Scientific session of the Division of General Physics and Astronomy of the Russian Academy of Sciences, in commemoration of the 80th birth anniversary of Academician A S Borovik-Romanov (29 March 2000)

A scientific session of the Division of General Physics and Astronomy of the Russian Academy of Sciences (RAS) was held at the P L Kapitsa Institute for Physical Problems, Moscow on March 29, 2000. The session was devoted to the 80th anniversary of academician A S Borovik-Romanov's birth. The following reports were presented at the session:

(1) **Ginzburg V L** (P N Lebedev Physical Institute, RAS, Moscow) "Superconductivity: The day before yesterday — yesterday — today — tomorrow";

(2) **Dmitriev V V** (P L Kapitsa Institute for Physical Problems, RAS, Moscow) "New NMR modes in superfluid ³He-B".

(3) **Smirnov A I** (P L Kapitsa Institute for Physical Problems, RAS, Moscow) "Magnetic resonance of intrinsic and extrinsic defects in a spin-Peierls magnet CuGeO₃".

An enlarged version of the first report is published in this issue on p. 573. Abridgments of the two other reports are given below.

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New NMR modes in superfluid ³He-B

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The superfluidity of ³He is related to Cooper pairing into a state with spin and orbital moment equal to unity. The presence of a spin in Cooper pairs and the complex form of the order parameter lead to a variety of dynamic magnetic properties of superfluid phases, and magnetic measurements (in particular, nuclear magnetic resonance) are actually the main methods of studying superfluid ³He. For magnetic methods to be used effectively, a clear understanding of spin dynamics in superfluid phases is required. Shortly after the superfluidity of ³He was discovered, Leggett [1] derived a set of equations that described the coupled motions of the magnetization **M** and the order parameter:

$$\dot{\mathbf{M}} = g\mathbf{M} \times \mathbf{H} + \mathbf{R}_{\mathrm{D}} \,, \tag{1}$$

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$$\dot{\mathbf{d}} = \mathbf{d} \times g\left(\mathbf{H} - \frac{\mathbf{M}}{\chi}\right),$$
 (2)

where $\mathbf{d} = \mathbf{d}(\mathbf{k})$ is the order parameter in the vector representation, \mathbf{k} is the unit vector in orbital space, χ is the susceptibility, g is the gyromagnetic ratio, \mathbf{H} is the applied magnetic field, and \mathbf{R}_D is the dipole moment. In the B phase of superfluid ³He, which is the main object of this paper, $\mathbf{d}(\mathbf{k}) = \hat{\mathbf{R}}\mathbf{k}$, and $\hat{\mathbf{R}}$ is the matrix of rotation of the spin space (this matrix can be parametrized by the rotation angle θ about an axis \mathbf{n}). The dipole moment is due to the energy $F_D \propto \Omega_B^2 (2 \operatorname{Tr} \hat{\mathbf{R}} - 1)^2$ related to the dipole–dipole interaction of ³He nuclei and dependent on the mutual orientation of the spin and orbital spaces (here, Ω_B is the so-called Leggett frequency characteristic of the strength of dipole interaction). The dipole energy is minimum at $\theta =$ $\theta_0 \approx 104^\circ$.

It is the dipole energy that to a large extent determines stable spin-orbit configurations; therefore, to find the possible dynamic spin states, the method of minimization of the dipole energy proved to be quite efficient [2]. The orientation of spin and orbital spaces in this case can conveniently be characterized by unit vectors s and l (s is the unit vector in the magnetization direction and $\mathbf{l} = \hat{\mathbf{R}}\mathbf{s}$ is the unit vector of the orbital moment indicating the B-phase gap anisotropy direction in a magnetic field). For precessing states, the matrix $\hat{\mathbf{R}}$ and, hence, F_{D} are time-dependent. However, if $\Omega_B \ll gH$, this energy can be averaged over the periods of fast motions [3]; for such motions, it is suitable to choose the magnetization motion with a frequency $\omega_{\rm M}$, which is close to the Larmor frequency $\omega_{\rm L} = gH$, and the precession of **d** about the instantaneous direction of **M** with a frequency ω_d , which, according to Eqn (2), is equal to gM/χ . The results of averaging depend on the relationship between $\omega_{\rm L}$ and ω_d , and the minimum energy corresponds to the so-called 'resonance' case, when $\omega_{\rm M} = \omega_d$ and $M \simeq \chi H.$

Shortly after the discovery of superfluidity of ³He, a number of NMR modes were found and studied, such as (a) the NMR mode related to the spatial distribution (texture) of the order parameter; (b) the Brinkman–Smith mode; (c) longitudinal vibrations; and (d) the wall-pinned mode [4, 5]. All these modes correspond to the 'resonance' case or small vibrations near such special ('preferred') spin–orbit configurations. Volovik [2] predicted the existence of some more (still experimentally undetected) NMR modes corresponding to the precession of the magnetization with an equilibrium magnitude).

