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Superconducting fluctuation magnetoconductance in a tungsten carbide film

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Abstract

Magnetoconductance (MC) measurements have been performed on a 2140 Å thick tungsten carbide film at temperatures very close to the superconducting transition temperature T_c of the film. The data are dominated by superconducting fluctuations. A novel three-dimensional phenomenological model is proposed to explain the MC data, yielding good fits. The Larkin beta factor, β_{Larkin} , appeared as a fitting parameter. An expression proposed by Larkin for β_{Larkin} failed badly for temperatures extremely close to T_c . But at intermediate and high temperatures compared to T_c , Larkin's expression gave very good agreement in fits to the MC data. At temperatures very close to T_c , a crossover from three dimensions to two dimensions was observed in the behaviour of the MC of the film.

1. Introduction

Superconducting fluctuations (SCFs) have been observed for over 45 years since the pioneering experiments by Buchel and Hilsch [1] on amorphous bismuth films and later by Shier and Ginsberg [2]. Superconducting fluctuation conductivity causes the 'rounding' of the resistance transition curve above the superconducting transition temperature T_c in high-resistance thin films. Glover demonstrated that the SCF conductivity, also known as the excess conductivity or paraconductivity, follows a Curie–Weiss-type law [3]. The rounding is caused by fluctuations of the superconducting order parameter. Even above T_c , the fluctuations create superconducting Cooper pairs of finite lifetime that contribute to the conductivity. The Curie–Weiss-type conductivity law was derived microscopically by Aslamazov and Larkin [4]. Many theoretical and experimental papers followed on the subject, focusing on the two-dimensional (2D) aspects of the topic, including the early review papers by Glover [5]

and Skocpol [6] and Tinkham's book [7]. A particularly important experimental paper by Serin *et al* [8] reported on magnetoconductance (MC) measurements made on thin (500 Å) aluminium films. Both the theories and data for the zero-field conductivity and MC in two dimensions are well understood.

In contrast to the 2D results, the experimental and theoretical publications in 3D films and samples are limited, with the outstanding exception of studies of the high-temperature superconductors [9, 10]. For example, there is no simple 3D expression for magnetoconductivity using the *Aslamazov–Larkin* model when T is slightly greater than T_c and there is *no* satisfactory 3D expression for the magnetoconductivity using the *Maki–Thompson* model when T is considerably greater than T_c . Moreover, there is *no* prediction for the novel scattering time in three dimensions when normal electrons scatter off the 3D SCF regions, in contrast to the interesting 2D scattering formulation of Brenig *et al* [11, 12].

Probably the lack of theoretical interest results from the scarcity of published MC data on bulk samples and thick films, and there are many reasons for this. The MC measurements are nontrivial. First, most 3D metallic samples are thick and hence have very low resistances in their normal state at liquid helium temperatures. Sensitive ac lock-in detection techniques are required to measure the very small changes to zero resistance upon the approach to the superconducting transition temperature. To enhance the normal resistance values, one tries to use disordered alloys, but then one must worry that such samples have convenient T_c 's in the liquid He⁴ and He³ temperature range. Generally, the T_c 's are depressed to lower temperatures in such alloys. However, the most serious experimental problem is temperature stability during the magnetic field sweeps. Temperature drifts of more than several millikelvin during the sweeps will completely corrupt the MC data, with the temperature dependence of the resistance dominating over the magnetic field dependence of the resistance. In addition, we have been plagued with inhomogeneous samples that exhibit more than one superconducting transition temperature upon their cooling down to the superconducting state. Note that we define the MC in three dimensions as $\Delta\sigma = \sigma(B, T) - \sigma(0, T)$ in units of $(\Omega \text{ cm})^{-1}$.

In view of these experimental problems and challenges, there have been very few published 3D MC results on bulk samples. Almost all these data are taken at temperatures much greater than T_c , where the weak localization (WL) contribution and to a lesser degree the electron–electron interaction (EEI) contribution to the MC are as important or even more dominating than the SCF contribution. The relevant MC papers include measurements on disordered MgCuZn alloys by Meiners-Hagen and Gey [13], on disordered alloys of Ti–Al–(Sn, Co) by Wu and Lin [14], on Mg–Zn metallic glasses by Richter *et al* [15] and on amorphous Cu–Ti–Au alloys by Hickey *et al* [16].

The goal of this paper is to extend MC measurements on a ‘3D’ film as close as possible to T_c and to interpret the data in terms of the Larkin beta prefactor, β_{Larkin} .

2. The zero-field superconducting fluctuation conductivity

About 40 years ago, Aslamazov and Larkin (AL) calculated the influence of SCFs on electrical conductivity above the superconducting transition temperature T_c [4, 17]. The AL contribution results from the direct acceleration of the fluctuation-induced Cooper pairs above T_c . Close to T_c for a 2D film, Aslamazov and Larkin obtained the zero-field Curie–Weiss-type law expression for the conductivity:

$$\sigma^{\text{AL,2D}}(T) = e^2/[16\hbar \varepsilon(T)] = e^2/[16\hbar(T/T_c - 1)] \quad (1)$$

where $\varepsilon(T) = \ln(T/T_c) \approx T/T_c - 1$. In 2D, σ has units of $(\Omega/\square)^{-1}$ and one often divides the film conductivity by the film thickness d to compare the results in three dimensions. If

one wishes to extend the temperature range of equation (1) considerably above T_c , then one replaces $\varepsilon(T) = T/T_c - 1$ by $\ln(T/T_c)$ [3, 18].

The 2D AL conductivity expression should be contrasted with the 3D zero-field conductivity expression [3]:

$$\sigma^{\text{AL},3\text{D}}(T) = e^2 / \{32\hbar \xi(0) [\varepsilon(T)]^{1/2}\} \quad (2)$$

where $\xi(0) = 0.85(\xi_{\text{BCS}}l)^{1/2}$ is the zero-temperature coherence length that appears in the Ginzburg–Landau superconductivity coherence length $\xi_{\text{GL}}(T) = \xi(T) = \xi(0)/|\varepsilon(T)|^{1/2}$. l is the elastic mean free path. Recall that ξ_{BCS} is the Bardeen–Cooper–Schrieffer (BCS) coherence length given by $\xi_{\text{BCS}} \approx 0.18\hbar v_{\text{F}}/k_{\text{B}}T_c$ [7].

The Aslamazov–Larkin theory provided excellent agreement with measurements on thin amorphous films having *high* normal resistances per square. However, measurements on *clean* Al films by the Brookhaven National Laboratory group and the University of Rochester group showed SCF conductivity values to be much larger than the AL predictions [19–22]. Maki suggested another contribution to explain this large conductivity [23]. The Maki–Thompson (MT) contribution originates from the inertia of the superconducting pairs after decaying into pairs of quasiparticles with opposite momenta. Since elastic scattering by impurity potentials conserves time-reversal symmetry, these quasiparticle pairs continue to have nearly zero total momentum and to produce excess conductivity. The quasiparticle pair lifetime is limited by inelastic scattering, which breaks the quasiparticle pairs. Thus, the more disordered the film, the shorter will be the lifetime and hence the less important becomes the MT contribution. Thompson showed that the non-physical divergence in the 1D and 2D cases is prevented by the presence of any pair-breaking effect such as magnetic impurities or a magnetic field [24]. For the case of three dimensions, the MT conductivity term takes the form [25]

$$\sigma^{\text{MT},3\text{D}}(T) = e^2 / \{8\hbar \xi(0) [(\varepsilon(T))^{1/2} + \delta^{1/2}]\} \quad (3)$$

where $\delta(T)$ is the pair-breaking parameter given by [24–26]

$$\delta(T) = \pi\hbar / [8k_{\text{B}}T \tau_{\text{in}}(T)] = \pi e D_{\text{dif}} B_{\text{in}} / (2k_{\text{B}}T). \quad (4)$$

The MT contribution is important when the pair-breaking parameter $\delta(T)$ is small. This implies that the inelastic scattering time τ_{in} be large (weak scattering) or the inelastic field B_{in} be small as is the case for clean films. The relation between the inelastic magnetic fields B_{in} 's extracted from the MC data and the inelastic scattering times τ_{in} 's is given by $\tau_{\text{in}} = \hbar / (4e D_{\text{dif}} B_{\text{in}})$, with D_{dif} being the diffusion constant in units of $\text{m}^2 \text{s}^{-1}$.

