



# LABORATORY Spotlight

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## Proton NMR Studies of Spin Density Wave Fluctuations up to 1.9 GHz

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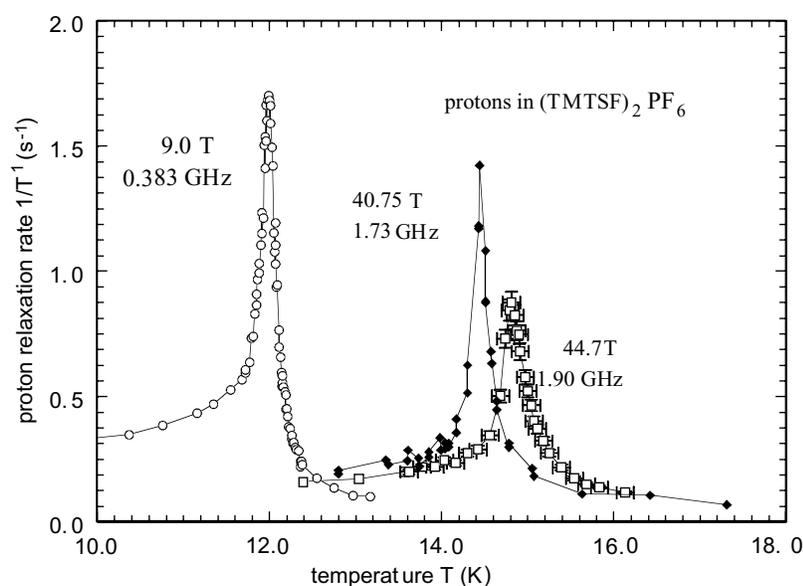
Spin density wave (SDW) order and fluctuations associated with this low temperature phase of quasi one-dimensional conductors, such as  $(\text{TMTSF})_2\text{PF}_6$ , have been widely investigated for a substantial length of time. In part, this is because they reflect the static and dynamic properties of the pinned density wave phase and provide a means to compare these properties with theoretical models for their origin. One NMR measurement of interest is the nuclear spin-lattice relaxation rate  $1/T_1$ .<sup>1-3</sup>

For the ordered phase of the SDW not far below the SDW transition, it is generally agreed that thermally excited phase fluctuations of the pinned SDW (phasons) are responsible for  $1/T_1$ . In this regime, the phason fluctuations generate a fluctuating magnetic field whose power spectrum is frequency dependent. Since the fluctuation motion of the SDW also corresponds to polarization charge fluctuations of the condensed electrons that make up the SDW, according to one model,<sup>4</sup>  $1/T_1$  is proportional to the sum of the imaginary part of the SDW dielectric constant over all wave vectors at the NMR frequency. Thus, through measurements of  $1/T_1$  as a

function of frequency, one investigates the dynamics of the pinned SDW charge fluctuations on different time scales.

Two other regions of temperature are also of interest in this field. One is the critical fluctuations close to the SDW transition whose temperature ( $T_{\text{SDW}}$ ) is near 12 K at low field. The other is the low temperature region below about 4 K, where there is an unresolved controversy regarding several possibilities, including a simple slowing of the dynamics,<sup>2</sup> additional phase transitions,<sup>1</sup> and opening a gap for a residual conduction electron density.<sup>5</sup>

One of the experimental goals in this field is to extend the frequency of the measurements as high as



**Figure 1.** Proton spin-lattice relaxation rate as a function of temperature at three values of the magnetic field. The peak in  $1/T_1$  occurs at  $T_{\text{SDW}}$  and the rapid variation near  $T_{\text{SDW}}$  is caused by critical fluctuations. At temperatures more than 1 K below  $T_{\text{SDW}}$ , the relaxation is caused by pinned SDW phase fluctuations.

possible. In a recent set of measurements in the Hybrid magnet at the NHMFL in Tallahassee, we carried out such measurements up to a world record of 1.9 GHz for proton NMR measurements. Here, we give a preliminary, qualitative report of some of this work and describe several special circumstances of the experiments.

Part of the results are summarized in Fig. 1, where  $1/T_1$  in the critical and SDW regimes is shown for three values of the applied field ( $B$ ) aligned approximately 30 degrees from the  $c^*$ -axis in the  $b^*c^*$  plane. The values of  $B$  and the corresponding NMR frequency are: 44.7 T (1.90 GHz), 40.75 T (1.73 T), and 9.0 T (.383 GHz at UCLA). For all three values of  $B$ , a sharp peak is seen where  $1/T_1$  is dominated by critical fluctuations near  $T_{SDW}$ . The value of  $T_{SDW}$  has a weak, quadratic dependence on  $B$ , with values that agree with transport measurements for  $B$  along  $c^*$ .<sup>6</sup> The latter were interpreted using a model of imperfect nesting. Since this temperature dependence disappears for  $B$  along  $b'$ ,<sup>6</sup> there should be an angular dependence to it. Our measurements at 30 degrees from  $c^*$  show a value close to that for  $c^*$ .<sup>6</sup> We plan to extend them to

other angles to investigate this aspect of nesting in the material.

