Observation of a direct magnetic coupling between nuclei in liquid ^{3}He and ^{169}Tm nuclei in a thulium ethyl sulfate crystal

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The cross-relaxation between the Tm spins in the van Vleck paramagnet TmES and the ^{3}He spins in a layer of liquid helium-3 surrounding the crystal is observed by using the method of pulsed nuclear magnetic resonance at 1.5 K.

In 1966, Wheatley and his coworkers discovered an anomalously low thermal resistance between the powder of cerium-magnesium nitrate and liquid ^{3}He at extremely low temperatures.[1] Leggett[2] proposed a theory, in which this effect was interpreted as being the result of a dipole-dipole interaction between the electronic spins of the paramagnetic powder and the nuclear spins of ^{3}He ($I = 1/2$). In 1981, Richardson and his coworkers, while studying the relaxation of ^{3}He absorbed on particles of a fluorocarbon polymer, confirmed the presence of a coupling between the nuclear spins of the ^{3}He and ^{19}F systems.[3] All experiments performed to date were intended to study the limiting magnetic coupling between magnetic moments of different magnitude. Using van Vleck paramagnets, known for their strong anisotropy of magnetic properties,[4] we can, however, study the resonant interaction of spins at the boundary between ^{3}He and a solid. We were able to observe such cross-relaxation between the nuclear spins of ^{169}Tm ($I = 1/2$) in thulium ethyl sulfate ($Tm(C_2H_5SO_4)_3 \cdot 9H_2O$) and the nuclei of the liquid ^{3}He surrounding the crystal.

The TmES crystal has hexagonal symmetry. The effective gyromagnetic ratio of ^{169}Tm nuclei depends on the angle θ which the external magnetic field **H** forms with the hexagonal c axis of the crystal in the following way: $\gamma = (\gamma_\parallel^2 \cos^2\theta + \gamma_\perp^2 \sin^2\theta)^{1/2}$. Here $\gamma_\parallel/2\pi = -0.480$ kHz/Oe and $\gamma_\perp/2\pi = -26.12$ kHz/Oe. At $\theta_X = 7.05°$ the gyromagnetic ratio of ^{169}Tm and ^{3}He nuclei are equal. We investigated the longitudinal relaxation time T_1 of ^{169}Tm and ^{3}He crystals at a temperature of 1.5 K. The measurements were performed with the help of a coherent NMR relaxometer at a frequency of 13.4 MHz: for different ($\theta \sim \theta_X$) orientations of the external field **H** relative to the c axis of the crystal we recorded the amplitude of the spin echo signal A_τ as a function of the repetition rate of the probing rf pulses τ, and the relaxation time T_1 was estimated from the equation $A_\tau = A_\infty [1 - \exp(-\tau/T_1)]$. The rf field $\mathbf{H}_1$ was oriented perpendicular to the c axis of the crystal. Because of the amplification of the field $\mathbf{H}_1$ by the $4f$ electronic shell, the optimum durations of the sounding pulses for thulium nuclei (~ 1 μs) were much shorter than for ^{3}He nuclei (~ 10 μs). Under the conditions of cross-relaxation ($\theta = \theta_X$) we were able to distinguish the signals of the spin echo of ^{169}Tm and ^{3}He nuclei not only due to the difference in the durations of the rf pulses but also due to the strong difference in the transverse relaxation times: $(T_2(^3\text{He})/T_2(^{169}\text{Tm}) \sim 10^3$. During the course of the experiments, we noticed that favorable conditions for observing the effect of cross-relaxation ^{3}He–^{169}Tm cannot always be completely reproduced. The factors responsible for the incomplete reproducibility

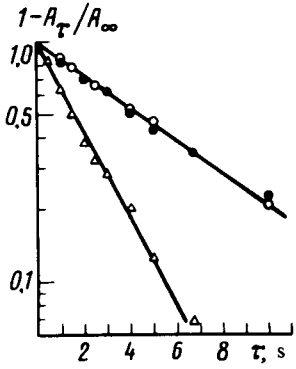

FIG. 1. Relaxation of longitudinal magnetization of liquid ^{3}He nuclei surrounding a TmES crystal at a temperature of 1.5 K: △—$\theta = \theta_X = 7.05°$; $T_1 = 2.4$s; ○, ●—$\theta = 7.5°$; $6.5°$; $T_1 = 6.5$ s; θ is the angle between the c axis of the crystal and the external magnetic field.

