Supplementary Material: Optimization of the DOSY sequence

The experimental conditions for performing the DOSY-NMR studies on lipid NP (very slow diffusing systems) had to be optimized in order to obtain the adequate level of sensitivity in standard NMR probes. Therefore, an equation was deduced to optimize the diffusion delay time (Δ) in the experiment. For the egsteSL sequence used (Antalek et al., 2002), the decay of the sample magnetization stored on the z-axis during the diffusion delay time can be written as:

$$I = I_0 e^{(-\Delta/T_1)} e^{(-(\gamma G\delta)^2(\Delta-\delta/3)D)} \qquad \text{(Eq. 1)}$$

In this equation I represents the finally observed signal intensity or integral, Δ the diffusion delay time, G the gradient power level, δ the duration of the gradients, D the diffusion of the molecule and T_1 the spin-lattice relaxation.

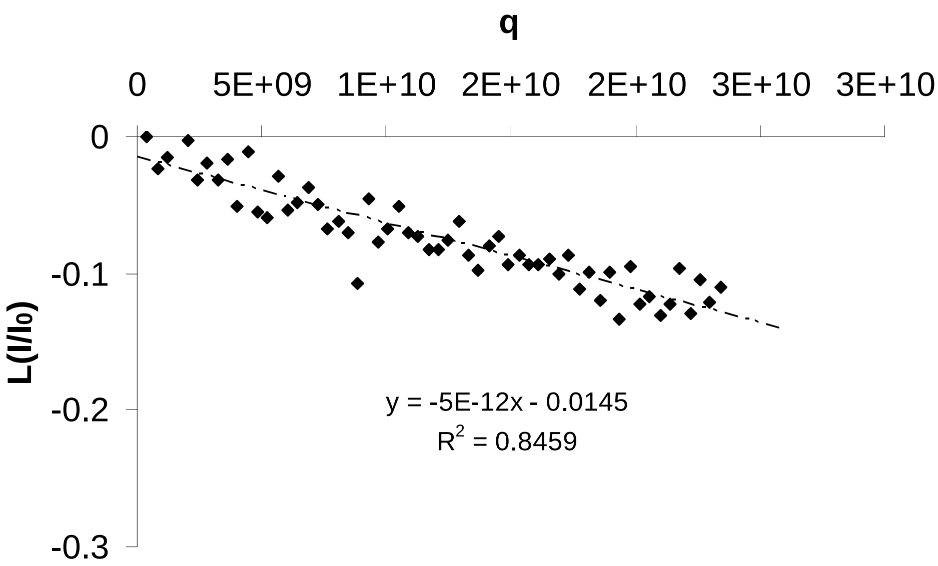
The optimum value of Δ which maximizes the sensitivity (Δ_{max}) corresponds to the value that maximizes the difference in the observed intensity (I) between the trace of the experiment acquired with the minimum gradient (G0), and the trace acquired with the maximum gradient (Gmax) (typically the maximum available in the NMR probe) (Eq. 2):

$$\Delta_{max} = \frac{1}{3D\gamma^2\delta^2(G_{min}{}^2 - G_{max}{}^2)}(\gamma^2 G_{min}{}^2\delta^3 D - \gamma^2 G_{max}{}^2\delta^3 D - 3Ln\frac{1+(\gamma G_{max}\delta)^2 DT_1}{1+(\gamma G_{min}\delta)^2 DT_1})$$

In practice, accurate values of Δmax can be obtained for most samples considering G_0=0, a simplification that lead to the following equation:

$$\Delta_{max} = \frac{L(1+T_1(\gamma G\delta)^2 D)}{(\gamma G\delta)^2 D} + \delta/3 \qquad \text{(Eq. 3)}$$

For very slow diffusion values, the equation converges to a value of $\Delta_{max} \approx T_1$. The optimum values calculated from this equation are in agreement with some previous numerical simulations performed by us. A typical graph from nanoparticles with a diameter equal to 80 nm can be seen in the following graph:



In this graph, I is the integral for each experiment with a given G gradient, I_0 is the integral for G_0 and $q= -(\gamma G\delta)2(\Delta-\delta/3)$ is the total gradient strength. The slope of the linear fitting of this plot results in the diffusion of the system.

Reference:

B. Antalek. Using pulsed gradient spin echo NMR for chemical mixture analysis: how to obtain optimum results. Concepts in Magnetic Resonance 4:225-258, 2002.