NEUTRON PHYSICS INVESTIGATIONS OF FUNDAMENTAL PROCESSES

OF STATISTICAL PHYSICS BY

BETA-NMR AND MULTIPLE SMALL ANGLE SCATTERING

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β-NMR is magnetic resonance and relaxation of polarized beta-active nuclei.

It was invented by F.L.Shapiro in 1958 just after discovery of parity violation and first measurements were published by D. Connor (1958, USA) and Yu.G.Abov, O.N.Ermakov, A.D.Gulko at al. (1962, ITEP, USSR).

Angular distribution of β-radiation $w(\vartheta) \square 1 + ap(t)\cos\vartheta$

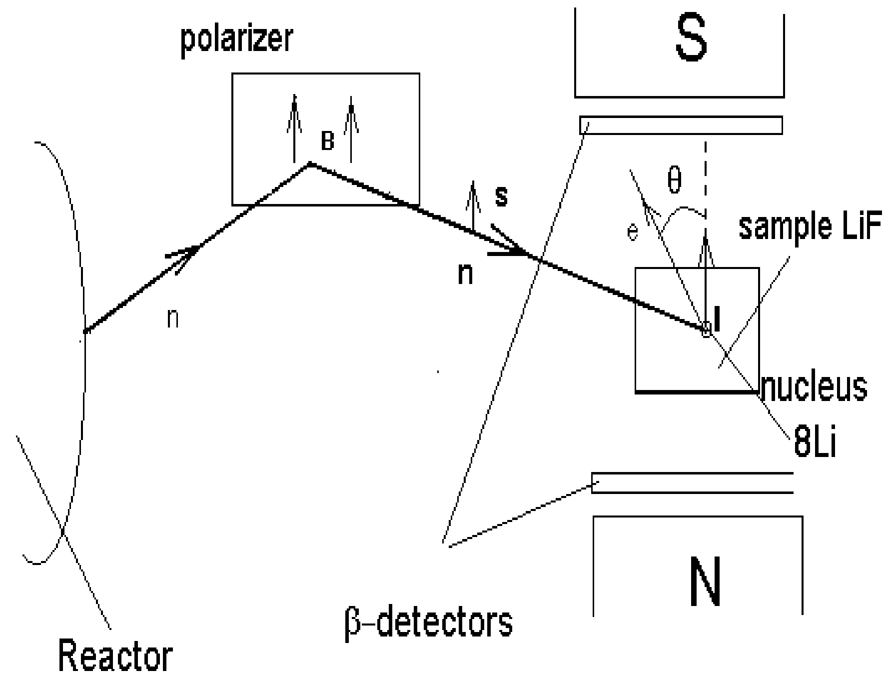
Main measurable value is the beta-decay asymmetry

$$\varepsilon(t) = \frac{N(\vartheta=0,t) - N(\vartheta=\pi,t)}{N(\vartheta=0,t) + N(\vartheta=\pi,t)} \Leftrightarrow ap(t),$$

t=0 corresponds to creation of β-nucleus in the sample via (n, γ)-reaction.

Example:

$$^{7}Li(\vec{n},\gamma)^{8}\overrightarrow{Li}, \quad a \approx 0.1, \quad \mathrm{T}_{1/2} \approx 0.84s$$



The dependencies of ε on time, external magnetic static $\mathbf{H}_0$ and alternating $\mathbf{H}_1$(t) fields, temperature T and other controllable parameters give information about the processes with participation of beta-nuclei. **Last reviews:**

Yu.Abov, F.Dzheparov, A.Gulko, D.Lvov. Appl. Magn.Res. **45**,1205,2014.

W.A.MacFarlane. Solid State NMR **68**, 1, 2015

Reference example of modern statistical physics problem for beta-NMR: random walks in disordered media

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}Li in the single crystal ^{7}Li^{19}F. The system is accessible for direct experimental β-NMR study due to unique coincidence of g-factors of stable nuclei ^{6}Li and β-active nuclei ^{8}Li (β-nuclei). The process under study: nuclear polarization $p_{00}(t)$ from initially polarized ^{8}Li nucleus (with number i=0) can transfer to the nearest nonpolarized ^{6}Li nuclei ($i \neq 0$) and then migrates over other ^{6}Li nuclei and might return back to the ^{8}Li.

