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NMR shifts in ^{3}He in aerogel induced by demagnetizing fields

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Introduction. As it was shown by C. Kittel [1], demagnetizing fields may result in an additional frequency shift in magnetic resonance experiments at large values of magnetization. The spin susceptibility of liquid ^{3}He is small, and the Kittel shifts in bulk samples were observed only in spin polarized ^{3}He [2, 3]. At ultralow temperatures the shift was also observed in ^{3}He films [4, 5] due to a few atomic layers of solid paramagnetic ^{3}He adsorbed on the surface which follow the Curie–Weiss law. In this case the nuclear magnetic resonance (NMR) signal from ^{3}He, containing liquid and solid components, is observed as a single line (due to a fast spin exchange [5]) with the shift as weighted average. The Kittel shift also may be observed in ^{3}He in nanostructures, e.g., in aerogels. In globally isotropic aerogel the average shift is zero, but it may appear in the anisotropic sample.

Here we study the Kittel effect in liquid ^{3}He in two different anisotropic nanostructures: nematic and planar aerogels. Nematic aerogel consists of nearly parallel strands along the symmetry axis z, while in planar aerogel the strands are random in a plane normal to z.

Theory. The magnetic susceptibility of ^{3}He in aerogel is a sum of those in liquid (χ_l) and solid (χ_s) ^{3}He:

$$\chi = \chi_l + \chi_s = \chi_l + C_s/(T - \Theta),$$

where C_s is the Curie constant, Θ is the Curie temperature of solid ^{3}He, $\chi_i \equiv M_i^{\text{aero}}/H$, $\mathbf{H}$ is the magnetic field, M_i^{aero} is the magnetic moment of liquid ($i = l$) or solid ($i = s$) ^{3}He per unit volume of the *aerogel* sample. In normal liquid ^{3}He χ_l does not depend on T and may only decrease in superfluid ^{3}He, so at low T the NMR signal from solid ^{3}He can prevail over that from liquid ^{3}He. The common NMR line has the shift [5]:

$$\Delta\omega' = (\chi_l \Delta\omega_l + \chi_s \Delta\omega_s)/(\chi_l + \chi_s), \tag{1}$$

where $\Delta\omega_l$ is a shift in liquid ^{3}He, $\Delta\omega_s$ is a Kittel shift in solid ^{3}He. Here all the shifts are measured from the Larmor frequency $\omega_L = \gamma H$, where γ is the gyromagnetic ratio of ^{3}He.

The shift in solid ^{3}He at a separate strand is $\Delta\omega_s = \pi\gamma M_s^{\text{cyl}} \left(2 - 3\sin^2\varphi\right)$, where M_s^{cyl} is a magnetization of solid ^{3}He on a cylinder surface, φ is an angle between $\mathbf{H}$ and the cylinder axis [3]. Strands of nematic aerogel are parallel to one another, so, if we neglect interactions between strands, then the mean shift is

$$\Delta\omega_s = \pi\gamma \frac{\chi_s}{s_V \delta} H \left(2 - 3\sin^2\varphi\right), \tag{2}$$

where s_V is the surface area per unit volume of the aerogel, δ is a thickness of solid ^{3}He layers.

Strands of planar aerogel are parallel to the distinguished plane. After averaging over angular distribution of the non-interacting strands we get

$$\Delta\omega_s = \pi\gamma \frac{\chi_s}{s_V \delta} H \left(\frac{3}{2}\sin^2\varphi - 1\right), \tag{3}$$

where φ is an angle between $\mathbf{H}$ and z.

Samples and methods. We used 2 nematic aerogel samples made from nafen [6] consisting of Al_2O_3 strands with diameters $d \approx 9$ nm [7]: nafen-243 with overall density $\rho = 243\,\text{mg/cm}^3$, porosity $p = 93.9\,\%$, and nafen-910 with $\rho = 910\,\text{mg/cm}^3$, $p = 78\,\%$. The planar aerogel was made from mullite aerogel consisting of strands with $d \approx 10$ nm [8]. It has $p = 88\%$ and $\rho = 350\,\text{mg/cm}^3$. The characteristic sizes of all samples is 4 mm. Spin diffusion measurements in normal ^{3}He in these samples [8, 9] confirm their strong anisotropy.

Experiments were done using continuous wave NMR in fields 24–402 Oe (NMR frequencies are 78–1303 kHz) at pressures s.v.p.–29.3 bar. The purity of ^{3}He was about 0.01 % in experiments with nafen and 0.07 % with planar aerogel that corresponds to preplating of the samples with ~ 0.1 and ~ 0.6 atomic layers of ^{4}He respectively. The necessary temperatures were obtained by a nuclear demagnetization cryostat and measured by a quartz tuning fork.