Thus, the total 3D zero-field conductivity is given by the sum of the normal conductivity $\sigma^{\text{normal},3\text{D}}$, the AL term and the MT term:

$$\sigma^{\text{total},3\text{D}}(T) = \sigma^{\text{normal},3\text{D}} + \sigma^{\text{AL},3\text{D}}(T) + \sigma^{\text{MT},3\text{D}}(T). \quad (5)$$

The small contributions from the weak localization and electron–electron interaction theories have been neglected for temperatures close to T_c .

Note that if one uses units of $(\Omega \text{ cm})^{-1}$ for the conductivity and magnetoconductivity data, one must divide all the theoretical equations by a factor of 1/100 to convert from the unit of m to cm. The 3D expressions are always in units of $(\Omega \text{ m})^{-1}$.

3. Superconducting fluctuation magnetoconductivity in two dimensions

3.1. The 2D Aslamazov–Larkin MC theories

The magnetoconductance in two dimensions, $\Delta\sigma^{\text{AL},2\text{D}}$, for highly disordered films exhibiting SCFs by the AL term was first derived by Abrahams, Prange and Stephen (APS) [28]. Using

a microscopic calculation, Redi reconfirmed their expression [29]; we use Redi's notation, which differs slightly in the definition of the variable z used by APS:

$$\Delta\sigma^{\text{AL,2D}}(B, T) = \sigma^{\text{AL,2D}}(B = 0, T)\{8z^2[\psi(1/2 + z) - \psi(1 + z) + 1/(2z)] - 1\} \quad (6)$$

where $\sigma^{\text{AL,2D}}(B = 0, T)$ is the zero-field AL expression for conductivity, ψ is the digamma function and $z = B_{\text{SCF}}/B$. Useful formulae for approximating the digamma function can be found in [30, 31]. Equation (6) has the desirable property that in the low-field limit, the MC follows a quadratic field dependence given as

$$\Delta\sigma^{\text{AL,2D}}(B \rightarrow 0, T) = -\sigma^{\text{AL,2D}}(B = 0, T)B^2/(8B_{\text{SCF}}^2) \quad (7)$$

and at large fields the MC *saturates* at the value of

$$\Delta\sigma^{\text{AL,2D}}(B \rightarrow \infty, T) = -\sigma^{\text{AL,2D}}(B = 0, T)[1 - 4B_{\text{SCF}}/B]. \quad (8)$$

If the high-field MC data saturate with a $1/B$ dependence, then this behaviour is useful in identifying the dimensionality since in three dimensions, the MC saturates differently with a $1/B^{1/2}$ dependence. Many years ago Usadel first predicted this $1/B$ saturation dependence for 2D films [32]. This field dependence is useful since the zero-temperature coherence length $\xi(0)$ can be deduced from the 'high' field MC data knowing the value for B_{SCF} .

According to Redi [29], the characteristic 'superconducting fluctuation' field, B_{SCF} , is defined as

$$B_{\text{SCF}} = c\varepsilon(T)k_{\text{B}}T/(\pi e D_{\text{dif}}) \quad (9)$$

where $\varepsilon(T) = \ln(T/T_c) \approx T/T_c - 1$ and c is a numerical factor. Redi suggested that $c = 2$ [29], APS defined $c \approx 6$ [28], Tinkham implied that $c \approx 3.4$ from his definition of B_{c2} [7] and Bergmann [18] and Wiesmann *et al* [33] set $c = 4$ according to their expression that defines D_{dif} from the slope values of dB_{c2}/dT . We have fixed $c = 3.4$ throughout this paper.

B_{SCF} can also be expressed in terms of the Larkin beta factor β_{Larkin} [34]: β_{Larkin} appears as a prefactor in the MT magnetoconductance expressions:

$$\beta_{\text{Larkin}} = \pi^2/\{4 \ln(T/T_c)\} \quad T \geq T_c. \quad (10)$$

For temperatures much greater than T_c , Larkin suggested [36]

$$\beta_{\text{Larkin}} = \pi^2/\{6[\ln(T/T_c)]^2\} \quad T \gg T_c. \quad (11)$$

The characteristic field B_{SCF} near T_c can be expressed in terms of β_{Larkin} as

$$B_{\text{SCF}} = c\pi k_{\text{B}}T/(4e D_{\text{dif}}\beta_{\text{Larkin}}). \quad (12)$$

But even more interestingly, B_{SCF} can be reformulated in the following way, first suggested by APS [28]:

$$B_{\text{SCF}} = 0.78\Phi_0(T/T_c - 1)/\{2\pi[\xi(0)]^2\}. \quad (13)$$

Here Φ_0 is the fluxoid equal to $h/2e = 2.07 \times 10^{-15} \text{ T m}^2$. In this representation, B_{SCF} has almost the identical mathematical form that B_{c2} has, since [7]

$$B_{c2} = \Phi_0(1 - T/T_c)/\{2\pi[\xi(0)]^2\}. \quad (14)$$

Recall that B_{c2} is the magnetic field that destroys superconductivity in the mixed state of a type II superconductor *below* T_c . In analogy, B_{SCF} is the magnetic field that destroys the SCF regions *above* T_c , and B_{SCF} acts as a 'mirror image or reflection' of B_{c2} about the vertical axis passing through T_c . In order to derive equation (14), we have used the following relations: $D_{\text{dif}} = v_{\text{F}}l/3$, $\xi_{\text{BCS}} = 0.18\hbar v_{\text{F}}/(k_{\text{B}}T_c)$ where $T_c \approx T$, $\xi(0) = (0.85\xi_{\text{BCS}}l)^{1/2}$ and $\Phi_0 = h/2e$.

Note that equation (14) is valid for temperatures slightly greater than T_c . We prefer using the general expression given by equation (9) for B_{SCF} .

Recalling that B_{SCF} vanishes as T approaches T_c , we get the surprising result from equation (7) that extremely small magnetic fields should be able to saturate the SCF magnetoconductivity owing to the B^2/B_{SCF}^2 dependence. This prediction was recently pointed out by Meiners-Hagen and Gey [13]. This behaviour certainly works against our physical intuition where one would postulate that the Cooper pairs would be more strongly bounded to one another as the temperature approaches T_c and that stronger rather than weaker magnetic fields would be required to quench the SCF regions.

