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## The Volume Dependence of the Electron Level Density and the Critical Temperature in Superconductors

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*Abstract.* Experimental work on the volume dependence of the critical magnetic field in superconductors and on the volume change at the transition is summarized and discussed. The best values for  $d \ln T_c / d \ln v$  and  $d \ln \gamma / d \ln v$  ( $T_c$  = transition temperature,  $\gamma T$  = electronic specific heat per mole,  $v$  = molar volume) are estimated.  $d \ln \gamma / d \ln v$  is many times greater than the free electron value. No correlation between  $d \ln T_c / d \ln v$  and  $d \ln \gamma / d \ln v$  could be seen, but the relation  $d \ln (T_c / \theta) / d \ln v = \text{const.} \ln (\theta / T_c)$  is approximately obeyed for the soft superconductors ( $\theta$  = Debye temperature).

### 1. Introduction

This paper is a summary of some experiments on superconductivity. It appears in this memorial volume because it was a question of PAULI's about a possible change in sound velocity in superconductors which started the experiments leading to our share of the work described here. Our first experiment in this series was a search, in collaboration with BÖMMEL, for a change in sound velocity at the superconducting transition<sup>1</sup>). Since no change was seen we examined the difference in modulus of rigidity statically<sup>2</sup>), and then embarked on a closely related investigation of the difference in volume between the normal and superconducting states.

From such measurements the pressure dependence of the superconducting critical field can be calculated with an accuracy which in many cases is superior to that obtained by direct measurements. As a further step one may calculate two quantities of interest for solid state theory. These are the volume dependence of the superconducting transition temperature, and of the density of states at the Fermi level in the normal metal. The first of these ought to be calculable from any theory which is to explain superconductivity, and the other should be calculable for metals where enough is known about the electronic energy states.

Apart from the volume change work carried out here a great number of direct measurements of the pressure dependence of the critical field have been made in other laboratories. Unfortunately, there are wide

discrepancies between the various results, and one is left in some doubt about which may safely be compared with theoretical conclusions. It seems time, therefore, to summarize all existing work in this field, and to try to indicate its reliability.

## 2. Results

The calculation of the pressure dependence of the critical magnetic field,  $H_c$ , and the further calculation of the volume dependence of the electronic specific heat,  $\gamma T$ , per mole and of the critical temperature,  $T_c$ , from experimental data have been discussed sufficiently elsewhere<sup>3</sup>). Here we shall just recapitulate the final result.

We write  $H_c = H_0 f(t)$  where  $t = T/T_c$ , and we assume that  $f(t)$  is a function which is unaltered by the deformation. Then

$$\frac{v}{H_0} \cdot \frac{\partial H_c}{\partial v} = \frac{v}{H_0} \cdot \frac{\partial H_0}{\partial v} (f(t) - t f'(t)) + \frac{1}{2} \left\{ \frac{v}{\gamma} \frac{\partial \gamma}{\partial v} - 1 \right\} t f'(t) \quad (1)$$

where  $v$  is the molar volume.

Clearly then, we only need  $\partial H_c/\partial p$  at  $T = 0$ , and at  $T = T_c$  to evaluate the following two quantities which have been called  $g$  and  $s$  in previous papers:

$$s = d \ln T_c / d \ln v \quad \text{and} \quad g = d \ln \gamma / d \ln v. \quad (2)$$

The assumption that the form of  $f(t)$  is independent of deformation cannot be completely true, but it is a good enough approximation as SWENSON<sup>4</sup>) has shown recently. Once theory and experiment become sufficiently refined to take into account multiple, and possibly anisotropic energy gaps, then this assumption may have to be abandoned.

In the table we have listed all measurements of  $(\partial H_c/\partial p)_{T=0}$  and  $(\partial H_c/\partial p)_{T=T_c}$  known to us. Many different techniques have been used: direct hydrostatic pressure using liquid helium or hydrogen as a pressure transmitting fluid, ice bomb, uniaxial tension of single crystals, and observations of length changes on destruction of superconductivity. These latter have been done using a bifilar strip<sup>26</sup>), an optical lever or a condenser for indicating the change in length. In general, the direct  $\partial H_c/\partial p$  measurements are more reliable near  $T = T_c$  than at lower temperatures, while the values derived from length changes (the work by CODY, MÜLLER and ROHRER, OLSEN and ROHRER, and ROHRER) are good near  $T = 0$  but have an uncertainty proportional to  $H_c^{-1}$  so that they become poor close to  $T_c$ .

