THE INFLUENCE OF THERMAL CONDUCTION ON THE FIELD DEPENDENCE IN SPIN–LATTICE RELAXATION EXPERIMENTS

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It is demonstrated that heat transfer in magnetic samples and the surrounding liquid helium leads to a field dependence of the observed relaxation time in low-frequency relaxation measurements which deviates strongly from that of the intrinsic spin–lattice relaxation.

1. Introduction

The dispersion and absorption curves, obtained in low-frequency relaxation experiments on paramagnetic crystals in a liquid-helium bath having a temperature above the helium transition temperature T_λ , strongly deviate in many cases from the shape of the Casimir and Du Pré curves. The deviations are considerably smaller for $T < T_\lambda$. In a previous paper [1] we introduced a thermodynamic model for the relaxation (the thermal conduction model) involving the heat properties of the environment of the crystal. With this model it was possible to explain satisfactorily the shape of the susceptibility curves for $T > T_\lambda$ as well as for $T < T_\lambda$. Assuming a direct process for the spin–lattice relaxation ($\tau_s \propto T^{-1}$), it was calculated that the temperature dependence of the time constant τ_{abs} obtained from the absorption curve in the conventional way may differ greatly from T^{-1} . It was found that $\tau_{\text{abs}} \propto T^{-\alpha}$ with $1 < \alpha \leq 6$, the large values for α being due to bad thermal conduction in the lattice or the surrounding medium.

In the present paper the influence of the above-mentioned thermal effects on the field dependence of the relaxation time constant is investigated. The field dependence of τ_{abs} and of the parameter d , which describes the deviations of the measured absorption curve with respect to the absorption curve according to the Casimir and Du Pré theory, shows a number of

characteristic features for many compounds. As a typical example experimental results on $\text{YbCl}_3 \cdot 6\text{H}_2\text{O}$ taken from ref. 2 are presented (fig. 1). Corrections were made for the anisotropy of the relaxation time constant. It is seen that the deviation parameter d in the strong-field range increases rapidly with increasing field and that the slope of the τ vs. H curve may change in sign in this field range. The radius of the sample has a great influence on τ_{abs} and d .

Using the thermal conduction model we calculate in this paper that the experimental results $\tau_{\text{abs}}(H)$ and $d(H)$ at strong fields may be due to bad heat conduction in the lattice and the surrounding medium.

2. The thermal conduction model and the calculations

A diagram of the thermal conduction model is given in fig. 2. The spin system (specific heat C_H) is coupled to the lattice (C_L , thermal conductivity λ_L) by the heat transfer coefficient α . The thermal contact resistance between lattice and bath (C_g, λ_g) is denoted by R_K . The expression for the susceptibility of a sample with a characteristic dimension r_0 is given in ref. 1. Apart from scaling factors the dispersion and absorption are fully determined by the dimensionless parameters D and R (ratios of specific heats) and P , Q and S (ratios of coefficients of heat transfer). The field-dependent parameters are C_H and α ($= C_H/\tau_s$)

Fig. 1. Relaxation time constants and deviation parameters of $\text{YbCl}_3 \cdot 6\text{H}_2\text{O}$ as a function of the magnetic field at 4.2 K. The data are taken from ref. 2. $\circ$ = powdered sample; $\square$ = single crystal (orientation angle $\theta = 30^\circ$). For the powdered sample we subtracted 0.15 from the value of d and we fitted the $\tau_{\text{abs}}(H)$ curve to that of the single crystal at 1 kOe in order to compensate for the anisotropy effect.

and therefore also D , R and P . It is usual to write C_H as

$$C_H = \frac{b}{T^2} + \mu_0 C \frac{H^2}{T^2},$$

where C is the Curie constant and b a constant related to an internal field by $b = \mu_0 C H_{\text{int}}^2$. If D_0 and R_0 denote the values of D and R at $H = 0$ then

$$D = C_H/C_L = D_0[1 + H^2/H_{\text{int}}^2],$$

Fig. 2. The thermal conduction model and the dimensionless parameters.

$$R = C_g/C_H = R_0[1 + H^2/H_{\text{int}}^2]^{-1}.$$

It is assumed that the spin–lattice relaxation time τ_s is a constant τ_{s0} at weak magnetic fields and that it is determined by the direct process at strong fields so that

$$\tau_s^{-1} = \tau_{s0}^{-1} + AH^4T.$$

This can be also written as

$$\tau_s = \tau_{s0}[1 + \epsilon H^4/H_{\text{int}}^4]^{-1},$$

where $\epsilon = AH_{\text{int}}^4 T \tau_{s0}$ is independent of the field. For P the following expression can be derived:

$$P = P_0[1 + H^2/H_{\text{int}}^2][1 + \epsilon H^4/H_{\text{int}}^4],$$

where P_0 is the value of P at $H = 0$. The dimensionless parameters P and D increase rapidly with increasing field for $H > H_{\text{int}}$, while R decreases. In ref. 1 it is shown that a large magnitude of P and D results in strongly broadened absorption curves for which τ_{abs} may deviate considerably from τ_s , so these phenomena can also be expected in the present calculations.