Approximately 1 K below the transition, it is seen that  $1/T_1$  decreases monotonically with frequency/field, which indicates that the power spectrum of the density wave fluctuations continues to decrease with frequency up to 1.9 GHz. Although we had anticipated a change in this behavior that could be attributed to a larger amplitude of phason fluctuations at wave vectors that are too short to be screened and damped by thermally excited quasiparticles, it did not occur.

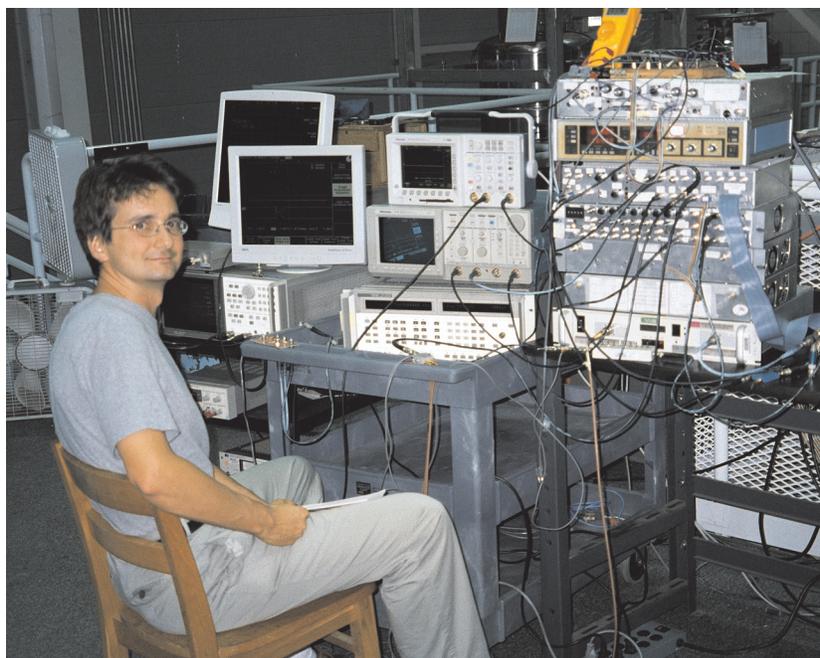
There are two aspects of  $1/T_1$  near the transition that should be noted. The first is that somewhat away from the transition in the high temperature side, its value as a function of the temperature difference from  $T_{SDW}$  is essentially independent of frequency, including other measurements that go down to 15 MHz.<sup>3</sup> This means that unlike the fluctuations in the ordered phase, those in this part of the critical regime are essentially independent of frequency. Also, they do not display the angular dependence seen in the ordered phase.<sup>2,7</sup> This suggests that they are dominated by amplitude fluctuations rather than phase fluctuations. Another

point is that the peak amplitude at the transition appears to decrease as a function of frequency. Unfortunately, on the basis of the present measurements, we cannot tell whether this decrease is a real property of the material or an artifact of the measurements. The problem is that the peak is very narrow and we had relatively large temperature fluctuations during the run. Subsequently, at UCLA we have identified the origin of the thermal fluctuation problem and decreased it by more than a factor of 20. If future measurements confirm the decrease in  $1/T_1$ , the challenge will then be to determine whether its origin is critical slowing that becomes evident in higher frequency measurements, an effect of the high field on the nesting conditions, or some other mechanism.

A challenging part of these measurements was to complete them in the very short running time available. One of the strategies we used was a continuous saturation method for measuring  $1/T_1$ . It permitted us to carry out individual measurements about 4 times faster than more conventional methods available with our instrumentation and signals. As a consequence, the 68 points shown at high fields in Fig. 1 include three conventional



**Figure 2.** NMR probe and measurement team. L to R: Phil Kuhns, Arneil Reyes, Patrik Vonlanthen, and Gil Clark



**Figure 3.** Patrik Vonlanthen, the .005-2 GHz NMR spectrometer, and related instruments used for the experiments.

ones that required a total of 20 minutes and 65 that were done in 130 minutes.

A key element of the success of our measurements was the level of cooperation and collaboration provided by the staff at the NHMFL. Although the probes and part of the spectrometer were brought from UCLA for the experiments, the rest of the NMR instrumentation and essential collaboration in the measurements were provided by the NHMFL condensed matter physics NMR (CMP-NMR) group. We were also fortunate to have very effective support from the magnet operators, the cryogenics group, the machine shop, the electronics shop, the instrument shop, and those responsible for managing the activities of external users.

