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Nuclear spin dynamics in nuclear-ordered solid ^{3}He in the low field phase

T. Matsushita, R. Nomura, H. H. Hensley*, H. Shiga, and T. Mizusaki

Department of Physics, Faculty of Science, Kyoto University, Kyoto 606-01, Japan
E-mail:f54122@jpnkudpc.bitnet

* *Present address: Metabolic Magnetic Resonance Research Computer Center, University of Pennsylvania, Philadelphia, PA 19104, USA*

Pulsed NMR experiments have been performed on U2D2 solid ^{3}He with a single domain in high fields. The free induction signal decayed rapidly under certain conditions. The rapid decay was attributed to the onset of an instability of uniform precession. We propose a model for the instability due to the self-induced emission from the upper-mode to lower-mode magnon branch, which is similar to the Suhl instability in electronic ferromagnets. Under stable conditions of the spin motion, we observed the tipping-angle-dependent frequency shift and multiple spin echoes, which agrees well with Namaizawa's theory.

1. Introduction

Nuclear-ordered bcc solid ^{3}He in the low field phase is an ideal system to study spin dynamics in an antiferromagnet with uniaxial anisotropy, where Hamiltonian can be described by a sum of the isotropic exchange interaction and the nuclear dipole interaction. The elementary excitations in the ordered phase are well described by magnons and all the nuclear-magnetic properties can be studied on a microscopic basis.

Osheroff, Cross and Fisher (OCF) [1] proposed the structure for the low field phase, called the U2D2 phase, from their analysis of the observed cw-NMR spectrum and spin dynamic equations (OCF Equations). The OCF Eqs. have two modes of cw-NMR, the upper mode, ω_+, and the lower mode, ω_-,

$$\omega_\pm^2 = \frac{1}{2}\left\{\omega_L^2 + \Omega_0^2 \pm \right.$$

$$\left. \pm \sqrt{(\omega_L^2 - \Omega_0^2)^2 + 4\omega_L^2\Omega_0^2\cos^2\theta}\right\}, \qquad (1)$$

where $\omega_L = \gamma H_0$ is the Larmor frequency; γ is the gyromagnetic ratio; H_0 is the external magnetic field; Ω_0 is the zero-field antiferromagnetic resonance frequency; θ is the angle between I and H_0; and I is the anisotropy axis along the (100) axis of the bcc lattice. Further support for such an identification of the U2D2 structure was given by neutron scattering experiment [2]. Sasaki et al. [3] studied spin relaxation mechanisms by measuring the line width of cw-NMR and clarified the relaxation mechanisms due to the three- and four-magnon processes. Their result agreed well with Ohmi and Tsubota's theory [4]. A negative frequency shift of cw-NMR which was not explained by OCF Eqs. was observed [3,5] and recently explained as a thermal fluctuation effect by Fomin and Ohmi [6].

Kusumoto et al. [7] performed pulsed NMR on this system and found an anomalous free induction decay (FID) signal when the spin system was tipped away from the equilibrium configuration. There has been much theoretical work on the spin dynamics in the U2D2 in the non-linear regime. In high fields, where $\omega_L >> \Omega_0$, Namaizawa [8] calculated the tipping-angle-dependent frequency shift and also predicted multiple spin echoes similar to that observed in superfluid ^{3}He [9]. Tsubota [10] treated Kusumoto et al.'s results theoretically and attributed the anomalous FID to an onset of chaotic motion of the spin system in the low field region where $\omega_L \cong \Omega_0$. Ohmi et al. [11] and Shopova [12] predicted the instability of the uniform precession with respect to a long-wave spin wave mode in the case of samples with $\cos^2\theta > 1/5$.

In this paper, we report studies of the spin dynamics in the non-linear regime in a single crystal sample with a single magnetic domain in high fields where $\omega_L >> \Omega_0$. Preliminary results of non-linear spin dynamics have been published elsewhere [13,14]. We found a rapid decay of the FID signal under certain conditions and attributed the rapid decay to the onset of an instability of uniform motion of spins. We propose a mechanism for the instability of uniform precession and analyze the results. In the region of stable spin motion we observed the tipping-angle-dependent frequency shift and multiple spin echoes

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which were in good agreement with Namaizawa's theory.

2. Experimental methods

A single-crystal sample with a single magnetic domain was grown from superfluid ^{3}He at a temperature of 0.4 mK. The method used to grow the single-domain crystal and the sample cell were described in detail in Ref. 13. Our samples were at melting pressure. We grew the crystal in a long cylinder (the inner diameter was 1 mm and the length was 12 mm). The bottom part of the crystal had three domains but the upper part of the crystal usually contained one domain where our pulsed-NMR was performed.

