Thermodynamic parameters of atomically thin superconductors derived from the upper critical field

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Abstract
The amplitude of the ground-state superconducting energy gap $\Delta(0)$ and relative jump in the electronic specific heat at the transition temperature $\Delta C/\gamma T_c$ are the primary fundamental parameters of any superconductor. Several well-established techniques are available for measuring these values in bulk samples. However, a limited number of techniques can be used to measure these parameters in atomically thin superconductors. Here, we propose a new approach for extracting $\Delta(0)$ and $\Delta C/\gamma T_c$ in atomically thin superconductors by utilizing the upper critical field data from perpendicular, $B_{c2,\perp}(T)$ (when a magnetic field is applied perpendicular to the film surface) and parallel, $B_{c2,\parallel}(T)$ (when a magnetic field is applied in the direction parallel to the film surface), external field directions. The deduced parameters for few-layer-thick Al, Sn, NbSe$_2$, MoS$_2$, magic angle twisted trilayer graphene, and WTe$_2$ are well-matched values expected for strongly and moderately strongly coupled electron-phonon-mediated superconductors. In many reports, the enhancement of $B_{c2,\parallel}(0)$ above the Pauli–Clogston–Chandrasekhar limiting field (i.e. magnetic field required to break the Cooper pair) in atomically thin superconductors has been explained based on the assumption of exotic pairing mechanisms, for instance, Ising-type pairing. Here, we explain the observed $B_{c2,\parallel}(0)$ enhancement based on the geometrical enhancement factor that originates from the sample geometry. This approach does not assume the existence of novel exotic pairing mechanisms in atomically thin superconductors.

Keywords: characterizing technique for atomically thin materials, upper critical field, Pauli–Clogston–Chandrasekhar limiting field, superconducting energy gap, magic-angle twisted trilayer graphene

(Some figures may appear in colour only in the online journal)

1. Introduction

Recently, Cao et al [1], and Zhou et al [2] measured the temperature-dependent upper critical field in twisted trilayer graphene (TTG) superlattices when an external magnetic field was applied parallel to the film surface, $B_{c2,\parallel}(T)$, and reported the extrapolated ground-state upper critical field, $B_{c2,\parallel}(0)$, which was obtained from the fit of $B_{c2,\parallel}(T)$ data [1, 2] to the equation:

$$B_{c2,\parallel}(T) = B_{c2,\parallel}(0) \times \sqrt{1 - \frac{T}{T_c}},$$  \hspace{1cm} (1)

where $T_c$ and $B_{c2,\parallel}(0)$ are free-fitting parameters, which by several times exceeds [1, 2] the Pauli–Clogston–Chandrasekhar limiting field, $B_{CPP}(0)$, for this material:

$$1.5 \times B_{CPP}(0) \lesssim B_{c2,\parallel}(0) \lesssim 3 \times B_{CPP}(0),$$ \hspace{1cm} (2)
where $B_{CPP}(0)$ was assumed to be [1, 2]:

$$B_{CPP}(0) = 1.86 \times T_c. \quad (3)$$

Similar observations have been reported for atomically thin MoTe$_2$ [3–5], NbSe$_2$ [6–9], MoTe$_2$ [10, 11], few-layer stanene [12], PdTe$_2$ [13], TaS$_2$ [7], ion-gated SrTiO$_3$ [14] and WTe$_2$ [15, 16] (detailed reviews of recent studies in this field can be found elsewhere [17–22]).

Based on the observed violation of the Pauli–Clogston–Chandrasekhar limiting field (equation (2)), and the proposal by other research groups (see [1]) that magic-angle twisted trilayer graphene (MATTG) is a unique quantum matter material that can be considered as a material for next-generation quantum devices, including quantum computers.

Here, we point out that the first report of a many-fold increase in $B_{2,\parallel}(T)$ versus the decrease in film thickness $d_{sc}$ (from 450 to 20 nm) was reported by Blumberg and Douglass [23, 24] for Sn films in 1962. The reported increase in $B_{2,\parallel}(T)$ was so high that the maximum available magnetic field of $B_{appl} = 0.3 T \equiv 10 \times B_c(0)$ (where $B_c(0)$ is the ground-state thermodynamic critical field of bulk Sn [24]) allowed the measured $B_{2,\parallel}(T)$ data set for 20 nm thick film [23] only within close proximity to the transition temperature: $0.965 \leq \frac{T}{T_c} \leq 1.0$.

The first study, where the extrapolated $B_{2,\parallel}(0)$ exceeded the $B_{CPP}(0)$ (equation (3)), was reported by Tedrow et al. in 1970 [25], who found that a 5 nm thick Al film exhibits the following:

$$B_{2,\parallel}(0) = 4.84 T > B_{CPP}(0) = 1.86 \times T_c = 4.56 T. \quad (4)$$

Later [26], the same group confirmed this result for several Al films with thicknesses of $d_{sc} < 20$ nm, and in 1982, the same group [27] reported a record high $B_{2,\parallel}(0)$ in 4 nm thick Al films coated with 0.2 nm thick Pt film:

$$B_{2,\parallel}(T = 0.55 K) = 9 T > 4.5 \times T_c = 2.4 \times B_{CPP}(0). \quad (5)$$

Considering that Tedrow et al. [25–27] utilized the 50% normal state resistance criterion to define $B_{2,\parallel}(T)$ in their films, while in some recent papers, a much less restrictive criterion (for instance, 75% of the normal state resistance [16]) was used, it can be concluded that the $B_{2,\parallel}(0)$ enhancement observed in atomically thin Al films (equations (5) and [27]) is identical to the enhancement reported for various van der Waals and electric-field-gated atomically thin crystals [1–17].

This implies that the key parameter for the observed $B_{2,\parallel}(0)$ enhancement in atomically thin superconductors is the film thickness, $d_{sc}$, because an identical effect is observed in pure elemental Al and Sn, on the one hand, and in NbSe$_2$, MoTe$_2$, TTG, few-layer stanene, PdTe$_2$, TaS$_2$, electric-field-gated SrTiO$_3$, WTe$_2$ and MoS$_2$, on the other hand, it cannot be classified as exhibiting any similarity except being atomically thin.

