RELAXATION PHENOMENA IN $\text{CsCoCl}_3 \cdot 2 \text{H}_2\text{O}$

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Dynamic susceptibility measurements have been performed on a single crystal of $\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$ at liquid temperatures by means of a Hartshorn mutual inductance bridge. At the magnetic phase transition a maximum in $\tau_{\text{abs}}(H)$ has been observed. A jump in $\tau_{\text{abs}}(T)$ has been found at the λ -point of liquid helium.

Not much attention has been paid to relaxation phenomena at magnetic phase transitions up to now [1–6]. Much more information is available about e.g. the static magnetization in the transition region. We assume that the study on the dynamical susceptibility can give valuable additional information about ordered magnetic materials. Because one of us was involved in an extensive research on $\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$ [7], we decided to start relaxation measurements on this material.

The crystallographic space group of $\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$ is Pcca [8]. At a temperature of about 3.4 K magnetic ordering appears. The magnetic state has been described in [7]. Along the a -axis spins couple strongly anti-ferromagnetically and the moments are situated in the a - c plane, canted over approximately 10° from the c -axis ($D = 3.6$ K). They form linear Ising chains. In the a - c plane chains couple ferromagnetically ($J_c = +4.5$ K) and along the b -axis weakly anti-ferromagnetically ($J_b = -0.2$ K). At a critical field value H_c , applied in the a -direction, the weak coupling J_b is broken and in a small field range near H_c there appears a large change in the magnetization. With the aid of a Hartshorn mutual inductance bridge we have investigated the dynamical behaviour of this transition.

In the figs. 1 and 2 the dispersion and absorption, reduced on the zero field susceptibility, have been shown at a fixed frequency (275 Hz) as a function of the magnetic field in the temperature range $3.02 < T < 4.23$ K. At a temperature of about 3.4 K a peak in the dispersion and absorption is observed. By lowering the temperature these peaks become very large and shift to larger field values. At $T = 1.785$ K it is found that $\chi'/\chi_0(H=0) = 228$

Fig. 1. Field dependence of the dispersion at 275 Hz.

and $\chi''/\chi_0(H=0) = 43$ at the maximum. The maximum in dispersion and absorption (at about the same field) gives the critical field H_c . Our values of $H_c(T)$ are in very good agreement with those given in [7]. Fig. 2 shows the field dependence of the absorption. Two regions can be distinguished, separated by a relative minimum close to zero. For $T > 2.7$ K this minimum is zero, but for $T < 2.7$ K an increasing overlap of the two regions has been observed at decreasing temperature. In the lower field region the sharp peaks in the dispersion and absorption indicate that spin reversal occurs [7]. This process is possibly

Fig. 2. Field dependence of the absorption. The numbers of the curves correspond with the temperatures in fig. 1.

completed at about the field strength where $\chi''(H)$ has the minimum mentioned above. In the second field range the χ'' versus H curve shows a paramagnetic behaviour.

To investigate the relaxation behaviour we measured at various temperatures the dispersion and absorption as a function of frequency. For $T > T_\lambda$ (the λ -point of liquid helium) the curves deviate strongly from the ideal Casimir and Du Pré curves but for $T < T_\lambda$ a good agreement is found. This indicates that the heat conductivity within the sample influences largely the relaxation behaviour [9]. In fig. 3 $\tau_{\text{abs}}(H)$ is given at four temperatures; τ_{abs} is defined as $1/2\pi\nu_{\text{max}}$, with ν_{max} the frequency where $\chi''(\nu)$ has a maximum. At the critical field we found a maximum in τ_{abs} . At 3.23 K this maximum is 5.2 ms., at 3.33 K 17.7 ms. At temperatures close to $T_N(H=0)$, τ_{abs} is larger than 0.1 second and has not been measured up until now. The temperature dependence of τ_{abs} deviates strongly from common theories about paramagnetic spin-lattice relaxation times. In the temperature region where τ_{abs} could be determined we found a maximum in the τ_{abs} versus T curve at $T = T_N(H)$. For $T_\lambda < T < T_N(H)$ it has been observed that $\partial\tau_{\text{abs}}/\partial T > 0$. At T_λ a jump in the τ_{abs} versus T curve has been found as can be seen in fig. 3. Also the peak in the relaxation time changes considerably.

As a first interpretation of the observed relaxation phenomena we suppose that in the lower field range the spin reversal is responsible for this behaviour. Possibly this process takes place under the influence of lattice vibrations and in that case has to be considered as a special spin-lattice relaxation phenomenon. In the second field range we assume that the dynamical behaviour of the susceptibility is determined by other relaxation processes, to be compared with

Fig. 3. Field dependence of τ_{abs} at the temperatures: $\square$ 2.799 K, Δ 2.596 K, $\circ$ 2.186 K, ∇ 2.164 K.

well-known paramagnetic spin-lattice relaxation. For $T > T_\lambda$ it is not possible, for the whole field range, to describe the phenomena with one relaxation time. The heat transport in the sample already mentioned gives a broadening of the χ'' versus ν curve.

Measurements concerning the relaxation behaviour below T_λ will be published elsewhere.

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