In experiments using transverse NMR, the first two of the above-mentioned modes are usually observed. In the unperturbed ³He-B, the order-parameter texture is determined by the combined orienting effect of the magnetic field and the walls of the experimental cell (I is perpendicular to the surface at the walls and is parallel to $\mathbf{I} \parallel \mathbf{H}$ far from the walls). For small amplitudes of the radiofrequency (rf) field (standard continuous NMR mode), the order-parameter texture remains unaltered on the average, and mode (a) is excited, which corresponds to small oscillations of the magnetization and of the order parameter relative to the equilibrium configuration. The absorption line in this case exhibits a peak at the Larmor frequency and a long 'tail' in the frequency range $\omega_{\rm L} < \omega < \omega_{\rm L} + (2\Omega_B^2/5\omega_{\rm L})$. The Brinkman-Smith mode (b) corresponds to the simultaneous precession of **M** and **n** at the Larmor frequency (if the angle of the deviation of magnetization β is less than θ_0) for a fixed value of $\theta = \theta_0$ and is observed, as a rule, when the angles of the deviation of magnetization are sufficiently large and the orienting effect of the walls can be ignored. In this case, we have $\mathbf{I} \parallel \mathbf{H}$ over the whole volume of the cell. If $\beta > \theta_0$, the character of precession remains unaltered, but it occurs at a frequency that is shifted from the Larmor frequency by an amount $-(4\Omega_B^2/5\omega_L)(1+4\cos\beta)$.

It was shown in Ref. [6] that there are also other preferred cases for which the dipole energy is minimum and which are also called 'resonance' cases. In these cases, the magnetization is equal to $M_0/2$ or $2M_0$ (here, $M_0 = \chi H$) and, correspondingly, the order parameter moves half as fast or two times faster than M does. Depending on the conditions (e.g., on the shift from the Larmor frequency in the presence of an rf field), the minimum energy may be associated with different spin-orbit configurations. One of these states (with $M = M_0/2$) was recently obtained and studied experimentally at the Institute for Physical Problems and at Helsinki Technical University [7]. In these experiments, cells were used in the form of a cylinder with characteristic dimensions 4-7 mm closed at both ends; with the rest of the experimental device, the cells were connected through a long narrow channel.

The experiments were carried out at a fixed pumping frequency $\omega_{\rm rf}$ in the transverse continuous NMR mode at large amplitudes of the rf field $H_{\rm rf} \ge 0.01$ Oe (note that $\omega_{\rm M} = \omega_{\rm rf}$). At low temperatures, the above-mentioned texture-related NMR mode (a) was observed. However, it was found that at temperatures $T \ge 0.98T_{\rm c}$ ($T_{\rm c}$ is the temperature of transition of ³He into the superfluid state, which changes from 0.93 to 1.93 mK in the pressure range of 0-12 bar that was used in the experiments), the line shape changed qualitatively.

Figure 1 displays experimental curves of the dispersion signal recorded as the field passed through the line during slow warming of the ³He sample. Note that the features seen in the signals are well reproducible and exhibit hysteresis in most cases. Similar jumplike features are also present in the corresponding absorption lines; they also arise as the line is scanned in the opposite direction.

In order to understand the nature of the observed features, we performed a numerical simulation of the experiment. We solved the complete set of Leggett equations in the variables \mathbf{M} , \mathbf{n} , and θ for a spatially uniform case and simulated the passage through an NMR line with parameters close to those characteristic of real experiments. It turned out that the numerical experiment satisfactorily

Figure 1. Dispersion signals obtained when slowly warming the samples to the A-phase state. States with a nonequilibrium magnitude of the magnetization are marked with letter abbreviations. The dashed lines correspond to the zero level for each curve. The absorption is much smaller than the dispersion and is not shown. $\omega_{\rm rf}/2\pi = 461$ kHz. The arrow indicates the direction of the variation of the scanning field.

reproduced not only most features found in the real experiment, but even the succession in which they appear as the temperature increased.