Considering that the proper equation for $B_{PCC} (0)$ [28] is:

$$B_{PCC}(0) = \frac{\Delta(0)}{\sqrt{R} \times \mu_B} \quad (6)$$

where $\Delta(0)$ is the ground-state superconducting energy-gap amplitude, $\mu_B$ is the Bohr magneton, and $g$ is the Lande factor. It should also be stressed that equation (6) has been derived for the case of an infinitely large isotropic 3D superconductor. To the best of the author’s knowledge, detailed theoretical consideration of the angular dependence of $B_{PCC}(0, \theta)$ (where $\theta$ is an external field direction) for iso- and anisotropic superconductors with spatial confinement within several atomic layers has not been performed. Instead, in many reports on atomically thin superconductors, equation (6) is used to declare that the observed upper critical field $B_{2,\parallel}(0)$ exceeds $B_{PCC}(0)$ (calculated by equation (6)), atomically thin superconductors exhibit an exotic type of charge carrier pairing.

By looking more closely at equation (6), it can be concluded that equation (6) can be simplified into equation (3) only for clean s-wave weak coupling superconductors that exhibit:

$$g = 2, \quad (7)$$

and

$$2 \Delta(0) = 3.53 \times k_B T_c. \quad (8)$$

However, for many superconductors, even for s-wave lead, equation (8) is not satisfied [29]:

$$2 \Delta(0) = 4.5 \times k_B T_c \quad (\text{for lead}). \quad (9)$$

For pnictides, experimental data show [30] that some of these superconductors exhibit a gap-to-transition temperature ratio as large as:

$$2 \Delta(0) \gtrsim 9 \times k_B T_c. \quad (10)$$

Based on multiple statements [1, 31, 32] that magic-angle twisted bilayer graphene (MATBG) and magic-angle twisted trilayer graphene (MATTG) are very strongly coupled superconductors, one can conclude that these materials exhibit:

$$2 \Delta(0) \gtrsim 3.53 \times k_B T_c. \quad (11)$$

Thus, even with the assumption that the Lande $g$-factor is unaltered (equations (6) and (7)) in MATBG and MATTG, equation (2) transforms into:

$$B_{2,\parallel}(0) \lesssim B_{PCC}(0). \quad (12)$$

Based on this, the observed $B_{2,\parallel}(0)$ enhancement in MATTG [1, 2] agrees with the Pauli–Clogston–Chandrasekhar limiting field.

Apart from the problem of the Lande $g$-factor in atomically thin superconductors, here we present a model to deduce the ground-state superconducting energy gap, $\Delta(0)$, in these superconductors to at least partially clarify a multiplicative term in primary equation (6) of the Pauli–Clogston–Chandrasekhar limiting field.

The model is based on the analysis of both perpendicular, $B_{2,\perp}(T)$ (when a magnetic field is applied in the direction perpendicular to the film surface) and parallel, $B_{2,\parallel}(T)$, upper critical field data for atomically thin superconductors.
It should be noted that scanning tunnelling spectroscopy (STS) is the most widely used technique to measure the superconducting energy gap, \( \Delta(T) \), in atomically thin films [33]. However, the STS technique measures the out-of-plane component, \( \Delta_z(T) \), of the gap, whereas \( B_{c2,\perp}(0) \) in atomically thin superconductors depends on the in-plane, \( \Delta_{ab}(0) \), gap component.

For anisotropic superconductors in-plane, \( B_{c2,\parallel}(T) \), and out-of-plane, \( B_{c2,\perp}(T) \), the upper critical fields within the Ginzburg-Landau theory are given by [34]:

\[
B_{c2,\perp}(T) = \frac{\phi_0}{2\pi} \times \frac{1}{\xi_{ab}(T)},
\]

\[
B_{c2,\parallel}(T) = \frac{\phi_0}{2\pi} \times \frac{1}{\xi_{ab}(T)} \times \frac{1}{\xi_z(T)},
\]

where \( \phi_0 \) is the superconducting quantum flux. Recently, Farrar et al. [35] pointed out that the Bardeen–Cooper–Schrieffer (BCS) theory of superconductivity [36] considers two main Cooper pairs breaking mechanisms in atomically thin FeSe films under high magnetic field. The first mechanism is orbital pair breaking, which originates from the Lorentz force acting on the charge of the paired electrons/holes. The respective upper critical fields for this mechanism are designated as the orbital limiting upper critical field, \( B_{c2,orb}(T) \), and equations for these fields are given by equations (13) and (14) (where we omitted the use of superscript \( orb \) to simplify equations (13) and (14) and several of the following equations). The second mechanism is that the pairs break as a result of the spin-paramagnetic effect explained above (equation (6)), which is designated as \( B_{c2,\gamma}(T) \). Due to the fact that in many superconductors [37] (including atomically thin films [35]), \( B_{c2}(T) \) is affected by both pair-breaking mechanisms, there is a useful parameter \( \gamma_{\text{M}} \), defined as the magnetic field where both pair-breaking mechanisms, accounted for by the Maki parameter, \( \alpha_M \), and the spin-orbit constant, \( \lambda_{so} \), are present. Both \( \alpha_M \) and \( \lambda_{so} \) have angular dependence, and these parameters are utilized for the analysis of atomically thin superconductors (see, for instance, [35]).

In BCS theory of superconductivity [36]:

\[
\frac{1}{\xi(0)} = \frac{\pi}{\hbar} \times \Delta(0) / v_F,
\]

where \( v_F \) is the Fermi velocity and the ground-state out-of-plane upper critical field (14) is:

\[
B_{c2,\parallel}(0) \propto \Delta_{ab}(0) \times \Delta_z(0).
\]

It should be noted that even in bulk elemental fcc superconductors, for instance, in classical BCS superconductor Al [40–42], the superconducting energy gap exhibits crystallographic anisotropy, which is one of the important findings of the BCS theory. In a detailed study of the superconducting gap anisotropy in Al, reported by Blackford [42], it was shown that pure bulk Al exhibits \( \Delta_{max}(0) = 145 \mu\text{eV} \) and \( \Delta_{min}(0) = 198 \mu\text{eV} \). This implies that in bulk samples \( B_{c2,\parallel}(0) \), \( B_{c2,\perp}(0) \), and \( \Delta_{ab}(0) \) and \( \frac{\Delta_{ab}(0)}{\Delta_{max}(0)} \) can be different by a factor of \( \frac{\Delta_{max}(0)}{\Delta_{min}(0)} \approx 1.4 \), depending on sample orientation and measuring technique.