In the cubic metals and in polycrystalline specimens of anisotropic metals

$$\frac{\partial H_c}{\partial p} = 3 \frac{\partial H_c}{\partial p_0} \quad (3)$$

	Author		$(\partial H_c/\partial p)_{T=0}$ Oersted dyn <sup>-1</sup> cm <sup>2</sup> × 10 <sup>-9</sup>	$(\partial H_c/\partial p)_{T=T_c}$	$\frac{d \ln T_c}{d \ln v}$	$\frac{d \ln \gamma}{d \ln v}$	
Lead	K L S 5)			< -6			
	H M 6)			-9.6 ± 0.2			
	O R 3)		-6.3 ± 0.3	-11.0 ± 1.0		1.8 ± 0.4	
	CODY 7)		-9.1 ± 0.5	-10.9 ± 1.1			
	B J 38)		$dT_c/dp = -4.5 \pm 0.5 \times 10^{-n\dagger}$				
	M G 39)		-7.8 ± 0.2	-9.06 ± 0.15	3.2 ± 0.1	6 ± 1	
Vanadium	M R 8)		2.7 ± 0.2	4.1 ± 0.3	-3.5 ± 0.4	-0.4 ± 0.6	
Lanthanum	ROHRER 14)	$T_c=4.3^\circ\text{K}$	5.5 ± 0.5	1.1 ± 0.3	-0.5 ± 0.2	-3.4 ± 1.0	
		$T_c=4.6^\circ\text{K}$	6.2 ± 0.5	1.8 ± 0.3			
Tantalum	K S L 9)			< 0			
	HATTON 10)			$dT_c/dp = -0.4 \pm 0.15^\dagger$			
	FISKE 11)			-3.5 ± 0.4			
	O R 3)		7.0 ± 2.0	-4.0 ± 3.0			
	BOWEN 12)			$dT_c/dp = -0.24 \pm 0.05^\dagger$			
	CODY 7)			-2.6 ± 0.4			
	J S 13)			$dT_c/dp = -0.26^\dagger$	1.5 ± 0.1		
	ROHRER 14)	Ta II		-2.1 ± 0.3	-2.1 ± 0.3		
	S H 40)			-1.1 ± 0.2	-0.9 ± 0.05		4.0 ± 0.5
	$\alpha$ Mercury	K S L 9)			< 0		
GRENIER 16)			-4.8	-6.8			
			-0.4 ± 0.4	-4.0 ± 0.4			
		⊥	-2.2 ± 0.3	-1.4 ± 0.4			
HATTON 10)				-7.5 ± 2.0			
FISKE 11)			-6.8 ± 0.8	-8.3 ± 0.2			
ROHRER 14)			-7.7 ± 0.5	-5.7 ± 0.5	3.3 ± 0.7	10.2 ± 2	
			-0.9 ± 0.2	-3.9 ± 0.2	4.1 ± 0.4	9.7 ± 2	
		⊥	-3.4 ± 0.4	-0.9 ± 0.4	3.0 ± 0.6	11.0 ± 2	
S S 41)			-7.25 ± 0.1	-7.7 ± 0.1	[3.6 ± 0.1]	[6.2 ± 0.2]	
$\beta$ Mercury	S S 17)			-7.5			
	S S 41)		-7.1 ± 0.2	-8.7 ± 0.2	[6.1 ± 0.2]	[6.1 ± 0.2]	
Tin	S O 15)			-4.5			
	K L S 5)		-4.6	-8.7 ± 0.3			
	C J 37)			-6.3			
	GRENIER 16)		-5.0 ± 0.4	-6.4 ± 0.3	7.5 ± 0.5	1.7 ± 0.3	
			-3.8 ± 0.2	-6.0 ± 0.2	13.2 ± 0.5	2.8 ± 0.3	
		⊥	-0.6 ± 0.3	-0.2 ± 0.2	5.6 ± 0.5	1.2 ± 0.3	
	G M 18)			-6.6 ± 0.3			
	MUENCH 19)		-4.2 ± 0.6	-6.9 ± 0.3			
	HATTON 10)			-6.3 ± 0.5			
	O R 3)			$(\partial H_c/\partial p)_0/(\partial H_c/\partial p)_{T_c} = 0.58$			
FISKE 11)		-5.0 ± 1.0	-6.48 ± 0.14				
CODY 7)		-3.2 ± 0.6	-9.1 ± 0.9				
J S 13)			-7.3				
B J 38)			$dT_c/dp = -4.4 \pm 0.3^\dagger$				

