SOVIET PHYSICS JETP VOLUME 34(7), NUMBER 5 NOVEMBER, 1958

DETERMINATION OF SPIN-LATTICE RELAXATION TIME FROM PARALLEL-FIELD ABSORPTION CURVE

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Submitted to JETP editor November 30, 1957

J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 1090-1092 (May, 1958)

It is shown that the spin-lattice relaxation time can be determined from the paramagnetic absorption curve in parallel fields. The method has been verified using the normal paramagnetic material $MnSO_4 \cdot 4H_2O$. Results of measurement of ρ at low temperature for various values of the constant magnetic field are presented for the salts $CrK(SO_4)_2 \cdot 12H_2O$, $FeNH_4(SO_4)_2 \cdot 12H_2O$ and $CuSO_4 \cdot 5H_2O$ for which the values of ρ have hitherto been unknown.

1. As is well known (cf. Ref. 1), at sufficiently low frequencies paramagnetic absorption in parallel fields is due exclusively to spin-lattice relaxation and is described by the expression

$$\chi'' / \chi_0 m = \rho\nu F / (1 + \rho^2\nu^2), \tag{1}$$

where χ'' is the imaginary part of the complex magnetic susceptibility, χ_0 is the equilibrium susceptibility per unit mass, m is the mass of the sample, ρ is the spin-lattice relaxation time, ν is the frequency of the oscillation field, and F is a function of the fixed field. We have used Eq. (1) to measure ρ. The absorption curves were observed using the Zavoiskii method;[2] the apparatus is described in detail in Ref. 3.

2. For convenience we write Eq. (1) in the form

$$\chi''(H_c) = FD, \tag{2}$$

where (cf. Ref. 1)

$$F = H_c^2 (b/c + H_c^2)^{-1}, \tag{3}$$

$$D \equiv \rho\nu/(1 + \rho^2\nu^2), \tag{4}$$

$$\chi''(H_c) \equiv \chi''(\chi_0 m)^{-1}. \tag{5}$$

Here H_c is the fixed magnetic field, b is the heat capacity of the spin system, and c is the Curie constant.

The Debye function D is a function of frequency ν and the fixed field H_c since[4]

$$\rho = \rho_0 (b/c + H_c^2) / (b/c + pH_c^2), \tag{6}$$

where $p < 1$. It is apparent that D will be an increasing function of the field H_c if the frequency ν and region of variation of H_c are chosen in such a way that $\rho\nu < 1$; conversely this quantity is a decreasing function if $\rho\nu > 1$. For a certain value of ν and region of variation of H_c the function $D(H_c)$ will have a maximum. Consequently (cf. Ref. 5) the absorption curve $\chi''(H_c)$, given by Eq. (2), depending on the choice of frequency ν and region of variation of H_c, can be increasing (if $\rho\nu < 1$) and have a maximum (if $\rho\nu > 1$). This means that even if we do not analyze the absorption curve, but know its shape and the oscillator frequency, it is possible to find the spin-lattice relaxation time ρ. For an increasing absorption function $\chi''(H_c)$ we have $\rho < \nu^{-1}$; for an absorption function having a maximum, $\rho > \nu^{-1}$.

3. Suppose now that the absorption function $\chi''(H_c)$ is measured in the sample material

$$\chi''(H_c) = FD \tag{2}$$

and in a reference material (in which ρ_{ref} is known):

$$\chi''(H_c)_{ref} = F_{ref} D_{ref}. \tag{7}$$

From Eqs. (2) and (7) we have

$$D = \chi''(H_c) F_{ref} D_{ref} / \chi''(H_c)_{ref} F. \tag{8}$$

The right-hand side of Eq. (8) is known since the ratio of the quantities $\chi''(H_c)$ and $\chi''(H_c)_{ref}$ is equal to the ratio of the corresponding readings in the grid circuit (cf. Ref. 2). Thus, using Eq. (8) the quantity D can be found from measurements; whence, using Eq. (4), for ρ we have:

$$\rho = (1/\nu)\left(1/2D \pm \sqrt{(2D)^{-2} - 1}\right). \tag{9}$$

We rewrite Eq. (9) in the form

$$\rho\nu = K \pm \sqrt{K^2 - 1}, \tag{10}$$

where $K \equiv (2D)^{-1}$; it is easy to see that $K > 1$ both when $\rho\nu < 1$ and $\rho\nu > 1$. For an increasing absorption curve, or what is the same thing, for an increasing Debye function D, we have $\rho\nu < 1$,