3.2. The 2D Maki–Thompson MC theories

We now consider the 2D Maki–Thompson (MT) magnetoconductance. Important theoretical work on the 2D Maki–Thompson MC was published by Larkin [34], who showed that values for the inelastic magnetic fields, B_{in} 's, could be deduced from the MC data. Larkin suggested the following expression for the MC [34]:

$$\Delta\sigma^{\text{MT},2\text{D}}(B, T) = -(e^2/2\pi^2\hbar)\beta_{\text{Larkin}}\{\psi(1/2 + B_{\text{in}}/B) + \ln(B/B_{\text{in}})\} \quad (15)$$

with $\beta_{\text{Larkin}} \approx \pi^2/[4 \ln(T/T_c)]$. Note the simplicity of the Larkin expression, involving only one digamma function ψ and one ‘ln’ term. For small fields, equation (15) simplifies to

$$\Delta\sigma^{\text{MT},2\text{D}}(B \rightarrow 0, T) = -(e^2/2\pi^2\hbar)\beta_{\text{Larkin}}B^2 / (24B_{\text{in}}^2). \quad (16)$$

But in the high-field limit, $\Delta\sigma^{\text{MT},2\text{D}}(B, T)$ *diverges* owing to the $\ln(B)$ term. This prediction is *unphysical* since at high fields the MC is *finite* and equal to the difference between the normal conductivity value $\sigma(B)$ and the finite zero-field conductivity value $\sigma(0)$. Since the normal conductivity $\sigma(B)$ is generally much smaller than the large zero field conductivity arising from the SCFs, the MC is negative and generally very large but *finite* close to T_c . Note that the characteristic spin–orbit field, B_{so} , does not appear in the Larkin expression. The MT term will not be affected by the spin–orbit scattering since this term is concerned with the singlet part of the electron–electron interaction in the Cooper channel. Also, the effective spin–spin field, B_s , does not appear in the Larkin expression for its presence would imply magnetic moments which would ‘break up’ the Cooper pairs. The $\Delta\sigma^{\text{MT},2\text{D}}$ contribution would be suppressed in exactly the same way that the weak localization effects would be suppressed in the presence of the magnetic spin–spin scattering.

The divergence problem in two dimensions was resolved by Lopes dos Santos and Abrahams (LSA) [27], who suggested that the Larkin expression should be replaced by the expression

$$\Delta\sigma^{\text{MT},2\text{D}}(B, T) = -(e^2/2\pi^2\hbar)\beta_{\text{LSA}}\{\psi(1/2 + B_{\text{in}}/B) - \psi(1/2 + B_{\text{SCF}}/B) + \ln(B_{\text{SCF}}/B_{\text{in}})\} \quad (17)$$

where $\beta_{\text{LSA}}(T, \delta)$ differs slightly from Larkin’s β_{Larkin} , and is defined as

$$\beta_{\text{LSA}}(T, \delta) = \pi^2/\{4[\ln(T/T_c) - \delta]\}. \quad (18)$$

Here δ is the pair-breaking parameter given by equation (4).

Equation (17) has the desirable property that in the high field limit, the MC now *saturates* at

$$\begin{aligned} \Delta\sigma^{\text{MT},2\text{D}}(B \rightarrow \infty, T) &\approx e\pi k_{\text{B}}T/(8\hbar D_{\text{dif}}B) - \sigma^{\text{MT},2\text{D}}(B=0, T) && \text{or} \\ \Delta\sigma^{\text{MT},2\text{D}}(B \rightarrow \infty, T) &\approx (\pi^2/8c\varepsilon)(e^2/\hbar)(B_{\text{SCF}}/B) - \sigma^{\text{MT},2\text{D}}(B=0, T) && (19) \end{aligned}$$

and for the very small field limit,

$$\Delta\sigma^{\text{MT},2\text{D}}(B \rightarrow 0, T) \approx \left[-e^2 \beta_{\text{Larkin}} / (2\pi^2 \hbar) \right] \times \left[\{ \ln(T/T_c) - \delta \} / \ln(T/T_c) \right] \left[B^2 / (24 B_{\text{in}}^2) \right] \quad (20)$$

similar to the low field limit of equation (16).

A most important observation is that the LSA expression of equation (17) is simply the *difference* between *two* Larkin MC expressions given by equation (15), with the first expression having the inelastic field B_{in} appearing in the arguments of the digamma function and ‘ln’ term while the second expression has the SCF field B_{SCF} appearing in the arguments of the digamma function and ‘ln’ term. We will use this observation for proposing a 3D MT expression.

4. Superconducting fluctuation magnetoconductivity theories in three dimensions

4.1. The 3D Aslamazov–Larkin MC theories

Usadel derived a complicated expression for the 3D AL case involving a summation of digamma and trigamma functions in his equation (35) [32]. However, his expression for $\Delta\sigma^{\text{AL},3\text{D}}$ does saturate to $-\sigma^{\text{AL},3\text{D}}(0)$ in the limit of high fields with an additional small but important positive contribution $1/B^{1/2}$ at moderately large fields [32]. His expressions (equations (35), (22), (23) and (24)) are not easily adopted to fitting MC data [32].

Hikami and Larkin suggested an alternative 3D AL expression that involves a series that includes the Bernoulli numbers [10]. Unfortunately this series converges slowly and poorly owing to the slow convergence property of the Bernoulli numbers. In the small field limit where $B \ll B_{\text{SCF}}$, Usadel proposed [32] that

$$\Delta\sigma^{\text{AL},3\text{D}}(B \rightarrow 0, T) = - \left[e^2 / (8\hbar \xi(0) \varepsilon^{5/2}) \right] \left[\xi(0)^2 B / \Phi_0 \right]^2. \quad (21)$$

Hikami and Larkin derive an almost identical expression except that the ‘1/8’ term is replaced by the factor ‘ $3\pi^2/128$ ’ [10]. Experimental constraints make the observation of this B^2 dependence of the MC most difficult since typical B ’s are much less than 10 Gauss. Temperature fluctuations, trapped flux in the superconducting magnet and also the Earth’s magnetic field will all corrupt the very low field MC data.

Usadel has suggested the following high magnetic field limit for the 3D AL MC term [32]:

$$\Delta\sigma^{\text{AL},3\text{D}}(B \rightarrow \infty, T) \approx [0.24 e^2 / \hbar] [B_{\text{SCF}} / \Phi_0] [1 / \varepsilon^{3/2}] [\hbar / eB]^{1/2} - \sigma^{\text{AL},3\text{D}}(B = 0, T). \quad (22)$$

In this 3D case, the MC saturates as $1/B^{1/2}$, in contrast to the $1/B$ dependence in two dimensions.

We see no simple intuitive way of ‘extending’ the 2D formulae of APS–Redi, namely equation (6), to the 3D AL case.

4.2. The 3D Maki–Thompson MC theories and a novel phenomenological expression

For the *MT* case in three dimensions the theoretical situation is incomplete. Altshuler, Aronov, Larkin and Khmel’ nitskii (AALK) have suggested the following expression [35]:

$$\Delta\sigma^{\text{MT},3\text{D}}(B, T) = - \left[e^2 / (2\pi^2 \hbar) \right] \beta_{\text{Larkin}} [eB / \hbar]^{1/2} f_3(B / B_{\text{in}}). \quad (23)$$

Note the interesting factor $[eB / \hbar]^{1/2}$ which is the inverse of the magnetic length, $1/l_{\text{mag}}$. A length scale is required since the conductivity and magnetoconductivity in three dimensions always have units involving an inverse length, namely $(\Omega \text{ m})^{-1}$ or $(\Omega \text{ cm})^{-1}$. Again, assuming the absence of magnetic moments, the characteristic spin–orbit and spin–spin magnetic fields,

B_{so} and B_s , do not enter into the argument of $f_3(x)$ —only B_{in} , the inelastic scattering field. Baxter *et al* [36] have given a numerically convenient approximation for the $f_3(x)$ function, which is accurate over the entire range of x and retains the correct asymptotic limits:

$$f_3(x) = 2[2 + 1/x]^{1/2} - 2[1/x]^{1/2} - [1/2 + 1/x]^{-1/2} \\ - [3/2 + 1/x]^{-1/2} + (1/48)[2.03 + 1/x]^{-3/2} \\ f_3(x \rightarrow 0) \rightarrow (x^{3/2})/48$$

and

$$f_3(x \rightarrow \infty) \rightarrow 0.6049 - 2[1/x]^{1/2}. \quad (24)$$

Note at high fields, the MC is predicted to *diverge* as $B^{1/2}$ since $f_3(x)$ saturates to 0.605 and the prefactor involving the inverse magnetic length $[eB/\hbar]^{1/2}$ dominates. As in the 2D Larkin expression, this *divergence is unphysical*. One can fit equation (23) to the very low field MC data but the fitting task is difficult since the two fitting parameters, β_{Larkin} and B_{in} , can take on a very wide range of values and physical insight must be used to limit their values to realistic magnitudes.

