Anisotropy and Strong-Coupling Effects on the Critical-Magnetic-Field Curve of Elemental Superconductors

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An empirical method of separating strong-coupling effects from anisotropy effects in critical-magnetic-field curves of elemental superconductors is presented. Using Clem's critical-field expressions for a weak-coupling anisotropic superconductor, together with an empirical scaling of the superconducting energy gap, a set of relations is derived which allows an independent determination of the mean-squared energy-gap anisotropy parameter $\langle a^2 \rangle$ and a strong-coupling scaling parameter δ. These parameters depend on the experimental determination of two quantities, $2\pi\gamma(T_0/H_0)^2$ and $(dh/dt)_{t=1}$, where T_0 is the zero-field superconducting transition temperature, H_0 is the zero-temperature critical magnetic field, γ is the temperature coefficient of the normal electronic specific heat, and $(dh/dt)_{t=1}$ is the initial slope of the critical-field curve expressed in reduced variables. Values of $\langle a^2 \rangle$ are calculated using published critical-field data and are compared with those values obtained by independent means.

INTRODUCTION

The critical-magnetic-field curve, $H_c(T)$ vs T, of a superconductor has been calculated by Mühlschlegel[1] using the Bardeen-Cooper-Schrieffer (BCS) theory.[2] For the elemental superconductors deviations from this theoretical curve are generally small, 2–3% (the notable exceptions, Pb and Hg, deviate by only 5%), but are strikingly apparent when the deviation function $D(t)$ is plotted for various elements (see Fig. 1):

$$D(t) \equiv h - (1 - t^2), \tag{1}$$

where $h \equiv H_c/H_0$; H_0 is the zero-temperature critical magnetic field; $t \equiv T/T_0$; T_0 is the zero-field transition temperature.

Observed deviations from these critical-field calculations can be attributed to two approximations made in the original BCS theory. First, the BCS theory is a weak-coupling theory ($\Theta_D \gg T_0$; Θ_D is the Debye temperature) which ignores the detailed energy dependence of the electron-phonon interactions. Second, the theory also ignores the crystal lattice structure of the material which produces anisotropy in the superconducting energy gap. Strong-coupling effects (i.e., large electron-phonon interactions) generally increase the critical magnetic field of a superconductor, while anisotropy effects tend to decrease it.

The effects of energy-gap anisotropy have been included in the weak-coupling BCS theory by Clem,[3] who derived expressions for the critical-field curve of an anisotropic superconductor. Strict comparison of his theory to data for elemental superconductors is uncertain, however, because of competing strong-coupling effects. These strong-coupling effects can be empirically introduced into critical-field calculations by scaling the superconducting energy gap to values in excess of the BCS prediction. Energy-gap scaling modifies Mühlschlegel's critical-field expression for an isotropic superconductor in such a way as to give

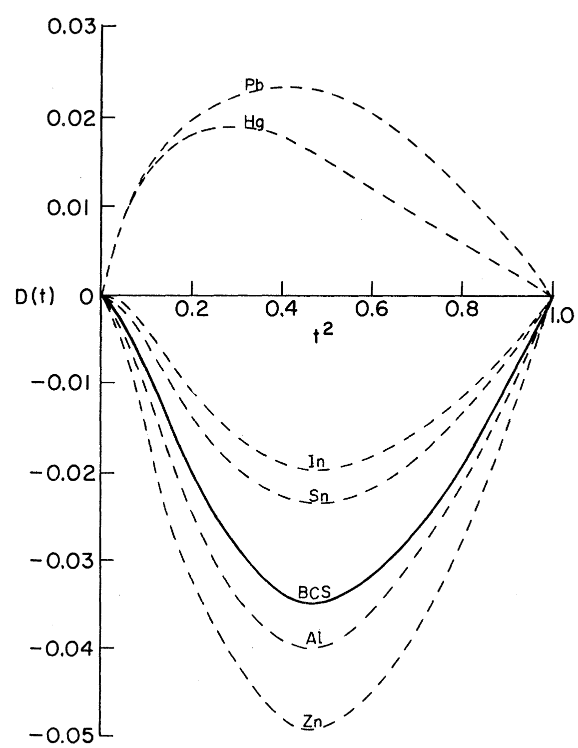

FIG. 1. Deviation function $D(t)$ for a various elements. Differences from the BCS prediction, although small, are strikingly apparent. Experimental data are taken from Refs. c, f, h, and p of Table I.