Because equations (14) and (16) have two independent multiplicative terms, it is impossible to fit \( B_{c2,\parallel}(T) \) data to equations (14) and (16) to keep both terms as free-fitting parameters. In other words, the utilization of equation (14) for \( B_{c2,\parallel}(T) \) data analysis strictly depends on the chosen mathematical approximation function for temperature-dependent anisotropy, \( \gamma(\xi) \):

\[
\gamma(\xi) = \frac{\xi_{ab}(T)}{\xi_z(T)} = \frac{B_{c2,\parallel}(T)}{B_{c2,\perp}(T)},
\]

\[
B_{c2,\parallel}(T) = \frac{\phi_0}{2\pi} \times \frac{1}{\xi_{ab}(T)} \times \frac{1}{\xi_z(T)} \times \gamma(\xi),
\]

for given material.

Surprisingly, this problem does not exist in atomically thin superconductors, because it was first shown by Tinkham and co-workers [43–45], for conditions of very thin films:

\[
d_{sc} \ll \xi(0),
\]

\[
d_{sc} \ll \lambda(0),
\]

where \( d_{sc} \) is the thickness of the superconducting layer, and equation (14) transforms into [43–45]:

\[
B_{c2,\parallel}(T) = \frac{\phi_0}{2\pi} \times \frac{1}{\xi_{ab}(T)} \times \sqrt{\frac{12}{d_{sc}}},
\]

and, thus, one can conclude that equation (16) transforms into:

\[
B_{c2,\parallel}(0) \propto \sqrt{\frac{12}{d_{sc}}} \times \Delta_{ab}(0),
\]

As a result, \( B_{c2,\parallel}(0) \) values described by equations (21) and (22) can be orders of magnitude larger than \( B_{c2,\parallel}(0) \) expected from equation (14), even if all fundamental parameters of the superconductor, such as \( \xi_{ab}(0) \), \( \xi_z(0) \), \( \Delta_{ab}(0) \) and \( \Delta_z(0) \), remain unchanged.

Thus, considering the anisotropy, \( \gamma(\xi) \), for atomically thin film:

\[
\gamma(\xi) = \frac{B_{c2,\parallel}(0)}{B_{c2,\perp}(0)} = \sqrt{\frac{12}{d_{sc}}} \times \frac{\xi_{ab}(0)}{\xi_{ab}(0)},
\]

and typical values for \( \xi_{ab}(0) \sim 50 \text{ nm} \) and \( d_{sc} = 1 \text{ nm} \) for MATBG [31, 46], we can calculate:

\[
\gamma(\xi) = 200,
\]

\[
B_{c2,\parallel}(0) = 200 \times B_{c2,\perp}(0).
\]
It should be noted that any type-II superconductor has three fundamental fields, i.e., the lower critical field, \(B_{c1}\), the upper critical field, \(B_{c2}\), and the Pauli–Clogston–Chandrasekhar field, \(B_{PCC}\). These fields are not independent of each other, and they are linked through primary superconducting parameters \(\lambda, \xi, \Delta, \) etc, of the material. Equation (23) demonstrates that atomically thin superconductors have an additional parameter, which is the film thickness, \(d_{sc}\). It should be stressed that equation (23) is not the only equation that utilizes the film thickness, \(d_{sc}\), to rescale one of the fundamental values (in this case, the upper critical field) for thin-film superconductors. The approach to utilize \(d_{sc}\) as an additional fundamental constant for the case of very thin superconductors was recognized nearly six decades ago, when Pearl [47] showed that the characteristic length, \(\lambda_{\perp, \text{eff}}(T)\), for the decay of screening currents of the vortex in thin film depends on the film thickness:

\[
\lambda_{\perp, \text{eff}}(T) = \frac{2\lambda^2(T)}{d_{sc}} = \lambda(T) \times \frac{2\lambda(T)}{d_{sc}}.
\] (26)

Equation (26) can be presented in a similar form to equation (23):

\[
\gamma_{\lambda}(0) = \frac{\lambda_{\perp, \text{eff}}(0)}{\lambda(0)} = 2 \times \left[\frac{\lambda(0)}{d_{sc}}\right].
\] (27)

The difference between equations (23) and (27) is the multiplicative prefactor \(\sqrt{3}\). However, these equations demonstrate through a general approach that different fundamental superconducting values in atomically thin superconductors depend on the ratio of the bulk sample value divided by the film thickness, \(d_{sc}\). We depict the ratio by the square brackets in equations (23) and (27).

In this approach, the geometrical enhancement factor (with respect to ratio) should be applied for two other fundamental fields in type-II superconductors, i.e., for the lower critical field, \(B_{c1}\), and for the Pauli–Clogston–Chandrasekhar limiting field, \(B_{PCC}\) (0). Based on the fact that the Pauli–Clogston–Chandrasekhar limiting field in many cases is close (by its value) to the upper critical field, here we propose to apply the same geometrical enhancement factor for \(B_{PCC}\) (0) as for the \(B_{c2}\) (0) field:

\[
\frac{B_{PCC}||(0)}{B_{PCC,\perp}(0)} = \sqrt{12} \times \frac{\xi_{ab}(0)}{d_{sc}},
\] (28)

and, thus, primary equation (3) (even under the assumption of s-wave weak-coupling superconductivity) transforms into:

\[
B_{PCC}|| (0) = \sqrt{12} \times \frac{\xi_{ab}(0)}{d_{sc}} \times B_{PCC,\perp} (0) \approx 370 \times T_c,
\] (29)

which, as observed in the experiment [1–17], exceeds by two orders of magnitude the enhancement in \(B_{PCC,\perp}(0)\) (equation (2)) for atomically thin superconductors. It should be stressed that equation (29) originates from the ginzburg-landau (GL) theory [48] and the sample geometry (as was first proposed by Tinkham and co-workers [43–45]), without any assumptions about the emergence of a new physical phenomenon, such as Ising superconductivity [1, 6, 7], in atomically thin superconductors.

Based on the above, one of the remaining tasks is to deduce \(\Delta_{ab}(0)\) in atomically thin films and to compare this value with its counterpart in bulk materials.