Is it possible to postulate an expression for the 3D Maki–Thompson MC process? We now present a phenomenological approach.

Recall that Lopes dos Santos and Abrahams solved the divergence MC problem in high magnetic fields *in two dimensions* by proposing an expression involving the *difference* of two Larkin expressions [27]. What happens if we ‘*extend*’ their 2D formulation *to the 3D case* by taking the *difference of two AALK expressions*, namely the difference of two $f_3(x)$ functions as follows:

$$\Delta\sigma^{\text{MT,ext.3D}}(B, T) = -[e^2/(2\pi^2\hbar)]\beta_{\text{Larkin}}[eB/\hbar]^{1/2} [f_3(B/B_{in}) - f_3(B/B_{\text{SCF}})]. \quad (25)$$

As extreme as this idea sounds, let us check the magnetic field limits of equation (25). For the case of small magnetic fields,

$$\Delta\sigma^{\text{MT,ext.3D}}(B \rightarrow 0, T) \\ = -[e^2/(2\pi^2\hbar)]\beta_{\text{Larkin}}[eB/\hbar]^{1/2} (1/48)[(B/B_{in})^{3/2} - (B/B_{\text{SCF}})^{3/2}]. \quad (26)$$

As long as B_{SCF} is considerably larger than B_{in} , equation (26) goes over to the low field $(B/B_{in})^2$ behaviour of the AALK expression and is well behaved.

In the opposite limit of high fields, one can use the asymptotic limit of $f_3(x) \approx 0.6049 - 2[1/x]^{1/2}$ to find the very surprising result:

$$\Delta\sigma^{\text{MT,ext.3D}}(B \rightarrow \infty, T) = -[e^2/(2\pi^2\hbar)]\beta_{\text{Larkin}}2[(eB_{\text{SCF}}/\hbar)^{1/2} - (eB_{in}/\hbar)^{1/2}]. \quad (27)$$

Again as long as B_{SCF} is considerably larger than B_{in} , equation (27) predicts a *finite saturation value* for the MC at high fields and is well behaved. But we *stress* that equation (25) needs to be put on firm theoretical grounds.

We denote equation (25) as the ‘*extended 3D Lopes dos Santos–Abrahams theory*’. There are three fitting parameters in equation (25): the Larkin beta factor β_{Larkin} , the inelastic field B_{in} and the superconducting fluctuation field B_{SCF} . If one accepts Larkin’s definition that $\beta_{\text{Larkin}} = \pi^2/[4\varepsilon(T)] = \pi^2/[4\ln(T/T_c)]$, then B_{SCF} can be expressed in terms of β_{Larkin} as $B_{\text{SCF}} = c\pi k_B T/(4eD_{\text{diff}}\beta_{\text{Larkin}})$, and hence there should be self-consistency between the β_{Larkin} values and the B_{SCF} fitted values. To simplify the fitting procedure, we have set β_{Larkin} to Larkin’s theoretical prediction of $\beta_{\text{Larkin}} = \pi^2/[4\ln(T/T_c)]$ and we have treated B_{in} and B_{SCF} as two free-fitting parameters in the MC expression of equation (25).

Recently, Meiners-Hagen and Gey have suggested a complicated expression for the 3D MT MC term [13].

In conclusion, more theoretical work is certainly needed on the 3D SCF problem.

5. Sample characterization and measurement techniques

The tungsten carbide (WC) films were supplied by Dr Ajit K Meikap and were fabricated by ion-assisted deposition; a gas mixture of C_2H_2 and Ar in a ratio of 4/1 was used at 10^{-4} Torr. Silver paint was used to attach electrical leads to the WC films. The WC film had a normal resistance of 13.82Ω at liquid helium temperatures. The geometric factor of the WC film, $f_g = 28.7 \times 10^{-6}$ cm, was used to convert resistance to resistivity. The film thickness was 2140 \AA .

Measurements were made by mounting the sample inside a pumped liquid helium probe. The sample was positioned in the middle of a 7 T superconducting magnet. Temperature stability was limited to ± 3 mK using a Neocera LTC-11 or LTC-21 temperature controller. Owing to the uncertainty in the zero field resistance magnitudes arising from temperature instability, the fitting parameters are known for an accuracy of only $\pm 25\%$. Unfortunately, temperature stability was not good enough to observe the B^2 dependence of the MC at very low fields. However, when bad temperature drifts did occur, the MC run was repeated until better quality data were obtained. The small magnitudes of resistances were measured using a Linear Research ac bridge to an accuracy of $\pm 1\%$. We used the smallest possible excitation voltages (smallest currents) in order to minimize both the destruction of superconductivity in the SCF regions and the Joule heating in the sample. Thus, a difficult compromise was made between accuracy of the data and maximizing the superconductivity effect.

6. Results and analysis

The WC film had a T_c of approximately 3.949 K where the resistance vanished, as shown in figure 1. This T_c was estimated by a linear extrapolation of the zero-field resistance cool-down data. The SCF region extended above 7 K, with maximum resistance occurring at 7 K. The maximum resistance results from a competition between the electron–electron interactions (EEI) process which causes resistance to increase with decreasing temperatures, in contrast to the SCF process that causes resistance to decrease to zero. Weak localization (WL) also contributes to the ‘rounding’ behaviour. If there are strong spin–orbit interactions as in our case, then there is an ‘anti-localization’ that causes a decrease of the resistance with decreasing temperature. For weak spin–orbit interactions, the WL contributes an increase of the resistance, along with the EEI contribution, with decreasing temperatures. These data are compared to a fit (solid curve in figure 1) using the 3D equations (2)–(5). One fitting parameter, the zero-temperature coherence length $\xi(0) \approx 75 \text{ \AA}$, was used as well as values of B_{in} extracted from the MC fits. Agreement is poor very close to T_c , but is quite acceptable at higher temperatures. The disagreement probably arises from the use of the 3D expressions solely rather than 2D expressions where a crossover in dimensionality takes place very close to T_c . Hikami and Larkin suggested an AL crossover expression, their equation (2.6) in [10], but this expression did not give a significant improvement.

A value for the diffusion constant D_{dif} is needed to evaluate B_{SCF} in equation (9) and in converting a characteristic magnetic field to a scattering time using the expression $\tau_x = \hbar/(4eD_{dif}B_x)$. D_{dif} was determined by cooling the film below T_c and measuring the magnetic field B_{c2} , which restored the resistance to one-half of the normal resistance value. Figure 2 shows B_{c2} data for the WC film. By using the expression $D_{dif} = ck_B/[\pi e(-dB_{c2}/dT)]$ with $c = 3.4$ (obtained by taking the temperature derivative of equation (9)), a value of $D_{dif} = 0.276 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$ was determined.