The non-linear spin dynamics can be studied in high fields, where $\omega_L >> \Omega_0$, and we can tip the spin system from the equilibrium configuration in spite of the tipping-angle-dependent frequency shift [8]. Our NMR frequencies were 10, 7 and 2.4 MHz. A single-domain sample is necessary for pulsed NMR at high fields, otherwise the FID signals from each domain would be mixed up. It is very important to observe pulsed NMR by using a homogeneous rf-field H_1 since the precession frequency of the magnetization depends on the tipping angle so that non-uniformity of H_1 would cause a dephasing of the FID. A large saddle type transmitter coil for H_1-field around a small receiver coil was used. We measured the inhomogeneity of the H_1-field and confirmed the dephasing effect of the H_1-inhomogeneity to be about 10 msec at 2.4 MHz, i.e. negligible in this experiment. We determined $\cos^2\theta$ and $\Omega_0(T)$ by taking cw-NMR for the bottom part of the sample at 2.4 MHz where the effect of the negative shift is small and the observed signal can be fit to Eq. (1). In this method of crystal growth, the crystal grown in the upper part of the sample cell had a small value of $\cos^2\theta$.

3. Experimental results and analysis

3.1. Typical FID signal

We directly observed the FID signal with a high speed digitizer without detecting the signal. Typical data of the FID at 7 MHz, $T/T_N = 0.5$ and the tipping angle $\beta_p = 44°$ with $\cos^2\theta = 0.005$ is shown in Fig. 1,*a*, where T_N is the nuclear ordering temperature at zero field, 0.93 mK in this paper. The beat frequency in the figure was $f - Nf_S$, where f is the precession frequency of the FID and f_S is the sampling frequency of the digitizer and N is some integer number. From this data we extracted two quantities, the FID amplitude, $I(t)$, and frequency, $\Delta f(t)$, measured from the Larmor frequency f_L, which are shown in Figs. 1,*b* and *c*, respectively. The FID frequency $\Delta f(t)$ clearly changes in time. The FID signal suddenly begins to decay at 0.5 msec. We fitted $I(t)$ to the following equation:

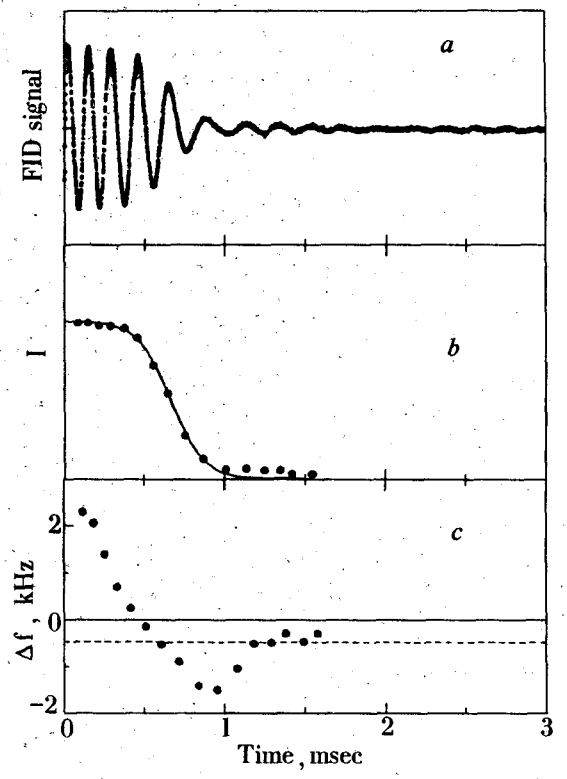

Fig. 1. Typical free induction signal. (*a*) A free induction decay (FID) signal at $f_L = 7$ MHz, $T = 0.5T_N$ and $\cos^2\theta = 0.005$ for $\beta_p = 44°$; (*b*) the FID amplitude $I(t)$; (*c*) change of the precession frequency counted from the Larmor frequency, $\Delta f(t)$. The dashed line is the cw-NMR resonance frequency.