The results obtained indicated that these features were related to transitions between various precession states and allowed us to perform their preliminary identification. It was found that several precession states with $M = M_0/2$ (halfmagnetization, or HM states) differing in the orientation and the character of motion of l and of the magnetization can arise in the presence of an rf field; some of them are marked in Fig. 1 by corresponding symbols, e.g., HM1, HM2, HM3). In addition, it proved that the state denoted ZM (zeromagnetization) corresponds to a precession with $\mathbf{l} \perp \mathbf{H}$ with a magnetization that is much smaller than the equilibrium value $(M < 0.3M_0)$. The possibility of existence of such an NMR mode was first considered in Ref. [8], but a later analysis showed that the ZM state observed in both experiment and upon numerical simulation did not correspond to the state predicted in Ref. [8], where the case of precession with $\mathbf{I} \parallel \mathbf{H}$ was considered. Note also that, in total, we observed six modifications of the HM state in the abovedescribed and later experiments [9, 10]; four of them cannot be described analytically even if, when minimizing the energy of the system, one takes into account the presence of an rf field and the shift from the Larmor frequency.



For the complete identification of the states revealed, we performed an experiment on the direct measurement of the magnetization magnitude. To this end, the sweeping of the external magnetic field H was stopped in a certain state of precession. Then, the rf field that maintained this field was turned off and, simultaneously, a powerful rf pulse that deviated the magnetization by an angle of 90° was applied to the sample, and the signal of free induction was recorded. This experiment was performed repeatedly with various phases of the rf pulse. Comparing the initial amplitude of the signals obtained with the initial amplitude of the signal of free induction recorded after the application of a similar pulse in the unperturbed normal ³He, we may judge the magnitude of magnetization and the angle of its deviation from the equilibrium position in a given state. It proved that in all the states marked as HM, we have $M = 0.50 \pm 0.03 M_0$ (Fig. 2). The angle of deviation of the magnetization is usually small (less than 10°) except for two (of the six revealed) HM states, where it becomes large for small shifts from the Larmor frequency. The magnitude of the magnetization in the ZM state varied from $0.3M_0$ to an immeasurable magnitude $(M < 0.02M_0)$ depending on the amount of the shift from the Larmor frequency and on the temperature, which agrees with the results of our numerical simulation. The simulation results permit us to understand the mechanism of the formation of states with a nonequilibrium magnetization magnitude: they arise upon the passage through the resonance, when the angle β grows and becomes close to 180°; then, the magnetization relaxes along a straight line M_{\perp} toward the state ZM or HM. At temperatures close to T_c , such a scenario is easily realized for a sufficiently large amplitude of the rf field. At lower temperatures, it is virtually impossible to deflect the magnetization to an angle markedly exceeding 104° because of the arising dipole frequency shift.

As follows from Eqn (2), the order parameter in the absence of an rf field precesses around the instantaneous direction of **M** with a frequency $\omega_d = gM/\chi = (1/2)\omega_L$. This leads to oscillations of \mathbf{R}_D with the same frequency; correspondingly, a component oscillating with a frequency $(1/2)\omega_L$ should appear in the motion of **M**. It can be shown that its amplitude is a fraction of M_0 of the order of $(\Omega_B/\omega_L)^2$. Such small magnetization oscillations in HM states were considered theoretically in Ref. [11], where it was found that, in the presence of an rf field, harmonics with frequencies $(1/2)\omega_M$, $(3/2)\omega_M$, $(5/2)\omega_M$ and ω_M , $2\omega_M$, $3\omega_M$ appear in the motions of all three components of **M**.

Note that the existence of harmonics at fractional frequencies is a very unusual feature for NMR experiments. Such fractional harmonics have indeed been recently revealed in experiment [10]. The experiment was performed as follows. An additional NMR coil with its axis directed along the applied dc magnetic field was wound around a cell with ³He; this coil served a detector of fractional harmonics. The amplitude of the harmonics was approximately 10⁵ times less than the signal at the fundamental frequency; in order to enhance the sensitivity, we used a cold NMR circuit and the coil itself was made of a superconductor. The resonance frequency of the circuit was chosen to be 1/2 or 3/2 of the rf pumping frequency. The HM states were formed by the transverse continuous NMR technique in the same way as was described above. Simultaneously with the passage of the resonance line, the signal at the chosen fractional frequency



Figure 2. Dispersion signal including the HM1 state. The inset shows the amplitudes of the signals of free induction from the HM1 state and normal ³He obtained after turning off the continuous pumping and the application of a 90° deviating rf pulse in the field marked by an arrow. The spike in the beginning portion of the signals of induction is related to an overdrive of the receiving system due to the deviating pulse. $\omega_{\rm rf}/2\pi = 334$ kHz; $T = 0.993T_{\rm c}$.

was recorded from the cold circuit. Figure 3 shows the dispersion signal (lower curve) obtained by the line passage in the mode of increasing of the external magnetic field and the total signal at the halved frequency simultaneously recorded from the cold circuit (upper curve). It is seen that, in the region of existence of the HM6 state, a noticeable signal at the halved frequency arises (in another experiment, a signal at frequencies multiple to 3/2 of the pumping frequency was also observed). Note that harmonics at fractional frequencies were also detected for the HM3 state. At the same time, no harmonics were revealed in other HM states. This is likely to be explained by the fact that, according to the numerical simulation, the order parameter moves in these states with a frequency that differs noticeably (by $\sim 0.2-2$ kHz) from $(1/2)\omega_{\rm rf}$ and this difference changes depending on the frequency shift. Because of the residual nonuniformity of the external magnetic field $[(2-5) \times 10^{-5} \text{ in given experiments}]$, these oscillations in various parts of the experimental cell should rapidly become misphased after the formation of the states.