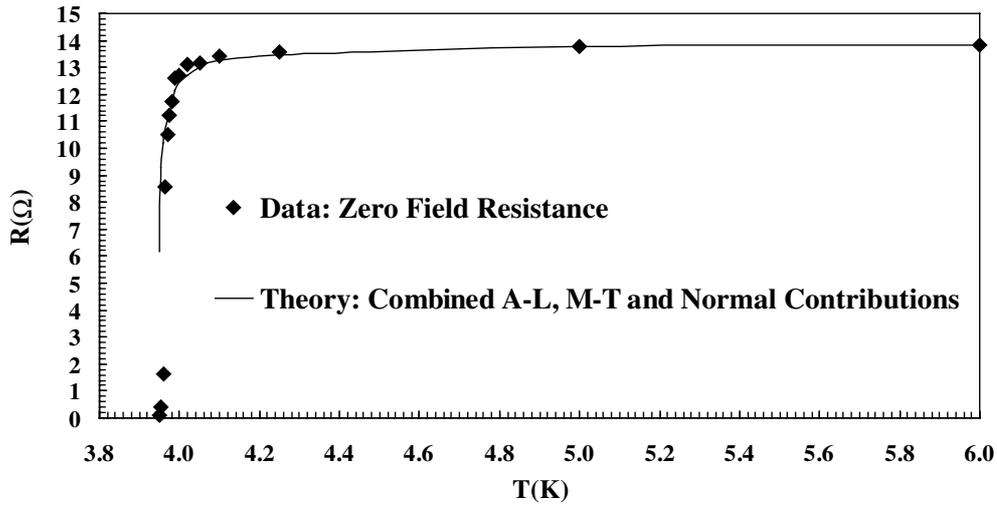


Figure 1. The zero-field resistance data for the tungsten carbide film. The ‘rounding’ above 6 K and drop in resistance results from a competition between the electron–electron interference and between superconducting fluctuations and ‘anti-localization effects’, arising from a large spin–orbit scattering field. The solid curve is a fit using equations (2)–(5) with $\xi(0) = 75 \text{ \AA}$. Very close to $T_c = 3.949 \text{ K}$, the agreement is poor.

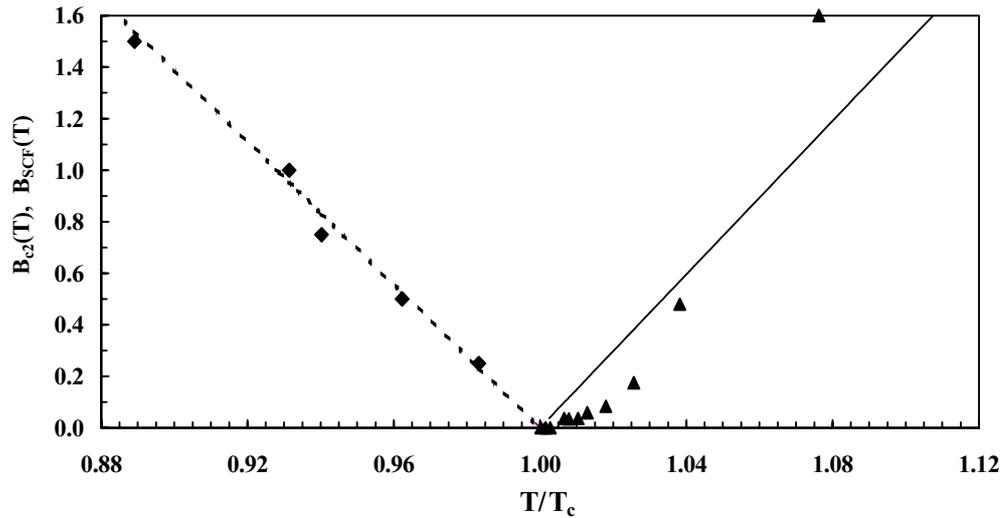


Figure 2. B_{c2} values measured below $T_c = 3.949 \text{ K}$ for the WC film. The criterion used for obtaining the B_{c2} 's was the one-half normal resistance method. The diffusion constant $D_{\text{dif}} = 0.276 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$ was extracted using the relation $D_{\text{dif}} = 3.4k_B / [\pi e(-dB_{c2}/dT)]$. The dashed line is a fit using equation (14) to B_{c2} with $\xi(0) = 47 \text{ \AA}$. The B_{SCF} data that appear above T_c were obtained from fits to the MC data. The solid line is derived from equation (9).

Over the entire temperature range of $3.950 \text{ K} \leq T \leq 8.00 \text{ K}$, the MC data are dominated by the SCF contribution. The MC data are particularly interesting at temperatures very close to $T_c = 3.949 \text{ K}$.

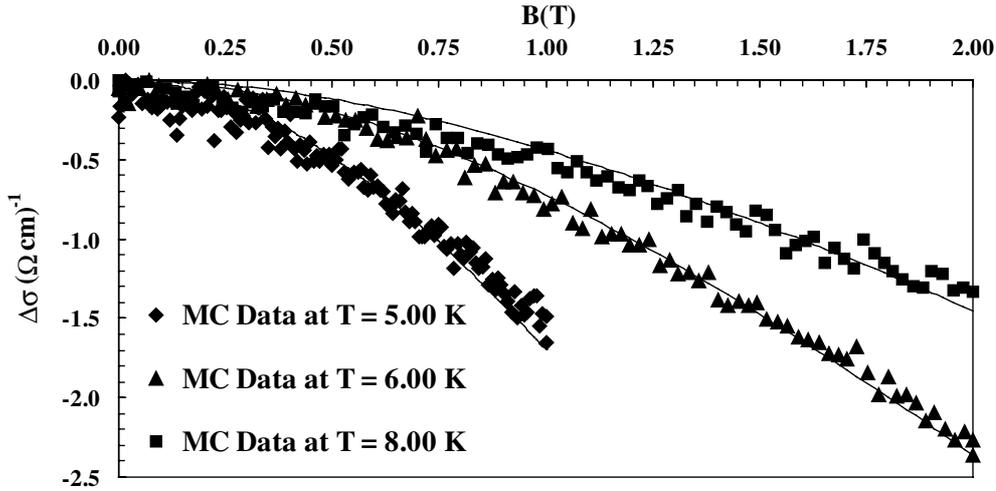


Figure 3. MC data taken at ‘high temperatures’ relative to $T_c = 3.949$ K. The MC values are small and negative. The data can be nicely described by a relatively large contribution from the ‘extended 3D’ Lopes dos Santos–Abrahams SCF expression, equation (25) and a much smaller contribution from the weak localization theory. The combined contributions are shown by the solid curves.

For all the MC data taken at or above $T = 3.975$ K, we have used the ‘extended 3D’ equation (25) that contains the three fitting parameters, β_{Larkin} , B_{in} and B_{SCF} . In order to simplify the fitting procedure, we assigned to β_{Larkin} the theoretical Larkin prediction, $\beta_{\text{Larkin}} = \pi^2/[4 \ln(T/T_c)]$. We then fitted each set of MC data, varying the magnitudes of B_{in} and B_{SCF} ; B_{in} determines the initial MC slope while B_{SCF} fixes the saturation magnitude of the MC. We then calculated an ‘experimental’ value for β_{Larkin} using equation (12) and the B_{SCF} fitting magnitude. We observed surprisingly good self-consistency, with the ‘experimental’ β_{Larkin} values being within a factor of 2 within the Larkin theoretical values. The only exceptions were the three closest temperature points of 3.950, 3.955, and 3.960 K where the Larkin expression for β_{Larkin} badly underestimated the observed β_{Larkin} values derived from the B_{SCF} values.