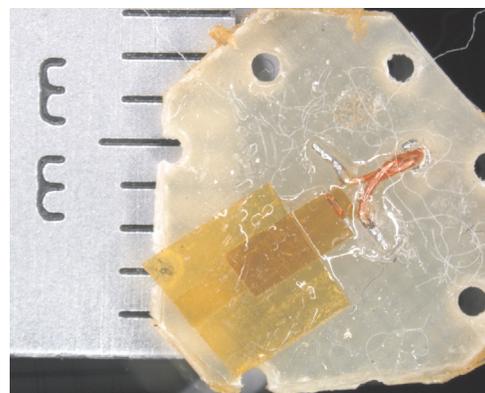
Some of the specialized NMR instrumentation used for the experiments along with the persons most closely associated with the project are shown in Figs. 2 and 3. Fig. 2 shows four of us on the platform at the top of the Hybrid magnet. The dewar and NMR probe are at the lower right. Since access close to the magnet is not permitted when the field is ramped up, the long, flexible epoxy glass rod, held by Phil Kuhns and attached to the probe goniometer control shaft, is used to make adjustments during the run. Fig. 2 shows Patrik Vonlanthen with the NMR spectrometer

and associated instruments. In the stack of chassis' at the right, the bottom one is the 1 to 2 GHz power amplifier provided by the CMP-NMR group. The next four include the rf power amplifiers and NMR spectrometer brought from UCLA.

During the recent run in the Hybrid the week of August 20, 2001, we also worked on a project that demonstrated the possibility of doing NMR on very small single crystals. In this case, the goal was to investigate the 1-D antiferromagnetic properties of  $\text{LiVGe}_2\text{O}_6$ . One of the challenges for this work was that the sample was very small and it had to be rotated about one axis at high field in the

NMR probe during the measurements. The probe design steps taken to carry out these measurements are shown in Figs. 4 through 6.

Fig. 4 shows the platform to which the NMR coil and its leads are attached. The platform, which is formed from a thin, epoxy-glass sheet, is attached to a holder that permits rotation of the sample in the magnetic field. A scale with markings one millimeter



**Figure 4.** Epoxy-glass platform with NMR coil, sample, and lead wires attached. The markings on the scale are spaced 1 mm.

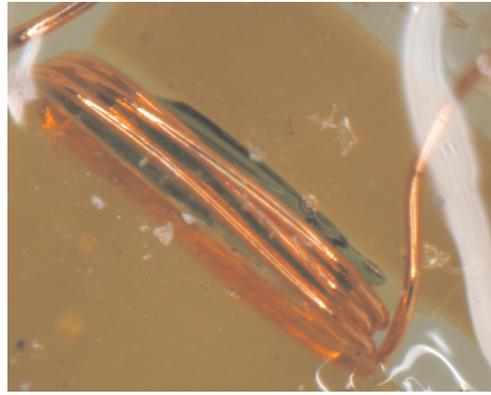


**Figure 5.** Expanded view of Fig. 4.

apart indicates the size of the platform. Fig. 5 is an enlargement of Fig. 4. At the upper right are the larger copper wire leads that connect the NMR coil to the rest of the probe circuit. The structure in the center is the NMR coil wrapped around the dark sample inside of it. The light streaks in the picture are reflections of light from the surface of a thin, clear epoxy layer that encapsulates the sample, the NMR coil, and part of the larger wire leads.

An enlarged view of the coil and the sample is shown in Fig. 6. The coil is three turns of 25 micron diameter wire placed closely on the sample to obtain the best possible signal. This thickness is about 1/2 to 1/3 the diameter of a human hair. The coil is wound around the long dimension of the sample to obtain the desired direction of the 737 MHz radio frequency magnetic field used for the NMR measurements. The sample itself is a single crystal of  $\text{LiVGe}_2\text{O}_6$  grown by Dr. Jean-Yves Henry of the CENG laboratory in Grenoble, France. Its dimensions are approximately 50 microns  $\times$  100 microns  $\times$  0.8 mm. It weighs approximately 15 micrograms and contains about  $4 \times 10^{16}$  of the  $^7\text{Li}$  spins whose NMR signal is measured in our experiments. At the high magnetic fields used, the signal-to-noise ratio of the  $^7\text{Li}$  NMR spin echo signal is approximately 20 for a single transient.

It is of interest to speculate what are the limits on small sample size one could reach by following the approach we have taken and what could be accomplished using nanofabrication technology. With the materials and coil geometry shown in Fig. 6, one could obtain usable results from a sample with  $1 \times 10^{16}$   $^7\text{Li}$  spins and extend that to  $1 \times 10^{15}$   $^7\text{Li}$  spins by averaging 100 transients. By using still smaller commercial copper wire for the NMR coil, we estimate one could obtain useful signals with 1.5 micrograms of sample



**Figure 6.** NMR coil (thin wires), sample (dark material inside the coil), and lead wires to the rest of the NMR coil circuit. The NMR coil wire diameter is 25 microns (one thousandth of an inch).

( $4 \times 10^{15}$  spins) without averaging and 150 nanograms ( $4 \times 10^{14}$  spins) by averaging 100 transients. Finally, we estimate that with coils produced using advanced nanofabrication technology, one could investigate samples of this material weighing as little as 1.5 nanograms ( $4 \times 10^{12}$  spins) by averaging 1,000 transients.

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