	Author		$(\partial H_c/\partial p)_{T=0}$ Oersted dyn <sup>-1</sup> cm <sup>2</sup> × 10 <sup>-9</sup>	$(\partial H_c/\partial p)_{T=T_c}$	$\frac{d \ln T_c}{d \ln v}$	$\frac{d \ln \gamma}{d \ln v}$
Indium	S O 15)			-6.2		
	K L S 5)		-4.9 ± 0.3	-7.6 ± 0.2		
	MUENCH 19)		-4.8 ± 0.7	-6.2 ± 0.5		
	HATTON 10)			-5.9 ± 0.6		
	O R 3)		-3.4 ± 0.5	-5.8 ± 0.8		
	CODY 7)		-4.53 ± 0.17	-5.8 ± 0.43		
		⊥ calc.	-0.09 ± 0.09	-0.22 ± 0.22		
	J S 13)			-6.8	[5.2]	
ROHRER 20)		-3.6 ± 0.2	-6.8 ± 0.3	5.1 ± 0.4	1.0 ± 0.2	
		-3.4 ± 0.2	-6.6 ± 0.3	5.3 ± 0.4	1.0 ± 0.2	
	⊥	-0.2 ± 0.1	-0.3 ± 0.1	5.0 ± 0.4	1.0 ± 0.2	
Thallium	L K 21)			0.8		
	MUENCH 19)			0.8 ± 0.4		
	HATTON 10)		$dT_c/dp = 1.5 ± 0.4^\dagger$			
	O R 3)	mainly	0	-2.0 ± 0.4		
		mainly ⊥	-0.8 ± 0.6	2.1 ± 0.3		
	FISKE 11)		2.7 ± 0.5	3.1 ± 0.15	[-2.5 ± 0.2]	[-4.2 ± 0.6]
	CODY 7)		1.83 ± 0.4	1.09 ± 0.3		
		calc.	-4.35 ± 0.8	-2.63 ± 0.5		
	⊥ calc.	3.09 ± 0.3	1.86 ± 0.2			
	B J 38)		$dT_c/dp = 0.4^\dagger$			
Aluminium	MUENCH 19)			-3.3 ± 0.3		
	GRENIER 22)			-3.7 ± 0.3	12.5 ± 2	
	OLSEN 23)		$(\partial H_c/\partial p)_\theta / (\partial H_c/\partial p)_{T_c} = 3$			
	OLSEN 41)		-3.0 ± 0.4	-4.7 ± 0.3		7 ± 5
Zinc	OLSEN 42)		-1.4 ± 0.2	-1.6 ± 0.3	5 ± 1	7 ± 5
Cadmium	A G 24)		-1.3 ± 0.3	-3.2 ± 0.5	[24 ± 4]	[-9 ± 4]

† Where  $(dT_c/dp)$  is given the units are 10<sup>-11</sup> deg dyn<sup>-1</sup> cm<sup>2</sup>; [] Values marked thus are for hydrostatic pressure.

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where  $p$  stands for a hydrostatic pressure and  $p_\theta$  is a uniaxial pressure in direction  $\theta$ . In anisotropic single crystals  $\partial H_c/\partial p_\theta$  is anisotropic, too. The table therefore gives data both for  $\partial H_c/\partial p$  for hydrostatic pressure and the extreme values of  $\partial H_c/\partial p_\theta$  parallel and perpendicular to the axis of symmetry of the crystal. These are indicated by || and ⊥ respectively, while the values corresponding to hydrostatic pressure are un-

marked. The errors shown have been estimated from the scatter of the observations. It seems likely that much greater systematic errors are present in some cases.

For some of the metals a few brief remarks are needed: *Lanthanum* has two crystallographic modifications (f. c. c. and h. c. p.). The measurements shown are on polycrystals of which the first contained roughly equal quantities of both phases, while the second was mainly cubic. The observed critical temperatures were too low and indicate the presence of magnetic impurities which may also influence the pressure effect (BALTENSBERGER<sup>27</sup>) and ROHRER<sup>14</sup>).