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

$$\nu_{ij} = \nu_0 r_0^s / r_{ij}^s, \quad \text{dipole transport <=> } s = 6$$

$$\text{Observable value } \left\langle p_{00}(t) \right\rangle_c = ?$$

$\left\langle p_{00}(t) \right\rangle_c$ is “proportional” to measurable beta-decay asymmetry in beta-NMR

Theoretical problem is very hard due to random distribution of the spins in crystal

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

Why the process is attractive for studies?

Primary equations are very simple, but modern theoretical physics can not predict neither exact solution nor long time asymptotics. We will see below, that solution of the problems requires development of **physics of unrenormalizable interactions**. It arises when main constructive approach – approximation of continuum media is applied. Example of simplest problem:

$$\left\langle \exp\left(-\sum_j \nu_{ji} t\right)\right\rangle = \exp\left(-\left(\beta t\right)^{1/2}\right)$$

The Förster constant $\beta \propto \nu_{ij}\left(\text{mean distance}\right) \Leftrightarrow$ natural time scale.

Moreover, **calculation of transition rates is complex modern problem of spin many body physics as well**.

Path integrals and occupation number repsentations

To measure a complexity of the problem we can construct the path integrals for averaged propagator. We will discuss simplified form, sufficient for study of long time asymptotics.

$$\frac{d}{dt}\widetilde{P}_{\mathbf{xy}} = -\sum_{\mathbf{z}}\left(n_{\mathbf{z}}\nu_{\mathbf{zx}}\widetilde{P}_{\mathbf{xy}} - n_{\mathbf{x}}\nu_{\mathbf{xz}}\widetilde{P}_{\mathbf{zy}}\right) = -\left(A\widetilde{P}\right)_{\mathbf{xy}}, \quad \widetilde{P}_{\mathbf{xy}}(t=0) = \delta_{\mathbf{xy}}.$$

Initial polarization (excitation) is placed at any site here, and then it hops in filled sites, while for exact description it must be placed initially at site, filled by a spin only.

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

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

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

Example of superfield representation

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

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

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

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

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

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

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.

Solution: we use infinite **crystal** with large **disordered unite cell,** containing $100<N_d<4000$ spins of ^{6}Li. 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.

F.Dzheparov, JETPL **82**, 521, 2005;

F.Dzheparov, D.Lvov, V.Shestopal, J.Supercond. Nov. Magn. **20**, 175, 2005.

The method incorporates all information, discussed above, its results are in satisfactory agreement with experimental data and indicate, that long time asymptotics has diffusion nature:

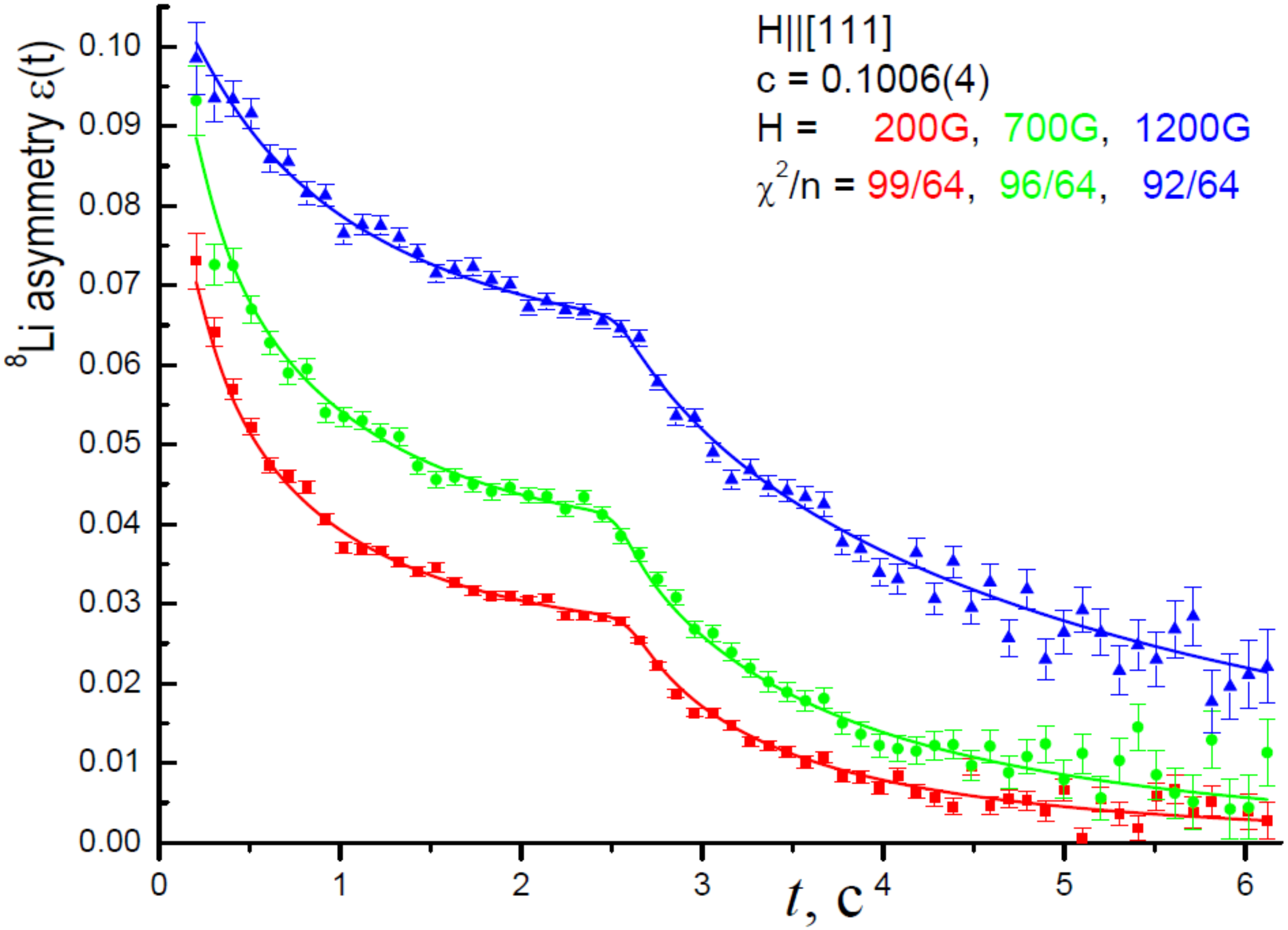
$$P_{00}(t) \sim t^{-3/2}.$$

Theoretical reviews:

F.Dzheparov. J.Phys.Conf.Ser. **324**, 012004, 2011.

F.Dzheparov. Spin Dynamics in Disordered Solids. In "Encyclopedia of Complexity and Systems Science", Springer 2015. DOI 10.1007/978-3-642-27737-5_513-3

Dependence of kinetics of depolarization of ^{8}Li on external field H. Solid line – theory. Natural time scale: β≈10/s.
Step on the lines corresponds to end of neutron pulse.
Theoretical lines have no fitting parameters, related with spin dynamics.



Yu.G.Abov, A.D.Gulko, F.S.Dzheparov, et al. Phys. At. Nucl. **77**, 682, 2014.

Similar measurements can not be fulfilled within conventional NMR!

- Spin dynamics is the most developed branch of nonequilibrium statistical mechanics now. To prove the thesis we can indicate, that inversion of evolution was realized here [W-K.Rim, A.Pines, J.Waugh. Phys. Rev. Lett. **25**, 218, 1970] in the system, which was proved as excellent thermodynamical one in brilliant studies, described in famous book of A.Abragam and M.Goldman: Nuclear magnetism. Order & Disorder. Oxford 1982.

  It should be stressed, that well known neutron spin echo is not inversion of evolution of thermodynamical system and reproduces only much more simple Hahn's echo (1950) of magnetic resonance.
- Other example is ergodic theorem, proved for systems, similar to ^{8}Li-^{6}Li, discussed above [F.Dzheparov. JETP **89**, 753, 1999].
- We hope that our beta-NMR study of delocalization of nuclear polarization in model disordered system ^{8}Li-^{6}Li produce substantial development of existing state of art both in the spin dynamics of random media and in random walks in disordered systems.

