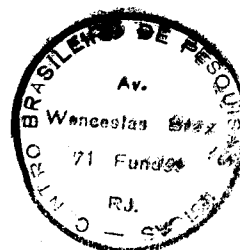


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THERMAL BEHAVIOUR OF THE ESR RELAXATION  
TIME IN SLIGHTLY DIRTY SUPERCONDUCTORS

Georges Schwachheim, Sydney Francisco Machado and Constantino Tsallis



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by

GEORGES SCHWACHHEIM  
SYDNEY FRANCISCO MACHADO  
CONSTANTINO TSALLIS

Centro Brasileiro de Pesquisas Físicas/CNPq  
Av. Wenceslau Braz, 71 - Botafogo  
Rio de Janeiro - RJ, BRAZIL

ABSTRACT

The thermal behaviour of the ESR relaxation rate in slightly dirty superconductors is discussed for both exchange and spin-orbit interactions between the conduction electrons and the impurities. The sensibility to the electronic density of states is exhibited by using, in a modified BCS framework, an heuristic analytic form which avoids two of three defects of a previous similar treatment. The sudden increase (decrease) of the relaxation rate immediately below the critical temperature for the exchange (spin-orbit) case is confirmed. Reasonable agreement with experimental data in  $\text{LaRu}_2:\text{Gd}$  is obtained.

RESUMÉ

On discute le comportement thermique du taux de relaxation de la résonance de spin électronique dans des supraconducteurs contenant des impuretés diluées, dans les cas d'interactions d'échange ou de spin-orbite entre les électrons de conduction et les impuretés. On montre la sensibilité à la densité d'états électroniques en utilisant, dans un contexte BCS modifié, une forme analytique heuristique qui évite deux parmi trois défauts d'un traitement semblable antérieur. On confirme l'augmentation (diminution) rapide du taux de relaxation juste en dessous de la température critique dans le cas d'échange (spin-orbite). Un accord raisonnable est obtenu avec les résultats expérimentaux existant pour  $\text{LaRu}_2:\text{Gd}$ .

## I- INTRODUCTION

Both experimental <sup>(1,2)</sup> and theoretical <sup>(1,3-10)</sup> approaches of electron spin resonance (ESR) in dirty superconductors have been attempted in last years; however none of them seems to have unambiguously clarified the corresponding microscopic situation as, in both experimental and theoretical grounds, exist several difficulties <sup>(8,11)</sup> which do not look like very easy to overcome.

The present work might be considered as the continuation of paper <sup>(10)</sup>, to which we shall continuously refer and where a quick discussion of the above bibliography is presented. In that paper a calculation is performed of the contributions to the ESR line width due to exchange and spin-orbit scatterings of conduction electrons by dilute impurities, through the normal-superconducting phase transition. That calculation was performed in a modified BCS <sup>(12)</sup> framework, and in principle the results concern only the type I-superconductors; however, it should not be very surprising if they held (at least qualitatively) even for type II - superconductors. The main result of that theory is that, immediately below the critical temperature  $T_0$ , the linewidth suddenly increases (decreases) with respect to its value in the normal state, for the exchange (spin-orbit) mechanism. This result had already been obtained by Maki <sup>(9)</sup> within a quite different framework, and seems to be supported by the experiences performed by Hebel and Slichter <sup>(1)</sup> and by Rettori et al <sup>(2)</sup>.

In the theory developed in Ref. (10) the electronic density of states  $\rho$  plays a key-role; however it is unsatisfactory in at least three points, namely:

- 1)  $\rho$  presents, for energies above the gap value, some unphysical oscillations
- 2)  $\rho$  does not contain the BCS density of states  $\rho_s$  as a particular case;
- 3) for the spin-orbit mechanism, the linewidth takes, for sufficiently low temperatures ( $T \ll T_0$ ) unphysical negative values.

In the present paper we introduce an heuristic density of states  $\rho$  which avoids the two first difficulties; unfortunately we have not succeeded in avoiding the third one, so the theory remains unacceptable for  $T \ll T_0$ , at least for the spin-orbit mechanism.