The *Tantalum* results have been the cause of a good deal of discussion, and there now is little doubt that those due to SWENSON are the most reliable. SWENSON has frequently pointed out<sup>28</sup>) the great importance of using specimens of high purity having sharp transitions even when length changes are used for deducing  $\partial H_c/\partial p$ . This is especially true for the hard superconductors, and of these, tantalum has been the most striking example of the errors which may be caused if poor specimens are used.

In *Thallium*  $T_c$  increases with pressure up to about 2500 atmospheres, and then decreases. The values of  $\partial H_c/\partial p$  listed are for  $p \rightarrow 0$ . The measurements marked || and  $\perp$  are for preferentially oriented specimens but it is not known how close the values are to the || and  $\perp$  values for single crystals.

For *Aluminium* the measurements leading to a value for  $\partial H_c/\partial p$  at  $T = 0$  were made using adiabatic demagnetization and an ice bomb technique to obtain the pressure. They must be regarded as at best preliminary, and used with extreme caution.

In *Cadmium* recent work by one of us indicates an anisotropy in  $\partial H_c/\partial p_\theta$ .

In the table we have also listed the two dimensionless quantities  $s = d \ln T_c / d \ln v$  and  $g = d \ln \gamma / d \ln v$  where  $v$  is the volume change considered. The volume changes considered here are not, however, those corresponding to hydrostatic pressure or uniaxial pressures because these volume changes are complicated and uninteresting physically<sup>20</sup>). Instead, the values for  $s$  and  $g$  given are for equal dilatations in all three directions, and for uniaxial dilatations where one dimension of the crystal is changed leaving the two dimensions at right angles unaltered. The results of uniaxial dilatation parallel and perpendicular to the main crystalline symmetry axis are shown on the lines in the table marked || and  $\perp$  respectively. It should be noted that the anisotropies thus obtained are different from, and in general less than those associated with uniaxial pressures. To avoid confusion we have only calculated  $s$  and  $g$  for those measurements we consider most accurate. The errors indicated are fairly conservative.

### 3. The Volume Dependence of the Density of States near the Fermi Level

$\gamma T$  is given by

$$\gamma T = (2/3) \cdot \pi^2 k^2 N(0) T \quad \text{erg/mole deg.} \quad (4)$$

where  $N(0)$  is the density of states at the Fermi surface and  $k$  is the Boltzmann constant. In the case of free electrons we have

$$N(0) \propto v^{2/3} n_a^{1/3} \quad (5)$$

where  $v$  is the molar volume, and  $n_a$  the number of free electrons per atom. We thus find that generally  $d \ln \gamma / d \ln v = d \ln N(0) / d \ln v$ , and that for free electrons  $g = 2/3$ . This is also the value obtained for more complex electronic structures where the Fermi surface overlaps Brillouin zone boundaries if the rigid-band approximation holds. In this approximation it is assumed that the number of electrons in each zone is unaltered by the deformation, and that the relative position of the Fermi surface is unaltered. This may not seem a very drastic assumption since the total number of states per mole of metal within each zone is independent of the shape of the unit cell of the crystal. A look at the values of  $g$  in the table, however, shows that this assumption must be far from correct in most of the metals investigated. It seems highly unlikely that more accurate experiments can reduce  $g$  to  $2/3$  in all cases.

An explanation of anomalous  $g$  values may, however, be given if it is assumed that the energy gap at the Brillouin zone is sensitive to changes in the lattice parameter. Such changes are of course to be expected and ROHRER<sup>14</sup>) has shown that the smaller the overlap into higher zones the larger will  $g$  become. It seems that this may explain the large value of  $g$  in Aluminium where the recently published models of the Fermi surface suggest that very small pockets are present in the higher zones<sup>29) 30)</sup>.

In the case of tantalum the newer measurements yield results much less startling than those published previously. The extreme sensitivity of the physical properties of tantalum to impurities would still seem to indicate that the Fermi level in tantalum must lie close to some anomaly in the density of states curve.

It is unfortunate that no other reliable methods are available at present for measuring  $g$ . Only in the case of aluminium would there be some slight hope of detecting a change in specific heat with pressure at low temperatures, and an investigation of the change in magnetic susceptibility with pressure would appear fairly hopeless. The only field where related information can be found is in the pressure dependence of the electrical resistivity where a great deal of experimental data is available. Unfortunately, however, we have been unable to find any clear cut correlation between these and the results of the type of work described in the present paper.