Future studies should increase the time domain accessible for measurements, now we have appropriate statistics for $\beta t \leq 25$, while reliable information on asymptotics starts at $\beta t \geq 50$.

Detail studies of dependencies of the kinetics on concentration and external static magnetic field are desirable as well to check our understanding of complex many body processes in spin dynamics, which define the rates ν_{ij} .

Other applications should be based on the advantages of beta-NMR relative to the standard NMR:

1) high sensitivity that makes it possible to perform measurements for N= $=10^6$–10^8 beta-active nuclei in a sample;

2) the sensitivity of the method does not depend on the value of external magnetic field, therefore it is possible to perform relaxation investigations in arbitrary fields, including low fields;

3) relaxation measurements do not require the application of radio-frequency fields, this is of importance in dealing with metallic samples and samples placed in metallic casings;

4) some beta-active nuclei have quadrupole moments while their stable isotopes do not (for example, ^{20}F, ^{108}Ag, and ^{110}Ag), this gives the possibility of measuring the electric field gradients at their positions – very important information for quantum chemistry;

5) defects are created near the beta-nuclei at their production via (n, γ)-reaction, that should be used in future as important property of the probe in substance on the time scale of hyperfine interactions.

Installation development

Extended investigations of fundamental processes of statistical physics as well as modern material studies are expected.

Successful work will require for the beginning

1) new detectors and electronics to operate with the flux $\sim 10^8$ beta-particle/second,

2) new NMR magnets to produce static fields up to 10T.

Multiple (ultra)small angle scattering is an excellent problem of statistical physics. For example, famous main phenomelogical equation of Moliere-Bethe theory

total scattering cross section

$P($

differential scattering cross section

$$\frac{dP(\theta,l)}{dl} = -c\sigma_t\left(P(\theta,l) - \int \frac{d^2\theta'}{\sigma_t}\sigma(\theta-\theta')P(\theta',l) \right)$$