## II - RELAXATION RATE AND DENSITY OF STATES

Let us define the ESR relaxations rate (or reduced line-width)  $R_{\pm}$  as follows

$$R_{\pm} \equiv \frac{(T_2^S)^{-1}}{(T_2^N)^{-1}}$$

where  $T_2^N$  and  $T_2^S$  are the transverse relaxation times in the normal and superconducting phases respectively, and where the plus (minus) sign holds for exchange (spin-orbit) interaction. Within the theoretical framework of Ref. (10) we have that

$$R_{\pm} = \frac{\int_0^{\infty} dx [f(x)]^2 (1 \pm x^2) \cosh^{-2} \delta x}{\int_0^{\infty} dx f(x) \cosh^{-2} \delta x} \quad [1]$$

with  $\delta \equiv \Delta(T)/2k_B T$  and  $f(E/\Delta) \equiv \rho(E)/2\rho_F$ , where  $\Delta(T)$  is the temperature-dependent variational BCS energy gap (see, for example, Ref. (13)),  $E$  is the electronic energy measured from the Fermi level,  $\rho(E)$  is the density of electronic states in the superconducting phase and  $\rho_F$  is its value at the Fermi surface in the normal phase (without consideration of the spin degeneracy). The heuristic reduced density of states  $f(x)$  is demanded to satisfy the following reasonable restrictions:

a) for  $E \gg \delta$  we must reobtain the standard BCS density of states, i.e.

$$x \rightarrow \infty \Rightarrow f(x) \sim f_s(x)$$

$$\text{where } f_s(x) \equiv \frac{x}{\sqrt{x^2 - 1}} \quad \text{if } x > 1$$

$$\equiv 0 \quad \text{otherwise}$$

b)  $f(x)$  is maximal at  $x = 1$ ;

c)  $f(x)$  strongly vanishes at the limit  $x \rightarrow 0$ ;

- d)  $f(x)$  identically vanishes if  $x \leq 0$ , and is otherwise positive;
- e)  $f(x)$  conserves the total number of states, even after elimination of its divergent part, in other words
- $$\int_0^1 dx f(x) + \int_1^{\infty} dx [f(x) - 1] = \int_1^{\infty} dx [f_s(x) - 1] = 1$$
- f)  $f(x)$  leads, through the integration process, to  $R_{\pm} = 1$  in the limit  $\delta \rightarrow 0$ ;
- g)  $f(x)$  monotonically increases in the interval  $(0, 1)$  and monotonically decreases in the interval  $(1, \infty)$ ;
- h) the family of densities of states  $f(x, h)$  (where  $h$  is a parameter) must contain  $f_s(x)$  as a particular case (which, by anticipating our future choice, will be  $h \rightarrow \infty$ ).

Let us remark that only the two last restrictions are new with respect to the set of restrictions already used in Ref. (10). Our proposal will be

$$\begin{aligned} f(x) &= (a - bx - cx^2) e^{-d/x} && \text{if } x \leq x_0 \\ &= f_s(x) = \frac{x}{\sqrt{x^2 - 1}} && \text{if } x \geq x_0 \\ &= 0 && \text{if } x \leq 0 \end{aligned}$$

where, as we shall see, it will always be  $x_0 \gg 1$  ( $x_0 = 1$  corresponds to the particular case  $f_s(x)$ ) and where the minus signs attached to  $b$ ,  $c$  and  $d$  have been introduced for future convenience. Furthermore we shall impose continuity of the function and of its first derivative at the junction point  $x_0$ , i.e.

$$(a - bx_0 - cx_0^2) e^{-d/x_0} = \frac{x_0}{\sqrt{x_0^2 - 1}} \quad [2a]$$

$$[2cx_0^3 + (b+cd)x_0^2 + bdx_0 - ad]e^{-d/x_0} = \frac{x_0^2}{(x_0^2 - 1)^{3/2}} \quad [2.b]$$

Let us call  $h$  the maximal value (or height) of  $f(x)$ , which, as a consequence of restrictions (b) and (g), will occur at  $x = 1$ , i.e.

$$h = (a - b - c)e^{-d} \quad [2.c]$$

Furthermore, restriction (b) leads to

$$b + 2c - d = 0 \quad [2.d]$$

and restriction (e) may be rewritten as follows

$$\int_0^{x_0} dx (a - bx - cx^2) e^{-d/x} = \sqrt{x_0^2 - 1} \quad [2.e]$$

Let us now synthesize: we have 6 unknown parameters ( $a, b, c, d, x_0$  and  $h$ ) associated to our choice of  $f(x)$  and 5 relations between them ( $[2.a]$  to  $[2.e]$ ), so we have only one degree of freedom which, for commodity, we shall associate to an arbitrary value of the height  $h$ , which in principle may take any value superior to one (the limiting situation  $h \rightarrow \infty$  will reproduce the BCS density of states  $f(x)$ ). It is easy to verify that the whole set of restrictions (a) to (h)



