

# Nuclear Quadrupole Resonance Studies of a Quantum Organic Magnet

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3 August 2011

## Abstract

The nuclear quadrupole resonance (NQR) frequency of Chlorine atoms in the quantum organic magnet  $\text{NiCl}_2 - 4\text{SC}(\text{NH}_2)_2$ , known as DTN, was located using a superheterodyne spectrometer. The NQR frequency provides direct information about the local magnetic ordering, the magnitude of the local field, and its direction. The free induction decay signal occurred when the sample was struck with an RF pulse at 30.180 MHz and we believe this to be the NQR frequency.

## I. INTRODUCTION

The nuclear quadrupole resonance (NQR) frequency of Chlorine atoms in the quantum organic magnet dichloro tetrakis-thiourea nickel (DTN), with chemical formula  $\text{NiCl}_2 - 4\text{SC}(\text{NH}_2)_2$ , is a physical characteristic that can be used to study its fine structure. DTN shows Bose-Einstein condensation (BEC) of its magnetic spin system and is predicted to show Bose-Glass behavior, which has only been shown in a few physical systems. [1, 2] Bose-Glass is similar to BEC but has had disorder deliberately introduced into the system.

While much research has been done on DTN [3, 4], especially establishing it as a BEC candidate [5-8], the Chlorine NQR frequency is unknown and cannot be calculated with any accuracy. Locating the NQR frequency will provide more information into the orientation of the internal magnetic fields and provide a basis for studying the transition from a Bose-Einstein condensate to a Bose-Glass.

The NQR frequency provides direct information about the local magnetic ordering with respect to the local crystal axes ( $\epsilon, \eta, \zeta$ ) as shown in Fig. 1a. The local crystal axes determine the symmetry axes of the local electric field gradient and the interaction between the nuclear electric quadrupole moment  $Q$  and the local field gradient  $q$ . The Hamiltonian for this interaction is

$$H = \frac{e^2 q Q}{4I(2I-1)} (3I_\zeta^2 - I^2)$$

where  $I$  is the nuclear spin of Chlorine. The transitions between the  $I_\zeta = \pm \frac{3}{2}$  levels and the  $I_\zeta = \pm \frac{1}{2}$  levels, shown in Fig. 1b, give the nuclear quadrupolar frequency, in MHz, as

$$\omega_Q = \frac{3e^2 q Q}{4I(2I-1)} \frac{1}{\hbar}$$

In the presence of a local magnetic field  $B_L$  produced by the ordering of Nickel electronic spins,  $\omega_Q$  is split into components given by

$$\omega'_Q = \omega_Q \pm \gamma B_L \cos\theta$$

where  $\theta$  is the angle between  $\hat{\zeta}$  and the internal magnetic field and  $\gamma$  is the gyromagnetic ratio. This splitting is shown in Fig. 1c. The studies will therefore give information not only about the magnitude of the local field  $B_L$  but also its direction. [9]

