Anomalous spin echoes in highly polarised liquid helium mixtures

Geneviève Tastevin [1], Nathalie Piegay, François Marion, Pierre-Jean Nacher

Laboratoire Kastler Brossel, E.N.S., 24 rue Lhomond, F75005 Paris, France

Abstract

NMR experiments are performed on laser polarised liquid helium mixtures in low field (2mT) above 1K. Concentrations range from 0.5% to 10% and nuclear polarisations up to 0.1. For large magnetisations, following a 90°-τ-180° RF pulse pair the spin echo signal detected at time 2τ is significantly smaller than expected from attenuation due to diffusion in the applied static field gradient. Unexpected echoes are also observed at times 3τ, 4τ, 5τ... The first feature may be related to other effects of large dipolar fields in bulk samples (dynamic instabilities, altered transverse decay). Numerical simulations on lattices have been performed to quantitatively interpret all spin echo results.

Key words: spin-polarised He3-He4 solutions ; NMR ; dipolar field ; spin diffusion

Classical dipolar fields can have a major influence on spin dynamics in highly magnetised samples. Strongly altered spin echo (SE) or free induction decay (FID) signals have been obtained by pulsed NMR in brute force polarised systems [1–3]. Fast liquefaction of laser polarised noble gases produces liquid samples with high magnetisation densities in any magnetic field. Accurate NMR measurements can be performed at low field, with well controlled static inhomogeneities, to elucidate the interplay of dipolar and external fields. In anisotropic samples long-lived magnetisation modes and spectral clustering [4,5] (resp. abrupt decays [6]) are obtained by FID at low (resp. large) flip angles. Bulk samples [4] also exhibit instabilities at large flip angles, a generic feature related to turbulent spin motion as shown by recent theoretical work [7]. Preliminary investigations of the evolution of the average magnetisation at larger time scales indicated that refocussing 180° NMR pulses fail to generate SE in pure liquid ^{3}He films [6] or lead to non exponential decay of echo train decay in bulk ^{3}He-^{4}He solutions [8]. We report on a systematic study of SE signals induced by a 90°-τ-180° pulse sequence as a function of magnetisation density and applied field gradient, at low field (2 mT), for spherical samples (8.8 mm in diameter) of laser polarised liquid ^{3}He-^{4}He mixtures above 1K. Experimental results are compared to computer simulations data.

The sample preparation method is described elsewhere [9]. Magnetisation densities are parametrized using the dipolar frequency $F_{dip} = (\gamma/2\pi)\mu_0\mu_n NM$, where γ is the gyromagnetic ratio, μ_n the nuclear moment, N the number density, and M the nuclear polarisation of ^{3}He nuclei in the liquid. For these measurements ^{3}He concentrations ranging from 0.5% to 12% and nuclear polarisations up to M=0.1 provide F_{dip} up to 10 Hz. Static gradients G, applied along the field axis, induce Larmor frequency spreads (over the cell of diameter d) $\Delta F_G = (\gamma/2\pi)Gd$ varying from 30 to 400 Hz. Under these conditions, the FID signal lifetime is independent of F_{dip} and changes with G as expected [9].

In a classical system, the 180° pulse is expected to induce a single SE at time 2τ of amplitude A_1 such that $(A_1/A_0)_{diff} = \exp(-2\gamma^2 G^2 D\tau^3/3)$ where D is the diffusion coefficient and A_0 the FID amplitude after the 90° pulse. In the polarised mixtures, the SE amplitude at time 2τ is significantly depleted (Fig.1, main plot) and additional SE observed at times $n\tau$, with $n \geq 3$ (Fig.1, insert). While large dipolar fields may lead to