number of scatterers per cm^3

probability to find a neutron at angle θ

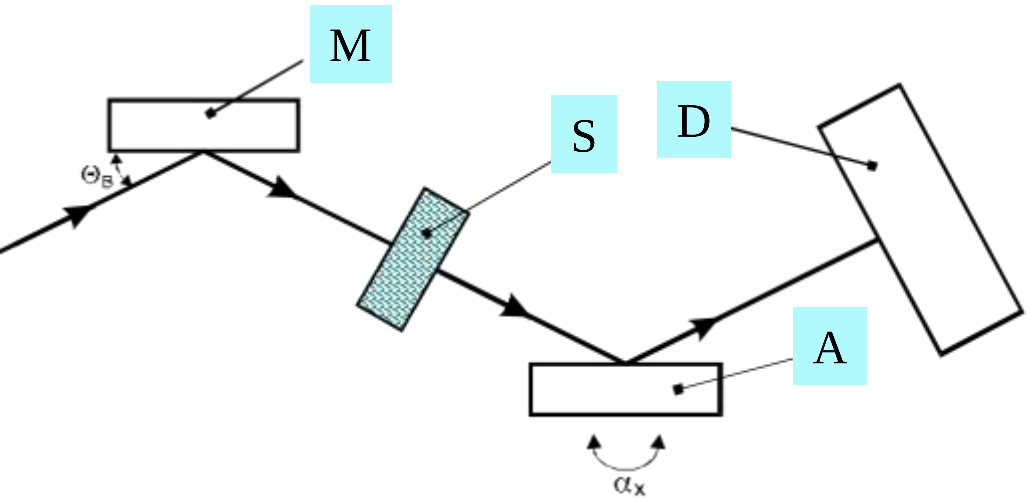
distance from the sample surface

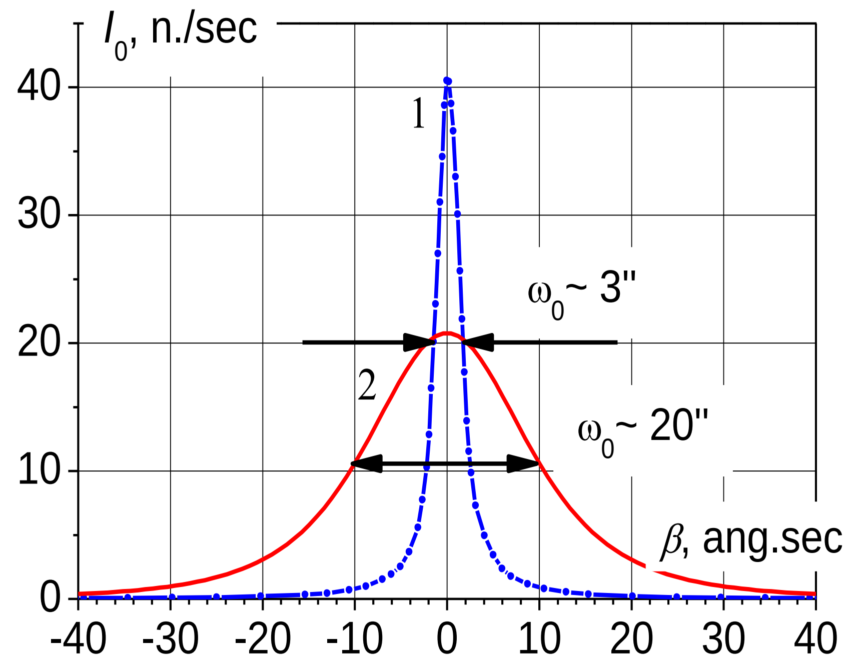
has exact foundation based only on eikonal approximation for the scattering amplitude of the sample [F.Dzheparov, D.Lvov. JETPL **72**, 360, 2000].

P(**θ**,*l*) is the probability of scattering at angle **θ** for neutrons traveled the path *l*.

Applicability of the conception of scattering amplitude for the sample in realistic conditions (out of the Fraunhofer zone) was proved as well for double crystal spectrometers [Dzheparov, Zabelin, Lvov. J. Suf. Invest. **5**, 665, 2009] and standard installations [Dzheparov, Lvov. Sol. St. Phys. **56**, 142, 2014].

Double crystal diffractometer schematic.





M and **A** are monochromator and analyzer crystals, **S** is a sample and **D** is a detector. α_x is an angle of rotation, θ_B is the Bragg angle. Intensity of neutrons on the detector **D** is measured as a function of rotational angle a_x of the analyzer.

1) Instrumental line of DCD $I_{ins}(\beta)$ at $\lambda = 1{,}75$ Å
2) becomes SANS curve when a sample is placed between crystals

Correlation effects in multiple small angle scattering.

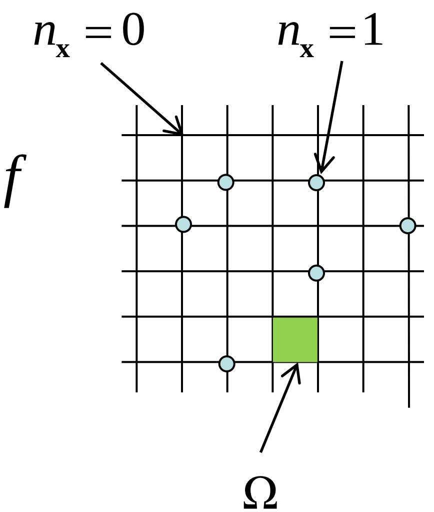
The neutron-optical potential of a medium through which neutrons propagate

occupation numbers

$$U(\mathbf{r}) = \sum_{\mathbf{x}} n_{\mathbf{x}} U_0(\mathbf{r} - \mathbf{x}, \alpha_{\mathbf{x}}) \qquad \langle n_{\mathbf{x}} \rangle = f$$

potential of grain

grain parameters



In the continuum limit $f \to 0, \;\; \mathbf{\Omega} \to 0, \;\; f/\mathbf{\Omega} = c = \mathrm{const}$