TABLE I. Summary of critical-magnetic-field data for various elements. Values of $\langle a^2\rangle$ and δ are calculated from Eqs. (13) and (14) in the text using these data. The values of $\langle a^2\rangle$ thus obtained are compared with values derived from impurity studies ($\langle a^2\rangle_I$) when such data are available. [Note: No value of $-2\pi\gamma(T_0/H_0)^2(dh/dt)_1$ less than 1.836 can be explained by this modified anisotropy model; hence, Hg does not fit the model.]

Element	T_0 (K)	H_0 (G)	γ (erg/cm^3 K^2)	$-\left(\frac{dh}{dt}\right)_1$	$2\pi\gamma\left(\frac{T_0}{H_0}\right)^2$	$-2\pi\gamma\left(\frac{T_0}{H_0}\right)^2\left(\frac{dh}{dt}\right)_1$	$2\pi\gamma\left(\frac{T_0}{H_0}\right)^2\left(\frac{dh}{dt}\right)_1^2$	$\langle a^2\rangle$	$\langle a^2\rangle_I$	δ	Reference[a]
Al	1.179	104.9	1367	1.719	1.085	1.865	3.206	0.016	0.011[b]	1.006	c
Cd	0.5151	28.05	533	1.680	1.129	1.897	3.186	0.033		0.999	d
Ga	1.083	59.2	512	1.72	1.076	1.851	3.183	0.008		0.998	e
Hg	4.154	410.9	1312	2.060	0.842	1.735	⋯	⋯		⋯	f
In	3.409	281.5	1088	1.882	1.002	1.886	3.549	0.027	0.021[b]	1.113	g
Mo	0.9134	96.23	1956	1.698	1.107	1.880	3.192	0.025		1.001	d
Pb	7.177	802.6	1705	2.134	0.856	1.826	3.897	0.00		1.222	h
Sn	3.722	305.5	1086	1.842	1.013	1.866	3.437	0.016	0.019[b]	1.078	f
Ta	4.482	830	5600[i]	1.850	1.006	1.861	3.443	0.014		1.080	j
Th	1.390	159.2	2200	1.775	1.053	1.869	3.317	0.019	0.020[k]	1.041	l
Tl	2.39	171	870[m]	1.790	1.068	1.912	3.441	0.041		1.079	n
Zn	0.850	54.0	718	1.692	1.118	1.892	3.201	0.031	0.030[o]	1.004	p

[a]Except where otherwise noted, the values in this table are obtained from the references shown in this column.
[b]Reference 8.
[c]E. P. Harris and D. E. Mapother, Phys. Rev. 165, 522 (1968).
[d]D. G. Hamblen, thesis (University of Illinois, 1969) (unpublished).
[e]W. D. Gregory, J. P. Sheahen, and J. F. Cochran, Phys. Rev. 150, 315 (1966).
[f]Reference 4.
[g]D. U. Gubser, D. E. Mapother, and D. L. Connelly, Phys. Rev. B 2, 2547 (1970).
[h]D. L. Decker, D. E. Mapother, and R. W. Shaw, Phys. Rev. 112, 1888 (1958).
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[n]E. Maxwell and O. S. Lutes, Phys. Rev. 94, 333 (1954).
[o]From an analysis of the data of D. Farrell, J. G. Park, and B. R. Coles, Phys. Rev. Letters 13, 328 (1964); see Ref. p.
[p]D. U. Gubser and J. E. Cox (unpublished).