One of the techniques available to achieve this was introduced by Talantsev et al [49, 50], who proposed an equation for the self-field critical current density, \(J_c(sf, T)\), in type-II thin-film superconductors:

\[
J_c (sf, T) = \frac{\phi_0}{4\pi\mu_0} \times \frac{\ln (\kappa_c (T)) + 0.5}{\lambda_{ab}^3 (T)},
\] (30)

where \(\mu_0 = 4\pi \times 10^{-7} N/A^2\) is the permeability of free space, \(\kappa_{ab}(T)\) is the in-plane London penetration depth and \(\kappa_c (T) = \lambda_{ab}(T)/\lambda_c(T)\) is a temperature-dependent Ginzburg–Landau parameter. Later [51, 52], equation (equation (30)) was extended to both type-I and type-II superconductors:

\[
J_c (sf, T) = \frac{\phi_0}{4\pi\mu_0} \times \frac{\ln (1 + \sqrt{2} \times \kappa_c (T))}{\lambda_{ab}^3 (T)},
\] (31)

Considering that \(\lambda_{ab}(T)\) can be calculated from \(\Delta_{ab}(0)\) (all expressions below are given for s-wave superconductors):

\[
\lambda_{ab} (T) = \frac{\lambda_{ab} (0)}{\sqrt{1 - \frac{\Delta_{ab} (0)}{\Delta_{ab} (T)}}},
\] (32)

where \(k_B\) is the Boltzmann constant. The temperature-dependent superconducting gap can be expressed by the equation given by Gross et al [53]:

\[
\Delta_{ab} (T) = \Delta_{ab} (0) \times \text{tanh} \left[ \frac{\pi k_B T_c}{\Delta_{ab} (0)} \times \sqrt{\frac{\Delta C}{\gamma T_c} \left( \frac{T_c}{T} - 1 \right)} \right],
\] (33)

where \(\Delta C\) is the relative jump in electronic specific heat at \(T_c\) (where \(\gamma\) is the so-called Sommerfeld constant) and \(\eta = 2/3\) (for s-wave superconductors). Thus, the primary parameters of the superconducting state of atomically thin superconductors, 

(a) ground-state superconducting energy gap, \(\Delta_{ab}(0)\);
(b) relative jump in electronic specific heat at the transition temperature, \(\Delta C/T_c\);
(c) ground-state in-plane London penetration depth, \(\lambda_{ab}(0)\);
(d) transition temperature, \(T_c\);
(e) gap-to-transition temperature ratio, \(\frac{2\Delta_{ab}(0)}{k_BT_c}\),

can be deduced from the fit of the measured \(J_c(sf, T)\) to equations (30)–(33). Thus, the superconducting parameters of thin films of Ga [50], FeSe [50], InN [54], TaS\(_2\) [50], PdTe\(_2\) [55], MoS\(_2\) [5, 50], \(\alpha\)-MoO\(_2\)C [50], NbSe\(_2\) [50, 56], MATBG [46] and IrTe\(_2\) [57] were deduced.

However, \(J_c(sf, T)\) measurements in atomically thin superconductors are perhaps one of the most challenging experiments and, more commonly, magnetoresistance data, \(R(T, B)\),
We demonstrate the model applicability for atomically thin superconductors:

(a) ground-state superconducting energy gap, \( \Delta_{ab}(0) \);
(b) relative jump in electronic specific heat at the transition temperature, \( \frac{\Delta C}{C_f} \);
(c) ground-state in-plane superconducting coherence length, \( \xi_{ab}(0) \);
(d) transition temperature, \( T_c \);
(e) gap-to-transition temperature ratio, \( \frac{\Delta_{ab}(0)}{k_B T_c} \).

We demonstrate the model applicability for atomically thin superconductors Al, Sn, NbSe2, MoS2, TTG and WTe2.

2. Model description

The model is based on two primary ideas.

(a) The first is the superconducting coherence length, \( \xi_{ab}(T) \), which is linked to the London penetration depth, \( \lambda_{ab}(T) \), through the Ginzburg–Landau parameter, \( \kappa_c(T) = \frac{\lambda_{ab}(T)}{\xi_{ab}(T)} \), for which we adopted the temperature dependence proposed by Gor’kov [58]:

\[
\kappa_c(T) = \kappa_c(0) \left( 1 - 0.2429 \left( \frac{T}{T_c} \right)^2 + 0.0396 \left( \frac{T}{T_c} \right)^4 \right).
\]

(b) Based on equation (34), the temperature-dependent coherence length, \( \xi_{ab}(T) \), can be represented as follows:

\[
\xi_{ab}(T) = \xi_{ab}(0) \times \left( 1 - 0.2429 \left( \frac{T}{T_c} \right)^2 + 0.0396 \left( \frac{T}{T_c} \right)^4 \right) \times \left( \frac{1}{1 - \frac{1}{2k_B T} \int_{-\infty}^{0} \frac{d\varepsilon}{\cosh^2 \left( \frac{\sqrt{\varepsilon^2 + \Delta_{ab}(T)^2}}{2k_BT} \right)} \right),
\]

where temperature-dependent superconducting energy gap, \( \Delta_{ab}(T) \), is given by equation (34).

(c) By substituting equation (35) in equations (13) and (21), one can obtain a system of two equations that describe the upper critical field in atomically thin films (i.e. films for which the conditions of equations (19) and (20) are satisfied):

\[
B_{2,\perp}(T) = \frac{\phi_0}{2\pi} \times \frac{1}{\xi_{ab}(T)} = \frac{\phi_0}{2\pi} \times \frac{1}{\xi_{ab}(0)} \times \left( 1 - 0.2429 \left( \frac{T}{T_c} \right)^2 + 0.0396 \left( \frac{T}{T_c} \right)^4 \right) \times \left( \frac{1}{1 - \frac{1}{2k_B T} \int_{-\infty}^{0} \frac{d\varepsilon}{\cosh^2 \left( \frac{\sqrt{\varepsilon^2 + \Delta_{ab}(T)^2}}{2k_BT} \right)} \right).
\]

Equation (36) is used to fit \( B_{2,\perp}(T) \) in MATBG [46] and thin-film Ni3AlSn3NiO2 [59], while equation (37) is proposed herein.

