Signatures of Nematic Superconductivity in Doped Bi$_2$Se$_3$ under Applied Stress

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(Dated: February 1, 2019)

The $M_x$Bi$_2$Se$_3$ family are candidates for topological superconductors, where $M$ could be Cu, Sr, or Nb. Two-fold anisotropy has been observed in various experiments, prompting the interpretation that the superconducting state is nematic. However, it has since been recognized in the literature that a two-fold anisotropy in the upper critical field $H_{c2}$ is incompatible with the naive nematic hypothesis. In this paper we study the Ginzburg-Landau theory of a nematic order parameter coupled with an applied stress, and classify possible phase diagrams. Assuming that the $H_{c2}$ puzzle is explained by a pre-existing "pinning field", we indicate how a stress can be applied to cancel the pinning field and unmask the actual anisotropy of the superconducting state itself. We also explore the Josephson tunneling between the proposed nematic superconducting state and an s-wave superconductor. Both the anisotropy and temperature dependence of the critical current are markedly different from the case of tunneling between s-wave superconductors.

I. INTRODUCTION

Bi$_2$Se$_3$ is a topological insulator$^{12}$. When intercalated with electron-donating atoms such as Cu$^{16}$, Sr$^{18-19}$ or Nb$^{20}$, it becomes superconducting at low temperature. Following their identification as candidates of topological superconductors$^{9,11}$, the family of materials has recently garnered a lot of interest$^{12}$. Experimentally, two-fold anisotropy in the superconducting phase has been observed in NMR Knight's shift$^{14,15}$, specific heat$^{14,15}$, magnetic torque$^{6}$, magneto-transport and upper critical field$^{11,14,16-18}$. This is incompatible with the crystal lattice symmetry, and has led to the hypothesis that the superconducting state is nematic in nature, i.e. it spontaneously breaks both lattice rotational and gauge symmetries. While the superconductivity is suppressed by an applied hydrostatic pressure$^{19,20}$, the two-fold anisotropy is observed as far as superconductivity holds$^{20}$.

The parent material Bi$_2$Se$_3$ forms a rhombohedral crystal, with a lattice point group $D_{3d}$; see FIG 1. According to the nematic hypothesis, the complex superconducting order parameter $\vec{\eta}$ is believed to transform under the irreducible representation $E_u$, which is parity-odd and two-dimensional. It can be thought of as a nematic director in the basal plane, since $\vec{\eta}$ and $-\vec{\eta}$ can be identified up to a global $U(1)$ phase shift of $\pi$. Deep in the superconducting phase, the finite $\vec{\eta}$ defines a preferred orientation and spontaneously breaks the point group symmetry. Experiments that detect the anisotropy in the basal plane all report a two-fold symmetry, consistent with the presence of a nematic director.

However, the story with the upper critical field $H_{c2}$ is more complicated. Experimentally a two-fold anisotropy was observed$^{14,16,18}$ with the applied field parallel to the basal plane, seemingly inline with other measurements. However, this result is actually incompatible with the nematic hypothesis, which indicates a six-fold anisotropy for $H_{c2}$.$^{21,22}$

To understand this, one considers the onset of superconductivity in high field. Instead of a finite $\vec{\eta}$ providing a preferred orientation, the magnetic field and the lattice anisotropy together determine the orientation of the infinitesimal $\vec{\eta}$ when it first emerges. This consideration leads to the prediction of a six-fold anisotropy associated with the six-fold improper rotation symmetry of $D_{3d}$.

To reconcile the $E_u$ pairing scenario and the observed two-fold anisotropy of $H_{c2}$, some nematic “symmetry-breaking field” (SBF) must already be present in the sample. Stress or strain were named as possible candidates$^{10}$. However, this really casts doubt onto the interpretation of the anisotropy: the pre-existing SBF alone may be responsible for the observed anisotropy, and if there is...
somehow a way to cancel this SBF, the pristine superconducting state itself may as well be s-wave and fully respects all crystal symmetry.

Indeed, for the case of Sr, evidence of nematicity above the superconducting transition $T_c$ has been reported by Kuntsevich et al., who reported a small lattice distortion in their high-quality Sr$_2$B$_2$Se$_3$ single crystal sample, and found a correlation between this lattice nematicity with the two-fold anisotropy in magnetoresistance observed in their sample. This is in contrast with the case of Cu, where no nematicity was observed above $T_c$.