qualitative agreement with the observed critical-field curves of strong-coupling superconductors.[4,5]

In this paper, Clem's expressions for the critical-field curve of an anisotropic superconductor are similarly scaled to include strong-coupling effects. The effects of strong coupling and energy-gap anisotropy are given in this model by two parameters—an anisotropy parameter $\langle a^2\rangle$ and a scaling parameter δ. Since the effects of these two parameters on the critical-field curve are different at the two temperature extremes, $T=0$ and $T=T_0$, it is possible to establish from critical-field data alone two independent expressions which can be individually solved for the parameters $\langle a^2\rangle$ and δ. The parameter $\langle a^2\rangle$, as evaluated using this model and published critical-field data, is consistent (see Table I) with the $\langle a^2\rangle$ values derived from an analysis of the depression of T_0 with dilute nonmagnetic impurities.

MODIFIED ANISOTROPY MODEL

The shape of the critical-field curve at low temperatures, $t<0.20$, is given by the thermodynamic relation[4]

$$H_c^2=H_0^2-4\pi\gamma T^2, \tag{2}$$

where γ is the temperature coefficient of the normal electronic specific heat per unit volume. Expressing (2) in reduced variables and expanding to second order in t^2 gives

$$h=1-2\pi\gamma(T_0/H_0)^2t^2-\tfrac{1}{2}[2\pi\gamma(T_0/H_0)^2]^2t^4+\cdots\ . \tag{3}$$

The quantity $2\pi\gamma(T_0/H_0)^2$ is seen to be related to the curvature of the reduced critical-field curve as t approaches 0.

To calculate this quantity in the weak-coupling BCS limit, we begin with the well-known BCS prediction for the ratio of the superconducting energy gap at $T=0$ to the critical temperature:

$$\Delta(0)/kT_0=1.764\ . \tag{4}$$

The condensation energy at $T=0$ is given in this limit by

$$H_0^2/8\pi=\tfrac{1}{2}N(0)\Delta^2(0)\ , \tag{5}$$

where $N(0)$ is the electronic density of states at the Fermi level. Using the relation for the temperature coefficient of the normal-state electronic specific heat,

$$\gamma=\tfrac{2}{3}\pi^2N(0)k^2\quad(k\equiv\text{Boltzmann's constant}), \tag{6}$$

Eq. (5) becomes

$$H_0(\pi k^2/6\gamma)^{1/2}=\Delta(0). \qquad (7)$$

Combining (4) and (7) yields the weak-coupling BCS value of

$$2\pi\gamma\,(T_0/H_0)^2=1.057. \qquad (8)$$

At high temperatures, $t\approx 1$, the shape of the critical-field curve is characterized by the initial slope. In the weak-coupling BCS limit, Mühlschlegel[1] shows that

$$\left(\frac{dh}{dt}\right)_{t=1}\equiv\left(\frac{dh}{dt}\right)_1=-1.737. \qquad (9)$$

To introduce empirically strong-coupling effects into the critical-field calculations, (4) is scaled to values larger than the weak-coupling BCS prediction, as is experimentally observed, by the inclusion of a scaling parameter δ:

$$\Delta(0)/kT_0=1.764\delta \quad (\delta\geq 1). \qquad (4a)$$

Using this scaled energy gap and the Mühlschlegel expression[1] for the critical-field curve of a weak-coupling superconductor, Finnemore and Mapother[4] (FM) calculated critical-field curves for various values of δ and obtained qualitative agreement with the experimental critical-field curves of moderately strong-coupling superconductors (see Fig. 2).

Such calculations, however, increase the value of the critical fields at low temperatures too much, as is seen by the fact that the condensation energy at $T=0$ is still given by the weak-coupling BCS formula, Eq. (5). For strong-coupling superconductors, (5) should be replaced by

$$H_0^2/8\pi=\tfrac{1}{2}N(0)\Delta^2(0)(1-e^{-2/N(0)V}), \qquad (5a)$$

where V is the BCS interaction parameter. Equation (5a) follows directly from the BCS theory if the usual weak-coupling approximation

$$\sinh[1/N(0)V]\approx\tfrac{1}{2}e^{1/N(0)V} \quad [N(0)V\ll 1] \qquad (10)$$

is not used.[6] For strong-coupling superconductors, where $N(0)V>0.25$, this weak-coupling approximation is not valid.