are now satisfied. In Figure 1 typical densities of states are presented. The substitution of a given  $\rho(x)$  into relation [1] gives the relaxation rate  $R$  as a function of  $\delta$  (typical examples are indicated in Table 1). In order to have  $R \pm$  as a function of temperature  $T$ , we must explicitly use

$$\delta(T) \equiv \frac{\Delta(T)}{2k_B T} = \frac{\Delta(0)}{2k_B T_0} \frac{\Delta(T)/\Delta(0)}{T/T_0} \equiv \frac{\Delta(0)}{2k_B T_0} z(T/T_0)$$

Typically it is  $\Delta(0)/2k_B T_0 \approx 3.5$  (see, for example, Ref. (13)) and  $z(T/T_0)$  may be straightforwardly obtained from the BCS theory (see, for example, Ref. (13)) and has been represented in Fig. 2.

### III - COMPARISON WITH EXPERIENCE AND CONCLUSION

In order to exhibit the "bridge" between the present theory and experience, we shall use the results obtained by Rettori et al<sup>(2)</sup> in  $\text{La Ru}_2: \text{Gd}$ , which are represented (after renormalization by the normal-phase linear behaviour) in Fig. 3. As both exchange and spin-orbit mechanisms are present in the experimental line width we propose

$$R(T) = W R_+(T, h) + (1 - W) R_-(T, h)$$

where  $W$  gives the relative weigh of those contributions. We have in this way 2 parameters ( $W$  and  $h$ ) which we shall determine by comparison with experience. A reasonable fit has been obtained for  $W \approx 0.29$  and  $h \approx 3$  (see Fig. 3) (for  $\Delta(0)/2k_B T_0$  we have used the typical value 3.5).

Let us conclude by saying that the present work:

- a) confirms<sup>(10)</sup> the great sensibility of the thermal behaviour of the relaxation rate with respect to the density of states;
- b) exhibits the possibility of the sudden increase or decrease of the relaxation rate immediately below the critical temperature, being

due to density of states effects together with the fact that the interaction Hamiltonian<sup>(10)</sup> behaves, with respect to time inversion into the electronic degrees of freedom, differently for exchange or spin-orbit mechanisms (its sign changes in the exchange case whereas it remains invariant in the spin-orbit case);

- c) does not explicitly take into account the external ESR static magnetic field  $H$ , however we believe that essentially this should not bring down other effects than the standard dislocation of the critical temperature (this is to say, in the present theory we may interpret  $T_0$  as  $T_0(H)$ );
- d) leads to a reasonable fit with the experimental results<sup>(2)</sup> in  $\text{La Ru}_2\text{: Gd}$ ;
- e) unfortunately has not been able to avoid the unphysical negative values of the relaxation rate that appear at sufficiently low temperatures in the spin-orbit case.

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CAPTION FOR FIGURES AND TABLES

Fig. 1 - Three typical reduced densities of states (we recall that  $x \equiv E/\Delta$  and  $f \equiv \rho/2\rho_F$ ): (a)  $h = 1.5$  (hence  $a = 5.216$ ,  $b = 2.815$ ,  $c = 0.4729$ ,  $d = 0.6502$  and  $x_0 = 1.584$ ), (b)  $h = 2$  (hence  $a = 14.49$ ,  $b = 3.409$ ,  $c = 3.427$ ,  $d = 1.342$  and  $x_0 = 1.239$ ), (c)  $h = 3$  (hence  $a = 2292$ ,  $b = 1855$ ,  $c = 40.13$ ,  $d = 4.884$  and  $x_0 = 1.092$ )

Fig. 2 - The renormalized BCS energy gap  $z \equiv T_0 \Delta(T)/T \Delta(0)$  as a function of the reduced temperature  $T/T_0$ .

Fig. 3 - The thermal behaviour of the relaxation rate.

Experimental points<sup>(2)</sup>:  $\text{LaRu}_2$ : Gd samples.

Solid line: present theory with  $h = 3$  and  $W = 0.29$ .

Table 1 - Typical values of the relaxation rate as a function of the reduced energy gap  $\delta$  (we recall that  $\delta \equiv \Delta/2k_B T$  and that  $R_+$  ( $R_-$ ) corresponds to the exchange (spin-orbit) case).

