ELECTRON SPIN RESONANCE MEASUREMENTS OF NiCl2-THIOUREA

R.D. McDonald (NHMFL, LANL), M. Jaime (NHMFL, LANL), C. Batista (T11, LANL), J. Singleton (NHMFL, LANL), A. Paduan-Filho (Instituto de Fisica, Universidade de Sao Paulo, Brazil)

The electron spin resonance spectrum of NiCl2-thiourea was measured at a temperature of 1.5 K, in the frequency range 7 to 40 GHz, with the magnetic field applied parallel to the tetragonal \( c \)-axis. These measurements were performed in order to verify the magnetic energy level structure, i.e. the values of the spin, \( S \), g-factor, \( g \), the exchange coupling, \( J \), and singlet-doublet separation energy, \( D \). A cylindrical gold cavity, measured in transmission, with E-field coupling so as to resonate in the Transverse Magnetic (TM) modes was used. The use of TM modes ensures a component of oscillating magnetic field perpendicular to the cylindrical axis of the cavity, which is parallel to the applied magnetic field, a necessary condition for observing electron spin resonance. The loaded Q-factor was in excess of 6000 for all the TM modes.

In NiCl2-thiourea the Nickel has a spin of one, however, the tetragonal symmetry causes a zero field splitting of the singlet \(|0\rangle\) and triplet states \(|\pm1\rangle\) which is greater than the antiferromagnetic exchange coupling, \( D > J \). As a result long range magnetic order does not occur at zero field, but magnetic field applied parallel to the \( c \)-axis will cause a crossing of the spin up and singlet states, which in turn gives rise to magnetic order at finite field. The magnetic susceptibility data of Armando Paduan-Filho et al. [1], indicates an antiferromagnetic ground state below a temperature of 1 K, in the field range 2-12 T. This suggests the following values of \( g = 2.26 \), \( D = 7.6 \) K and \( J = 4.5 \) K. The \( \Delta S \pm 1 \) selection rule implies that two ESR lines will be observed in the experimental frequency range, the first heading to zero frequency at 7.7 T at a rate of -31.64 GHz/T, the second emerging at 7.7 T at a rate of 31.64 GHz/T. This prediction is plotted as the red line in Figure 1.

Figure 2 shows the dips in Q-factor as a function of magnetic field corresponding to the ESR absorption for a selection of modes. As can be observed, although the lines have significant width, at frequencies below approximately 20 GHz only one line is observed. The blue squares in Figure 1 plot the frequency of the absorption dips as a function of field. Above 20 GHz, the gradients of the ESR lines agree with prediction. Moreover, the field at which the two lines combine is close to 7.7 T. Both of these facts suggest that the parameters of Armando Paduan-Filho et al. [1] are a reasonable basis for a preliminary understanding of this material's magnetic properties. The occurrence of a single ESR line below 20 GHz is not explicable by thermal broadening of isolated spin energy levels. It is hence expected to be linked to the energy scale of the spin interactions. It should also be noted that 20 GHz is equivalent to 1 K, the upper temperature of the antiferromagnetic order.

References