(d) The second novel idea of this approach (equations (36) and (37)) is that these two data sets, \( B_{2,\perp}(T) \) and \( B_{2,\parallel}(T) \), are simultaneously fitted to equations (36) and (37). This type of fitting is also known as global data fitting. As a result, a common set of free-fitting parameters, \( \Delta_{ab}(0) \), \( \frac{\Delta C}{C_f} \), \( \xi_{ab}(0) \), \( T_c \), \( \frac{\Delta_{ab}(0)}{k_BT_c} \) and \( \Delta_{sc} \) can be deduced from the experimental \( B_{2,\perp}(T) \) and \( B_{2,\parallel}(T) \) data sets. For cases, when only \( B_{2,\parallel}(T) \) data sets were reported, we used equation (37) with a fixed \( \Delta_{sc} \) to the reported layer thickness to extract the fundamental superconducting parameters for these films.

3. Results

3.1. Sn films

In 1962, Blumberg and Douglass [23, 24] discovered a many-fold increase in \( B_{2,\parallel}(T) \) versus the decrease in the superconductor thickness, \( d_{sc} \), when they studied thin films of Sn. In figure 1, we show \( B_{2,\parallel}(T) \) data and fit them to equation (37) for a Sn film with \( d_{sc} = 36 \text{ nm} \) reported by Bloomberg [23]. Because the \( B_{2,\parallel}(T) \) data set for this film was measured within a narrow temperature range 0.79 < \( \frac{T}{T_c} \) < 1.0, to make it possible to deduce the ground-state energy gap, \( \Delta(0) \), for this film, we fixed \( \frac{\kappa_c}{\kappa_f} \) to its reported value of 1.68 [29] in equation (37). This approach is based on the similarity proposed by Talantsev et al [46], who reduced the number of free-fitting parameters in equations (30)–(33) by fixing \( \frac{\kappa_c}{\kappa_f} \) for \( J_c(s,T) \) data sets, when the fitted data sets do not contain enough raw data points to deduce \( \Delta(0) \) with the required accuracy.

The result, deduced \( \xi(0) = 61.8 \pm 1.1 \text{ nm} \) satisfies the condition of \( d_{sc} < \xi(0) \) and deduced \( \frac{\Delta C}{C_f} = 3.7 \pm 0.3 \) is in excellent agreement with the value of \( \frac{\Delta C}{C_f} = 3.705 \) reported by Carbotte [29] for electron–phonon mediated superconductivity in bulk Sn.

To demonstrate that the global fit to equations (36) and (37) works for thin films of Sn in figure 2, we show experimental \( B_{2,\perp}(T) \) and \( B_{2,\parallel}(T) \) data sets for sample TD3 (\( d_{sc} = 13.6 \text{ nm} \) nm) reported by Harper and Tinkham [45]. For this fit (figure 2), we also fixed \( \frac{\kappa_c}{\kappa_f} = 1.68 \), whereas the other parameters were free. All deduced values (for instance, \( \xi(0) = 70.6 \pm 1.5 \text{ nm} \), \( \Delta(0) = 613 \pm 58 \text{ µeV} \), and \( \frac{\Delta C}{C_f} = 4.0 \pm 0.4 \)) are in remarkable agreement with values deduced from data reported by Bloomberg (figure 1 and
Few-nanometre-thick Al film represents, perhaps, the most studied object of elemental metallic superconductors due to its prominent dependence of the observed superconducting coherence length, $\xi$, on the film thickness, $d_{sc}$ [25–27, 60, 61]. The highest $T_c$ = 5.7 K was reported by Townsend et al. [60] for Al film with $d_{sc} = 2$ nm, while bulk Al exhibits $T_c = 1.12$ K [29].

In figure 3, we show the $B_{z, \parallel}(T)$ data set reported for Al film with $d_{sc} = 5$ nm by Tedrow and Meservey [62] and the data fit to equation (37). The deduced $\Delta(0)$, $\frac{\Delta(0)}{k_B T_c}$ and $\frac{2\Delta(0)}{k_BT_c}$ values show that this Al film is a strongly coupled superconductor. This result agrees with the general trend [25–27, 60–62], where the thinning of Al causes a significant enhancement in $T_c$, $\Delta(0)$ and $B_{c2, \parallel}(0)$.

It should be stressed that if for Pauli–Clogston–Chandrasekhar limiting field calculation, one uses deduced $T_c = 2.04$ K (figure 3) and weak-coupling equation (3), then,

\[ B_{PCC, weak-coupling}(0) = 1.86 \times T_c \]

\[ = 3.79 \times T > B_{c2, \parallel}(0) = 4.2 \times T \]  

This means that in this Al film, the Pauli–Clogston–Chandrasekhar limiting field was violated. However, if proper equation (6) and deduced $\Delta(0) = 522 \mu$eV is used for calculations, then,

\[ B_{PCC}(0) = \frac{\Delta(0)}{\sqrt{R} \times \mu_B} = 6.4 \times T > B_{c2, \parallel}(0) = 4.2 \times T \]  

Thus, this Al film complies with Pauli–Clogston–Chandrasekhar limiting field because $B_{c2, \parallel}(0)$ is significantly lower than $B_{CPP}(0)$.

Practically identical values for $\Delta(0)$, $\frac{\Delta(0)}{k_BT_c}$ and $\frac{2\Delta(0)}{k_BT_c}$ were deduced for the 4 nm thick Al film, for which raw $B_{c2, \parallel}(0)$ data were reported by Tedrow and Meservey in their figure 7 [27]. $B_{c2, \parallel}(0)$ data and fit for the Al film are shown in figure 4. Calculated $B_{CPP}(0)$, based on equation (6), is well above extrapolated $B_{c2, \parallel}(0)$:

\[ B_{PCC}(0) = \frac{\Delta(0)}{\sqrt{R} \times \mu_B} = 6.3 \times T > B_{c2, \parallel}(0) = 4.2 \times T \]
Figure 3. $B_{c2,\|}(T)$ data and fit to equation (37) for an Al film with thickness $d_{sc} = 5$ nm (raw data reported by Tedrow and Meservey [62]). Deduced parameters are $T_c = 2.040 \pm 0.001$ K, $\xi(0) = 54.3 \pm 0.1$ nm, $\Delta(0) = 522 \pm 21 \mu$eV, $\frac{\Delta T_c}{T_c} = 2.85 \pm 0.07$ and $\frac{\Delta \xi(0)}{2 \xi(0)} = 5.9 \pm 0.2$. Goodness of fit is 0.9994. 95% confidence bands are indicated by the shaded areas.