The three values with (\*) as well as the zeros have been obtained by extrapolating the analytical expression of  $R_-$ .

$\delta$	$h = 1.5$		$h = 2$		$h = 3$	
	R+	R-	R+	R-	R+	R-
0	1	1	1	1	1	1
0.1	1.477	0,688	1.522	0.744	1.583	0,815
0.2	1.877	0.412	1.951	0.525	2.072	0.661
0.3	2.217	0.161	2.305	0.334	2.478	0.529
0.5	2.771	$\approx 0$	2.845	0.011	3.096	* 0.328
0.7	3.206	$\approx 0$	3.225	$\approx 0$	3.522	* 0.206
1	3.715	$\approx 0$	3.594	$\approx 0$	3.911	* 0.079
2	4.734	$\approx 0$	3.959	$\approx 0$	4.143	$\approx 0$
3	5.196	$\approx 0$	3.792	$\approx 0$	3.770	$\approx 0$
5	5.355	$\approx 0$	3.110	$\approx 0$	2.663	$\approx 0$
7	5.084	$\approx 0$	2.456	$\approx 0$	1.672	$\approx 0$
10	4.451	$\approx 0$	1.716	$\approx 0$	0.777	$\approx 0$
20	2.581	$\approx 0$	0.572	$\approx 0$	0.067	$\approx 0$
30	1.503	$\approx 0$	0.219	$\approx 0$	0.008	$\approx 0$
50	0.561	$\approx 0$	0.042	$\approx 0$	0.0002	$\approx 0$