$$I(t) = \frac{I_0}{1 + A\exp(\Gamma t)} \,. \qquad (2)$$

The solid line in Fig. 1,*b* is the fit to Eq. (2) with the fitting parameters A and Γ. The decay behavior of the FID signal was very similar to that observed in superfluid ^{3}He–A. The OCF Eqs. are identical to the Leggett equations [16] for superfluid ^{3}He–A. Equation (2) was introduced in Ref. 15 and 17 in order to explain the instability of the uniform precession in ^{3}He–A; because of the similarity of the equations of spin dynamics in U2D2 and ^{3}He–A, we adapted Eq. (2) for our analysis of $I(t)$. We derived Eq. (2) from our instability model proposed in Sec. 4. We attribute the rapid decay of the FID signal to the onset of instability of the uniform motion of the spin system. The instability occurs under certain conditions,

such as low temperatures, low fields, and large tipping angles. When instability occurs, the fit of $I(t)$ to Eq. (2) is good. Although the fit of $I(t)$ is not good when the FID does not show any unstable behavior, we still use Eq. (2) in order to trace the change of the decay behavior under various conditions.

3.2. *Rapid-decay rate* Γ *and the onset of instability of the uniform precession*

We made a systematic study of the variation of Γ with temperature and tipping angle β_p at 10, 7 and 2.4 MHz for various samples with different $\cos^2\theta$. Figures 2,*a-c* show the temperature dependence of Γ for various β_p at 10 MHz and $\cos^2\theta = 0.000$ (*a*), 7 MHz and $\cos^2\theta = 0.108$ (*b*), and 2.4 MHz and $\cos^2\theta = 0.062$ (*c*). The Γ at 10 MHz does not change much as a function of temperature for both $\beta_p = 30°$ and 90° even though Γ for $\beta_p = 90°$ is larger than that for $\beta_p = 30°$. The lower value of Γ indicates that the precession is more stable. In the case of 7 MHz, Γ at a fixed β_p was essentially constant at higher temperatures, but at a certain temperature T_{onset} it increased rapidly and then decreased slightly at lower temperatures. We identify T_{onset} as an onset temperature of the instability. As β_p increased, Γ increased. Thus, the precession became more unstable with increasing β_p. The onset temperature T_{onset} also depended strongly on β_p. At 2.4 MHz, Γ increased (note the change of the scale for the vertical axis) and T_{onset} shifted to higher temperatures, compared with that for 7 MHz. The T_{onset} for $\beta_p = 60°$ was around the nuclear-ordering temperature T_N. From these data we conclude that the uniform precession of the FID becomes unstable under conditions such as lower temperatures, lower fields, and larger β_p. We could not make a systematic study of the $\cos^2\theta$-dependence of the instability, but the instability occured more easily for samples with larger values of $\cos^2\theta$.

3.3. *Spin dynamics in the stable region*

In Sec. 3.2 we have separated a region in the parameter space (ω_L, β_p, T and $\cos^2\theta$), where uniform spin precession was stable, and an other region, where uniform precession became unstable. Here we describe briefly what happens to the spin dynamics in the stable region. Firstly, in the stable region the FID signal stays for a few msec and also the functional form of $I(t)$ does not match well with Eq. (2). The precession frequency $\Delta f(t)$ does not change much in time for small β_p and changes slowly even for large

