	1.064	4.243	7/9	1.140
3.162	3/5	1.130	∞		1.122
3.464	2/3	1.136			
3.464	-1/5	1.204			

Described conception of spin dynamics is developed by our group for description of spin delocalization in disordered systems.

It is extended now to satisfactory description of correlation functions for many problems both for ordered and magnetically diluted paramagnetic spin systems.

The theory had produced the interpolating formula (by joining short and long time asymptotic forms)

$$P_{00}(t) = F(t) = \exp(-\sqrt{\beta_1 t}) + \xi \frac{1-\exp(-\sqrt{\beta_1 t})}{(\mu\beta(t+\tau))^{3/2}} \left(1 + \frac{\varphi}{\sqrt{\mu\beta(t+\tau)}}\right),$$

$$\xi = \xi_0 = I(I+1)/[S(S+1)] = 3, \quad \mu\beta = 4\pi\,(c/\Omega_c)^{2/3}\left(\prod_{\alpha=1}^{3} D_\alpha\right)^{1/3},$$

$$\beta = (256/243)\pi^3 c^2 \nu_0 r_0^6/\Omega_c^2, \;\; \beta_1 = \beta\cdot\nu_1/\nu_0, \;\; c \ll 1.$$

$$\varphi = 2.09, \;\; \mu\beta\tau = 5.11, \;\; \mu = 0.71$$

$P_{00}(t)$ holds to within $(\beta t)^{1/2}$ at small βt, and it holds to within $(\beta t)^{-2}$ at large βt if exact diffusion tensor D_α is used. In reality D_α are taken from numeric simulation.

There is no fitting parameters here.

D_α are main values of the diffusion tensor.

The relation for $P_{00}(t)$ does not include correlation of local field.

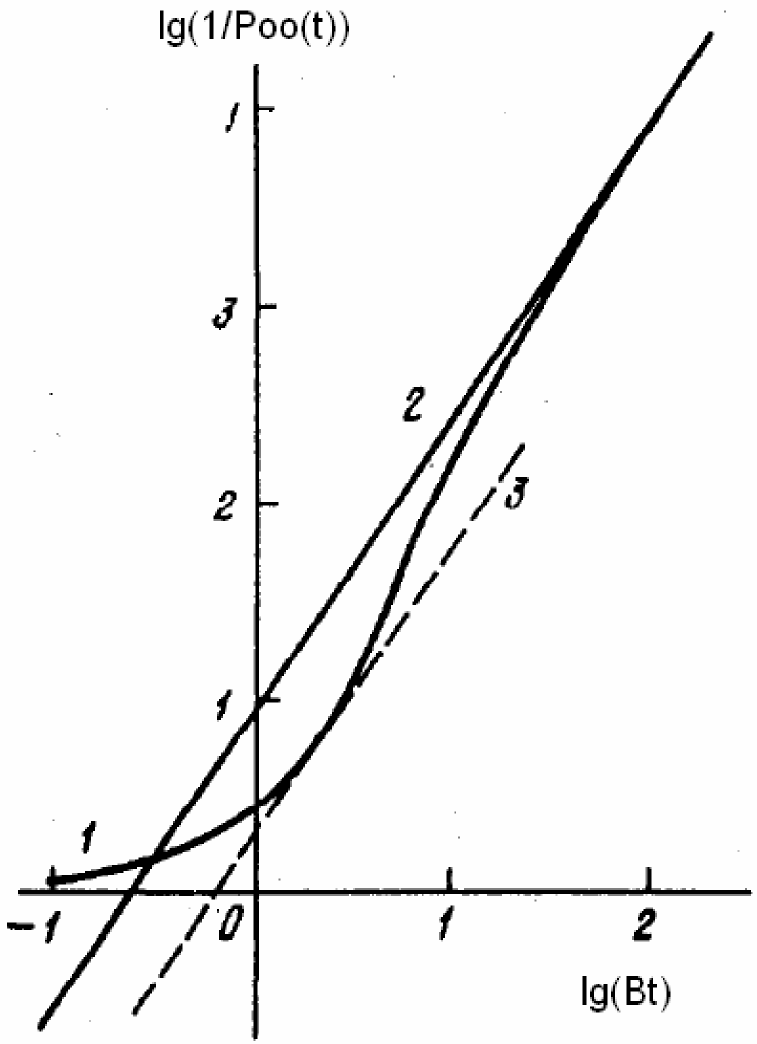
Preasymptotic effect - reoscillation. Illustration for the case of ordered system (simple cubic lattice, exact solution).