In this paper, we formulate the Ginzburg-Landau theory of a nematic superconducting order parameter to our knowledge has never been observed experimentally.

In light of these recent developments, a better understanding of how superconductivity interacts with an applied stress or strain is clearly desirable. Even if the SBF is not mechanical in nature, an externally applied stress presents a way to cancel the SBF and unmask the true, if any, anisotropy of the order parameter.

Historically, much of the phenomenology of a superconducting order parameter with non-trivial symmetry has been known as part of the lore of the unconventional superconductors (see references and references therein.) Pertinent to the present discussion is the fact that, due to the competition of the SBF and the crystal field, the superconducting transition can be split into two if the order parameter is multi-component.

In this paper, we formulate the Ginzburg-Landau theory of a nematic superconducting order parameter to include externally applied stress, and explore the phenomenology as a guide to future experiments. We identify the splitting of the superconducting transition, as well as another possible transition at a lower temperature. We classify possible phase diagrams, and indicate experimental signatures that may help distinguish, in the presence of a pre-existing SBF, a true nematic superconducting state from a single-component one.

In addition, we consider the critical tunnel current in the Josephson junction between the proposed nematic superconducting state and another s-wave superconductor. The idea of using the Josephson effect as a probe for unconventional pairing symmetry has long been considered. The Ginzburg-Landau approach is also applicable to describe the Josephson tunneling. If the nematic hypothesis does hold, the critical current shows a non-trivial anisotropic dependence on the SBF, as well as a temperature dependence that differs from the s-wave-to-s-wave scenario. This provides another experimental test for the nematicity.

II. GINZBURG-LANDAU FREE ENERGY

Let us first give a brief recap of the Ginzburg-Landau free energy for a nematic superconductor in the absence of any external perturbation.

We define the coordinate axes as follows: the $z$-axis is aligned with the principal $C_3$ axis of the lattice, the $x$-axis is along one of the $C_2$ axes in the basal plane, and the $y$-axis is chosen to form a right-handed set of coordinate system.

We assume that the superconducting order parameter is a complex two-component quantity $\eta$. It will be parameterized as:

$$\eta = \begin{pmatrix} \eta_x \\ \eta_y \end{pmatrix} = \sqrt{\beta} \begin{pmatrix} \cos \theta \\ e^{i\phi} \sin \theta \end{pmatrix}.$$  \hspace{1cm} (1)

Here $\eta_x$ is kept real by a suitable global $U(1)$ phase rotation. See Fig. 11 for illustration.

The point group $D_{3d}$ can be built from three operations: $2\pi/3$-rotation about the $z$-axis (denoted $C_3$), $\pi$-rotation about the $x$-axis (denoted $C_2'$), and mirror reflection about the $yz$-plane (denoted $M$). The odd-parity proposal would have $\eta$ transforming in the $E_u$ representation, similar to $(x, y)$.

However, we may equally well consider the possibility where $\eta$ transforms in the $E_u$ representation instead, similar to the pair $(yz, -xz)$. The Ginzburg-Landau free energy and most of the subsequent analysis remain unchanged in this scenario, except a minor modification for the Josephson tunneling, which does not affect our qualitative conclusion. For either case, one can consistently define $\eta_x$ to be the component invariant under $C_2'$.

The quantity $\eta$ is to be considered as a headless vector, since a sign in $\pm \eta$ can be regarded as a $U(1)$ phase factor. Therefore one may restrict $\theta$ to the interval $(-\pi/2, \pi/2]$, keeping in mind that $\theta$ and $\theta + \pi$ are always degenerate.