At the ‘highest’ temperatures of 5.0, 6.0 and 8.0 K, there is an additional small but important MC contribution from the WL process. A useful summary of the 3D WL expressions can be found in [37]. There are two fitting parameters in the WL expressions: B_{in} , that already appears in the ‘extended 3D’ equation and B_{so} , the spin–orbit field. Owing to the heavy nucleus of the tungsten atom, we have assigned a large value to $B_{\text{so}} = 10$ T. The fits are shown in figure 3, where B_{in} takes a typical value of 0.6 T. Since the data were taken at ‘moderately’ small fields, no saturation trends were observed in the MC, and hence it was difficult to determine accurate values for B_{SCF} .

At ‘intermediate’ temperatures of 4.05, 4.10 and 4.25 K, only the ‘extended 3D’ equation (25) was used since the WL contribution was negligible. Again the data exhibit no definite trends to saturate as seen in figure 4, but values for both B_{in} and B_{SCF} could be determined. And there was consistency between the theoretical and experimental β_{Larkin} values.

Data and fits at the four ‘low’ temperatures of 3.980, 3.990, 4.000 and 4.020 K are shown in figure 5; agreement is very good. From the B_{SCF} values, the zero-temperature coherence length $\xi(0)$ was estimated to be 75 Å. Again there is self-consistency between the theoretical β_{Larkin} values and the experimental values derived from B_{SCF} .

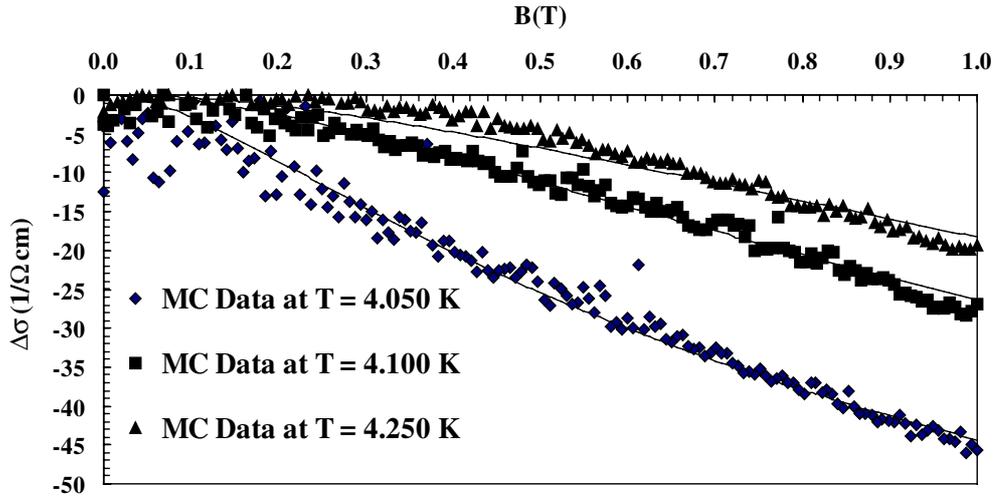


Figure 4. Fits to the MC data taken at ‘intermediate’ temperatures above $T_c = 3.949$ K. The weak localization contribution is negligible and only the ‘extended 3D’ SCF expression of Lopes dos Santos–Abrahams, equation (25), contributes as shown by the solid curves.

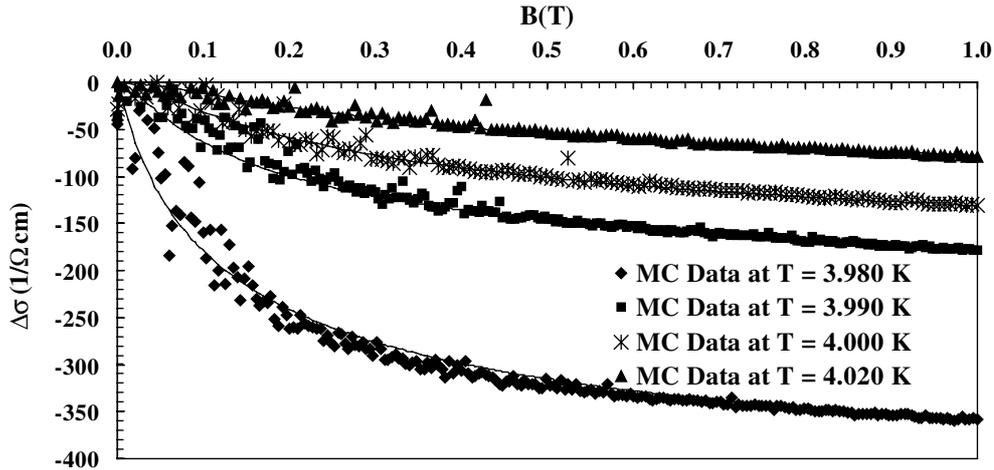


Figure 5. Fits of the MC data taken at ‘low’ temperatures above $T_c = 3.949$ K. The tungsten carbide film is still in the 3D regime and the ‘extended 3D’ Lopes dos Santos–Abrahams 3D expression, given by equation (25), still fits the data nicely as shown by the solid curves. One of the fitting parameters, B_{in} , was observed to tend to zero as the measuring temperatures approached T_c , suggesting that equation (25) might not be valid very close to T_c .

At the ‘low’ temperature of $T = 3.975$ K, the saturation of the MC was again observed as illustrated in figure 6. Here a comparison is made between the AALK theory, equation (23), that shows no saturation property at high magnetic fields and the ‘extended 3D’ equation (25) that exhibits saturation. The two different theoretical behaviours are striking. The three fitting parameters in the ‘extended 3D’ expression are $\beta_{Larkin} = 376$, $B_{in} = 0.000\,005$ T and $B_{SCF} = 0.03$ T.

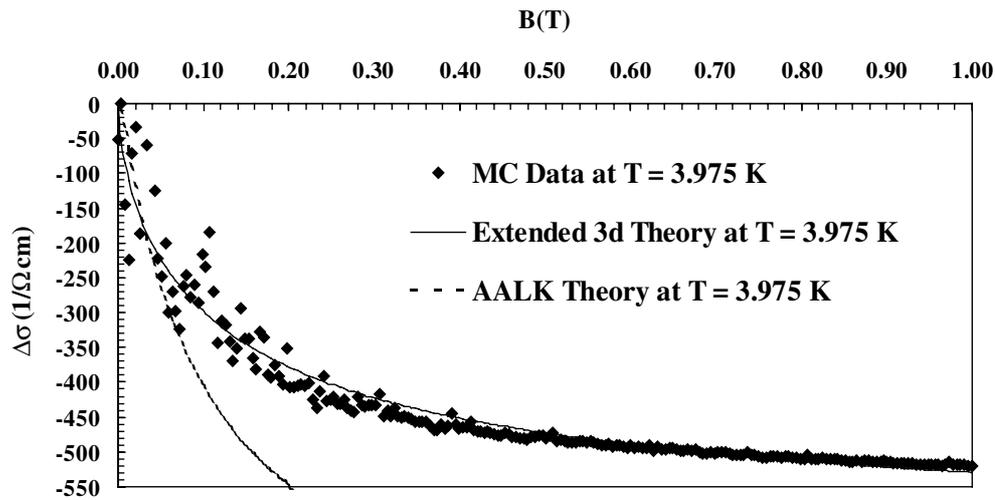


Figure 6. Fits to the MC data taken at the ‘low’ temperature of $T = 3.975$ K, close to $T_c = 3.949$ K. The poorest fit, shown by the dashed-dotted curve, comes from the AALK expression of equation (23). The best fit, represented by the solid curve, uses the ‘extended 3D’ expression of Lopes dos Santos–Abrahams, equation (25). The fitting value for B_{in} was extremely small, $0.000\,005$ T compared to $B_{SCF} = 0.037$ T; and $\beta_{Larkin} = 376$.