[1] E-mail: tastevin@lkb.ens.fr

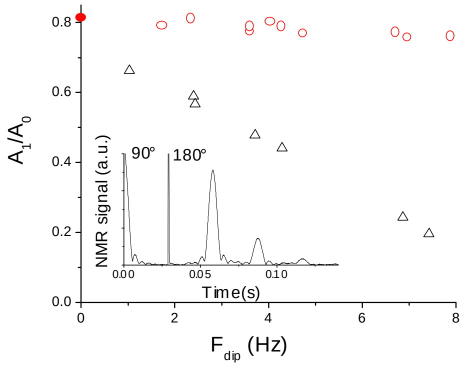

Fig. 1. Selected measurements in a 0.92% mixture at 1.14K. Insert : NMR signal for $F_{dip} = 7\ Hz$ and $\Delta F_G = 340\ Hz$. Main plot : Normalised amplitudes of the first SE as a function of the dipolar frequency (see text). Triangles (circles) correspond to $\Delta F_G = 33.3\ (340)\ Hz$ and τ=110.8 (23.6) ms, i.e. to identical diffusion attenuation (solid dot).

multiple SE [1] a single echo is expected for the particular 90°-180° pulse pair, as has been argued in the large gradient limit [11,12] and observed [3]. Effects of radiation damping on transverse precession have been checked to be negligible in our experiment. The potential influence of imperfect 180° pulses [3,11] must also be ruled out, since rectangular pulses of uniform RF field B_1 are used such that $(\gamma/2\pi)B_1 = 1.25\ kHz$.

The variation of A_1/A_0 with the sample magnetisation has been quantitatively studied as a function of applied gradient and diffusion coefficient. The first SE depletion becomes smaller when D increases (e.g. if concentration or temperature are lowered) or when a stronger gradient is applied (see Fig.1, main plot), which is consistent with established features of NMR instabilities [7,9]. Computer simulations are performed

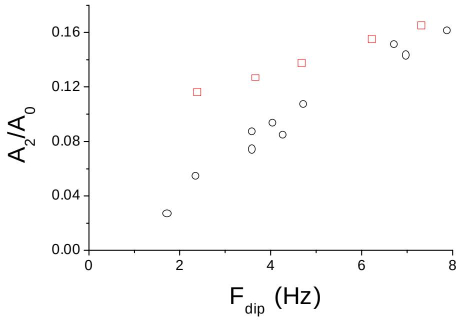

Fig. 2. Amplitude of the 2^{nd} spin echo, scaled to that of the FID, as a function of F_{dip}. Squares (circles) correspond to $\Delta F_G = 83.2\ (340)\ Hz$ and τ=59.9 (23.6) ms (same mixture as in Fig.1).

using a cubic lattice model with periodic boundary conditions. The use of fast Fourier transform drastically reduces the computation load to derive the full time evolution of the magnetisation under the combined influence of external and dipolar fields. The non linear Bloch equations are solved by the Runge-Kutta method.The underlying coarse graining approximation is valid as long as spatial variations of magnetisation from site to site are smooth enough. Finite size samples are modelled with the magnetisation being set to zero on a suitable number of lattice sites located at the edges of the unitary replication cell. This model has been shown to predict a F_{dip} and ΔF_G dependence of FID lifetimes which accurately reproduce the experimental data [9]. Dynamic analysis of magnetisation maps show that the depleted amplitude of the first SE arises from transverse magnetisation being transfered back to the longitudinal axis (with null average over the sample volume). However the model gives a A_1/A_0 variation with F_{dip} which quantitatively smears out too rapidly with increasing D. It also fails to generate multiple SE for a 90°-180° pulse pair.

Further theoretical and experimental work is needed to fully elucidate the complex evolution of magnetisation under combined action of dipolar field and applied gradients. Large magnetisation densities contribute to enhance the signal-to-noise ratio in NMR experiments, but operating conditions must be carefully chosen to avoid biases (e.g. for diffusion coefficient measurements [8]). Our results have been obtained in situations where NMR evolution is dominated by classical dipolar couplings, but similar effects can have a strong influence on magnetisation dynamics in Fermi liquids [11] or degenerate systems at low temperature.

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