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ULTRASONIC SATURATION OF NUCLEAR MAGNETIC ENERGY LEVELS [1]

BY

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Le processus de relaxation directe du Na^{23} d'un monocristal de NaCl placé dans un champ magnétique intense a été étudié expérimentalement. La densité d'énergie de phonons à la double fréquence de Larmor a été augmentée artificiellement à l'aide d'ondes ultrasonores jusqu'à ce que la saturation des niveaux énergétiques magnétiques se produise. La population des niveaux a été étudiée en appliquant la technique de résonance magnétique pulsée. Les résultats observés peuvent être interprétés par l'interaction du moment quadripolaire du Na avec le déplacement des ions de Cl environnants, environ une charge élémentaire leur étant attribuée. Ce nombre est environ cent fois trop petit pour expliquer le taux de relaxation indirecte, tel qu'il fut calculé par Van Kranendonk.

Recently Proctor and Tantilla have described an experiment in which pure nuclear electric quadrupole energy levels were saturated by ultrasonic excitation at the pure quadrupole transition frequency [1]. In a similar experiment we have observed a change in the population of the nuclear magnetic energy levels of Na^{23} in a single crystal of NaCl as a result of ultrasonic excitation. The cubic symmetry of the NaCl

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crystal is removed by the sound, causing time varying electric field gradients at the Na^{23} nuclei which interact with their quadrupole moment. This may induce transitions corresponding to m changes of both ± 1 and ± 2. In our experiment the sound frequency was twice the magnetic resonance transition frequency, which corresponds to $\Delta m = \pm 2$. Our experiment was performed in a steady magnetic field of 4220 oersteds, under which conditions the nuclear magnetic resonance occurred at a frequency of 4.75 Mc/sec. We measured the thermal relaxation time to be 8 seconds at room temperature.

In a magnetic field Na^{23} has four nuclear magnetic energy levels corresponding to m values of $-\frac{3}{2}, -\frac{1}{2}, +\frac{1}{2}$ and $+\frac{3}{2}$. There are 3 transitions between these and they are at identical frequencies in ideal NaCl crystals. In this experiment the population difference between the $m = +\frac{3}{2}$ and $m = -\frac{3}{2}$ levels was measured by the amplitude of the nuclear induction signal that followed a short pulse of radio-frequency magnetic field at the larmor frequency [2]. The nuclear signal was induced in a receiver coil oriented perpendicular to the transmitter coil and containing the NaCl crystal sample. The sample was a cylinder of halite 1.5 cm in diameter and 3 cm long. It was cemented to an identical crystal which in turn was cemented to an X-cut quartz transducer. The quartz had a broad resonance centered at 9.5 Mc/sec when loaded and was driven by a variable frequency oscillator. This oscillator was turned on for a period of 8 seconds, then after a delay of .03 second the $+\frac{3}{2}, -\frac{3}{2}$ population difference was measured. This cycle was repeated every 17 seconds. We assume that the sound generated by the quartz was scattered into an isotropic distribution at the free end of the halite sample since this end had been made irregular. The magnetic fields generated by currents in the ultrasonic system, a possible source of difficulty in the experiment of Proctor and Tantilla, could not affect the population since they were at twice the larmor frequency.

We observed a decrease in the $m = +\frac{3}{2}$, $m = -\frac{3}{2}$ population difference for a small range of ultrasonic frequencies

centered exactly at twice the nuclear frequency. With 1.2 watts of radio-frequency power delivered to the quartz transducer the population difference could be reduced to 26% of its equilibrium value. The difference of frequency values for which the population decrease was $\frac{1}{2}$ of the maximum decrease was found to be 4 kc/sec. The 4 kc width of this effect is to be compared to the nuclear magnetic resonance line width of 2.3 kc/sec [3]. The difference in these widths can be explained in terms of imperfections in the crystal lattice giving rise to random but permanent electric field gradients. These gradients cause the $\Delta m = \pm 2$ transitions to be spread by the quadrupole interaction [4]. In nuclear magnetic resonance two transitions are of the type $m = \frac{1}{2}$ to $m = \frac{3}{2}$ and these will be spread by the same amount as the $\Delta m = \pm 2$ transitions. The $m = \frac{1}{2}$ to $m = -\frac{1}{2}$ transition, however, will have only dipolar broadening and will weight the nuclear magnetic resonance line toward the center frequency.

Measurements are now under way to examine the quantitative and directional properties of this phenomenon. It is hoped that the thermal relaxation mechanism proposed by Van Kranendonk [5] can be studied in this way.

Van Kranendonk calculates the relaxation rate due to the indirect process, using a model in which the electric quadrupole moment of the nucleus in question interacts with the displacements of the nearest neighbor ions. He introduces a dimensionless factor γ, the number of elementary charges to be assigned to each of the nearest neighbor ions, to account for co-valent, shielding, and anti-shielding effects, and other shortcomings of the model. He finds by comparing his results to the known value of the thermal relaxation time that γ must be approximately 50 for Na^{23} in NaCl. By measurements of the sonic energy density, and the use of a simple expression for the direct process relaxation rate, we have arrived at a tentative value for γ which is very close to one. It is of course true that we may not be measuring Van Kranendonk's γ.

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