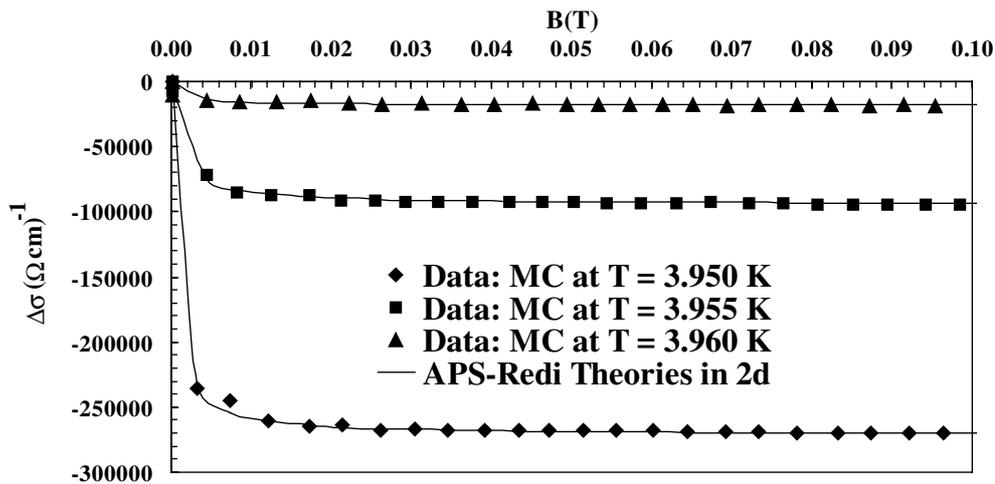


Figure 7. MC data taken ‘just above’ $T_c = 3.949$ K. Note the extremely small fields required to destroy the SCF regions, suggesting a crossover to the 2D regime. The fits, shown by the solid curves, use the 2D formalisms of Abrahams, Prange and Stephen and of Redi according to equation (6). The ‘extended 3D’ Lopes dos Santos–Abrahams expression, equation (25), breaks down in this temperature range.

But the really fascinating and puzzling MC data for the WC film appear at temperatures *just above* T_c as shown in figure 7. First, note the extremely small magnetic fields needed to quench the SCFs. The other outstanding characteristic is the enormously large negative values to which the MC saturates. These data do not closely resemble the behaviour of the higher temperature data of figures 4–6.

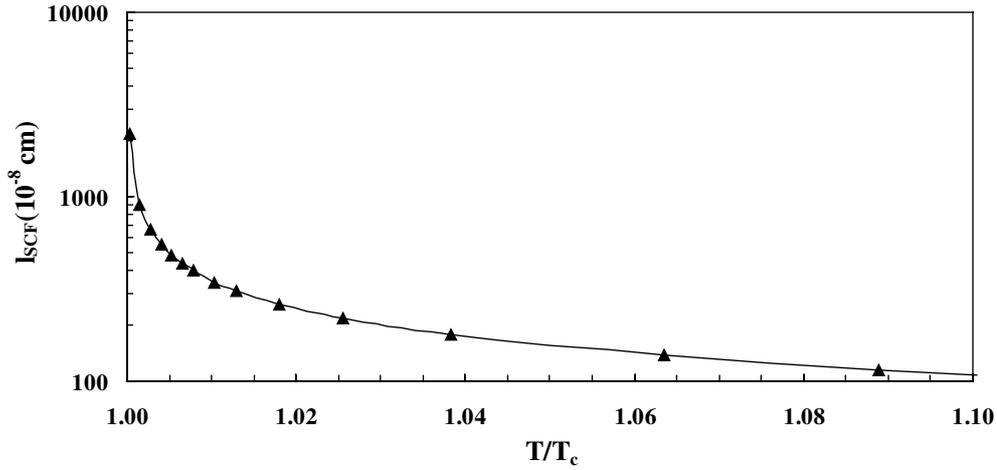


Figure 8. The dominating length scale $l_{\text{SCF}} = [\tau_{\text{SCF}} D_{\text{dif}}]^{1/2} = [\hbar/(4eB_{\text{SCF}})]^{1/2}$ as a function of T/T_c . Note the rapid increase in magnitude of the characteristic length l_{SCF} as $T \rightarrow T_c$, owing to $B_{\text{SCF}} \rightarrow 0$. At approximately 5 mK above T_c , this length exceeds the film thickness of 2140 Å, causing a crossover from 3D to 2D behaviour as T_c is approached. Recall that this is a theoretical prediction: the actual crossover is observed approximately 15 mK above T_c .

This anomalous behaviour suggests that there is a *crossover* from 3D to 2D behaviour. Let us assume that the physical behaviour is controlled by the characteristic field B_{SCF} rather than the inelastic field B_{in} and that the dominating length scale is now $l_{\text{SCF}} = [\tau_{\text{SCF}} D_{\text{dif}}]^{1/2} = [\hbar/4eB_{\text{SCF}}]^{1/2}$ rather than the inelastic length scale $l_{\text{in}} = [\hbar/4eB_{\text{in}}]^{1/2}$. If l_{SCF} now *exceeds* the sample thickness d , then the 2D physics will dominate. A plot of l_{SCF} versus T is shown in figure 8 using $D_{\text{dif}} = 0.276 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$ for the WC film and equation (9) for B_{SCF} . One observes that for temperatures sufficiently close to T_c , the characteristic length l_{SCF} indeed equals the film thickness of 2140 Å. Thus, the 2D MC expressions of APS–Redi, namely equation (6), *should* describe the data of figure 7. Such a 3D to 2D crossover was predicted by Serin *et al* [8] and Abrahams and Woo [38] many years ago, but few people have paid attention to their intriguing prediction. The fits in figure 7 use the 2D AL MC equation (6), with essentially one fitting parameter, B_{SCF} . The second fitting parameter, $\sigma^{\text{A-L,2D}}(B=0, T)$, is known from the zero-field resistance value. In principle, we should be using units of $(\Omega/\square)^{-1}$; but we continue to use the 3D units of $(\Omega \text{ cm})^{-1}$ in figure 7. The fits are very good. According to Abraham *et al* [28], Redi [29] and Usadel [32], the MC should saturate, including a small positive contribution $1/B$, for the 2D case, and this was indeed the behaviour observed. However, Larkin’s expression for $\beta_{\text{Larkin}} = \pi^2/[4 \ln(T/T_c)]$ badly underestimates the experimental β_{Larkin} values deduced from the B_{SCF} ’s. No 2D MT contribution was included since equations (17) and (18) behaved badly with the modified Larkin factor, $\beta_{\text{Larkin}} = \pi^2/\{4[\ln(T/T_c) - \delta]\}$, diverging and then changing sign as $T \rightarrow T_c$.

Interestingly, Serin *et al* suggested that the crossover criterion be given by $\xi(T) \approx d$, where d is the sample thickness [8]. Within a numerical constant, their criterion is equivalent to our criterion that $[\hbar/(4eB_{\text{SCF}})]^{1/2} \approx d$. Below T_c , $\xi(T)$ is interpreted as the coherence length that describes the typical separation between the two oppositely spin-paired electrons in the Cooper pair. Above T_c , $\xi(T)$ *might* be interpreted as the typical separation between the two oppositely spin-paired electrons in the *fluctuating* Cooper pair or perhaps *may* be interpreted as a typical linear size dimension of a SCF region.