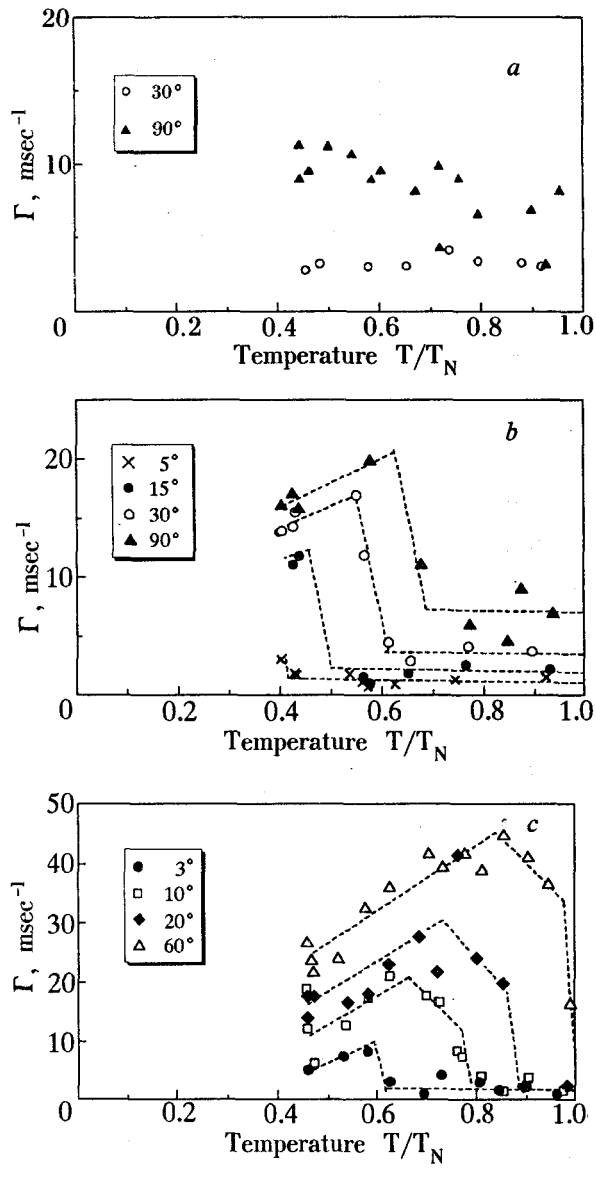

Fig. 2. The temperature dependence of the decay constant Γ for various tipping angles β_p. (*a*) f_L =10 MHz, $\cos^2\theta = 0.000$; (*b*) f_L = 7 MHz, $\cos^2\theta = 0.108$; (*c*) f_L = 2.4 MHz, $\cos^2\theta = 0.062$. The dashed lines are a guide for the eye. We define T_{onset} as a temperature where Γ increases rapidly.

β_p. Therefore, we can easily extrapolate the precession frequency to right after the rf-pulse. We denoted the value $\Delta f(t \to 0)$ as the tipping-angle-dependent frequency shift $\Delta f(\beta_p)$ and measured it as a function of β_p. In Fig. 3, $\Delta\omega(\beta_p) = 2\pi\Delta f(\beta_p)$, normalized to Ω_0^2/ω_L, is plotted against β_p for 10 MHz and 7 MHz, and $T/T_N = 0.5$; $\cos^2\theta = 0.005$. Some of the data at 7 MHz were taken in the unstable region. The tip-

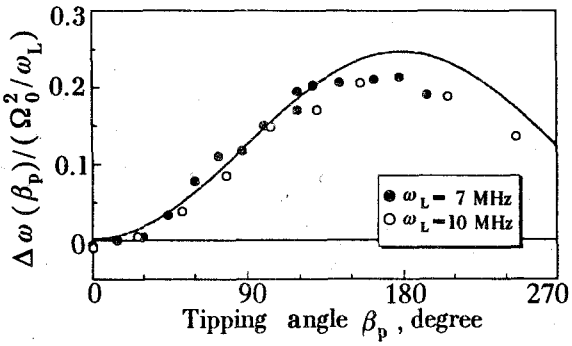

Fig. 3. The tipping-angle-dependent frequency shift. Solid circles are taken at 7 MHz and open circles at 10 MHz; $T = 0.5T_N$, $\cos^2\theta = 0.005$. The curve is Namaizawa's theory.

ping-angle-dependent $\Delta f(\beta_p)$ is compared with Namaizawa's theory given by

$$\Delta\omega_N(\beta_p) = \left(\frac{\Omega_0^2}{\omega_L}\right)\frac{1}{8}\left\{(5\cos^2\theta - 1)\cos\beta_p + \sin^2\theta\right\}. \quad (3)$$

The curve in Fig. 3 is Namaizawa's theoretical curve. The observed $\Delta f(\beta_p)$ around $\beta_p = 180°$ is smaller than the theoretical one, partly because the actual tipping angle may be different from β_p especially around 180° due to the tipping-angle-dependent frequency effect. There is an obvious small discrepancy for small β_p, since $\Delta f(\beta_p)$ is negative shifted for this sample and the OCF Eqs. predict no negative shift for any value of β_p for our samples with a small value of $\cos^2\theta$. Therefore, agreement with the theory is good except for the small discrepancy of the negative shift at small β_p. Namaizawa also predicted that after two-rf-pulses, we would observe many echoes, called multiple spin echoes (MSE). The MSE are generally formed after two rf pulses when the precession frequency depends on the z-component of the magnetization. In the U2D2 solid, $\Delta f(\beta_p) = \Delta f(M_z/M)$ depends on M_z, where M is the total magnetization and M_z is its z-component. In Fig. 4,*a–c*, the MSE after two 40° rf-pulses observed at 10 MHz for $\cos^2\theta = 0.00$ under a static field gradient dH_0/dz of about 10 G/cm are shown for $T/T_N =$ = 0.6, 0.8 and 0.9. We observed the MSE in the same time domain where the FID signal was studied. Therefore the instability of the uniform precession is not caused by the gradient of the applied magnetic field. The MSE are very sensitive to the dipole torque and a detailed analysis of the MSE will be reported elsewhere. The change of the MSE with temperature is due to the temperature dependence of $\Omega_0(T)$. The tipping-angle-dependent frequency shift and the observation of the MSE are all in good agreement with Namaizawa's theory [8] which was developed from OCF Eqs. for the case of high fields. Therefore we conclude that the spin motion in the stable region can be well described even in the non-linear regime by the OCF Eqs.