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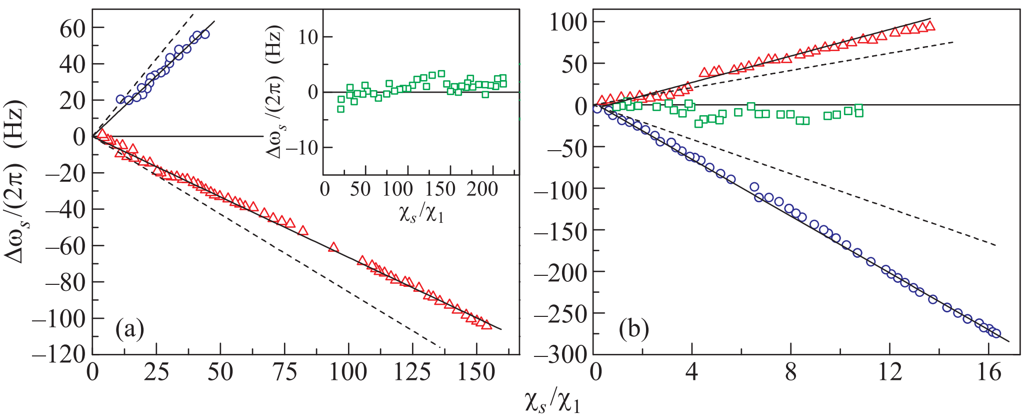

Fig. 1. (Color online) The Kittel shift in solid ^{3}He in nafen-910 (a) and in planar aerogel (b) versus χ_s. Circles: $\varphi = 0$, triangles: $\varphi = \pi/2$, squares: $\varphi \approx 55^\circ$ ($\sin^2\varphi \approx 2/3$). The dependencies are implicit functions of T, e.g., triangles in panel (a) correspond to temperatures from 23 mK down to ~ 0.6 mK. Solid lines are linear fits with the ratio of slopes ≈ 1.9 (a) and ≈ 2.1 (b). Dashed lines are theoretical predictions (see text). (a) – Circles and triangles: $P = 7.1$ bar and $\omega_L/(2\pi) = 361.4$ kHz, squares: $P = 29.3$ bar and $\omega_L/(2\pi) = 78.4$ kHz. (b) – $P \approx 0$ bar. Triangles and squares ($\omega_L/(2\pi) = 588.6$ kHz) were recalculated to match the Larmor frequency of circles ($\omega_L/(2\pi) = 1303.2$ kHz)

Results and discussions. In all samples the measured susceptibility, determined from the intensity of the NMR absorption line, has a clear Curie–Weiss behavior. From Eqs. (2), (3) it follows that the Kittel shift $\Delta\omega_s \propto H$, so high magnetic fields allow us to investigate the Kittel effect originating from the aerogel anisotropy.

We studied in more detail the Kittel effect in nafen-910, where superfluidity of ^{3}He is completely suppressed [10] and $\Delta\omega_l = 0$ at any T. Using Eq. (1) we can determine $\Delta\omega_s$ from measurements of $\Delta\omega'$ and χ_s/χ_l versus T (Fig. 1a). The shift is positive at $\varphi = 0$, negative at $\varphi = \pi/2$, and the ratio of the absolute values of the shifts is ≈ 1.9 (should equal 2 according to Eq. (2)), while the data in the inset demonstrate the absence of the shift at $\sin^2\varphi = 2/3$. Using Eq. (2) and estimations $\chi_l \approx 4.25 \cdot 10^{-8}$ emu, $s_V \approx 102\,\mathrm{m}^2/\mathrm{cm}^3$, $\delta \approx 5.5$ Å [11] (with accuracy of ± 20 %) we obtain the expected values for $\Delta\omega_s$ (dashed lines in Fig. 1a) which are surprisingly close to the experimental results.

Measurements of $\Delta\omega_s$ in planar aerogel (see Fig. 1b) also agree with theory (Eq. (3)). The shift for $\varphi = 0$ is negative and ≈ 2.1 larger (should also be 2) than that for $\varphi = \pi/2$ which is positive. At $\sin^2\varphi = 2/3$ the shift is zero as expected from Eq. (3). Estimation of the shift in planar aerogel gives the same order of magnitude as in experiments (we used $\chi_l \approx 3.11 \cdot 10^{-8}$ emu, $s_V \approx 47\,\mathrm{m}^2/\mathrm{cm}^3$, $\delta \approx 2.8$ Å [11]).

Conclusions. We have observed NMR shifts due to the Kittel effect in ^{3}He confined in aerogel-like nanostructures with different types of the global anisotropy and demonstrated that values of the shift well agree with the theoretical expectations. At ultralow temperatures even in moderate magnetic fields these shifts may be large enough to mask the ^{3}He superfluid transition but can be avoided by using lower magnetic fields or by choosing the proper angle between the axis of the anisotropy and the magnetic field.

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