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An Explanation of the Temperature-Dependent Upper Critical Field Data of H_3S on the Basis of the Thermodynamics of a Superconductor in a Magnetic Field

Gulshan Prakash Malik[1,2]

[1]Gurugram, India
[2]Theory Group, School of Environmental Sciences, Jawaharlal Nehru University, New Delhi, India
Email: gulshanpmalik@yahoo.com

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Abstract

Excellent fits to a couple of the data-sets on the temperature (T)-dependent upper critical field (H_{c2}) of H_3S (critical temperature, $T_c \approx 200$ K at pressure ≈ 150 GPa) reported by Mozaffari, *et al.* (2019) were obtained by Talantsev (2019) in an approach based on an ingenious mix of the Ginzberg-Landau (GL), the Werthamer, Helfand and Hohenberg (WHH), and the Gor'kov, etc., theories which have individually been employed for the same purpose for a long time. Up to the lowest temperature (T_L) in each of these data-sets, similarly accurate fits have also been obtained by Malik and Varma (2023) in a radically different approach based on the Bethe-Salpeter equation (BSE) supplemented by the Matsubara and the Landau quantization prescriptions. For $T < T_L$, however, while the (GL, WHH, etc.)-based approach leads to $H_{c2}(0) \approx$ 100 T, the BSE-based approach leads to about twice this value even at 1 K. In this paper, a fit to one of the said data-sets is obtained for the first time via a thermodynamic approach which, up to T_L, is as good as those obtained via the earlier approaches. While this is interesting per se, another significant result of this paper is that for $T < T_L$ it corroborates the result of the BSE-based approach.

Keywords

H_3S, Upper Critical Field (H_{c2}), Variation of H_{c2} with Temperature, Clausius-Clapeyron equation in a magnetic field, Behavior of H_{c2} for Temperatures Close to 0 K

1. Introduction

The temperature (T)-dependent values of the upper critical field H_{c2} of two samples of H_3S reported by Mozaffari, *et al.* [1] were explained by the authors on the basis of both the Ginzberg-Landau (GL) theory and the Werthamer, Helfand and Hohenberg (WHH) theory [2]. Based on a mix of these and other well-known theories, e.g., Gor'kov theory [3], accurate fits to the same data-sets were obtained by Talantsev [4] via four alternative equations. Each of these equations invoked two or more parameters from the following sample-specific set of the properties of the superconductor (SC): Σ_1 = {Critical temperature (T_c), gap, coherence length and penetration depth at T = 0, jump in sp. ht.}. In the following, we refer to this phenomenological approach as Approach I.

Fits as good as those obtained via Approach I for the above data-sets have also been obtained in [5] and [6] via a T-, chemical potential (μ)- and applied field (H)-incorporated equation for pairing [7]. This equation—derived in [8] and subjected to a correction in [7]—is obtained via a 4-d Bethe-Salpeter equation (BSE) which is temperature-generalized via the Matsubara prescription at the expense of the 4th dimension. The resulting 3-d equation is subjected to the Landau quantization scheme which causes the transverse components of momentum to be quantized into Landau levels. The 1-d equation thus obtained depends on the set Σ_2 = {μ, T, H, the Debye temperature of the SC, the magnetic interaction parameter, the effective mass of the electron, and the Landau index}, which is radically different from Σ_1. Hereafter we shall refer to the BSE-based approach as Approach II.

The purpose of this note is:

a) To present an approach which is neither dependent on the theories employed in Approach I nor on the rather specialized methodology of Approach II, but is based for the first time on the thermodynamics of an SC in a magnetic field via an adaptation of the Clausius-Clapeyron equation (CCE).

b) To shed light on the contrasting results that Approaches I and II yield for the values of $H_{c2}(T)$ for T close to 0 K. To elaborate, the lowest temperature (T_L) for which the empirical values of $H_{c2}(T)$ were reported in [1] are: Sample 1, T_c = 173.7 K: T_{L1} = 55.09 K; Sample 2, T_c = 191 K: T_{L2} = 105.1 K. While both the approaches lead to almost equally good fits to the empirical data up to T_{L1} and T_{L2}, the values of $H_{c2}(T)$ that they lead to for $T < T_{L1}$ or T_{L2} are significantly different. In Approach I, for values of T from about 10 to 0 K, the $H_{c2}(T)$ vs. T plot is almost parallel to the T-axis; the value of $H_{c2}(0)$ for each of the two samples is ≈ 100×10^4 G (100 Tesla). In Approach II, on the other hand, for the same range of T, there is an upward swing of the $H_{c2}(T)$ vs. T plot, leading to values of H_{c2} even at T = 1 K that are about twice the corresponding values in Approach I.