Sheahen[5] showed that the quantity $(1-e^{-2/N(0)V})$ is related to the parameter δ introduced in (4a) by

$$\delta=(1-e^{-2/N(0)V})^{-1}. \qquad (11)$$

It follows from (5a), (6), and (11) that

$$H_0(\pi k^2/6\gamma)=\Delta(0)/\delta^{1/2}. \qquad (7a)$$

Combining (4a) and (7a) yields[7]

$$2\pi\gamma(T_0/H_0)^2=1.057/\delta. \qquad (8a)$$

Figure 2 demonstrates how the weak-coupling BCS critical-field curve is affected by scaling of the energy gap in this manner.

Using the scaled energy gap (4a) and Mühlschlegel's expression for the critical-field curve near $t=1$, the initial slope becomes

$$\left(\frac{dh}{dt}\right)_1=-1.737\delta. \qquad (9a)$$

This result agrees with that given by FM but differs slightly from that given by Sheahen,[5] since Sheahen did not use the exact BCS expression for the energy gap. His result for $(dh/dt)_1$ does not reduce to the weak-coupling BCS limit for $\delta=1$ as does (9a).

Effects of anisotropy produced by the crystal lattice structure of the element are introduced into the weak-coupling BCS theory by using an energy gap of the form

$$\Delta_k=\epsilon_0(1+a_k), \qquad (12)$$

where ϵ_0 is the average energy gap and a_k is an anisotropy parameter for an electron in the k state.

Clem[3] shows that (4), (7), (8), and (9) transform

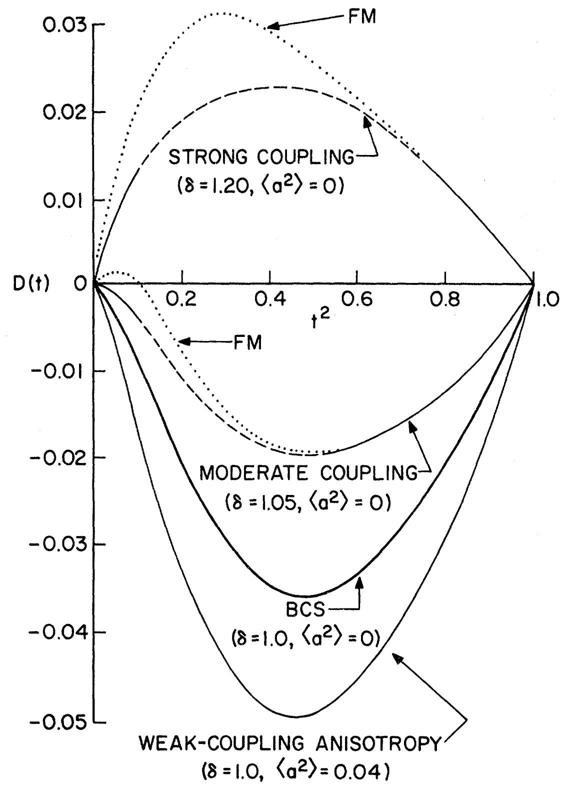

FIG. 2. Changes in the BCS prediction for the deviation function due to the inclusion of anisotropy and of energy-gap scaling. Finite values of $\langle a^2\rangle$ lower the deviation function, while the scaling modifications, used to introduce strong-coupling effects, increase the deviation function. The dashed lines represent an interpolation between the calculated values near the two temperature extremes. The dotted lines show the scaling modifications used by Finnemore and Mapother, Ref. 4.