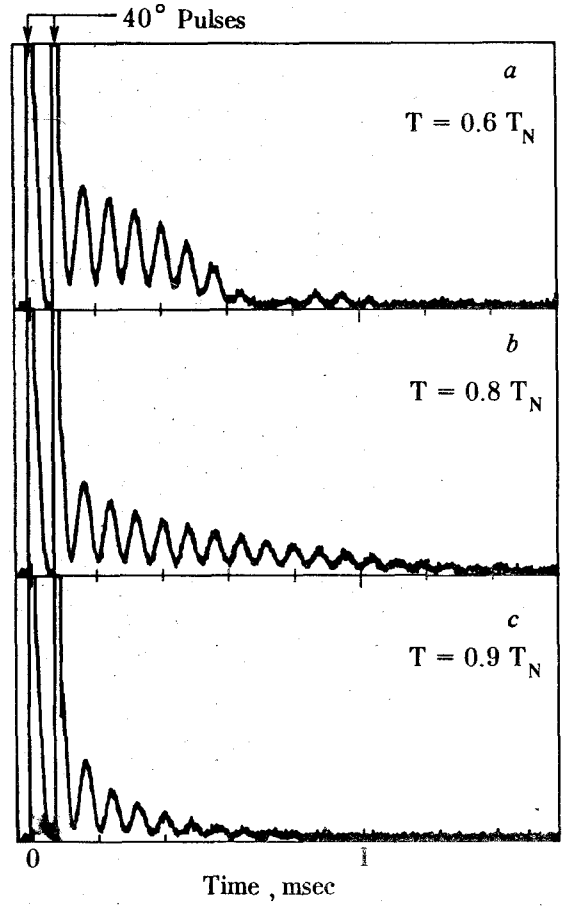

Fig. 4. Typical multiple spin echoes. The multiple spin echoes are taken at 10 MHz at various temperatures for $\cos^2\theta = 0.00$ after two 40° rf pulses.

3.4. Spin dynamics in the unstable region

In this Section we summarize some of the characteristic behavior of the FID in the unstable region. When the instability occurred during the FID, the precession frequency $\Delta f(t)$ of the FID changed rapidly in time. Figure 5 shows $\Delta\omega(t) = 2\pi\Delta f(t)$ at various temperatures at 2.4 MHz and $\beta_p = 60°$ for $\cos^2\theta =$ = 0.062. The vertical axis is $\Delta\omega(t)$ normalized by the Namaizawa shift $\Delta\omega_N$ (60°). The sample is the same as in Fig. 2,*c*. The change of $\Delta f(t)$ is rather rapid, but it seems likely that $\Delta f(t)$ starts from the Namaizawa shift at $t = 0$ and then goes to a large negative value, amounting to about 20 kHz at the lowest temperature of $T = 0.47T_N$, as large as the dipole shift, and in the

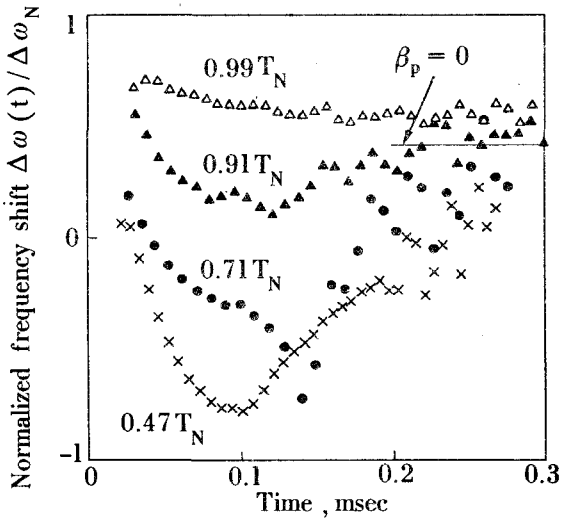

Fig. 5. Time evolution of the precession frequency $\Delta f(t)$ in the unstable region at various temperatures at 2.4 MHz and $\beta_p = 60°$ for $\cos^2\theta = 0.062$ (the same sample as in Fig. 2,*c*).The line indicated by $\beta_p = 0$ is the cw-NMR frequency normalized by the Namaizawa shift $\Delta\omega_N$.

