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SQUID DETECTION OF EPR IN DILUTE CMN*  

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Abstract.— We demonstrate the applicability of SQUID magnetometers to the detection of EPR at milli-Kelvin temperatures. Data on a sample of dilute 1% CMN is presented. This method of detection of EPR is particularly well suited for systems with long spin-lattice relaxation times.  

The very high sensitivity of a SQUID magnetometer makes it an ideal instrument for every low temperature research where heat dissipation has to be low. One such application has been the detection of NMR with a SQUID. We present here a new application, that of detecting EPR at temperatures below 1 K. This novel technique is demonstrated on dilute polycrystal CMN, a system whose magnetic behavior has been well studied. The extension of conventional EPR to ultra low temperatures has been limited primarily because of the technical difficulties of sensitivity and heat dissipation. With a SQUID magnetometer it is possible to overcome some of these difficulties and hence to be able to observe EPR with simple techniques, especially in small magnetic fields.

The apparatus consists of a standard SQUID magnetometer operating in the feedback mode. A steady field \( H_0 \) is trapped in a niobium tube located inside the mixing chamber of a \(^3\)He-\(^4\)He dilution refrigerator. The sample, about 5 mg of powdered 1% CMN diluted with LMN, is placed inside the niobium tube and the magnetization of the sample is coupled to the SQUID by means of a flux transformer. Perpendicular to \( H_0 \) is a two-turn saddleshaped coil for producing an r.f. field \( H_1 \). Actually the set-up is similar to the one for SQUID NMR detection /1/. To reduce the coupling of \( H_1 \) to the SQUID, a low-pass filter is used as described in ref. /2/. It consists of a small resistor in parallel with the SQUID input. Such a filter avoids the problems of the silver tube shield used originally. To minimize transmission line resonances, the main problems encountered in this method, the r.f. coil inductance must be kept small and the line is terminated with 50 ohms. The sample resonance is detected by sweeping the r.f. field \( H_1 \) with a Wavetek Signal Generator Model 2001, with the SQUID output simply displayed on a x-y recorder. Figure 1 shows a typical result at \( 1/T = 38.5 \text{ K}^{-1} \).

Fig. 1: A typical EPR trace obtained for dilute 1% CMN. The SQUID output corresponds to a change seen in the parallel magnetization of the sample. This data was taken at a temperature of 26 mK, in a fixed magnetic field of 29.4 Oe. The frequency sweep was made over a time span of about 100 s.

Because the sample is in the form of a fine powder, the line represents a range of resonances from \( g_\parallel \approx 0 \) to \( g_\perp = 1.84 \). The maximum signal occurs at a frequency corresponding to \( g_\perp = 1.84 \). A single crystal would have shown a narrower line. The resonance was investigated for fields of 7.4 Oe to 64.8 Oe trapped in the niobium tube. The \( g \)-value that we obtain for the maximum signal is 1.84 ± .1 taken from the graph in figure 2.

The SQUID magnetometer responds to changes of magnetic flux which in this case is caused by a change of

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magnetization-$\Delta M$ of the sample on resonance such that

$$\Delta M = \frac{M_0 W \tau_1}{1 + W \frac{\gamma H}{1 + \frac{\gamma H}{2} \tau_2}}$$

(1)

where $W$ is $\gamma H^2$, $\tau_1$ the spin-lattice relaxation time, $\tau_2$ the spin-spin relaxation time, $M_0$ is the equilibrium magnetization, and $H$ the r.f. field which was varied between 1.28 Oe and 12.8 Oe.

For a typical flux noise figure of order $10^{-3}$, the fractional change in the magnetization $\Delta M / M_0$ is $1.6 \times 10^{-7}$ for a field of 100 Oe at 10 mK. A comparison can be made between conventional EPR and the method described here using an analysis similar to the NMR case presented in ref. /1/, the SQUID being most useful for low frequencies.

This method of detecting EPR was also applied to the measurements of spin-lattice relaxation times in the dilute 1% CMN sample. This was done by observing the change in magnetization after r.f. power on resonance was turned off. Relaxation in CMN is determined by a bottleneck between the phonons and the bath where the sample is placed. Our relaxation results are in agreement with other measurements /3/ obtained with different techniques they will be discussed in another paper.

Of major interest is the fact that our results demonstrate that EPR can be done down to 10 mK using a simple SQUID magnetometer. Because of the high sensitivity of such a magnetometer, very little power has to be dissipated to detect the resonance. This method is most useful for systems with broad lines and long spin-lattice relaxation times such as in the dilute CMN used here.

References