Figure 4. $B_{c2,\|}(T)$ data and fit to equation (37) for Al film having thickness $d_{sc} = 4$ nm (raw data reported by Tedrow and Meservey [27]). Deduced parameters are $T_c = 2.136 \pm 0.001$ K, $\xi(0) = 67.9 \pm 0.4$ nm, $\Delta(0) = 517 \pm 36$ meV, $2\Delta(0)/k_B T_c = 5.6 \pm 0.4$. Goodness of fit is 0.9998. 95% confidence bands are shown by shaded areas.

3.3. Al film overcoated with Pt

Tedrow and Meservey [27] reported three $B_{c2,\|}(0)$ data sets for 4 nm thick Al films overcoated with thin Pt layers of different thicknesses, $d_{pt}$. Of the three reported $B_{c2,\|}(0)$ data sets, only one with $d_{pt} = 0.1$ nm has sufficient raw $B_{c2,\|}(0)$ data points to be fitted to equation (37) (figure 5). The deduced $\Delta(0) = 451 \pm 50$ meV is in close proximity to the values deduced for the other Al films (figures 3 and 4). Calculated Pauli–Clogston–Chandrasekhar limiting field:

$$B_{PCC,\|}(0) = \frac{\Delta(0)}{\sqrt{8} \times \mu_B} = 5.5 T < B_{c2,\|}(0) = 7.8 T \quad (41)$$

appears to be lower than that of the extrapolated $B_{c2,\|}(0)$. One of the two possible interpretations of this result is that Pt overcoating can suppress the Lande $g$-factor. However, it is much more natural to explain this result based on the geometrical field enhancement factor (equations (23) and (28)):

$$B_{PCC,\|}(0) = \sqrt{12} \times \frac{\xi(0)}{d_{sc}} \times B_{PCC,\|}(0) = 31.5 \times 5.5 T = 173 T, \quad (42)$$

which exceeds extrapolated $B_{c2,\|}(0) = 7.8 T$ (figure 5) by 22 times.

3.4. Electric-field-gated WTe$_2$ monolayer

Sajadi et al [15] reported $B_{c2,\perp}(T)$ and $B_{c2,\|}(T)$ data sets for the WTe$_2$ monolayer sample in their figures 2(b) and (c). Although these data sets were measured at slightly different doping states (i.e., $n_e = 2.0 \times 10^{13}$ cm$^{-2}$ for $B_{c2,\perp}(T)$ and $n_e = 1.8 \times 10^{13}$ cm$^{-2}$ for $B_{c2,\|}(T)$), we performed a global fit of these data sets to equations (36) and (37) in figure 6. By performing this fit, it was assumed that the superconducting layer thickness, $d_{sc}$, is a free-fitting parameter because it is difficult to establish this parameter in an electric-field-gated sample.
Goodness–of–fits are (a) 0.9718 and (b) 0.9764. 95% confidence data reported by Sajadi (a).

Enhancement factor (equations (16)–(18)): Chandrasekhar limiting field. The calculated Pauli–Clogston–Chandrasekhar coupling superconductivity in this electric-field-gated mono-

Figure 6. (a) $B_{c2,\perp}(T)$ and (b) $B_{c2,\parallel}(T)$ data and global fit to equations (36) and (37) for WTe$_2$ monolayer doped at (a) $n_e = 2.0 \times 10^{11}$ cm$^{-2}$ and (b) $n_e = 1.80 \times 10^{11}$ cm$^{-2}$ (raw data reported by Sajadi et al [15]). Deduced parameters are $d_{sc} = 2.95 \pm 0.12$ nm, $T_c = 0.70 \pm 0.03$ K, $\xi_{ab}(0) = 125 \pm 4$ nm, $\Delta_{ab}(0) = 105 \pm 25$ $\mu$eV, $2\Delta_{ab}(0)/k_B T_c = 3.5 \pm 0.9$ and $\Delta C/\gamma T_c = 1.4 \pm 1.0$. Goodness–of–fits are (a) 0.9718 and (b) 0.9764. 95% confidence bands are indicated by the shaded areas.

All the deduced parameters agree well with the weak-coupling superconductivity in this electric-field-gated monolayer superconductor. The calculated Pauli–Clogston–Chandrasekhar limiting field:

$$B_{PCC,\perp}(0) = \frac{\Delta(0)}{\sqrt{\pi} \times \mu_B} = 1.3 T < B_{c2,\parallel}(0) = 3.1 T,$$

(43)
appeared to be lower than that of the extrapolated $B_{c2,\parallel}(0)$. However, similar to the case of the Al film overcoated with Pt (figure 5), this result can be explained based on the geometrical enhancement factor (equations (23) and (28)):

$$B_{PCC,\parallel}(0) = \sqrt{12} \times \frac{\xi_{ab}(0)}{d_{sc}} \times B_{PCC,\perp}(0) = 55 T,$$

(44)

which exceeds extrapolated $B_{c2,\parallel}(0) = 3.1 T$ by 18 times (figure 6). Thus, this superconductor complies with Pauli–Clogston–Chandrasekhar limiting field.

3.5. Electric-field-gated MoS$_2$ monolayer

Saito et al [4] reported $R_{sheet}(T,B)$ curves for a 20 nm thick MoS$_2$ single crystal in which the superconducting state was induced by electric-field gating. From the reported $R_{sheet}(T,B)$ curves measured at a gate voltage of 5.5 V (the original curves are shown in figures 3(a) and (b) in [4]), the authors utilized 50% of the normal state resistance, we deduced $B_{c2,\perp}(T)$ and $B_{c2,\parallel}(T)$ data sets, which are shown in figure 7 together with the global data fit to equations (36) and (37). By performing the fit, it was assumed that the superconducting layer thickness, $d_{sc}$, is a free-fitting parameter.

Deduced $\frac{2\Delta_{ab}(0)}{k_B T_c}$ = 8.1 ± 1.4 (at $V_{gate} = 5.5$ V, figure 7) is high. However, we should note that Talantsev et al [46] reported $\frac{2\Delta_{ab}(0)}{k_B T_c} = 6.1 \pm 1.5$ for electric-field-gated MoS$_2$ (with $V_{gate} = 2.2$ V), which was extracted from the analysis of $J_c(s,T)$ measured by Costanzo et al [63]. Thus, as deduced by us, the ratio of $\frac{2\Delta_{ab}(0)}{k_B T_c}$ = 8.1 ± 1.4 (at $V_{gate} = 5.5$ V) is in
the same ballpark, which is further confirmation of strongly coupled superconductivity in electric-field-gated MoS₂.