to

$$\epsilon_0/kT_0 = 1.764(1 - \tfrac{3}{2}\langle a^2\rangle), \tag{4b}$$

$$H_0(\pi k^2/6\gamma)^{1/2} = \epsilon_0(1 + \tfrac{1}{2}\langle a^2\rangle), \tag{7b}$$

$$2\pi\gamma(T_0/H_0)^2 = 1.057(1 + 2\langle a^2\rangle), \tag{8b}$$

$$\left(\frac{dh}{dt}\right)_1 = 1.737(1 - \langle a^2\rangle), \tag{9b}$$

where $\langle a^2\rangle$ is the mean-squared anisotropy. These equations describe an anisotropic superconductor in the weak-coupling limit. Figure 2 shows how the weak-coupling BCS critical-field curve is affected by anisotropy.

Combining the scaling modifications with the anisotropy expressions gives as a final result

$$\epsilon_0/kT_0 = 1.764\delta(1 - \tfrac{3}{2}\langle a^2\rangle), \tag{4c}$$

$$H_0(\pi k^2/6\gamma)^{1/2} = (\epsilon_0/\delta^{1/2})(1 + \tfrac{1}{2}\langle a^2\rangle), \tag{7c}$$

$$2\pi\gamma(T_0/H_0)^2 = (1.057/\delta)(1 + 2\langle a^2\rangle), \tag{8c}$$

$$\left(\frac{dh}{dt}\right)_1 = -1.737\delta(1 - \langle a^2\rangle). \tag{9c}$$

Equation (8c) describes the shape of the critical-field curve near $t = 0$ including both the effects of strong coupling and anisotropy, while (9c) describes the shape near $t = 1$. The left-hand sides of these two equations are experimentally determinable from critical-field data. They provide a set of two independent equations involving two unknowns, $\langle a^2\rangle$ and δ, which can be solved to yield

$$2\pi\gamma\left(\frac{T_0}{H_0}\right)^2\left(\frac{dh}{dt}\right)_1 = -1.836(1 + \langle a^2\rangle) \tag{13}$$

and

$$2\pi\gamma\left(\frac{T_0}{H_0}\right)^2\left(\frac{dh}{dt}\right)_1^2 = 3.188\delta. \tag{14}$$

From (13) one can calculate the $\langle a^2\rangle$ value of an elemental superconductor and from (14), δ can be determined.

DISCUSSION

Table I lists the pertinent experimental data for some of the more accurately measured elemental superconductors. Values of $\langle a^2\rangle$ and δ, which are predicted using (13) and (14), are shown as well as values of $\langle a^2\rangle$ obtained by independent methods.

Determination of $\langle a^2\rangle$ from (13) requires that the quantities $2\pi\gamma(T_0/H_0)^2$ and $(dh/dt)_1$ be known to a high precision. A 1% uncertainty in their product leads to an error in $\langle a^2\rangle$ of 0.01, which for most elements is comparable to the absolute value of $\langle a^2\rangle$ itself. Since the product depends on four experimental quantities [γ, T_0, H_0, and $(dh/dt)_1$], it is necessary to know each of them to an accuracy of about 0.2% in order to obtain a reliable value for $\langle a^2\rangle$. Systematic errors are reduced if all of these quantities are determined self-consistently from the same set of data on a given sample as from a complete mapping of the critical-field curve.

Critical-field data for the elements Al, Sn, In, Th, and Zn are accurate enough to provide a good test for (13). Values of $\langle a^2\rangle$ obtained from the critical-field data compare favorably with values obtained independently from an analysis of the depression of T_0 caused by dilute nonmagnetic impurities.[8] In studies of such dilute alloys, the quantity derived is not $\langle a^2\rangle$, but rather $\lambda^i\langle a^2\rangle$. λ^i is an impurity-dependent constant of order unity which is not experimentally determinable and is usually set equal to unity for at least one impurity with the λ^i's for other impurities chosen to give consistent values of $\langle a^2\rangle$. Since $\lambda^i \leq 1$, one would expect $\langle a^2\rangle$ determined from critical-field studies to be equal to or greater than that obtained from impurity studies. For Al and In, $\langle a^2\rangle$ obtained from critical-field data is slightly greater than that obtained from alloy studies; however, even with these excellent critical-field data, the error in $\langle a^2\rangle$, determined from (13), is still about the size of the discrepancy ($\pm$ 0.005). The $\langle a^2\rangle$ comparisons of Sn, Th, and Zn all agree within the experimental accuracy of the data.

