LETTER TO THE EDITOR

THERMAL CONDUCTION EFFECTS IN SPIN–LATTICE RELAXATION EXPERIMENTS ON YTTERBIUM CHLORIDE HEXAHYDRATE

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The anomalous behaviour of the spin–lattice relaxation observed for single crystals of ytterbium chloride hexahydrate at fields stronger than 5 kOe is due to the poor heat transfer in the liquid-helium bath. The thermal conduction effects can be explained by means of a thermal conduction model for the relaxation.

1. Introduction

In a previous paper [1] we calculated the effect of thermal conduction in magnetic crystals and the surrounding medium on the field dependence of the spin–lattice relaxation time observed in dynamic susceptibility measurements and derived from the angular frequency at which the absorption curve is at a maximum.

We demonstrated that due to bad heat transfer the field dependence of the time constant so obtained may deviate strongly from that of the intrinsic spin–lattice relaxation time τ_{SL} .

At strong magnetic fields the time constant is much larger than τ_{SL} and even its field derivative becomes positive. Furthermore the calculated dispersion and absorption curves then deviate strongly from those predicted by the Casimir and Du Pré theory. Dynamic susceptibility measurements on ytterbium chloride hexahydrate ($\text{YbCl}_3 \cdot 6 \text{H}_2\text{O}$) show these characteristics for $H \gtrsim 5$ kOe [2] and in ref. 1 we suggested that this anomalous behaviour is likely to be due to bad heat transfer in the liquid helium. The aim of this paper is to show that this suggestion is correct.

We performed two series of dynamic susceptibility measurements on the same single crystals of $\text{YbCl}_3 \cdot 6 \text{H}_2\text{O}$ as were used for the measurements of Soeteman et al. [2]. In the first series a crystal was placed in direct contact with the liquid-helium bath. In the second series the sample was thermally isolated and

these measurements provide us with the time constants to be used in the thermal conduction model (see ref. 1 and references therein). With this model we calculated the effect of the bad heat transfer in the liquid helium on the field dependence of the relaxation time constant and were able to explain the anomalous strong-field behaviour in the first series observed.

2. Measurements and calculations

The measurements on a single crystal of $\text{YbCl}_3 \cdot 6 \text{H}_2\text{O}$ (size $6.0 \times 4.1 \times 2.0$ mm) were performed at 4.2 K and in fields up to 20 kOe. The field was applied parallel to the preferred magnetic axis. The results of both series of measurements are presented in fig. 1. The time constants of the liquid-helium measurements (τ_{liq}) are qualitatively in agreement with those of Soeteman et al. The deviation parameter (d_{liq}), which is a measure for the deviations with respect to the Debye shape, becomes large at strong fields. The measurements on the thermally isolated crystal (in fact in helium gas with $p < 10^{-4}$ Torr) show ideally shaped curves throughout the whole field range ($d_{vac} \approx 0$). The time constants (τ_{vac}) are shortened by a factor $C_L/(C_H + C_L)$ compared to the spin–lattice relaxation time τ_{SL} , whereby C_L is the lattice specific heat and C_H the spin specific heat at constant field. Because $C_L/(C_H + C_L)$ can be easily

Fig. 1. Relaxation time constants and deviation parameters of $\text{YbCl}_3 \cdot 6 \text{H}_2\text{O}$ at $T = 4.2$ K versus the applied magnetic field. ∇ : τ_{liq} , d_{liq} , liquid-helium contact; $\circ$: τ_{vac} , d_{vac} , thermally isolated crystal; $\bullet$: τ_{SL} , spin-lattice relaxation time ($\tau_{SL} = \tau_{vac}(C_H + C_L)/C_L$); $\circ$: τ_{calc} , d_{calc} , calculations for the spherical geometry ($r_0 = 1.5$ mm); $\triangle$: τ_{calc} , d_{calc} , calculations for the cylindrical geometry ($r_0 = 1.5$ mm); $\square$: τ_{calc} , d_{calc} , calculations for the plate-like geometry ($r_0 = 1.0$ mm). For $H \gtrsim 4$ kOe, $\tau_{calc} = \tau_{liq} = \tau_{SL}$ and $d_{calc} = 0$.

derived from our measurements, τ_{SL} can be determined. It is seen from the figure that in the strong-field range the difference between the values for τ_{SL} and τ_{liq} becomes large. Using the thermal conduction model we calculated the influence of heat transfer in the crystal and the liquid-helium bath. The spin system is characterized by the specific heat at constant magnetization: $C_M = 8.7 \text{ J/m}^3 \text{ K}$ ($T = 4.2$ K), $C_H - C_M = \mu_0 CH^2/T^2$, with $C = 2.46 \times 10^{-1} \text{ K}$ (Curie constant). The lattice specific heat was derived from the low-frequency final value of the dispersion curve for the isolated crystal. It was found that $C_L = 1.04 \times 10^3 \text{ J/m}^3 \text{ K}$ at $T = 4.2$ K, which is in reasonable agreement with the lattice specific heat of $\text{GdCl}_3 \cdot 6 \text{H}_2\text{O}$, found as $1115 \text{ J/m}^3 \text{ K}$ at $T = 4.2$ K [3]. We took the arbitrary value of 100 W/Km for the thermal conductivity of the lattice (λ_L), which can usually be expected for aqueous compounds. The properties of liquid helium can be found in text books. We used $C_g = 5.60 \times 10^5 \text{ J/m}^3 \text{ K}$ and $\lambda_g = 3.02 \times 10^{-2} \text{ W/Km}$. The thermal contact between crystal and bath was assumed to be very good.

The calculations were performed for three ideal configurations representing the shape of the crystal: a plate-like configuration with $r_0 = 1.0$ mm, a cylindrical configuration with $r_0 = 1.5$ mm and a spherical configuration with $r_0 = 1.5$ mm.

The results of the influence of the thermal conduction of the spin-lattice time constant is presented in the figure (τ_{calc}). It can be seen that the spherical approximation gives the best results. The deviation parameter is in all three cases somewhat larger than the experimental one.

In the calculations the thermal conductivity of the lattice was taken as 100 W/Km . A reduction of this value by a factor 10 does not have any influence on the calculated time constants and deviation parameters. Obviously the lattice thermal conductivity is not critical. We also varied the value of r_0 in the calculations. The rapid increase of τ_{calc} in the strong-field range takes place at stronger fields when r_0 decreases. Now we can explain why the time constants of Soeteman et al. increase at strong fields whereas in our liquid-helium measurements such an increase is not found for fields up to 20 kOe. Only a part of their crystal was used for our measurements. The value for the effective radius used in the calculations is in a reasonable agreement with the crystal size.

3. Conclusion

The conclusion from our measurements and calculations is that the observed anomalous behaviour of the relaxation time constant of $\text{YbCl}_3 \cdot 6 \text{H}_2\text{O}$ for $H \gtrsim 5$ kOe when the crystal is placed in liquid helium is not due to an intrinsic relaxation process but a result of bad heat transfer in the liquid-helium bath. The spin–lattice relaxation time constant varies in this field range with $H^{-\alpha}$, where $\alpha \simeq 2$.

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References

- [1] J. Flokstra, G.J. Gerritsma and L.C. van der Marel, *Physica* 92B (1977) 347.
- [2] J. Soeteman, L. Bevaart and A.J. van Duyneveldt, *Physica* 74 (1974) 126.
- [3] K.H. Hellwege, F. Kch, K. Niemann and W. Pfeffer, *Z. Physik* 162 (1961) 358.