Correlations in the scatterers distribution

$$\mathbf{\Omega}^{-2} \langle n_{\mathbf{x}} n_{\mathbf{y}} \rangle = C_2(\mathbf{x}, \mathbf{y}) \neq c^2 \qquad \mathbf{x} \neq \mathbf{y}$$

pair correlation function

Scattering in eikonal approximation

Scattering amplitude

impact parameter

$$f(\mathbf{q}) = \frac{k_0}{2\pi i} \int d^2\rho \left[S(\boldsymbol{\rho}) - 1 \right] \exp(-i\mathbf{q}\boldsymbol{\rho}),$$

$$U(\mathbf{r}) = \sum_{\mathbf{x}} n_{\mathbf{x}} U_0(\mathbf{r} - \mathbf{x}, \alpha_{\mathbf{x}})$$

$$S(\boldsymbol{\rho}) = \exp\left(-i \int_{-\infty}^{\infty} \frac{dz}{\hbar \mathrm{v}} U(\mathbf{r}) \right), \qquad \mathbf{r} = (\boldsymbol{\rho}, z)$$

wave vector

neutron velocity

The normalized angular distribution of neutron momentum is studied

$$D(\mathbf{q}) = \boldsymbol{\Sigma}(\mathbf{q}) / \boldsymbol{\Sigma}_0, \quad \boldsymbol{\Sigma}(\mathbf{q}) = \left| f(\mathbf{q}) \right|^2, \quad \boldsymbol{\Sigma}_0 = \int d^2 q \boldsymbol{\Sigma}(\mathbf{q}).$$

The theoretical analysis can conveniently be carried out for the Fourier transform

$$D(\mathbf{u}) = \Sigma(\mathbf{u}) / \Sigma_0, \quad \Sigma(\mathbf{u}) = \int d^2 q \exp(-i\mathbf{q}\mathbf{u}) \Sigma(\mathbf{q}), \quad \Sigma_0 = \Sigma(\mathbf{u} = \mathbf{0})$$

In the case of a system with an uncorrelated random scatterer distribution, the exact result can be obtained using the relationship (continuum media limit)

$$\left\langle \exp\left(\sum_{\mathbf{x}} n_{\mathbf{x}} R_{\mathbf{x}} \right) \right\rangle = \exp\left[c \int_V d^3x \left\langle e^{R_{\mathbf{x}}} - 1 \right\rangle_{\alpha} \right].$$

Averaging with the use of this formula gives

$$D(\mathbf{u}) = D_0(\mathbf{u}) = \exp\left(-cl(\sigma_0 - \sigma(\mathbf{u})) / p_0^2 \right)$$

This equation coincides with the standard result of the Moliere theory.

For a high scatterer concentration the many-body distribution function cannot be factorized into a product of one-particle functions.

In the **presence of correlations** we expand $D(\boldsymbol{\xi})$ in powers of occupation numbers.

$$\exp\left(\sum_{\mathbf{x}} n_{\mathbf{x}} R_{\mathbf{x}}\right) = \prod_{\mathbf{x}} \left[1 + n_{\mathbf{x}}(e^{R_{\mathbf{x}}} - 1)\right] =$$

$$= 1 + \sum_{\mathbf{x}} n_{\mathbf{x}} \left(e^{R_{\mathbf{x}}} - 1\right) + \frac{1}{2} \sum_{\mathbf{x} \neq \mathbf{y}} n_{\mathbf{x}} n_{\mathbf{y}} \left(e^{R_{\mathbf{x}}} - 1\right)\left(e^{R_y} - 1\right) + \ldots \tag{1}$$