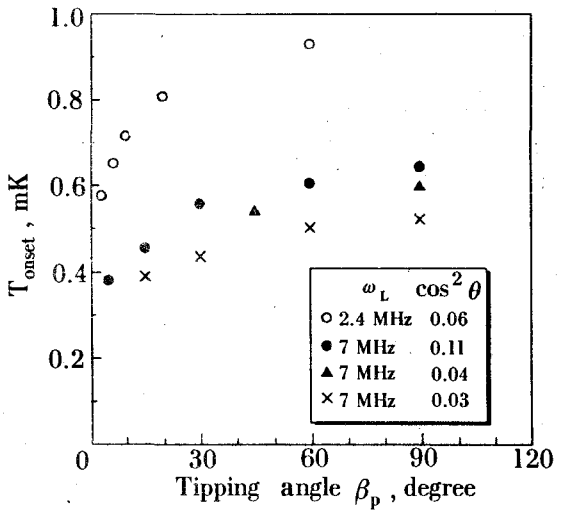

Fig. 7. Tipping-angle-dependence of the onset temperature at 2.4 MHz and 7 MHz. Data for various samples are shown at 7 MHz.

end it decays to the cw-NMR frequency indicated by the $\beta_p = 0$ line in the Fig. 5. As the temperature increases, the negative shift becomes smaller and at the highest temperature of $T = 0.99T_N$, where the instability is almost depressed, $\Delta f(t)$ decays smoothly from the Namaizawa shift to the cw-NMR frequency. We numerically solved the OCF Eqs. by introducing a phenomenological relaxation term such as the one in Ref. 10 to find that $\Delta f(t)$ changed smoothly from the Namaizawa to cw-NMR shift and never showed a large negative shift during the relaxation process within the OCF Eqs.

When the instability occurred at $T = 0.5T_N$ for 7 MHz and $\cos^2\theta = 0.005$, we investigated the β_p-dependence of Γ. The values of Γ are plotted against β_p in Fig. 6. It is clear that Γ increases as β_p increases

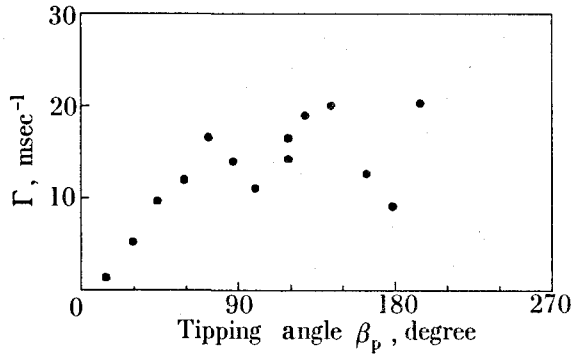

Fig. 6. The rapid decay rate against tipping angle β_p. Data are taken at 7 MHz and $T = 0.5T_N$ for $\cos^2\theta = 0.005$.

and may have some minimum around 180°. In Fig. 7, the onset temperatures T_{onset} are plotted against β_p for 2.4 MHz and 7 MHz. For 7 MHz, we did a systematic study of $\cos^2\theta$-dependence of T_{onset}. With our method of crystal growth, the upper part of the sample grown usually had a small $\cos^2\theta$ value and thus the range of $\cos^2\theta$ investigated is limited. However, as $\cos^2\theta$ increases, the instability is enhanced and T_{onset} increases systematically. The results of Figs. 6 and 7 will be discussed and compared with our model of instability.

4. Discussion

4.1.Comparison with instability theories

In this Section we discuss the existing theories of the instability for the uniform spin precession. So far two mechanisms have been proposed. Tsubota [10] found chaos in the uniform motion by solving numerically the OCF Eqs. in the low field limit where the non-linear dipole torque is comparable with the Zeeman torque. However, when we made pulsed NMR at high fields, Tsubota's chaotic behavior was completely suppressed and thus the onset of chaotic motion should not account for our instability.

Ohmi et al. [11] and Shopova [12] calculated the instability of the uniform precession when the spin system is tipped away from the equilibrium condition. The dispersion of the spin wave mode for $\omega_k^+(\beta_p)$ for small values of k and a finite tipping angle β_p is approximately given by

$$\left(\omega_k^+(k) - iDk^2\right)^2 = -\frac{1}{16}\left(\frac{\Omega_0}{\omega_L}\right)^2 \times$$

$$\times (5\cos^2\theta - 1)(1 - \cos\beta_p)(3 - \cos\beta_p)c^2k^2 \quad (4)$$

where D is the spin diffusion constant and c is the spin wave velocity. If $(5\cos^2\theta - 1) > 0$, $\omega_k^+(k)$ becomes purely imaginary and uniform precession with $k = 0$ is unstable to the creation of $k \neq 0$ spin wave modes, but if $(5\cos^2\theta - 1) < 1$, the uniform mode is stable. This instability was observed [15] experimentally in superfluid ^{3}He–A and was theoretically explained by a similar model [17]. All the samples we have investigated satisfy the condition $(5\cos^2\theta - 1) < 0$ and thus the uniform mode should be stable. Our observation of the onset of instability clearly disagrees with the theory.

