ULTRASONICALLY INDUCED NUCLEAR SPIN TRANSITIONS IN ANTIFERROMAGNETIC $KMnF_3$†

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Nuclear spin transitions in solids have been previously induced by ultrasonically modulating the interaction of the nuclear quadrupole moment with the crystalline electric field.[1]

We believe we have succeeded in producing nuclear spin transitions of fluorine nuclei via ultrasonic modulation of the magnetic interaction in $KMnF_3$ in the antiferromagnetic state. Shulman and Knox[2] have shown that $KMnF_3$ has an antiferromagnetic transition at 88.3°K.

In the present experiment the crystal is thermally cycled through the transition temperatures in a high magnetic field in order to align the antiferromagnetic domains. Then the crystal was brought to 4.2°K by immersion in liquid helium. The $KMnF_3$ single crystal is bonded to the end of a silvered, single-crystal, quartz sound pipe which extends from a metallic cylinder within which the exciting quartz transducer is bonded to the sound pipe. The $KMnF_3$ crystal is mounted so that the [100] direction is parallel to H_0, the steady magnetic field. The metallic cylinder and silvered quartz provide shielding of the $KMnF_3$ from the stray rf magnetic fields from the exciting current to the quartz transducer. The $KMnF_3$ crystal itself sits in the middle of a marginal oscillator coil, the axis of which is parallel to the [010] direction.

The procedure used to observe the effect is as follows: The sample assembly is placed in a magnetic field of 2.39 kG and the ultrasonic transducer is excited at the fluorine Larmor frequency of 25.33 Mc/sec, which signifies an internal field at the ^{19}F site of 3.82 kG. After an ultrasonic irradiation period of three seconds (during which the ultrasonically induced spin transitions are produced), the steady magnetic field is swept by discharging a condenser through auxiliary coils in such a manner that the fluorine nuclear line is swept through the resonance of a marginal oscillator oscillating at 25.73 Mc/sec. The change in the level of the marginal oscillator caused by the resonance is detected and displayed on the vertical axis of an oscilloscope trace. The horizontal sweep of the oscilloscope is proportional to the pulsed magnetic field. This arrangement is used because the nuclear resonance line is too broad to observe with the usual pulsed nmr techniques. The difference between the frequencies of the ultrasonic excitation and the marginal oscillator eliminates magnetic saturation by the continuously running marginal oscillator. The time lag between the completion of ultrasonic saturation and the sampling by the marginal oscillator is 10 milliseconds which is much less than T_1 for the fluorine, which was measured to be 56 ± 8 milliseconds. The signal height of the marginal oscillator output is proportional to the equilibrium magnetization at the end of the ultrasonic saturation.

In Figs. 1 and 2 is shown the ^{19}F nuclear resonance signal with no voltage applied to the transducer. The second trace in Fig. 1 shows the ultrasonic saturation when 0.6 volt excitation is applied to the quartz transducer. The second trace in Fig. 2 shows the effect of 165 volts of excitation but with the sample acoustically isolated by two pieces of cardboard bonded between the sample and the sound pipe. Figure 2 shows that nonacoustic effects are too small to explain the large acoustic saturation observed in Fig. 1. We were unable to measure the ^{19}F linewidth by ultrasonic saturation because of the shifting of the line when the ultrasonic frequency was driven through the resonance. At the present we do not understand this slight shifting of the line.

Silverstein[3] has recently independently published a theoretical prediction of the above effect and has applied it to ultrasonic attenuation in MnF_2 in the antiferromagnetic state. Robinson[4] had attempted the experiment on $CuCl_2 \cdot 2H_2O$ some five or six years ago. Our attempts on MnF_2 for

FIG. 1. Ultrasonic saturation of ^{19}F resonance; upper trace with no excitation on transducer, lower trace with 0.6 volt of excitation.

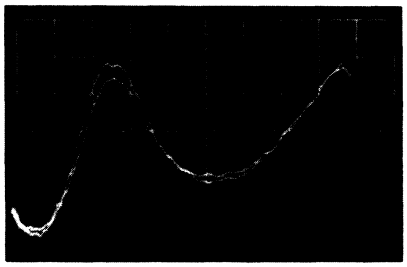

FIG. 2. Fluorine resonance with sample ultrasonically isolated from sound source; one trace with no excitation on transducer, the other with 165 volts of excitation.

the past three years have not met with success because of the high ultrasonic frequencies (162 Mc/sec) required.

Rather than apply Silverstein's theory to the present case we give a rudimentary equivalent of it. In $KMnF_3$ one can write the interaction between a Mn^{2+} electron spin and a ^{19}F nuclear spin in the form

$$H(\vec{a}) = \vec{I}\cdot A(\vec{a})\cdot\vec{S}, \qquad (1)$$

where $\vec{a}$ is the separation vector between the Mn^{2+} and the F^- ions. The tensor $A(\vec{a})$ will, in general, contain dipolar and orbital fields and the Fermi contact term.

The interaction in (1) is modulated by the ultrasound whose amplitude is $\vec{B}(\vec{r})e^{i\omega t} = \vec{B}(\vec{r}, t)$, where $\vec{r}$ is a crystal position vector. The change in the separation vector brought about by the ultrasonics is $\vec{\delta}(\vec{a}, \vec{B}, t) = \nabla\vec{B}(\vec{r}, t)\cdot\vec{a}$, and the modulation of the interaction is

$$H(\vec{B},\vec{a},t) = \nabla H(\vec{a})\cdot\vec{\delta}(\vec{a},\vec{B},t) = \nabla(\vec{I}\cdot A\cdot\vec{S})\cdot\vec{\delta}(\vec{a},\vec{B},t). \qquad (2)$$

In the above interaction we get energy-conserving transitions corresponding to the absorption or emission of one phonon of the nuclear Larmor frequency from the ultrasonic field and the occurrence of a nuclear-spin transition $\Delta m = \mp 1$ with no electron-spin transition. This comes from a term, for example in (2), of the type $I_x S_z(\nabla A_{xz}\cdot\vec{\delta})$. For transitions of this type the electron is merely a mechanism by which the nucleus is coupled to the ultrasonic field.

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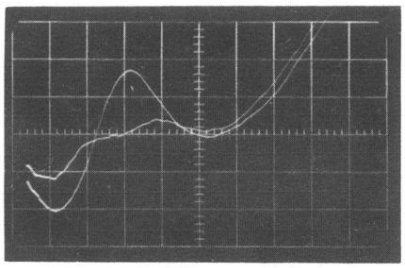
[1]W. G. Proctor and W. H. Tanttila, Phys. Rev. 101, 1757 (1956); W. G. Proctor and W. Robinson, Phys. Rev. 104, 1344 (1956). A very comprehensive review is given by V. A. Shutilov, Akust. Zh. 8, 383 (1962) [translation: Soviet Phys.—Acoust. 8, 303 (1963)].

[2]R. G. Shulman and K. Knox, Phys. Rev. 119, 94 (1960).

[3]S. D. Silverstein (to be published).

[4]W. A. Robinson (private communication).



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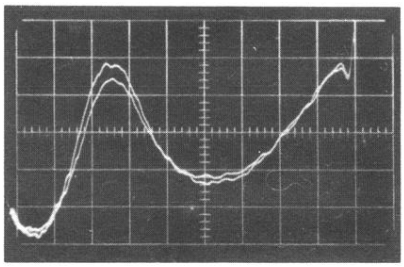

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