The Ginzburg-Landau expansion of free energy is obtained by writing down all the terms that are invariant under $D_{3d}$, global phase shift of $\eta$, and time reversal. Up to $O(\eta^6)$, the most generic form is:

$$\mathcal{F}_0 = \alpha |\eta|^2 + \beta'_1 |\eta|^4 + \beta'_2 (\eta \cdot \eta) (\eta^* \cdot \eta^*) + \gamma'_1 |\eta|^6 + \gamma'_2 (\eta_x + i\eta_y)^3 (\eta^*_x - i\eta^*_y)^3 + (\eta_x - i\eta_y)^3 (\eta^*_x + i\eta^*_y)^3 + \gamma'_5 |\eta|^2 (\eta \cdot \eta) (\eta^* \cdot \eta^*)$$ \hspace{1cm} (2)

Here $\alpha$ is taken to be

$$\alpha = \kappa \left( T - T_0 \right),$$ \hspace{1cm} (3)

where $T_0$ is the superconducting transition temperature.
without external field. We ignore the temperature dependence of all other coefficients.

Let us first consider the free energy up to $O(\eta^4)$. To this order $F_0$ is in fact invariant under arbitrary rotations in the $xy$-plane. Two solutions for $\bar{\eta}$ are possible: the complex chiral state that is rotationally invariant but spontaneously breaks time-reversal symmetry, and the real nematic state which breaks the rotational symmetry but is time-reversal invariant. The sign of $\beta_2$ decides which phase is favored.\(^{11,12}\)

Since the nematic state is by assumption the true solution, we take the appropriate sign $\beta_2 < 0$. The minimum of $F$ now lies along $\phi = 0$, and in effect $|\bar{\eta}|^2 = |\eta|^2$. Therefore we define $\beta \equiv \beta_1 + \beta_2$, and require $\beta > 0$ for stability.

Now let us move on to include terms of $O(\eta^6)$. The $\gamma_4$ term essentially has the same $\theta$- and $\phi$- dependence as the $\beta_2$ term. Since a transition from nematic to chiral state at a lower temperature is never observed, we explicitly require $\gamma_4 < 0$. On the other hand, it can be shown that all extrema of the $\gamma_2$ term correspond to real values of $\bar{\eta}$. Consequently, one may assume $\bar{\eta}$ is real and simply set $\phi = 0$. In terms of $\theta$ and $D$, the free energy reads:

$$F_0 = \alpha D + \beta D^2 + [\gamma_1 + \gamma_2 \cos (6\theta)] D^3,$$

where $\gamma_1 \equiv \gamma_1^\prime + \gamma_3$ and $\gamma_2 = 2\gamma_2^\prime$. For stability we require $\gamma_1 > 0$ and $\gamma_2 > |\gamma_2|$. The $\gamma_2$ term breaks the full rotational symmetry down to a discrete six-fold symmetry. This is the first instance of crystal anisotropy entering the expansion.\(^{13,14}\)

A. Coupling to Stress

We will focus on stress in the $xy$-plane. Generally planar stress is a rank-two symmetric tensor $\vec{\varepsilon}$ with three independent real parameters. Under $D_{3d}$, this tensor decomposes into two parts: the scalar $\text{Tr} \vec{\varepsilon}$, and the traceless part which is organized to form the two-component quantity

$$\varepsilon = \begin{pmatrix} \varepsilon_{xx} - \varepsilon_{yy} \\ -2\varepsilon_{xy} \end{pmatrix} = \begin{pmatrix} \varepsilon_1 \\ \varepsilon_2 \end{pmatrix} = \varepsilon \begin{pmatrix} \cos (2(-\Phi + \Phi_0)) \\ \sin [2(-\Phi + \Phi_0)] \end{pmatrix},$$

which transforms in the representation $E_g$ under $D_{3d}$.

We choose to parameterize $\varepsilon$ as above because $\Phi$ would correspond to the physical rotation angle about the $z$-axis. When the stress is rotated by an arbitrary angle $\delta$, $\varepsilon$ changes via $\Phi \rightarrow \Phi + \delta$. The angle $\Phi_0$ defines the direction relative to the $x$-axis that corresponds to $\Phi = 0$, and we leave it unspecified for now, to be chosen for convenience later. The angle $\Phi$ defines a nematic direction in the basal plane, and subsequently we may refer to it as the “direction” of stress. This breaks the point group rotation symmetry.

For the sake of completeness, we state that one can form another $E_g$ pair using out-of-plane stress components: the pair $(\varepsilon_{yz}, -\varepsilon_{xz})$ can be used in place of $(\varepsilon_1, \varepsilon_2)$.