4.2. New instability model due to the self-induced emission mechanism

According to the results on the cw-NMR line widths, the relaxation mechanisms are explained by the three- and four-magnon processes. In high fields and for samples with small values of $\cos^2\theta$, the relaxation rate of the upper mode is dominated by three-magnon processes. Figure 8 shows the dispersion curves for the upper and lower magnons. The spin wave dispersions curves for the upper mode, $\omega_+(k)$, and for the lower mode, $\omega_-(k)$, are given by

$$\omega_\pm^2(k) = \omega_\pm^2 + c^2k^2, \quad (5)$$

where $\omega_\pm$ is given by Eq. (1). In the three-magnon process, the upper-mode magnon with $k = 0$ shown as the open circle decays into two lower-mode magnons shown as solid circles. Due to the energy and momentum conservation laws, $\omega_-(k) = \omega_+/2$ and the momentum direction of the two magnons should be opposite. When the upper-mode magnon decays via the three-magnon process, particular lower-mode magnons are injected during the decay process. According to Eq. (15) in Ref. 4, the three-magnon relaxation rate $\Gamma^{(3)}$ for the upper mode is proportional to the occupation density of the above particular magnons, $n(\omega_-(k)) \equiv n_k$, and is related to the stimulated emission. The lower-mode magnon has a finite lifetime but when the upper mode magnons decay faster than the decay rate of the lower magnon, n_k increases drastically. The $\Gamma^{(3)}$ increases proportionally to n_k, and thus the upper mode with $k = 0$ (the uniform precession) becomes unstable. Similar phenomena were studied in electronic ferromagnets [18]. Onset of an instability, sometimes called Suhl's instability, was observed, and similar arguments were used for the explanation of the instability. We modify Ohmi and Tsubota's theory [4] by including explicitly the effect of the magnons decayed from the upper into the lower mode. Equation (15) of Ref. 4 can be written as

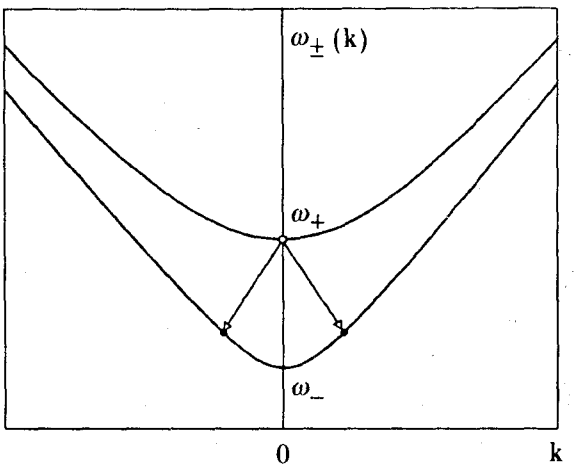

Fig. 8. Schematic diagram of the three-magnon process. The solid lines are the dispersion curves for the upper-mode magnon ($\omega_+(k)$) and the lower-mode magnon ($\omega_-(k)$). An upper-mode magnon with $k = 0$ decays into two lower-magnons with $k \neq 0$.

$$\dot{N}_0 = -\Gamma' n_k N_0 \quad (6)$$

where

$$\Gamma' = \frac{\gamma^2\hbar}{8\pi^2\chi c^3}\omega_L^3\left(\frac{\Omega_0}{\omega_L}\right)^4 \cos^2\theta\,(1 - \cos^2\theta)$$

for $\omega_L >> \Omega_0$. We denote the density of the upper-mode magnons introduced by the rf-pulse by N_0; n_k is the occupation number of the lower-mode magnon defined in Eq. (15) in Ref. 4. By using the energy conservation relation, N_0 is related to the change of the lower mode magnon decanting from the upper mode, $(\dot{n}_k)_{(+\to-)}$, as follows

$$\hbar\omega_L\dot{N}_0 = \Delta\dot{M}_z H = -G\hbar\omega_L(\dot{n}_k)_{(+\to-)}, \quad (7)$$