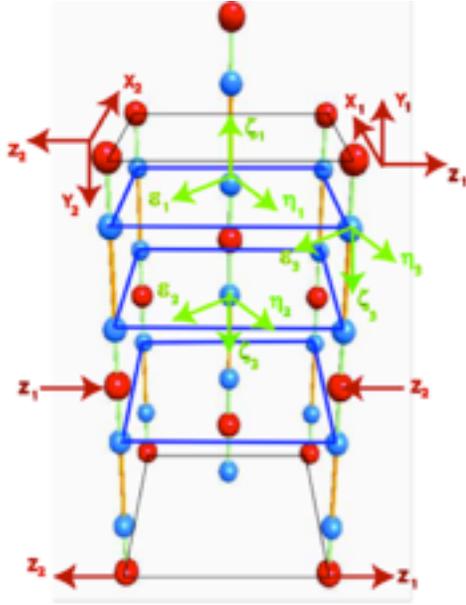
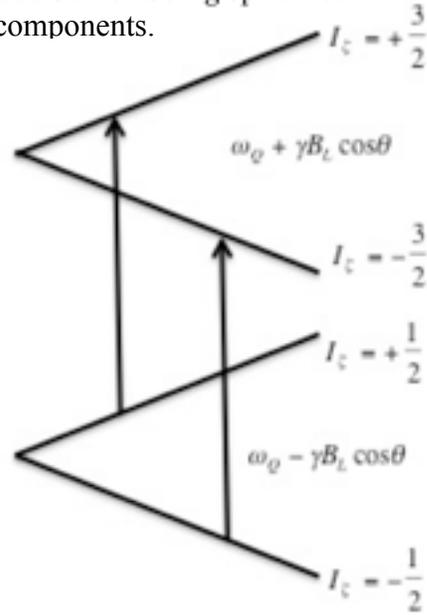
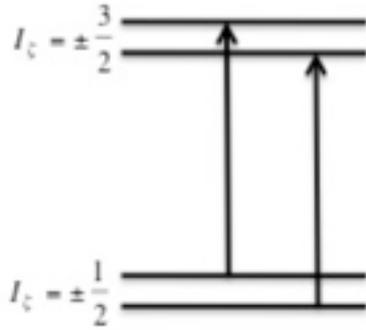


Figure 1a: (left) The structure of DTN in the ordered state. The local crystal fields are shown as  $(\epsilon, \eta, \zeta)$  for three lattice sites. The z-axes show the anti-ferromagnetic ordering.

1b: (bottom left) The transition between the  $\pm 3/2$  and  $\pm 1/2$  levels.

1c: (bottom right) The transition between the spin levels after being split into components.



Typical NQR frequencies for Chlorine in ionic crystals range from 25 to 42 MHz and that was used as the starting point for this experiment. [9] Here we present the method used to find the NQR frequency. We observed the correct response signal at 30.180 MHz.

## II. METHODS

A single crystal of DTN was placed in a helium environment inside an inductance coil. The coil was placed into a quadrature hybrid circuit [10] connected to a series of tunable

capacitors and superheterodyne NMR spectrometer [11] as shown in Fig. 2. The spectrometer used an Agilent 8648A signal generator, a model 550L RF power amplifier, and a PC operating a LabVIEW program designed to control the timing for RF pulses. The decay frequency was scanned for using a Tektronix TDS 3032 digital oscilloscope with an FFT application module.

We performed the broadband search for the NQR frequency by tuning the circuit for 29.970 MHz and generating pulses to excite the Chlorine spin. After allowing the oscilloscope to average the signal for several minutes we looked for the free induction decay and for any spikes on the oscilloscope's FFT. The tuning and signal averaging were repeated for increments of 10 kHz.