Critical-field data for Cd and Mo, both of which are weak-coupling superconductors, are also accurate enough to provide good tests for (13) and (14). As expected, no strong-coupling scaling is needed to explain the shape of the critical-field curve. Values of $\langle a^2\rangle$ obtained from impurity studies are not available to check the predictions of $\langle a^2\rangle$ obtained from Eq. (13). Critical-field data for Ga, Ta, and Tl are not precise enough to give reliable values for $\langle a^2\rangle$; however, the values calculated from them show no inconsistencies.

Pb and Hg provide extreme tests for the scaling model since they are the strongest-coupling superconductors. The scaling modifications necessary to explain the critical-field data of these elements push the original BCS theory far into a region where the BCS approximations do not hold. For strong-coupling superconductors, the superconducting properties are affected by the details of the phonon spectrum.[9] These details cannot be accounted for by the simple scaling concept employed here. Critical-field data for Pb, however, do appear to be interpretable by the scaling modifications giving a value for $\langle a^2\rangle$, obtained from (13), of zero. This prediction of small anisotropy is supported by dilute-nonmagnetic-alloy studies.[10] Hg is the only element listed in Table I which does not fit the scaling analysis. The reasons for this discrepancy are apparently connected with the unusual shape of its phonon spectrum.[11]

The results of Pb and Hg provide a clue as to where the scaling model can be applied. The phonon spectrum of Pb is fairly typical of most elemental superconductors containing two dominant peaks corresponding to transverse and longitudinal acoustical modes.[12] The phonon spectrum of Hg, however, contains just one dominant peak at a very low energy. Thus, because of its atypical phonon spectrum, Hg appears to be an isolated case whose superconducting properties are not susceptible to generalization to other elements. The scaling theory therefore appears applicable (i) for weak-coupling to moderately weak-coupling superconductors where details of the phonon spectrum are unimportant and (ii) for strong-coupling superconductors whose phonon spectra are not radically different from that of Pb.

As an example of applying these criteria, consider the element Tl. Tl is a moderately strong-coupling superconductor ($\Theta_D/T_0 \approx 35$) with a phonon spectrum similar in shape to that of Pb.[13] It therefore satisfies both conditions (i) and (ii) and is expected to fit the modified anisotropy model just presented. From data available, a $\langle a^2 \rangle$ value of 0.04 ± 0.04 is obtained which makes Tl potentially one of the most anisotropic superconductors. The large uncertainty is due to the inaccuracy of the critical-field data. The value of $\langle a^2 \rangle$ obtained from alloy studies[14] is 0.06, while $\langle a^2 \rangle$ obtained from an analysis of strain-induced effects[15] is 0.01. An accurate measurement of the critical-field curve of Tl could clear up this uncertainty in $\langle a^2 \rangle$.

CONCLUSION

An empirical method of separating strong-coupling effects from anisotropy effects in critical-field curves of elemental superconductors has been presented. It is demonstrated in this paper that a meaningful evaluation of $\langle a^2 \rangle$ can be obtained from accurate critical-field data on elemental superconductors without resorting to other, sometimes more ambiguous, methods of estimating this quantity.

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[7]Equation (8a) of this paper corresponds to Eq. (18) of Ref. 5 for the case of zero anisotropy; however, owing to an apparent misprint in that article, the quantity $(1-e^{-2/N(0)V})$ $(\equiv 1/\delta)$ erroneously appears in the denominator instead of the numerator. This can easily be verified by examining the entries in Table II of Ref. 5 which make use of Eq. (18).

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