Thus, as a result of the above experiments, a number of new NMR modes corresponding to stable precession states were revealed, with a magnetization magnitude substantially differing from the equilibrium value. Further experiments [9] showed that, once formed, these states can be cooled to significantly lower temperatures (at least to $0.7T_c$). In this connection, it is of interest to study HM states at even lower temperatures, where they should manifest phase stiffness, i.e., the free precession in an HM state should be spatially homogeneous even in a nonuniform magnetic field [7]. Such phase stiffness is known for the Brinkman–Smith mode. It is connected with the flowing of superfluid spin currents,

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Magnetic resonance of intrinsic and extrinsic defects in a spin-Peierls magnet CuGeO₃

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1. Introduction

A spin-Peierls transition is a brilliant phenomenon in the physics of low-dimensional magnets. This transition was revealed in crystals containing chains of magnetic ions with spin S = 1/2 coupled through an antiferromagnetic exchange interaction. The interchain interaction should be much weaker than the intrachain one. The phase transition into the spin-Peierls state consists in the dimerization of magnetic atoms into chains; in this process, the spins are grouped to form pairs in which the ion separation is shorter than between ions of neighboring pairs. The exchange interaction becomes alternating, i.e., the exchange integral alternately takes on the values $J(1 + \delta)$ and $J(1 - \delta)$. This transition is accompanied by a gain in the exchange energy that exceeds the loss in the elastic energy of the crystal [1]. The rearrangement of the lattice turns out to be correlated in all three dimensions, i.e., the dimers are located on an ordered sublattice. Below the point of the above transition, a so-called spin energy gap appears because of the alternation of the exchange interaction; this gap separates the ground singlet state from the spectrum of triplet excitations. Thus, the crystal becomes nonmagnetic at low temperatures; below the transition point, the susceptibility should exponentially vanish.

In spite of the great variety of quasi-one-dimensional magnetic structures, the spin-Peierls transition has been revealed only in a few organic compounds and one inorganic compound. The first and so far the only inorganic substance that exhibits the spin-Peierls transition is CuGeO₃ [2]. This compound can be obtained in the form of perfect single crystals, which permits one to perform exhaustive crystallographic and magnetic investigations, including elastic and inelastic neutron scattering. In addition, this compound allows substitution of nonmagnetic ions such as Zn^{2+} , Mg^{2+} or magnetic ions such as Ni²⁺ for the magnetic Cu²⁺ ions.

The main parameters that determine the magnetic properties of CuGeO₃ are as follows (see, e.g., Ref. [3]): the exchange integral inside the chains is $J_c = 10.2$ meV; the spin-Peierls temperature is $T_{SP} = 14.5$ K; dimerization in the chains of Cu ions located along the c axis of the orthorhombic crystal leads to the alternation of the exchange interaction with a parameter $\delta \approx 0.04$; and the spin energy gap at zero temperature is $\Delta \approx 2$ meV. The magnetic structure of CuGeO₃ is not ideally one-dimensional; the interchain exchange integrals are $J_b = 0.1J_c$ and $J_a = -0.01J_c$ [4]. It has also been supposed that there is an exchange integral approximately equal to $0.36J_c$ [5].

If the lattice of CuGeO₃ were rigid, this crystal would suffer a transition into an antiferromagnetic state at a Néel temperature $T_N \sim (J_c J_b)^{1/2} \sim 10$ K [6]. However, the spin-Peierls state proves to be energetically more favorable. Doping CuGeO₃ with both magnetic and nonmagnetic



Figure 3. Dispersion signal at a frequency $\omega_{\rm rf}$ (lower curve) and the signal amplitude at a fractional ($\omega_{\rm rf}/2$) frequency (upper curve). The arrow indicates the direction of the variation of the scanning field. $\omega_{\rm rf}/2\pi = 1699$ kHz; the *Q* factor of the cold circuit is 7000, $T \approx 0.98T_{\rm c}$.

which leads to the formation of uniformly precessing domains [12].

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