1 – $P_{00}(t)$.

2 - diffusion asymptote.

3 - false asymptote.

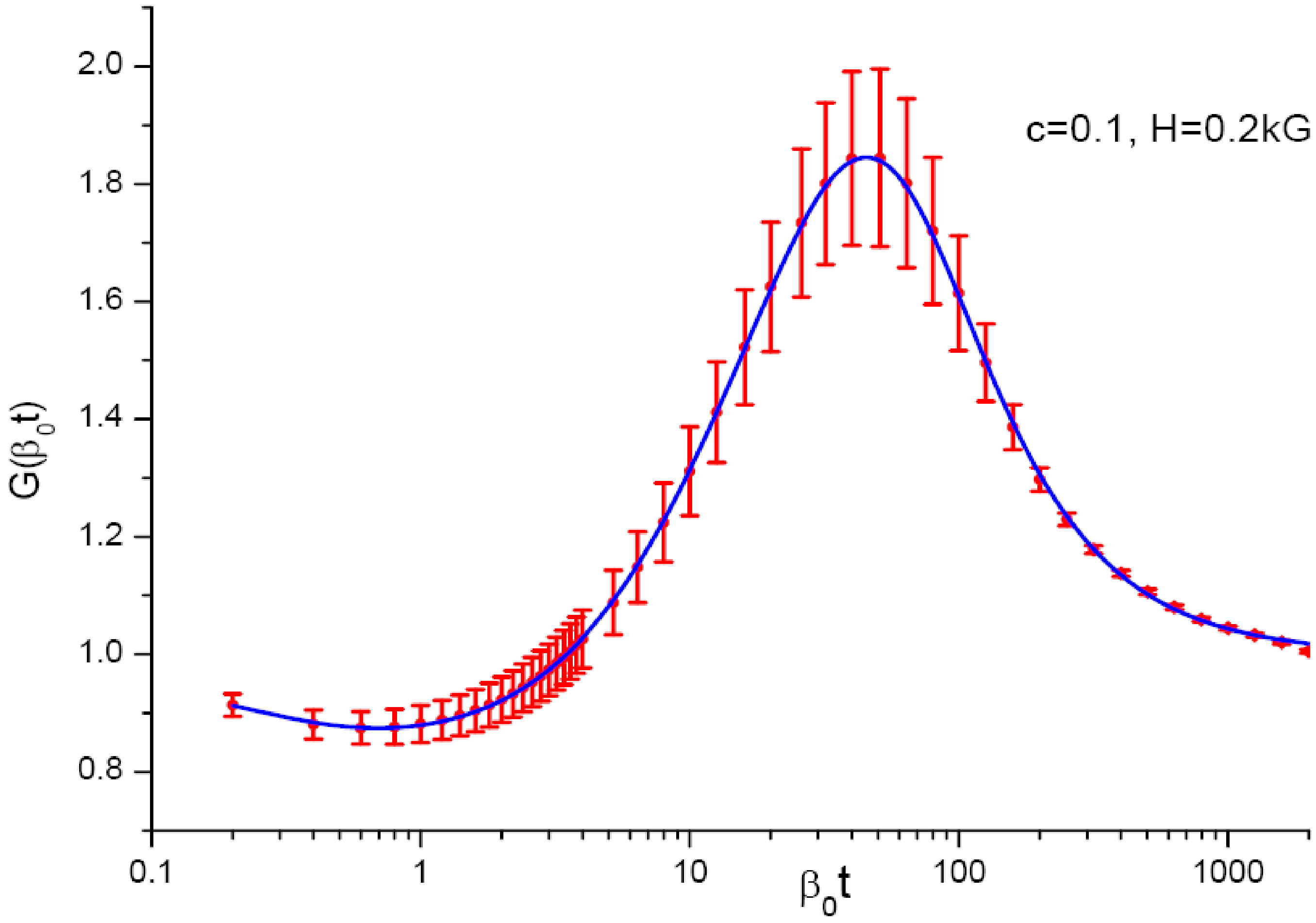
The reoscillation was unknown to the community both for ordered and disordered systems before our studies.