Since the most familiar form of the CCE is one that relates the change in the pressure of a system with T when it is undergoing a liquid ↔ gas or solid ↔ liquid phase transition, see, e.g., [9], we need to adapt this equation to the situation when pressure is kept constant and the solid ↔ liquid phase transition is brought about by H—as is the case for H_3S. This is done in the next section. Application of the thus obtained equation to Sample 1—which has been stated to be prepared from condensed liquid H_2S [1]—is taken up in Section 3. The final section sums up our findings.

2. The CCE in a Magnetic Field

When pressure is maintained at a constant value and the change from a normal (N)-state to the superconducting (S)-state, and vice-versa, takes place due to an applied field H, the differential of the Gibbs function for the two states are:

$$\text{S-State:} \quad dG\big|_S = S_s dT - VM_S dH \tag{1}$$

$$\text{N-state:} \quad dG\big|_N = S_N dT, \quad (\text{since } M_N = 0) \tag{2}$$

where S denotes entropy and V the volume of the sample, and M is the magnetic moment per unit volume.

It follows from the equality of $dG\big|_S$ and $dG\big|_N$ dictated by the coexistence of the two phases on the locus of the point of phase transition that

$$\left(S_S - S_N\right) dT = VM_s dH. \tag{3}$$

As usual, in this equation $\left(S_S - S_N\right) = L_m J/T$, where L_m is the latent heat that brings about the change of phase and J is the mechanical equivalent of heat (4.19×10^7 ergs/cal) employed to convert calories into ergs. There now remains M_S to be specified which satisfies the equation $B_S = H + 4\pi M_S$. If the SC we are dealing with were a type I SC which is a perfect diamagnet, we would have had the magnetic induction parameter $B_S = 0$ and therefore $M_S = -H/4\pi$. Since H_3S is known to be a type II SC, $B_S \neq 0$ and hence we need to parametrize B_S in terms of H and t in order to have a tractable problem. To this end, we assume that

$$B_S = \alpha t H / (1 + \alpha t), \tag{4}$$

where α is a constant and $t = T/T_c$. Thus, $M_S = -H/\left[4\pi(1+\alpha t)\right]$ and we now have (3) as

$$(1+\alpha t)\frac{dt}{t} = -\frac{VH}{4\pi L_m J} dH,$$

or,

$$t \exp(\alpha t) = C \exp\left(-\frac{VH^2}{8\pi L_m J}\right),$$

where C, a dimensionless constant since α, t and $\left(VH^2/L_m J\right)$ are dimensionless, can be fixed by putting $H = 0$ corresponding to $t = 1$. Thus,

$$Eq1(t, H, \alpha, L_m) \equiv t \exp\left[-\alpha(1-t)\right] - \exp\left(-\frac{VH^2}{8\pi L_m J}\right) = 0. \tag{5}$$

3. Application of the CCE in a Magnetic Field to a Sample of H_3S

Gleaned from (1), the specifications of the sample subjected to pressure of 160 GPa and the empirical values of its $H_{c2}(T)$ employed by us are as follows:

Diameter of the sample, d = 40 × 10^{-4} cm; thickness of the sample, th = 4 × 10^{-4} cm;

Volume of the sample (π d th) = 5.0265 × 10^{-6} cm^3;

T_c = 173.7 K;

Values of $H_{c2}(T)$ at 24 points—shown in **Figure 1**—for 55.09 K ≤ T ≤ 173.7 K: 62.45 × 10^4 G ≥ $H_{c2}(T)$ ≥ 0 G.

In particular,

$$\text{For } T_1 = 69.99 \text{ K}, \; H_{c2}(T_1) = 54.31 \times 10^4 \text{ G} \tag{6}$$

$$\text{For } T_2 = 141.6 \text{ K}, \; H_{c2}(T_2) = 12.62 \times 10^4 \text{ G} \tag{7}$$