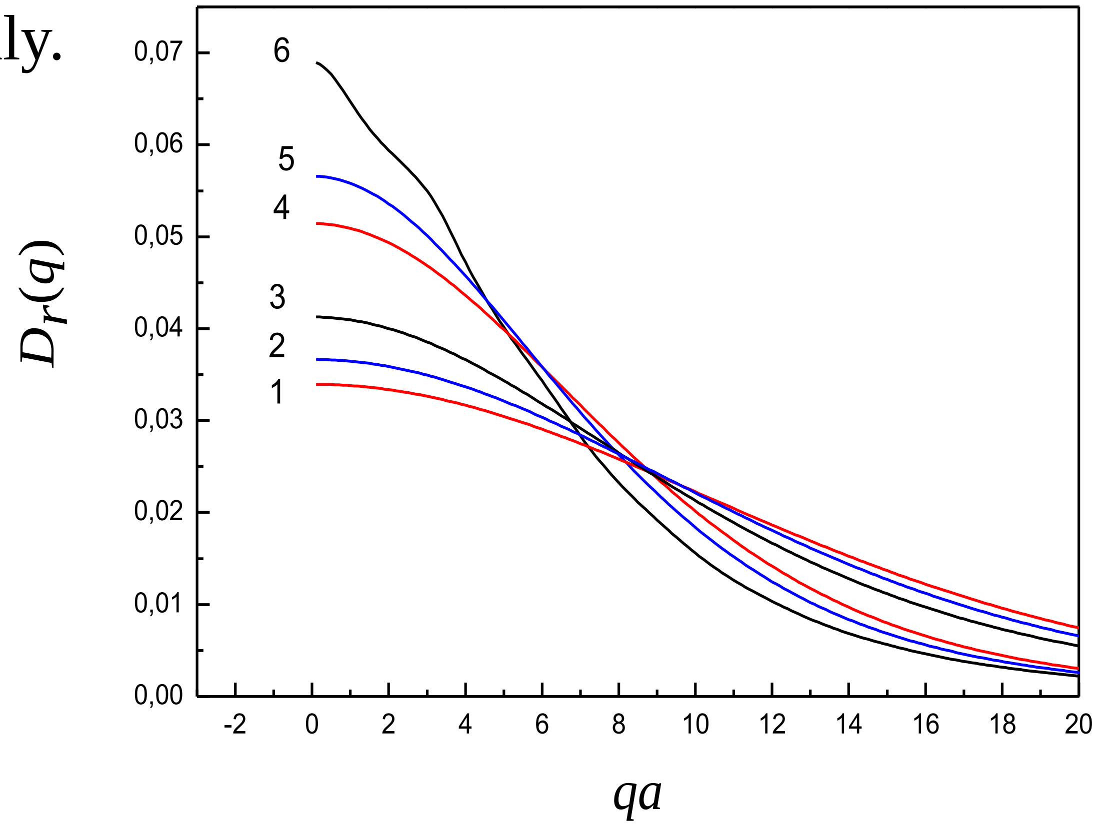
Introducing $Q_{\mathbf{x}}(\boldsymbol{\xi}, \alpha) = \exp(R_{\mathbf{x}}(\mathbf{0}, \boldsymbol{\xi}, \alpha)) - \mathbf{1}$ and averaging Eq.(1) term by term over the occupation numbers distribution, one obtains in the continuum limit

$$D(\boldsymbol{\xi}) = D_0(\boldsymbol{\xi}) \exp\left(\frac{c^2}{2} \int d^3x d^3y \kappa(\mathbf{x}, \mathbf{y}) \left\langle Q_{\mathbf{x}} Q_{\mathbf{y}} \right\rangle_{\alpha} + \right.$$

$$\left. + \frac{c^2}{2} \int d^3x d^3y (\left\langle Q_{\mathbf{x}} Q_{\mathbf{y}} \right\rangle_{\alpha} - \left\langle Q_{\mathbf{x}} \right\rangle_{\alpha} \left\langle Q_{\mathbf{y}} \right\rangle_{\alpha}) + O(c^3) \right)$$