of the results upon changing specimens include the shape of the specimen and the quality of its surface, the ratio of the volumes of the crystal and liquid ^{3}He, contamination of the crystal by paramagnetic impurities, contamination of helium-3 by oil vapor and helium-4, nonuniformity of the external field (required for observing the spin echo of ^{3}He), and the unavoidable nonuniformity of the field near the surface of the magnetized crystal. An additional circumstance complicating the observation of the cross-relaxation effect is that because of the strong anisotropy of the γ-tensor, the plane of precession of the nuclear spins of ^{169}Tm does not coincide with the plane of precession of the ^{3}He spins, but rather forms an angle of $\sim 75°$ with it.[5] Figures 1 and 2 show the results of one of the most successful experiments, in which a cylindrical specimen of TmEs (diameter and heighth ~ 3 mm) was placed in a cylindrical pyrex ampoule and clamped to the flat bottom of the ampoule by a pyrex piston. To ensure an approximately uniform gap (~ 0.1 mm) between the glass and crystal surfaces the crystal was bound with cotton thread. The ampoule with the rf coil wound around it was submerged into liquid ^{4}He. At $\theta \neq \theta_X$ the relaxation time T_I of ^{3}He nuclei (Fig. 1) was determined by the size of the gap between the crystal and the surface of the glass, and in this case was 6.5 s; at $\theta = \theta_X$ this time was approximately equal to the time T_I for ^{169}Tm nuclei. The restoration of the nuclear magnetization of thulium at $\theta \neq \theta_X$ was described by a single exponential function (Fig. 2) and at $\theta = \theta_X$ by two exponential functions. The initial section of the curve ($T_I \sim 0.4$ s) apparently corresponded to establishing an equilibrium between the ^{169}Tm and ^{3}He spin systems (the latter was heated to a smaller extent by short rf pulses), and later both spin systems gave up energy to the crystal lattice through the impurity paramagnetic centers.[6]

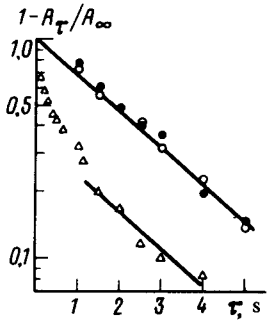

FIG. 2. Relaxation of the longitudinal magnetization of ^{169}Tm nuclei in a TmES crystal surrounded by a layer (~ 0.1 mm) of liquid ^{3}He at a temperature of 1.5 K; △—$\theta = \theta_X = 7.05°$; $T_1' \sim 0.4$ s, $T_1'' \sim 2.6$ s; ○, ●—$\theta = 7.5°$; $6.5°$; $T_1 = 2.6$ s.

We postulate that at a distance $r \sim 70$ Å from the surface (five lattice constants a_0) Tm^{3+} ions feel the same crystal field as in the bulk of the crystal, and their nuclei can be assigned the value $\gamma(\theta_X) = \gamma(^3\text{He}) = -2\pi \cdot 3.24 \times 10^3\ \text{s}^{-1}\text{Oe}^{-1}$. The probability of mutual orientation reversal of one of these ^{169}Tm spins and of the spin of the surface atom of ^{3}He will then be equal in order of magnitude to $w \sim \gamma^2\hbar/r^3 \sim 1\ \text{s}^{-1}$, consistent with our results. As is well known, dielectric van Vleck paramagnets are not used for nuclear magnetic cooling because of the very slow nuclear spin-lattice relaxation at superlow temperatures. The observed cross-relaxation indicates that there can be a direct transfer of spin temperature from the van Vleck paramagnet to the liquid ^{3}He, bypassing the lattice of the crystal. The ^{3}He spin system can probably be cooled to very low temperatures by first polarizing the nuclear spins of ^{169}Tm in a strong field $\mathbf{h} \perp c$ (at $T = 0.05$ K, in a field of $H = 70$ kOe, the intrinsic single-phonon relaxation time of thulium nuclei should be of the order of seven hours[7]), and then demagnetizing the crystal and rotating the external field toward the c axis of the crystal by an angle $(\pi/2 - \theta_X)$.

[1]W. R. Abel, A. C. Anderson, W. C. Black, and J. C. Wheatley, Phys. Rev. Lett. **16**, 273 (1966).
[2]A. J. Leggett and M. Vuorio, J. Low Temp. Phys. **3**, 359 (1970).
[3]L. J. Friedman, P. J. Millet, and R. C. Richardson, Phys. Rev. Lett. **47**, 1078 (1981).
[4]S. A. Al'tshuler and M. A. Teplov, Problemy magnitnogo rezonansa (Problems of Magnetic Resonance), Nauka, Moscow, 1978, p. 14.
[5]L. K. Aminov, M. S. Tagirov, and M. A. Teplov, Zh. Eksp. Teor. Fiz. **79**, 1322 (1980) [Sov. Phys. JETP **52**, 669 (1980)].
[6]A. Abraham and B. Bleaney, Proc. R. Soc. Lon. A **387**, 221 (1983).
[7]M. P. Vaĭsfel'd, Fiz. Tverd. Tela **14**, **737** (1972) (*sic*).

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