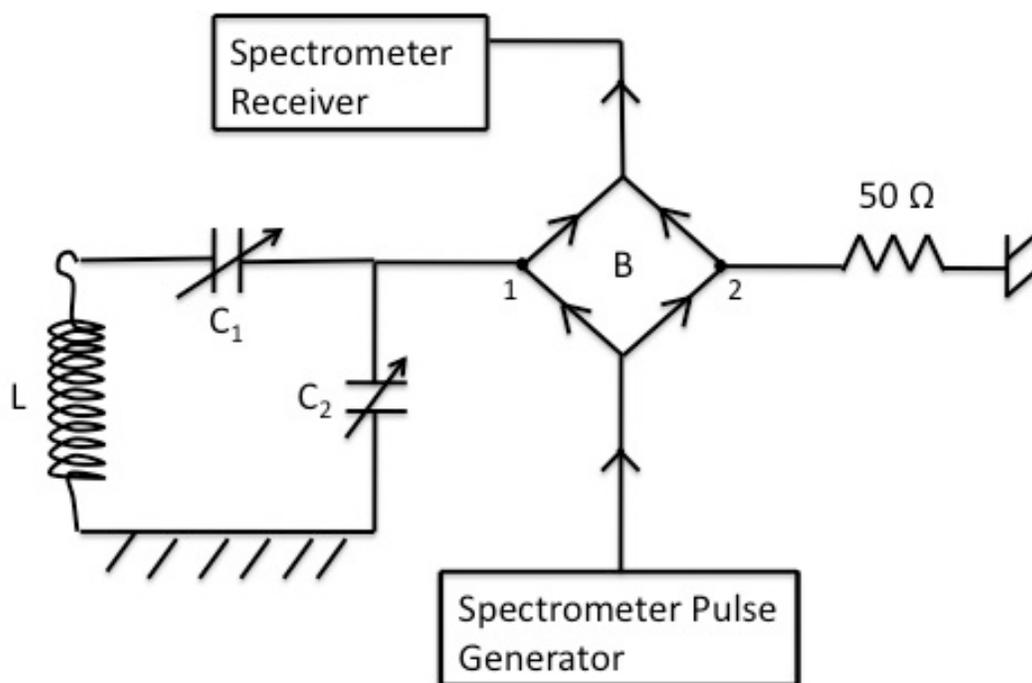


Figure 2: The quadrature hybrid circuit shown here was used in our experiment. The circuit used tunable capacitors,  $C_1$  and  $C_2$ , to match the impedances at junctions 1 and 2 on opposite sides of the bridge, B. The receiver detected the induced NQR signal after the pulse.

### III. RESULTS AND DISCUSSION

A strong resonating signal was noticed at 30.180 MHz, as shown in Fig. 3. Characteristic of free induction decay that occurs as the nuclear spins transition to their original levels [12, 13], the resultant signal indicates that this is a good candidate for the NQR frequency of DTN.

To validate this as the correct frequency, we performed a simple test. Placing a 1,000 Gauss magnet perpendicular to the inductance coil should effectively neutralize the signal with an acceptable amount of noise. This effect can be seen in Fig. 4. Passing this test increases the probability that 30.180 MHz is the true NQR frequency, but further tests determining the temperature dependence of this frequency and the shift in the frequency as various external fields are applied will provide stronger evidence.