Table 1

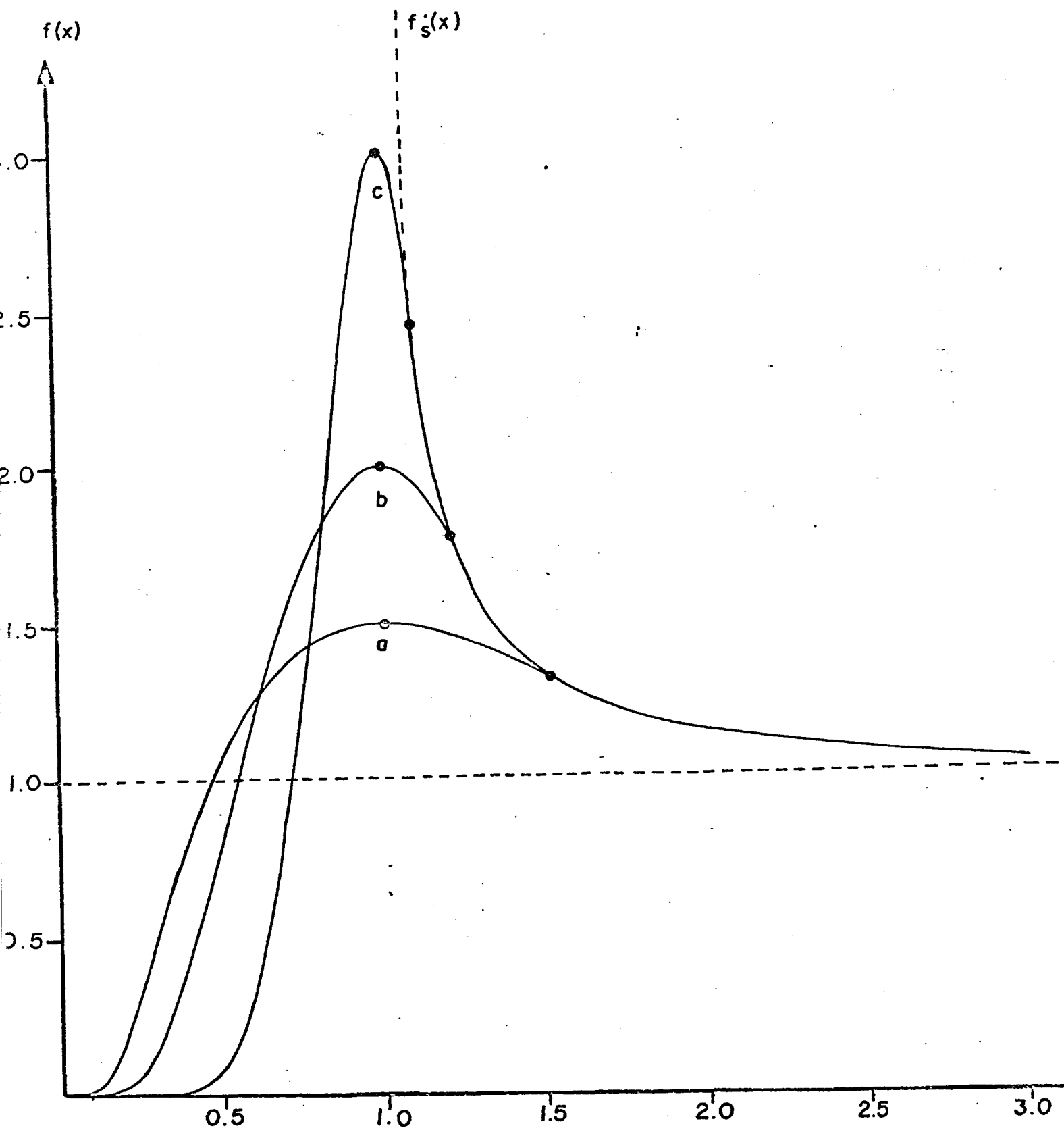


FIG. 1