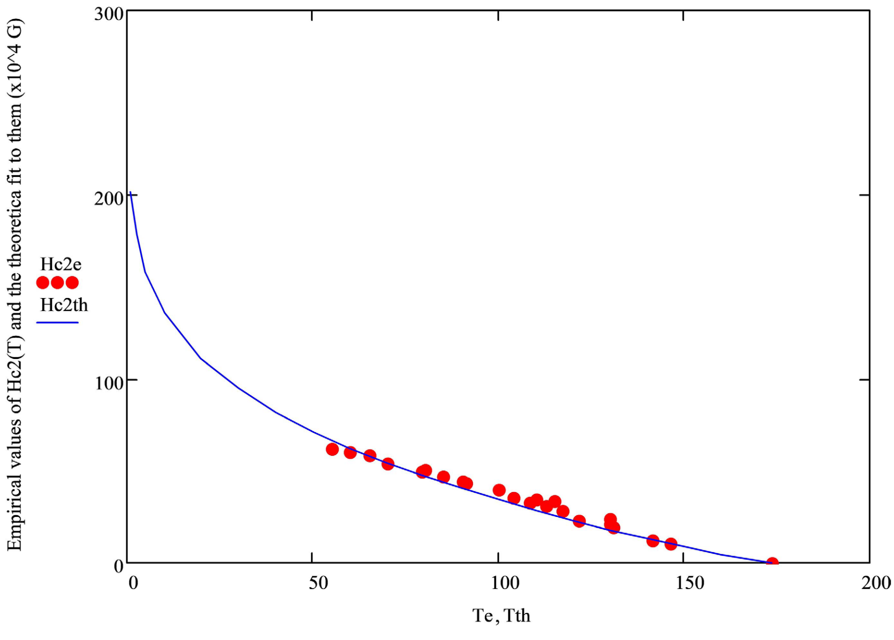
To find the unknowns α and L_m in (5), we simultaneously solve

$$Eq1\left(t_1 = T_1/T_c, H_{c2}(T_1), \alpha, L_m\right) \text{ and } Eq1\left(t_2 = T_2/T_c, H_{c2}(T_2), \alpha, L_m\right),$$

where T_1, T_2 and the corresponding values of H_{c2} are as given in (6) and (7), respectively. Thus,

$$\alpha = -1.01757, \; L_m = 4.6709 \times 10^{-3} \text{ cal} \tag{8}$$

We can now solve (5) to obtain the value of H_{c2} for any value of t. The results of this exercise carried out for 20 values of T between 1 and 173.7 K are given in **Figure 1** which also includes the empirical values of H_{c2} in the range of T noted above (6).



Te and Tth denote, respectively, the empirical and the theoretical values of temperature (K)

Figure 1. The continuous curve is the fit obtained via (5) to the empirical values of $H_{c2}(T)$—denoted by filled circles—as reported in [1] for 55.09 K ≤ T ≤ 173.7 K.

4. Discussion and Conclusion

The fit to the empirical data shown in **Figure 1** can be seen as providing a posteriori justification for the expression of B_S in terms of t and H noted in (4). We believe that this relation can also be validated empirically via a set-up similar to the one recently employed by Minkov *et al.* [10] to determine the trapped magnetic fluxes in H_3S and LaH_{10} in order to obtain the lower critical field H_{c1} of these SCs. It is interesting to note that the results plotted in **Figure 1** and obtained by assuming B_s to vary as in (4) and L_m to have a constant value can also be *formally* obtained by assuming M_S to have the value $-H/4\pi$ (i.e., by assuming H_3S to be a type I SC) and paying the price of making L_m t-dependent via $L_m = L_{m0}(1 + \alpha t)$, where $\alpha = -1.01757$ and $L_{m0} = 4.6709 \times 10^{-3}$ cal - vide (8).

We would like to note that the value of L_m given in (8) is not per gram, but for the whole of the tiny sample whose mass is not given.

Now a matter of detail. It is pertinent to ask: how does the plot given in **Figure 1** change if α and L_m are determined not via the choice of $(T_1, H_{c2}(T_1))$ and $(T_2, H_{c2}(T_2))$ noted in (6) and (7), respectively, but by the choice of two different points in the neighborhood of these points? The answer to this question is: indeed, the plot changes, but does so marginally. However, the feature of its upswing as $T \to 0$ K persists unmistakably. The slight shifting of the plot when points other than those specified in (6) and (7) are employed to fix α and L_m is not surprising in view of the uncertainties in the reported values of $(T, H_{c2}(T))$, which is evidenced by the values of $H_{c2}(T)$ given in the data as 21.02 and 23.88 T for the same value of T, viz., 130.1 K.

The $H_{c2}(T)$ data of H_3S addressed here via a thermodynamic approach have been dealt with earlier via approaches as different as the GL or the WHH, etc., theories, or via the field-theoretic approach provided by the BSE. These different approaches should be seen as complementing each other because they shed light on different features of the same phenomenon. Based on the simple framework of the thermodynamics of an SC in a magnetic field, a virtue of the present approach is that it does not require familiarity with the rather formidable theoretical apparatus employed by each of the earlier approaches.

To conclude, we have for the first time obtained in this paper via a thermodynamic approach a fit to the $H_{c2}(T)$ data of a sample of H_3S which, up to T_L, is nearly as good as those obtained earlier via several radically different approaches. For $T < T_L$, however, our findings are in accord with those of Approach II.

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Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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