The calculated Pauli–Clogston–Chandrasekhar limiting field:

\[ B_{PCC,\perp}(0) = \frac{\Delta(0)}{\sqrt{g_b}} = 23.3 \, T < B_{c2,\parallel}(0) = 39.5 \, T, \]  
(45)

is lower than extrapolated \(B_{c2,\parallel}(0)\). However, this result can again be explained based on the geometrical enhancement factor (equations (23) and (28)):

\[ B_{PCC,\parallel}(0) = \sqrt{1/2} \times \frac{\xi_{ab}(0)}{d_{sc}} \times B_{PCC,\perp}(0) = 946 \, T. \]  
(46)

The magnetic flux density, \(B_{PCC,\parallel}(0)\), indicated in equation (46) was created only a few times on Earth during extraordinary pulsed magnetic field experiments [64–66]. Thus, it is observed in experiment that \(B_{c2,\parallel}(T \to 0 \, K) = 39.5 \, T\) is still by more than one order of magnitude lower than the Pauli–Clogston–Chandrasekhar limiting field.

3.6. NbSe₂ bilayer

De la Barrera et al [7] reported \(B_{c2,\perp}(T)\) and \(B_{c2,\parallel}(T)\) data sets for NbSe₂ bilayer samples in their figure 3(e) [7]. We performed a global fit for these data sets using equations (36) and (37) in figure 8. By performing this fit, it was assumed that the superconducting layer thickness, \(d_{sc}\), is a free-fitting parameter.

The deduced values for the primary superconducting parameters, which are \(\Delta_{ab}(0), \frac{\Delta_0}{T_c}\) and \(\frac{2\Delta_{ab}(0)}{\xi_{ab}(0)}\), are within the expected values for moderately strongly coupled superconductors. It should be mentioned that figure 8 demonstrates the importance of a large number of experimental upper critical field data points, which cover, as widely as possible, the full temperature range 0.0 < \(T_c\) < 1.0. For this type of data set, a global fit to equations (36) and (37), it is possible to deduce \(d_{sc}\), \(T_c\), \(\xi_{ab}(0), \Delta_{ab}(0), \frac{\Delta_0}{T_c}\) and \(\frac{2\Delta_{ab}(0)}{\xi_{ab}(0)}\), in atomically thin superconductors with high accuracy.

The calculated Pauli–Clogston–Chandrasekhar limiting field:

\[ B_{PCC,\perp}(0) = \frac{\Delta(0)}{\sqrt{g_b}} = 10 \, T < B_{c2,\parallel}(0) = 29 \, T, \]  
(47)

is lower than the extrapolated \(B_{c2,\parallel}(0)\). However, this result can be explained based on the geometrical enhancement factor (equations (23) and (28)):

\[ B_{PCC,\parallel}(0) = \sqrt{1/2} \times \frac{\xi_{ab}(0)}{d_{sc}} \times B_{PCC,\perp}(0) = 137 \, T. \]  
(48)

3.7. Magic-angle twisted trilayer graphene

Finally, we present an analysis of several magic-angle twisted trilayer graphene (MATTG) samples for which raw \(B_{c2,\parallel}(T)\) data sets were recently reported by Cao et al [1]. For our analysis, we used \(B_{c2,\parallel}(T)\) data sets, which were deduced from experimental \(R(T)\) curves by applying 10% of the normal-state resistance criterion [1]. The importance of utilizing an as low as possible resistive criterion for \(B_{c2}(T)\) definition with following the analysis by equations (36) and (37) is based on the fact that one of the primary parameters that is deduced from the fit \(\frac{\Delta}{\xi}\) can be accurately measured only when,

\[ \frac{R(T)}{R_{\text{norm}}} \to 0, \]  
(49)

and this was clearly shown in recent precise experiments by Hirai et al [67]. In addition, Shang et al [68] in their precise experiments showed that the diamagnetic response from NbReSi superconductor was observed at the condition described by equation (49).

Another issue is that only \(B_{c2,\parallel}(T)\) data sets have been reported for MATTG [1, 2]. Based on this, we fixed the superconducting layer thickness \(d_{sc}\) in equation (36) to the assumed MATTG thickness of \(d_{sc} = 1 \, \text{nm}\). The MATTG devices for
which $B_{c2,\parallel}(T)$ data sets are analyzed herein have adjacent layers, which are sequentially twisted by $\theta$ and $-\theta$ angles, where $\theta \approx 1.57^\circ$ and $1.44^\circ$ (details can be found elsewhere [1,32]).

In figure 9, we show four $B_{c2,\parallel}(T)$ data sets chosen to cover the full range of observed $T_c$ values. As shown in figure 9, all deduced $\frac{\Delta \omega(0)}{k_B T_c}$ ratios are within the well-established range of this parameter variation for classical low-$T_c$ electron–phonon mediated superconductors [29]:

$$3.5 \leq \frac{2\Delta_0(0)}{k_B T_c} \leq 4.7.$$  \hspace{1cm} (50)

Thus, the first concern with regard to the current interpretation [1,2,32] of MATTG as a strong-coupling superconductor is that all deduced $\frac{2\Delta_0(0)}{k_B T_c}$ values demonstrated that MATTG exhibits much lower $\frac{2\Delta_0(0)}{k_B T_c}$ values in comparison to iron-based superconductors [30] and superhydride superconductors [69].

In addition, as reported by Cao et al [1], the effect of re-entering the superconducting state in MATTG at high magnetic fields has been observed in many superconductors [70], including single-layer graphene with deposited Sn nanodisks [71]. Thus, the effect of re-entering of the superconducting state is also not a unique MATTG property.

The remaining task is to show that MATTG (and presumably MATBG) does not demonstrate any unique superconducting properties that have not yet been observed in other superconducting materials to calculate $B_{PCC,\parallel}(0)$ and show that these fields are well above the observed $B_{c2,\parallel}(0)$ values. These calculations are presented in table I, from which it is clear that all MATTG samples exhibit an inequality:

$$B_{c2,\parallel}(0) \ll B_{PCC,\parallel}(0),$$  \hspace{1cm} (51)

because of the very large values of the $\sqrt[12]{\frac{\xi(0)}{\kappa}}$ ratio, similar to all other atomically thin superconductors.