#### 4. The Volume Dependence of the Critical Temperature

The volume dependence of the critical temperature is best discussed in the light of BARDEEN, COOPER and SCHRIEFFER'S<sup>31)</sup> well known formula

$$k T_c = 1.14 \hbar \omega \exp (-1/N(0)V) \quad (6)$$

where  $\omega$  is a characteristic phonon frequency,  $N(0)$  the density of states at the Fermi surface, and  $V$  an interaction energy.

It is of course well known from work on the isotope effect that a formula of this type reproduces the effect of changes of the lattice vibration frequencies correctly, and it is interesting to try to make use of it in interpreting the effect of volume change on  $T_c$ . On differentiating we find<sup>32)</sup>

$$\frac{d \ln T_c}{d \ln v} - \frac{d \ln \omega}{d \ln v} = \frac{1}{N(0)V} \frac{d \ln N(0)V}{d \ln v}. \quad (7)$$

Here we have moved  $-d \ln \omega / d \ln v$  which is actually GRÜNEISEN'S constant,  $\gamma_G$ , to the left of the equality, in order to isolate on the right hand side the term which contains the interesting physics of the BCS theory. It is here that physically significant correlations might be expected.

It is natural to suppose that the volume dependence of  $N(0)V$  should in some way be connected with that of  $N(0)$ . This is also the result obtained by MOREL<sup>33)</sup> who has carried out a theoretical calculation of the volume dependence of  $N(0)V$ . He found that changes in  $N(0)$  should be the main factor in determining  $d \ln N(0)V / d \ln v$ , and he obtained reasonable agreement with the experimental data then available. More experimental work has been done since, and it unfortunately appears that it has now become more difficult to reconcile MOREL'S calculation with the experimental results.

One is therefore tempted to look for purely empirical relations amongst the observations. ROHRER<sup>14)</sup> has recently pointed out that

$$d \ln N(0)V / d \ln v \approx 2$$

for all the soft superconductors except thallium and cadmium which are both strongly anisotropic. An equivalent statement of this relation is that  $s + \gamma_G$  which equals  $d \ln(T_c/\theta) / d \ln v$  is proportional to  $\ln(\theta/T_c)$  where  $\theta$  is the Debye temperature. In Figure 1 we have plotted  $s + \gamma_G$  against  $\ln(\theta/T_c)$  for the soft superconductors, and it will be seen that all except thallium lie approximately on a straight line. The failure of the correlation for thallium and cadmium need not be taken too seriously since we cannot calculate  $d \ln T_c / d \ln v$  for uniform deformation for either of these metals and we have had to use data for hydrostatic pressure. The results on zinc were obtained too late to be included in Figure 1. They fall a little below the line.



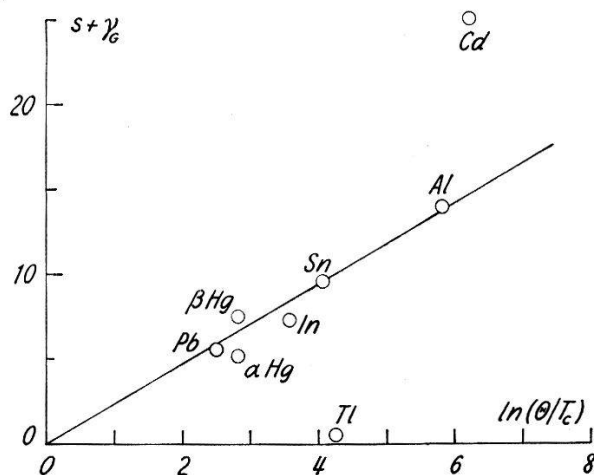


Fig. 1

$(s + \gamma_G) \equiv d \ln(T_c/\theta)/d \ln v$   
 plotted against  $\ln(\theta/T_c)$  for the soft superconductors

If we can trust the results we have collected for  $d \ln \gamma/d \ln v$  then such a correlation means that the changes in transition temperature with volume are almost independent of the changes in the density of states. On the other hand DAUNT'S<sup>34)</sup> and LEWIS'<sup>35)</sup> correlations and MATTHIAS'<sup>36)</sup> work on alloys has shown that the critical temperatures of different superconductors are at least to some extent connected with the density of states.

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