$$\kappa(\mathbf{x}, \mathbf{y}) = \frac{C_2(\mathbf{x}, \mathbf{y})}{c^2} - 1$$

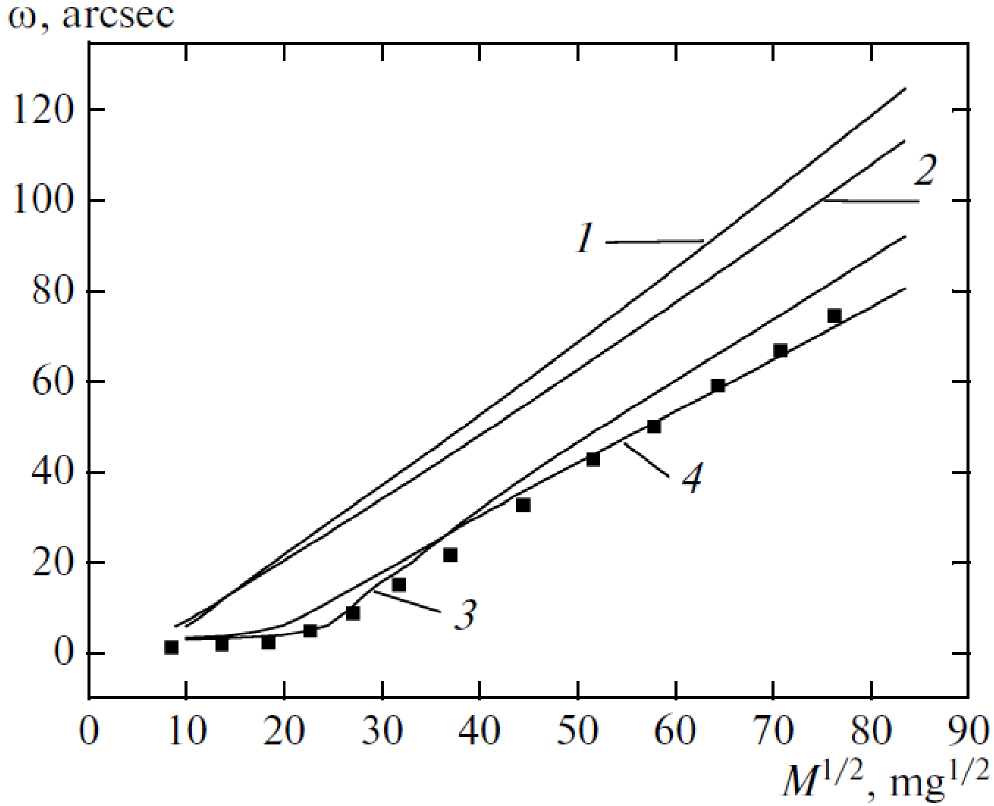
Intensity of scattered neutrons, taking into account two-particle correlations only.



Intensity $D_r(q)$ of scattered neutrons for samples with the Born parameter ν=U_0τ= 0.2 (τ is duration of the collision), effective thickness (or number of scattering) $l_e = l/l_c$, and different filling volumes of scatterers η: *1*) l_e = 40 and η = 0; *2*) l_e = 40 and η = 0.3; *3*) l_e = 40 and η = 0.5; *4*) l_e = 20 and η = 0; *5*) l_e = 20 and η = 0.3; and *6*) l_e = 20 and η = 0.5.

l_c is mean free path, η=0 correspond to Moliere-Bethe theory.

Modern results of our studies on MSANS for media with high density of catterers can be found in [Yu.Abov, F.Dzheparov, N.Elyutin et al. JETP **116**, 442, 2013; Phys.At.Nucl. **77**, 1187, 2014]. Example:



Plots of MSANS linewidth ω vs. square root of Al powder mass M (propotional to the scattering number l_c): (squares) experimental data; (curve 1) calculation using the Moliére theory for a monodisperse powder; (curve 2) same for polydisperse powder with grain size distribution; (curve 3) calculation using the proposed theory and the Poisson distribution of grain sizes; (curve 4) same for lognormal distribution of grain sizes.

1) Experimental results can be described, if parameters of scatterers are known (direct scattering problem). The study includes construction of simplest mathematical model of the sample, based on known distribution of spherical scatterer sizes and their impenetrability.

2) For the diffraction domain (where Born parameter $\nu = U_0 \tau << 1$, τ is duration of the collision) the mathematical model of the sample is not necessary – two particle correlations are sufficient.

3) Inverse problem for the same idealizations is not solved up to now and requires new studies.

4) Special study of importance of three-particle correlations for refraction domain (where Born parameter $\nu >> 1$) is desirable.

5) Extended experimental checking of the results for different model samples as well as material studies of new substances are expected.

6) The most interesting part of measurements for refraction domain and ultra-small angles can be fulfilled on standard double crystal set up. We are sure, that the installation, based on Universal Neutron Diffractometer, designed in our group (ITEP and MEPhI), and used for the studies, mentioned above, should be better for physical investigations at lest, because it can be adjusted for many new problems, arising during the studies.

We hope you will come to the conclusion that our studies are interesting and should be continued on the reactor PIK.
We invite you to cooperation in the studies!

Thank you for the attention!