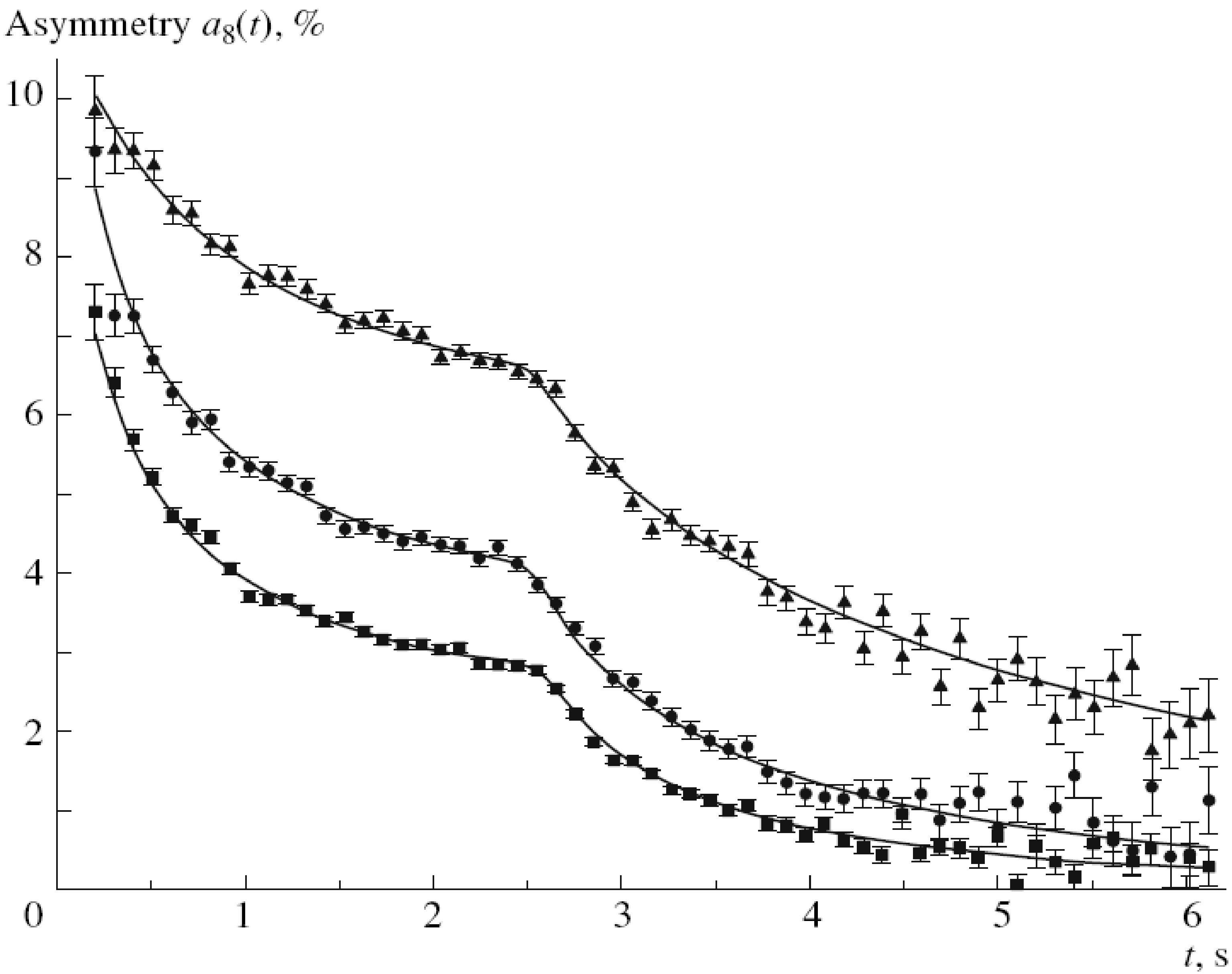


In order to incorporate results of numeric analysis we use

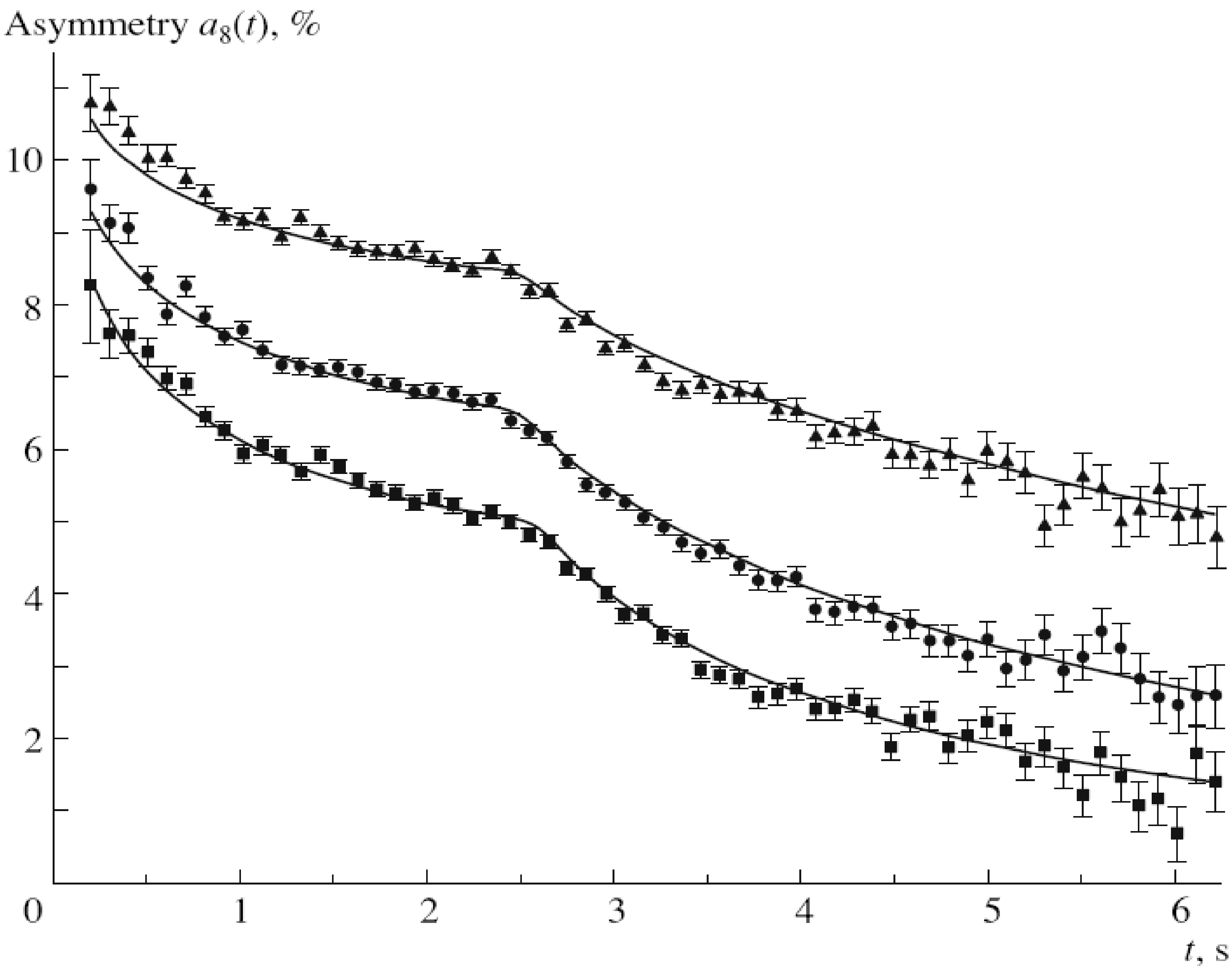
$P_{00}(t)=F(t)G(t)$,

where $F(t)$ is given by preceding formula, and correction function $G(t)$ is

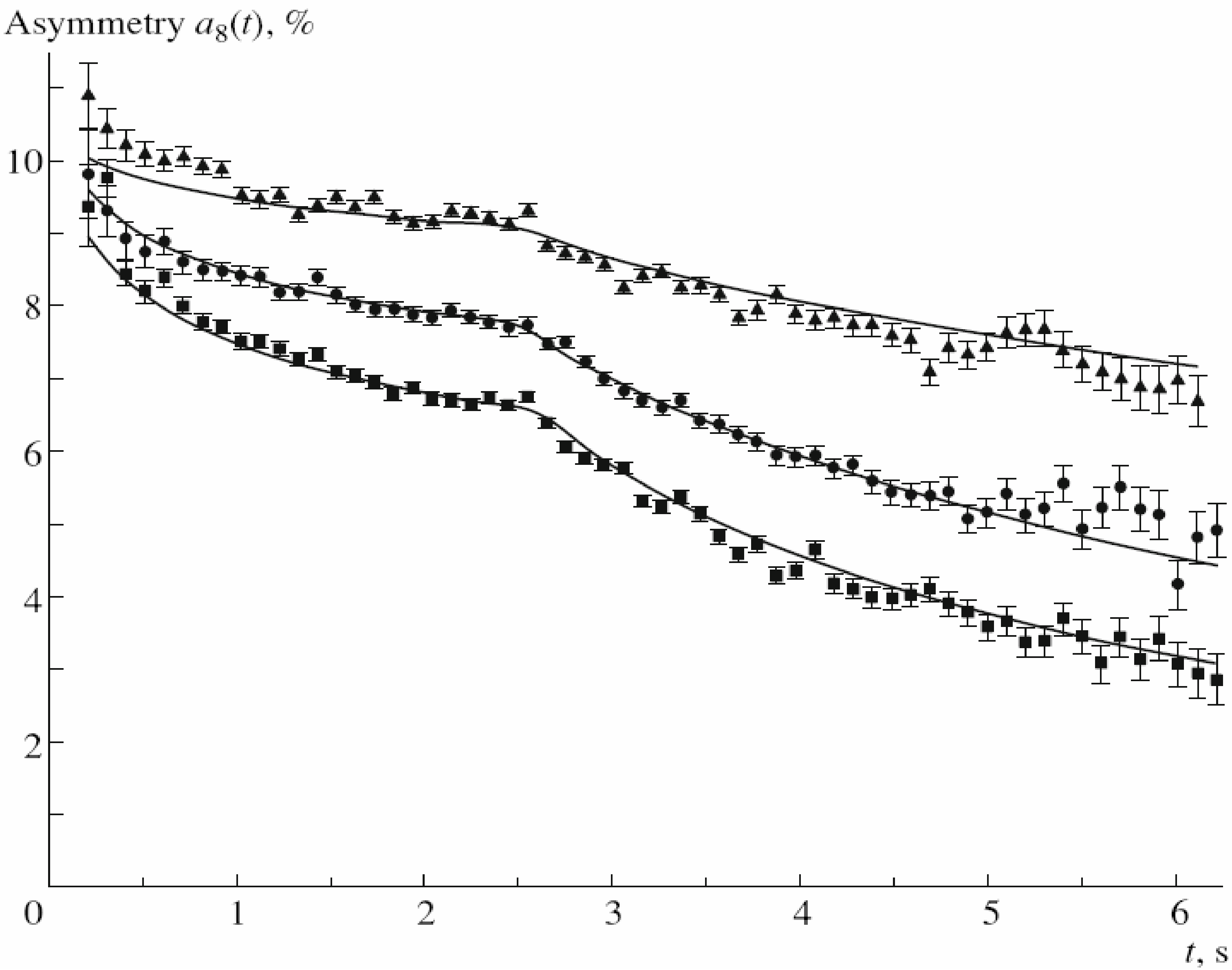




Concentration of ^{6}Li is c = 0.1006(4). Dependences for fields of strength 200, 691, and 1200 G (from bottom to top). Corresponding χ^2/n = 99/64, 96/64, and 92/64; the fitted parameter values are c = 10.10(4)% and a_0 = 10.62(6)%, c = 9.97(6)% and a_0 = 11.56(8)%, and c = 10.09(6)% and a_0 = 10.82(5)%.



Concentration of ^{6}Li is c = 0.0530(2). Dependences for fields of strength 200, 691, and 1200 G (from bottom to top). Corresponding χ^2/n = 74/64, 69/64, and 95/64; the fitted parameter values are c=5.32(2)% and a0 = 10.05(5)%, c = 5.32(2)% and a0 = 10.43(4)%, and c = 5.33(2)% and a0=10.86(3)%.



Concentration of ^{6}Li is c = 0.0330(3). Dependences for fields of strength 200, 691, and 1200 G (from bottom to top). Corresponding χ^2/n = 125/64, 55/64, and 168/64; the fitted parameter values are c = 3.43(3)% and a^0 = 9.86(4)%, c = 3.25(3)% and a^0 = 10.33(4)%, and c = 3.40(3)% and a^0 = 10.29(3)%

Conclusions

1. Kinetics of delocalization of nuclear polarization in disordered spin subsystem is studied in experiment and theory.

2. Existence of the spin diffusion in disordered spin subsystem is established.

3. Full numerical-analytical description of the kinetics is constructed.

4. Pronounced preasymptotical effects are revealed.

5. Field dependence of the kinetics is measured.

6. Field dependence of the kinetics is described taking into account both static and dynamic correlations of local field.

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Thank you for the attention!