Pulse Electron Spin Resonance Method For Investigation Of Atoms In Impurity-Helium Solids

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Abstract. We have constructed an X-band pulse electron spin resonance spectrometer for the investigation of hydrogen, deuterium, and nitrogen impurity-helium solids at low temperatures. The spectrometer is of a conventional homodyne detection design incorporating a sampling oscilloscope but uses a modified CW ESR resonant cavity.

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INTRODUCTION

Impurity-helium solids containing hydrogen, deuterium, and nitrogen atoms have been extensively studied through CW ESR spectroscopy.[1] These studies have been effective in determining local and average radical concentrations and in following the progress of exchange tunneling reactions. The CW method suffers from a high degree of inhomogeneous line broadening due to the nuclei and atomic radicals within the impurity clusters. This broadening obscures some of the weak interactions between the atomic radicals and the nuclei of neighboring molecules.

The standard pulse ESR methods of echo refocusing allow the study of weak couplings within inhomogeneously broadened systems. Two-pulse and three-pulse electron spin echo envelope modulation (ESEEM) sequences can be used to infer the local environment surrounding the atomic radicals.[2] To this end we have constructed a pulse ESR spectrometer specially equipped for the study of impurity-helium solids.

SPECTROMETER DESIGN

Our spectrometer operates in the X-band with a microwave frequency of 8930 MHz. Figure 1 shows a simplified block scheme. A 12 channel programmable pulse generator, (PulseBlaster model 125-12 by SpinCore) operating at a 125 MHz clock frequency provides logic signals to the PIN diode switch, bi-phase modulator, oscilloscope trigger and traveling wave tube (TWT) grid modulator. Microwaves from the tunable oscillator are amplified and split into pulse and reference paths. The pulse path passes though the bi-phase modulator and is gated by the PIN diode switch. The pulses are then amplified to ~200 mW by a medium power solid state amplifier. The pulse path then transitions from coax to waveguide, where the phase and attenuation of the pulse are manually adjustable. The pulses are amplified by a 1kW, 50dB TWT amplifier operating in the saturated regime. Ferrite isolators bracket the input and output of this amplifier. The amplified pulses pass through a rotary vane attenuator and a circulator before transitioning to a 20 cm length of stainless steel coax at the entrance to the cryostat. This prevents excessive heat conduction into the cryostat. Low loss coax traverses the remaining 107 cm to the resonant cavity at the bottom of the cryostat, which is situated between the poles of a Varian electromagnet. Echo signals return to the circulator through the same cabling and pass through a ferrite isolator before a transition to coax. The low noise amplifier (LNA) is protected by a diode limiter from the powerful microwave pulses. A quadrature mixer provides homodyne detection by referencing the echo signals to the microwave oscillator. The resulting in-phase and quadrature signals are sampled by a digital oscilloscope and transferred via GPIB to the computer,

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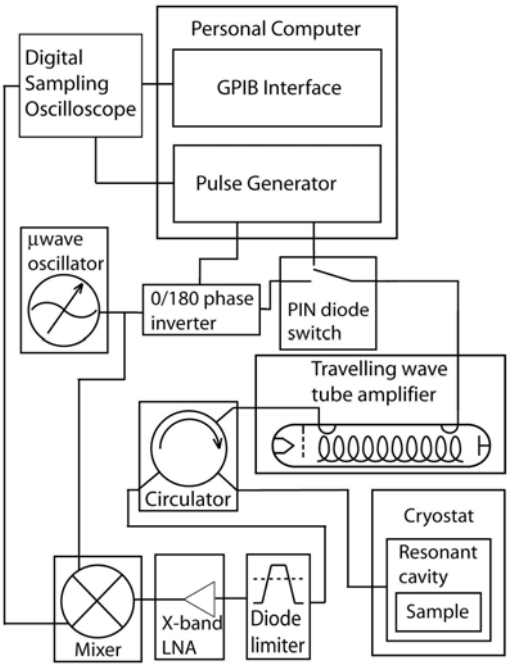

FIGURE 1. Block diagram showing key components of the pulse ESR spectrometer.

which is controlled by a C language program. The spectrometer design differs from other machines used to acquire electron spin echo data at cryogenic temperatures in three significant ways.[3] First, space constraints in our cryostat require the use of a coax rather than a waveguide connection to our cavity-type resonator. Second, our resonant cavity (fig. 2) is a modified CW ESR quartz-filled cavity designed to admit a funnel containing impurity-helium solid samples. The resonator operates in a TE_{011} mode that is coupled to the coax by a small fixed loop. To facilitate broadband excitation by short pulses, a chromium coated quartz ring is bonded with epoxy to thin fingers protruding from a brass pedestal. Adjusting the pedestal position then adjusts the Q factor of the cavity; we have found a Q of ~ 700 to be effective for detecting ESEEM modulation due to deuterium nuclei. At this setting a 90° pulse has duration 16 ns.

Finally, our spectrometer output is sampled by a sampling oscilloscope, not a boxcar integrator. Integrating a section of the echo peak recovers standard boxcar ESEEM data. Alternatively, for two–pulse ESEEM experiments the echo can be sliced into sections and the phase correction algorithm of Astashkin *et al*. applied.[4] For a typical echo width at half maximum of 250 ns, using this algorithm provides SNR improvements of 4.6 and 1.8 for modulation by

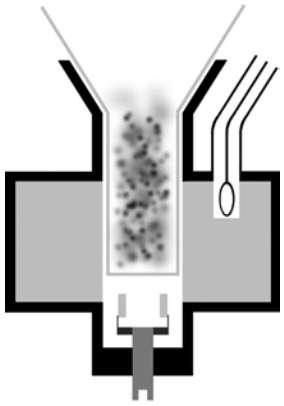

FIGURE 2. Cross section of our resonant microwave cavity, with funnel containing sample installed. Light gray shows the large synthetic fused silica ring and the small chromium plated quartz ring. Dark gray shows the brass screw. Black shows the copper walls and brass Q spoiler pedestal. The plane of the coupling loop, drawn as an ellipse, is actually perpendicular to the plane of the page. The loop connects the center conductor to the shield of the coax.

hydrogen and deuterium nuclei, respectively. This phase correction algorithm causes some distortion to two-pulse ESEEM data taken with short interpulse times τ. To avoid this distortion but retain signal to noise improvements, our two-pulse data is drawn from both boxcar and phase corrected data, with relative weightings changing with τ. Data transmission from the oscilloscope to the computer does not substantially slow the experiments because of the wait imposed by the long T_1 of atoms within impurity-helium solids, which is ~100 ms.

This spectrometer has been employed in the characterization of the molecular environment surrounding D atoms within deuterium-helium solids.[2]

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