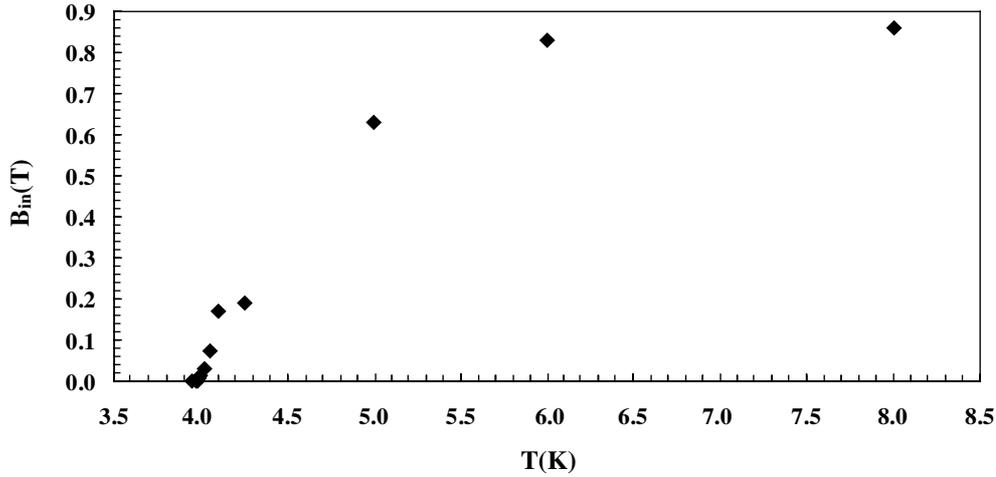


Figure 9. Experimental values for the inelastic scattering field B_{in} above T_c . Refer to the text for arguments of why B_{in} might or might not tend to 0 at T_c .

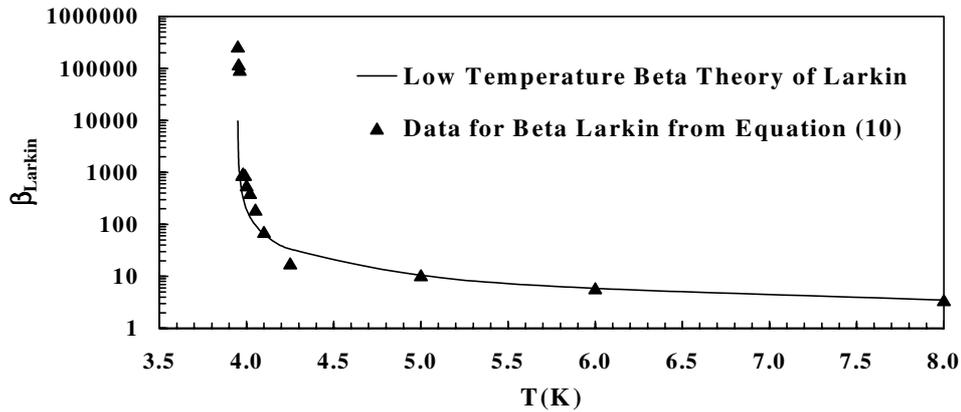


Figure 10. Magnitudes of the Larkin prefactor β_{Larkin} extracted from the MC fits. The solid curve is the Larkin prediction that $\beta_{Larkin} = \pi^2/[4\ln(T/T_c)]$. At temperatures very close to T_c , this expression gives a very poor fit.

Above $T \approx 3.960$ K, we have a *crossover* region from two dimensions to three dimensions where we are not aware of any theories. Thus, we have not presented the MC data at the temperatures of 3.965 and 3.970 K, nor were we successful in fitting these data with any of the above expressions.

Experimental values for B_{SCF} are summarized in figure 2 and compared with the predicted values according to equation (13) where $\xi(0) = 47$ Å was used. Experimental values for B_{in} are shown in figure 9. Lastly, in figure 10 we compare the theoretical Larkin $\beta_{Larkin} = \pi^2/[4\ln(T/T_c)]$ values with the experimental values derived using equation (9) and the B_{SCF} fitted values. Agreement is surprisingly good as long as $T \geq 1.005 T_c$.

7. Discussion

The ‘low’ temperature fits exhibited one very anomalous behaviour—namely that the values for B_{in} tend to 0 as the temperatures approached T_c . We found this observation difficult to accept, having been accustomed to fitting many sets of MC data taken on *normal* films. From such fits using the weak localization expressions, one extracts the total dephasing time $\tau_{\phi,\text{total}}(T) = [1/\tau_0 + 1/\tau_{\text{in}}(T)]^{-1}$; here τ_0 is the *saturated temperature-independent* dephasing time and $\tau_{\text{in}}(T)$ is the inelastic scattering time [39]. As Lin’s group has observed in many metallic *normal* films, the total dephasing time tends to *saturate to finite values* between 0.001 T and 0.02 T at very low temperatures [39]. Using their suggested observation that $(D_{\text{dif}}\tau_0)^{1/2} \approx 1000 \text{ \AA}$ [39], we would have anticipated $B_{\phi,\text{total}} \approx B_0 \approx 0.018 \text{ T}$, for example, several good orders of magnitude greater than our fitting value of $B_{\text{in}} = 0.000\,005 \text{ T}$ determined at the temperature of $T = 3.975 \text{ K}$. However, accurate values for B_{in} cannot be extracted from the very low temperature MC data owing to the dominating presence of the saturated dephasing field B_0 or τ_0 .

The problem might lie in the validity of the temperature ranges for both the 2D and 3D MT expressions of Lopes dos Santos–Abrahams, equations (17) and (25). Recall that as $T \rightarrow T_c$, then $B_{\text{SCF}} \rightarrow 0$. The values for B_{in} must also approach 0 since the condition $B_{\text{SCF}} > B_{\text{in}}$ must be satisfied for these equations to be meaningful. Maybe, these expressions *might* become invalid very close to T_c for some other physical reason, and hence the fitting values for B_{in} as well as B_{SCF} *might not* be physical for $T \rightarrow T_c$. Most likely, the values for B_{in} are meaningful if the second crossover condition is satisfied, namely $l_{\text{in}} = (\hbar/4eD_{\text{dif}}B_{\text{in}})^{1/2} < d$ or $B_{\text{in}} > 0.008 \text{ T}$. This criterion is met for $T \geq 1.01T_c$ or for our measuring temperatures of $T \geq 3.99 \text{ K}$.

From the theoretical viewpoint we must remember that our films become superconductive, consisting of SCF regions above T_c where normal electrons *have condensed* into Cooper pairs. As the transition temperature is approached, there could be *numerous and extensive* SCF regions, and hence many fewer normal electrons to participate in inelastic scattering events. Thus, this argument suggests that owing to the scarcity of normal electrons available to participate in inelastic scattering events, the inelastic scattering time approaches infinity, or that B_{in} approaches 0 as $T \rightarrow T_c$. But, there is also a *strong* counterargument too. There are indeed SCF regions but only a few in number. We now have a percolation picture where these regions become connected as $T \rightarrow T_c$, finally forming one continuous superconducting path across the sample. In this model, there are only relatively few SCF regions, and hence only a few Cooper pairs formed from condensation of a relatively few normal electrons. This argument describes a two-component model consisting of many normal electrons and a limited number of Cooper pairs. Hence this picture most likely predicts a finite inelastic electron scattering time and therefore a finite B_{in} as $T \rightarrow T_c$. There is no clear theoretical consensus on whether B_{in} remains finite or 0 as $T \rightarrow T_c$.

We hope that some of the above-mentioned problems and theoretical questions will stimulate further experimental and theoretical work.

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