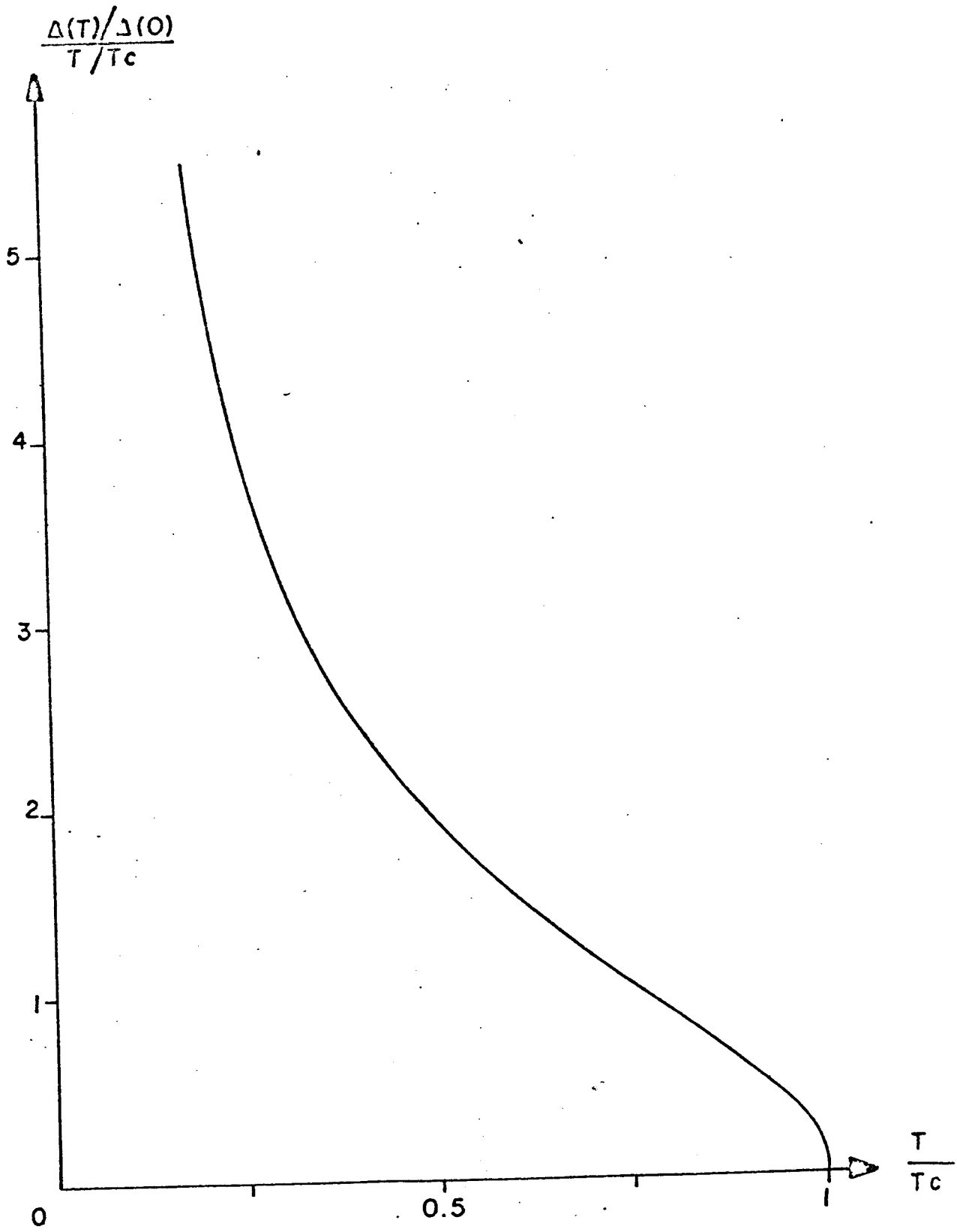


FIG. 2

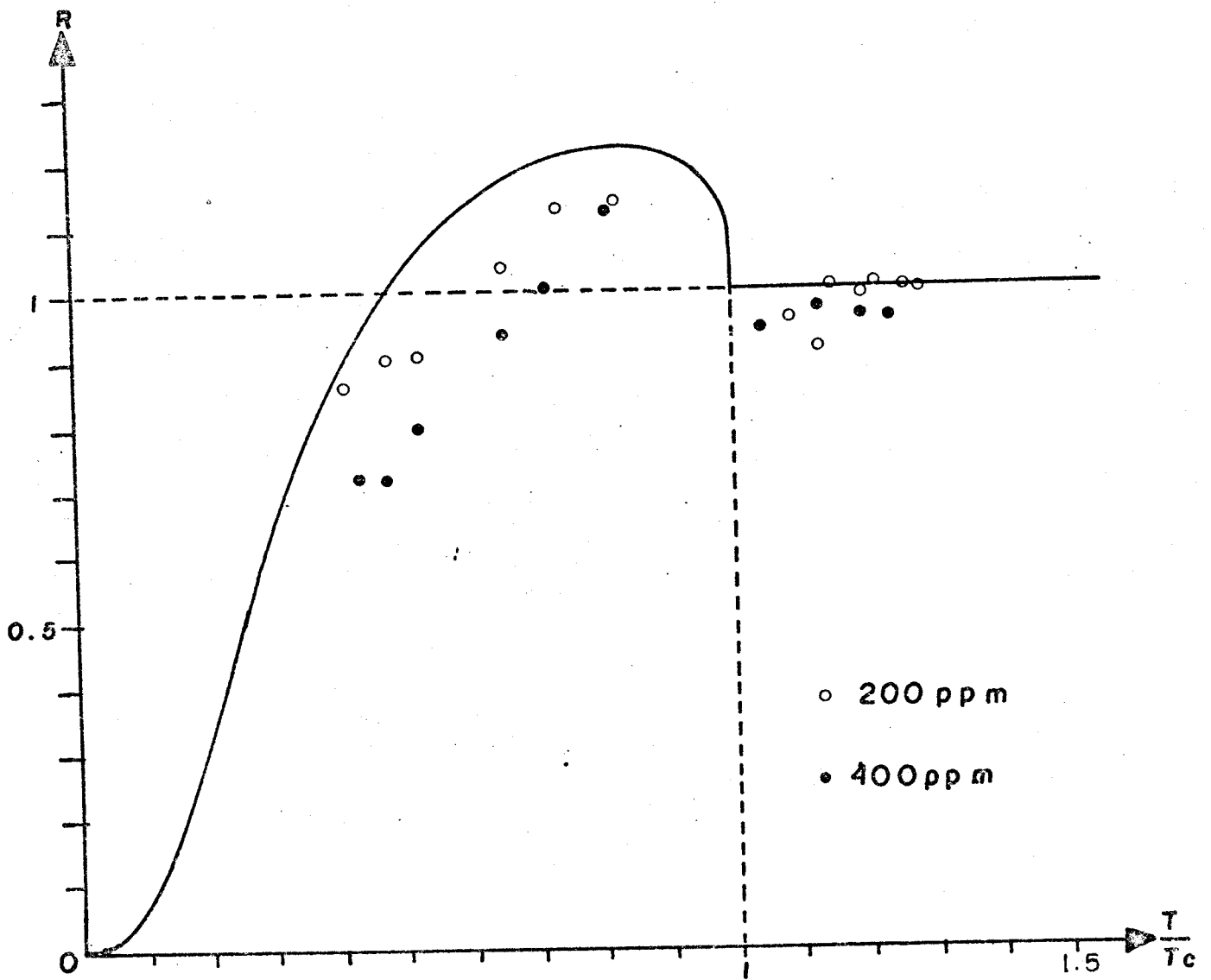


FIG. 3