Ferrofluid dynamics by ODD and EVEN Neutron Spin Echo studied

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Odd neutron spin echo (ODD-NSE)[1] combined with NSE(EVEN) [2] yield the excitations spectra in condensed matter $S(\omega,\mathbf{q})=S_{odd}(\omega,\mathbf{q})+S_{even}(\omega,\mathbf{q})$ from odd and even NSE-signals dependent on time t and momentum transfer **q**:
$S_{odd}\sim\int S(\omega,\mathbf{q})\sin(\omega t)d\omega$,
$S_{even}\sim\int S(\omega,\mathbf{q})\cos(\omega t)d\omega$.
In ferrofluids (FF) the frequencies $\omega \leq 1/\tau$ and relaxation times τ of soft modes characterise particles' interaction masked by viscous medium. We have studied the FF not magnetised (20°C, density d=1.69g/cm^3; 20%vol.of Fe_3O_4 stabilised in pentanole by oleic and dodecylbenzensulphone acids' bilayer; magnetisation $4\pi M_s$=860G). Each particle (diameter $D_P\sim$16nm, core $D_m\sim$10nm), surrounded by other particles (spacing ~20nm $\sim D_P$), can move only by their regrouping (cage effect). Indeed, the NSE-data (Fig.1) confirm a delayed correlations $S_{even}(t)$ damping started at t>t*~10ns for momentum transfer $q_{1,2,3}$ (0.31; 0.55; 0.91 nm^{-1}). At $q>1/D_P$ the even part shows a diffusion of single particle by cage slowed. The $S_{odd}(t)\sim t$ grow quite linear at $t \leq$ 1ns, their slope gives the energy transfer q-dependent. At t >10ns the curves converge.

In gaussian approximation $S_{even}(t) \sim \exp[-q^2\Gamma(t)/2]$ describes particle's self-correlation, where $\Gamma(t)$ is a squared particle's displacement for the time t. The $S_{odd}(t)$ do not adhere a simple model, $S_{odd}(t/\tau)\sim t\cdot\exp(-t/\tau)$. The normalised functions $F(t)=S_{odd}(t)/t$ display oscillations and damping at q~0.6-0.9nm^{-1}(Fig.2).

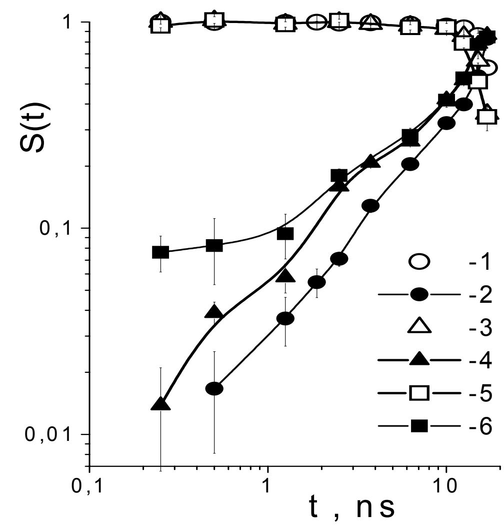

Figure 1: Even (1,3,5) and odd (2,4,6) functions: 20°C, q_1(1,2); q_2(3,4), q_3(5,6). Lines are spline-functions.

Note, neutrons excite particles' vibrations:
$F(t) = \langle\omega\rangle + \beta\cdot t^2 + \mu\cdot\cos(\Omega t)\exp(-\nu t)$
where fitting parameters are q-dependent (Tab.1). The $q_1\sim\pi/D_m$ is maximum wave number for acoustic wave, frequency $\Omega\sim$0.5rad./ns, in coupled particles chain (spacing ~ core diameter $D_m\sim$10nm). At $q_{2,3}>q_1$ the optical modes only permitted have frequency $\Omega\sim$0.3rad./ns, period $T_\Omega\sim$20ns, damping time $\tau=1/\nu\sim10\text{ns}\sim T_\Omega/2$.
The slow mode has frequencies $\langle\omega\rangle = \int\omega\cdot S(\omega,q)d\omega/\int S(\omega,q)d\omega\sim(3\text{-}5)\cdot10^7$rad./s. The periods $T=2\pi/\langle\omega\rangle\sim$100ns at $q_{2,3}$, and T~200ns at q_1 are much longer than the experimental time t=0-20ns. The estimated velocity of slow waves propagation U ~ $\langle\omega\rangle/q$ ~10cm/s is by 4 orders in magnitude smaller than sound velocity in liquids.