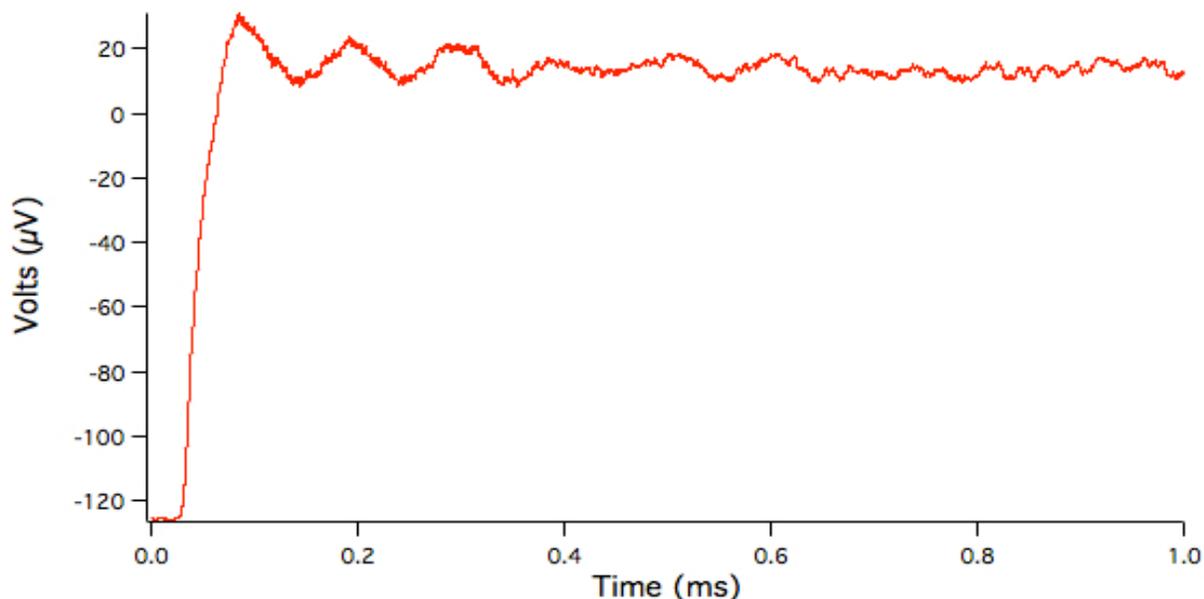


Figure 3: The NQR signal as it appeared on the oscilloscope. After the pulse, the signal slowly decays until there is no recognizable shape to the waveform.

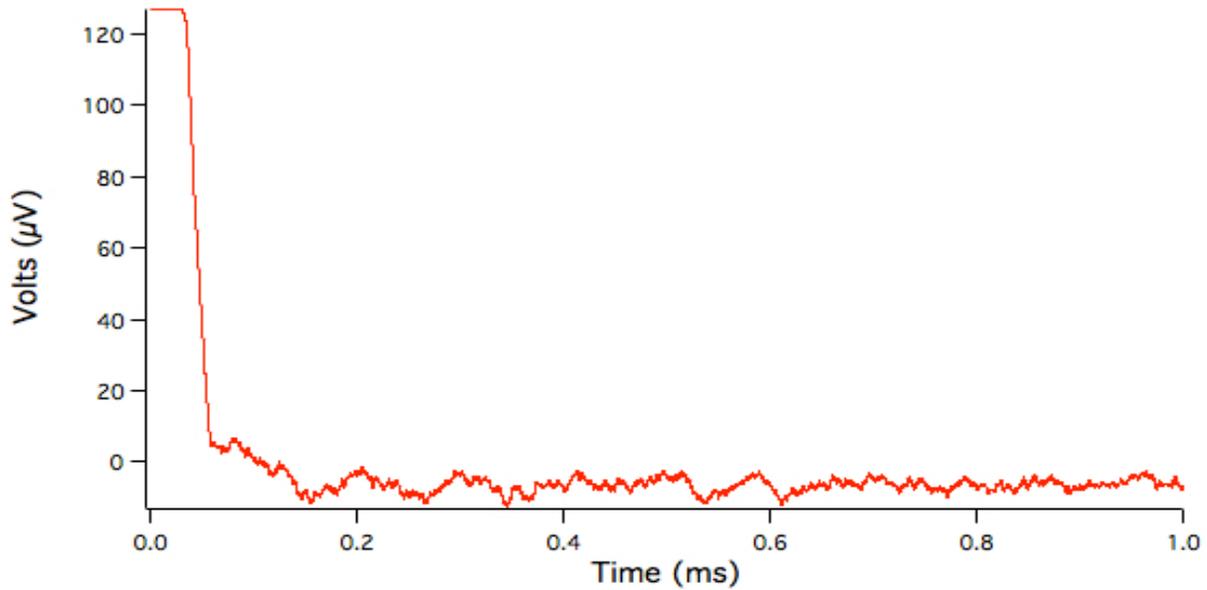


Figure 4: The NQR signal with a magnetic field perpendicular to the inductance coil. The magnetic field distorts the crystal's internal magnetic fields in such a way that the signal is weakened and the decay is not noticeable.

#### IV. CONCLUSION

While further tests determining 30.180 MHz to be the true NQR frequency have to be performed, having found it opens up research into the internal magnetic structure of DTN, especially in low temperatures and high magnetic fields. Moreover, this discovery gives us more information with which we can research the Bose-Glass state and the phase transitions into it.

#### ACKNOWLEDGEMENTS

This work was supported by NSF funding and performed at 2011 summer REU program at the University of Florida.

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