The field dependence of τ_{abs} and d was calculated for various values of the parameters. As a typical example the results for $D_0 = 1$, $R_0 = 1000$, $S = 10$, $Q = 0$ and $P = 10, 1, 0.1, 0.01$, respectively, are presented in fig. 3. Hence, the thermal resistance between crystal and bath is taken as zero. The arbitrary value of 1 was taken for ϵ , so that the direct process becomes effective at $H = H_{\text{int}}$. The different values for P_0 represent a variation of the crystal size. The values for the parameters correspond to realistic, experimental circumstances, although they do not apply to a particular compound. It is seen that the

Fig. 3. The field dependence of τ_{abs} and d of the calculated susceptibility curves. $H_r = H/H_{int}$, $D_0 = 1$, $R_0 = 1000$, $S = 10$, $Q = 0$, $\epsilon = 1$. $\circ = P_0 = 10$; $\triangle = P_0 = 1$; $\square = P_0 = 0.1$; $\bullet = P_0 = 0.01$. The dashed curve corresponds to the spin-lattice relaxation time τ_s .

smaller the value of P_0 , the larger the range for which τ_{abs} follows the spin-lattice relaxation time $\tau_s(H)$. At a certain field $\tau_{abs}(H)$ reaches a minimum value and then increases rapidly with increasing field. For $P_0 = 10$, however, τ_{abs} is considerably larger than τ_s even for $H < H_{int}$ and the minimum in $\tau_{abs}(H)$ does not occur. The deviation parameter becomes

very large in the field range around the minimum in the $\tau_{abs}(H)$ curve. At strong fields d again decreases.

3. Discussion

The calculated curves in fig. 3 show a great resemblance to the experimental results in fig. 1. Both figures exhibit a considerable size effect. The parameter P is proportional to the square of the radius of the crystal, so small crystals (e.g. powdered samples) lead to time constants almost equal to τ_s while large crystals give large deviations between τ_{abs} and τ_s . The shape of the calculated curves for strong fields is qualitatively in agreement with the characteristic shape of $\tau_{abs}(H)$ and $d(H)$ of $\text{YbCl}_3 \cdot 6\text{H}_2\text{O}$. The $\tau(H)$ curve of the powdered sample does not show a minimum at strong fields and also the deviation parameter is still small. It may be expected, however, that at still stronger fields a minimum will occur while d then reaches a large value.

At weak magnetic fields the calculated susceptibility curves are in agreement with the Casimir and Du Pré theory. In this case τ_{abs} is equal to τ_s and $d = 0$. Deviations occur when P has a large value. The calculated susceptibility curves are separated into two parts at very strong magnetic fields. The high-frequency part corresponds to the relaxation to the isolated lattice but when $C_H \gg C_L$ the effect on the susceptibility becomes very small. The low-frequency part represents the relaxation due to thermal conduction effects. (In the example of fig. 3 the rate-determining process in the energy transport is the thermal conduction in the liquid helium because $\lambda_L \gg \lambda_g$.) This relaxation process dominates in the susceptibility when $C_H \gg C_L$. The deviation parameter is then rather small. In the transition region between the weak and strong-field behaviour a maximum in the $d(H)$ curve occurs and τ_{abs} already deviates considerably from τ_s . Hence, τ_{abs} contains hardly any information about the spin-lattice relaxation for field values which are larger than the field where the deviation parameter increases rapidly. On the contrary, it is a time constant which characterizes the thermal transport in lattice or bath.

In our opinion it is unlikely that the time constants for the single crystal of $\text{YbCl}_3 \cdot 6\text{H}_2\text{O}$ represent a spin-lattice relaxation process for $H > 2$ kOe. It is necessary to study large single crystals of magnetically

concentrated compounds with the so-called “liquid-vacuum” method [3] in order to be able to separate the effect of thermal conduction from the intrinsic spin–lattice relaxation phenomena.

References

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