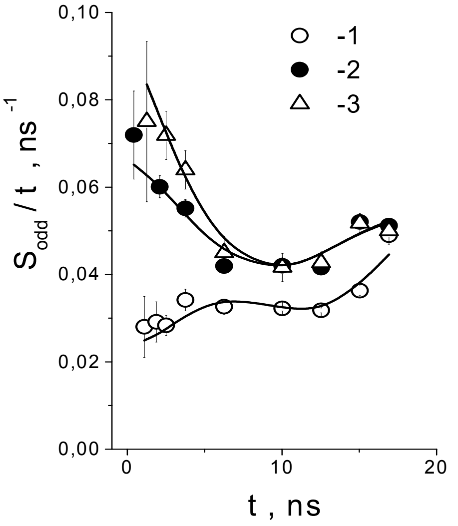

Figure 2: Function $S_{odd}(t)/t$ at $q_{1,2,3}$ (1,2,3). Lines are fitting functions F(t).

q , nm^{-1}	0.31	0.55	0.91
$<\omega>$, ns^{-1}	0.029 ±0.002	0.049 ±0.002	0.051 ±0.001
β , ns^{-3}	$(5\pm1)\cdot10^{-5}$	—	—
μ , ns^{-1}	- 0.0044 ±0.0011	0.017 ±0.010	0.042 ±0.011
Ω, ns^{-1}	0.50 ±0.02	0.32 ±0.02	0.30 ±0.02
ν , ns^{-1}	—	0.072 ±0.062	0.14 ±0.04

Tab. 1: Odd function: parameters.

The ratio $\mu/<\omega>\sim0.2$-0.8, growing with q, indicates the balance of fast and slow dynamics. The latter dominates at small q, when the odd-correlations are also delayed for $t^*\sim10$ns, that is described by the term $\beta\cdot t^2$ in function F(t). At the scale of radius $2\pi/q_3 \sim D_m/2$ fast and slow components in dynamics are comparable.

The $S(t)_{even}=\exp[-q^2\Gamma(t)/2]$ depend on particle's shift $\Delta X=\Gamma(t)^{1/2}=[-(2/q^2)\ln(S)]^{1/2}$ increasing slowly up to $\Delta X(t)\sim1$nm at t<15ns. Then it achieves the $\Delta X\sim3$nm ~ radius of core (Fig.3). At t<15ns the shift adheres the function $\Delta X(t)=[a^2+(vt)^2]^{1/2}$. Here a=0.4±0.1nm is the amplitude of fast oscillations, v=8.7±0.3 cm/s is the component of particle's velocity along q (data for $q=q_1$).

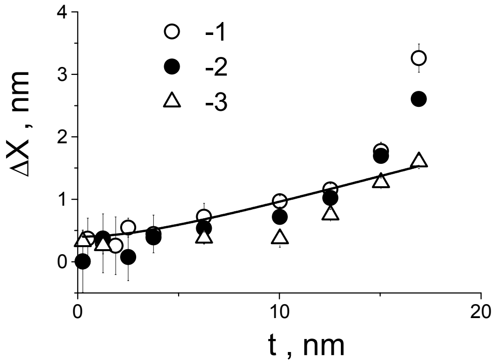

Figure 3: Particle's shift: data 1-3 at $q_{1,2,3}$, fitting function $\Delta X=[a_2+(vt)^2]^{1/2}$ for data1.

Vibrating with a small amplitude (~ solvent molecule), a particle moves slowly waiting the conditions for a jump (length ~ shell thickness ~ core radius). Its velocity v~10cm/s is smaller by an order in magnitude than thermal one, $V_T=(kT/m_P)^{1/2}$ ~100cm/s (m_P is particle's mass). Hence, a particle involves solvent molecules and neighbouring particles (1st, 2nd spheres, region ~100 volumes of particle). Comparing even and odd dynamics, we conclude: the damping time of particle's oscillations $\tau=1/\nu\sim T_\Omega/2\sim10$ns is comparable with induction period, after that it jumps from cage to a new position.

Even data show particle's motion as a fast vibration + slow moving and final jump. Odd data visualise the main dynamic component that is a soft mode. Without it a jump of particle to new position is not possible. Odd NSE-signal reflects the potential defining particles motion. In FF with massive particles we observe their fast vibrations and relaxation initial stage, $S_{odd}(t)\sim t$.

References

[1] Lebedev V.T., Torok Gy. Odd neutron spin echo // 3rd European Conf. on Neutron Scattering. Sept. 3-6, 2003, Montpellier, France.Abstract book, p.155.

[2] Mezei F. Neutron Spin-Echo: a new concept in polarised neutron techniques. // Z.Physik, 1972, v.255, p.146-160.