where we introduced the line width Γ_k for lower-mode magnons and $G = (1/2)\Gamma_k D(\omega_L/2)$; $D(\omega)$ is the density of states for lower magnons whose dispersion is given by Eq. (5). Combining Eqs. (6) and (7), we get

$$\dot{n}_k = (\dot{n}_k)_{(+\to-)} + (\dot{n}_k)_{\text{decay}} = \frac{\Gamma'}{G} n_k N_0 - \Gamma_k(n_k - \bar{n}_k), \quad (8)$$

where the last term is introduced phenomenologically and $\bar{n}_k$ is the value of n_k in thermal equilibrium. The steady-state solution for Eq. (8) is

$$n_k = \frac{\bar{n}_k}{1 - \Gamma' N_0/\Gamma_k G}. \quad (9)$$

When $\Gamma' N_0 / \Gamma_k G \to 1$, the quantity n_k diverges and the uniform precession becomes unstable. When we put all values for G, N_0 and Γ', we get the following condition for the onset of the instability.

$$\Gamma_k^2 = 2\cos^2\theta\,(1-\cos^2\theta)\left(\frac{\Omega_0}{\omega_L}\right)^4 \omega_L^2(1-\cos\beta_p)\,. \tag{10}$$

We can solve Eqs. (6) and (8) for N_0, neglecting the relaxation term in Eq. (8):

$$N_0(t) = \frac{N}{1 + A\exp(\Gamma' N t/G)}\,, \tag{11}$$

where $A = Gn_k(0)/N_0(0)$ is determined by the initial condition and $N = N_0(0) + Gn_k(0)$, where $N_0(0)$ and $n_k(0)$ are the initial conditions for $N_0(t)$ and $n_k(t)$. The above equation is the function to fit $I(t)$, with the decay rate Γ in Eq. (2) given by

$$\Gamma = \frac{\Gamma' N}{G} \cong \frac{\Gamma' N_0(0)}{G} \propto (1-\cos\beta_p)\,. \tag{12}$$

The data of Fig. 7 for T_{onset} are re-plotted in Fig. 9 against the quantity Γ_k^2 which is calculated from Eq. (10). The temperature dependence of Γ_k should be T^4. The difference between T_{onset} for 2.4 MHz and 7 MHz indicates that Γ_k would be k-dependent and Γ_k increases for larger values of k. The tipping-angle dependence of Γ shown in Fig. 6 agrees with Eq. (12) for small β_p, but the minimum of Γ around $\beta_p = 180°$ is not explained. All the instability data agree at least qualitatively with our model. Our argument is based upon the calculation of Ref. 4 which is only applicable to the relaxation for cw-NMR and should be re-examined further for a finite β_p. It has not been discussed what happens to the equation of motion of the spin after the instability sets in and during the time when we observe a large negative frequency shift in the rapid decay process.

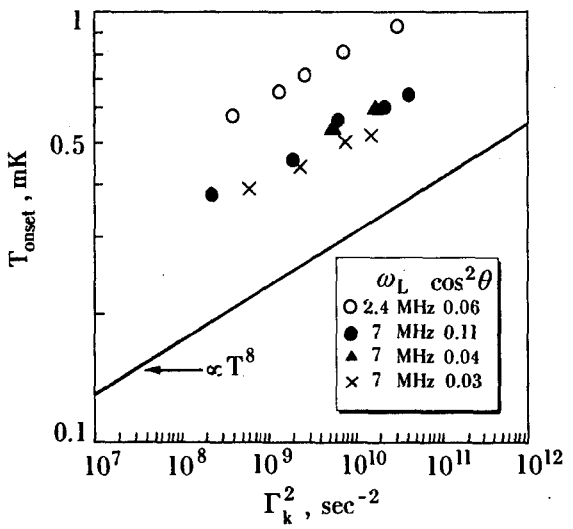

Fig. 9. Onset temperature T_{onset} vs. Γ_k^2. The Γ_k values are calculated from Eq. (10).

Summary

We studied spin dynamics in the U2D2 phase of solid ^{3}He in non-linear regime at high fields. We observed the onset of the instability of the uniform precession.

In the stable region, we observed a tipping-angle-dependent frequency shift and multiple spin echoes. These results agree well with Namaizawa's theory.

We investigated the condition for the onset of the instability. The uniform precession becomes unstable at lower temperatures, lower fields, larger tipping-angles and larger values of $\cos^2\theta$. The onset conditions agree well with our estimate for the instability criteria in our model. The observed decay rate when the instability was induced agrees qualitatively with our model.

Acknowledgment

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