4. Discussion

It is interesting to note that it is very likely that the first experimental observation of the emergence of superconductivity in twisted atomically thin nanosheets was reported by Pan et al [72]. This research group prepared bulk TaS$_2$ samples using randomly restacked chemically exfoliated TaS$_2$ monolayers. As a result, the randomly restacked bulk samples exhibit $T_c \sim 3$ K, compared to $T_c = 0.8$ K observed in ordered bulk 2H-TaS$_2$ compound. The randomness of the restacked samples was confirmed by the analysis of the angular dependence of the
upper critical field \cite{31}. One of the most natural explanations of this \(T_c\) enhancement is based on the idea that in billions of randomly restacked TaS\(_2\) monolayers, some neighboring monolayers are restacked at either magic-angle twist \cite{31} or at rhombohedral configuration \cite{2}. Thus, the superconducting state in bulk randomly restacked TaS\(_2\) samples emerges similarly to bi- and tri-layer graphene. It is important to note that Ma et al \cite{74} reported that extrapolated \(B_{c2,\perp}(0) = 9.5\) T is significantly exceeded \(B_{PCC}(0) = 5.8\) T (calculated by equations (6)–(8)) in bulk samples of randomly restacked TaS\(_2\) monolayers. This is not surprising if we assume that the superconductivity in restacked TaS\(_2\) originates from the magic angle or the rhombohedral twisted bi- or tri-layer TaS\(_2\).

There is a need to stress that even if MATTG and other atomically thin superconductors exhibit a violation of this \(T_c\) enhancement is based on the idea that in billions of randomly restacked TaS\(_2\) monolayers, some neighboring monolayers are restacked at either magic-angle twist \cite{31} or at rhombohedral configuration \cite{2}. Thus, the superconducting state in bulk randomly restacked TaS\(_2\) samples emerges similarly to bi- and tri-layer graphene. It is important to note that Ma et al \cite{74} reported that extrapolated \(B_{c2,\perp}(0) = 9.5\) T is significantly exceeded \(B_{PCC}(0) = 5.8\) T (calculated by equations (6)–(8)) in bulk samples of randomly restacked TaS\(_2\) monolayers. This is not surprising if we assume that the superconductivity in restacked TaS\(_2\) originates from the magic angle or the rhombohedral twisted bi- or tri-layer TaS\(_2\).

There is a need to stress that even if MATTG and other atomically thin superconductors exhibit a violation of this \(T_c\) enhancement is based on the idea that in billions of randomly restacked TaS\(_2\) monolayers, some neighboring monolayers are restacked at either magic-angle twist \cite{31} or at rhombohedral configuration \cite{2}. Thus, the superconducting state in bulk randomly restacked TaS\(_2\) samples emerges similarly to bi- and tri-layer graphene. It is important to note that Ma et al \cite{74} reported that extrapolated \(B_{c2,\perp}(0) = 9.5\) T is significantly exceeded \(B_{PCC}(0) = 5.8\) T (calculated by equations (6)–(8)) in bulk samples of randomly restacked TaS\(_2\) monolayers. This is not surprising if we assume that the superconductivity in restacked TaS\(_2\) originates from the magic angle or the rhombohedral twisted bi- or tri-layer TaS\(_2\).

### Table 1. Primary deduced parameters for MATTG samples shown in figure 9.

| Sample designation in figure 9 and in [1] | \(T_c\) (K) | \(\Delta_{ab}(0)\) (\(\mu\)eV) | \(\frac{2\Delta_{ab}(0)}{k_BT_c}\) | \(\frac{\Delta^c}{\Delta^s}\) | \(B_{c2,\perp}(0)\) (T) | \(B_{PCC,\perp}(0)\) (T) | \(B_{PCC,\parallel}(0)\) (T) |
|------------------------------------------|-------------|---------------------------|--------------------------|--------------------------|------------------|------------------|------------------|
| Panel a, Figure S1(a) (raw data available online in [1]) \(\theta = 1.57^\circ\) \(v = + 1.72\) | 0.624 ± 0.001 | 120 ± 8 | 4.5 ± 0.3 | 1.69 ± 0.04 | 3.1 | 1.5 | 1917 |
| Panel b, Figure S2(b) (raw data available online in [1]) \(\theta = 1.44^\circ\) \(v = + 1.68\) | 0.776 ± 0.001 | 158 ± 5 | 4.7 ± 0.1 | 2.28 ± 0.04 | 2.6 | 1.9 | 2903 |
| Panel c, Figure S2(b) (raw data available online in [1]) \(\theta = 1.57^\circ\) \(v = - 3\) | 0.975 ± 0.001 | 155 ± 5 | 3.7 ± 0.1 | 1.86 ± 0.06 | 3.6 | 1.9 | 2080 |
| Panel d, Figure S1(b) (raw data available online in [1]) \(\theta = 1.57^\circ\) \(v = + 2.16\) | 1.540 ± 0.001 | 231 ± 6 | 3.47 ± 0.09 | 1.79 ± 0.06 | 6.2 | 2.8 | 1794 |

5. Conclusion

A new model for deducing several primary superconducting parameters in atomically thin superconductors, which are in-plane ground-state superconducting energy gap, \(\Delta_{ab}(0)\), relative jump in electronic specific heat at the transition temperature, \(\Delta^c\), in-plane ground-state superconducting coherence length, \(\xi_{ab}(0)\), transition temperature, \(T_c\), and gap-to-transition temperature ratio, \(\frac{2\Delta_{ab}(0)}{k_BT_c}\), from the upper critical field data was proposed. The analysis was performed for a few atomic layers of thick Al, Sn, NbSe\(_2\), MoS\(_2\), MATTG and WTe\(_2\) and showed that all the films except electric-field-gated MoS\(_2\) exhibit moderately strong-coupling pairing.

The observed enhancement of the parallel upper critical field, \(B_{c2,\parallel}(0)\), in atomically thin superconductors, which for some samples exceeds the Pauli–Clugston–Chandrasekhar limiting field calculated perpendicular to the film surface direction of the applied magnetic field, \(B_{CPP,\perp}(0)\), is explained based on a geometrical enhancement factor that arises solely from small sample thickness. This enhancement is not related to the appearance of any new exotic pairing mechanism or new extraordinary physical phenomena, at least in all materials to date.
Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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Conflict of interest

The authors declare no competing interests.

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