TABLE I

Frequency Mcs	1.0		1.95		7.4		13	
H_c, Oe	D	$\rho \cdot 10^7$ sec	D	$\rho \cdot 10^7$ sec	D	$\rho \cdot 10^7$ sec	D	$\rho \cdot 10^7$ sec
800	0.30	3.4	0.45	3.2	0.38	3.4	0.20	3.4
1600	0.35	4.1	0.47	3.6	0.30	3.6	0.18	4.1
2400	0.41	5.2	0.50	5.1	0.23	5.2	0.14	5.2
3200	0.45	6.4	0.49	6.2	0.18	7.0	0.12	6.4
4000	0.47	7.5	0.45	8.0	0.16	7.6	0.10	7.5
4800	0.49	8.2	0.44	8.5	0.15	8.3	0.09	8.6
5600	0.50	10.0	0.42	10.4	0.14	9.5	0.08	9.6

TABLE II

Frequency Mcs	18		7.4		18	
Material	$CrK(SO_4)_2 \cdot 12H_2O$		$Fe(NH_4)(SO_4)_2 \cdot 12H_2O$		$CuSO_4 \cdot 5H_2O$	
H_c, Oe	D	$\rho \cdot 10^8$ sec	D	$\rho \cdot 10^8$ sec	D	$\rho \cdot 10^8$ sec
800	0.05	0.3	0.12	1.7	0.10	0.5
1600	0.10	0.5	0.14	2.0	0.18	1.1
2400	0.15	0.8	0.14	2.0	0.25	1.5
3200	0.19	1.0	0.15	2.1	0.31	1.9
4000	0.21	1.2	0.15	2.1	0.34	2.1
4800	0.22	1.3	0.16	2.2	0.35	2.3
5600	0.24	1.4	0.16	2.2	0.37	2.5
6400	0.24	1.4	0.16	2.2	0.38	2.6
	$\rho_0 = 0.13$ $\rho_\infty = 1.7$ $p = 0.08$		$\rho_0 = 0.9$ $\rho_\infty = 2.3$ $p = 0.4$		$\rho_0 = 0.13$ $\rho_\infty = 2.9$ $p = 0.08$	

so that the minus sign must be taken in (9); for a decreasing Debye function $\rho\nu > 1$ in which case the plus sign must be taken in (9).

4. The method proposed here for measuring ρ was verified experimentally in $MnSO_4 \cdot 4H_2O$. The results of the measurement of D and the calculation of ρ at various frequencies are shown in Table I. It is apparent from the table that the Debye function is an increasing function at 1 Mcs and a decreasing function at 7.4 and 13 Mcs; at 1.95 Mcs the function exhibits a maximum. The quantity ρ was computed from Eq. (9) using the proper sign (cf. above, end of Sec. 3). As is apparent from the table the values of ρ found at different frequencies are in good internal agreement. They are also are in good agreement with the values obtained by other methods (cf. Ref. 1).

5. Using the above method, measurements have been made at room temperature of the values of ρ in $CrK(SO_4)_2 \cdot 12H_2O$, $FeNH_4(SO_4)_2 \cdot 12H_2O$ and $CuSO_4 \cdot 5H_2O$. The results of these measurements are shown in Table II. No room-temperature measurements of ρ were ever made for these materials, nor was its value known at all for copper sulfate. The quantity ρ_0, the spin-lattice relaxation time at $H_C = 0$, and the parameter p were computed from the measurements using Eq. (6) while ρ_∞ is ρ_0/p and characterizes the spin-lattice relaxation time for $H_C \to \infty$.

6. In all cases the reference material was $Mn(NH_4)_2(SO_4)_2 \cdot 6H_2O$.

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Translated by H. Lashinsky