For the remainder of this paper, we will always assume that $\text{Tr} \vec{\varepsilon}$ is kept constant. The dependence of Ginzburg-Landau coefficients on $\text{Tr} \vec{\varepsilon}$ can therefore be entirely disregarded.

We will only consider coupling to $\vec{\varepsilon}$ at linear order. Both $D_{3d}$ and $U(1)$-invariance forbid any term bilinear in $\bar{\eta}$ and $\vec{\varepsilon}$. Instead, one must construct from $\bar{\eta}$ real, $U(1)$-invariant quantities that transform in $E_g$ under $D_{3d}$, and let $\vec{\varepsilon}$ couple to these quantities. Up to order $O(\eta^4)$, the following is the exhaustive list:

$$S = \begin{pmatrix} |\eta_x|^2 - |\eta_y|^2 \\ -\eta_x \eta_y^* - \eta_y \eta_x^* \end{pmatrix} \equiv \begin{pmatrix} S_1 \\ S_2 \end{pmatrix},$$

$$T = |\eta|^2 \vec{S},$$

$$\vec{U} = \frac{1}{2} \begin{pmatrix} (|\eta_x|^2 - |\eta_y|^2)^2 - (\eta_x \eta_y^* + \eta_y \eta_x^*)^2 \\ (\eta_x \eta_y^* + \eta_y \eta_x^*)^2 - (|\eta_x|^2 - |\eta_y|^2)^2 \end{pmatrix}.$$  

The additional terms entering the free energy are

$$F_\varepsilon = -g_0 \varepsilon \cdot \vec{S} - g_1 \varepsilon \cdot \vec{T} + g_2 \varepsilon \cdot \vec{U}. \quad (9)$$

We go beyond quadratic order in $\eta$ to study the interplay between the applied stress and crystal anisotropy.

All three terms in $[9]$ can be shown to be extremized by real values of $\eta$. Together with $[2]$, every term in the free energy favors a real $\bar{\eta}$. One is again allowed to set $\phi = 0$ from this point on.

Let us first look at $\varepsilon \cdot \vec{S}$. Define $g_0 = |g_0| > 0$. By appropriately choosing $\Phi_0 = 0$ or $\pi/2$, this term can be written as

$$-g_0 \varepsilon \cdot \vec{S} = -g_0 \varepsilon D \cos (2\theta - 2\Phi) \quad (10)$$

The free parameter $\Phi_0$ has been used up to fix the sign of $g_0$; now the $\vec{T}$ term must be taken as-is. Define $g_1 = \pm g_1^\prime$ with appropriate sign depending on the previous choice of $\Phi_0$, and one may write:

$$-g_1 \varepsilon \cdot \vec{T} = -g_1 \varepsilon D^2 \cos (2\theta - 2\Phi). \quad (11)$$

Similarly, the $\vec{U}$ term becomes:

$$g_2 \varepsilon \cdot \vec{U} = -g_2 \varepsilon D^2 \cos (4\theta + 2\Phi), \quad (12)$$

with $g_2 = \pm g_2^\prime$ appropriately defined to absorb the possible sign due to the choice of $\Phi_0$.

To sum up, the stress dependent part of Ginzburg-Landau free energy is

$$F_\varepsilon = -g_0 \cos (2\theta - 2\Phi) \varepsilon D - [g_1 \cos (2\theta - 2\Phi) + g_2 \cos (4\theta + 2\Phi)] \varepsilon D^2. \quad (13)$$

The coefficient $g_0$ is made positive by appropriate choice of $\Phi_0$. The other two coefficients $g_1$ and $g_2$ can be of either signs.
It can be seen that (4), (10), (11) and (12) are all invariant under six-fold (improper) rotation about the z-axis. Consequently, we are free to impose the restriction \( \Phi \in [-\pi/6, \pi/6] \), which amounts to redefining the \( x \)-direction relative to the external stress.

The full expression of Ginzburg-Landau free energy \( F \) employed in this paper is the sum of (4) and (13). Let us also introduce another notation:

\[
F = F_0 + F_\varepsilon = aD + bD^2 + cD^3,
\]

where \( a, b \) and \( c \) are functions of \( \alpha, \theta, \Phi \) and \( \varepsilon \).

We note that, for the case of externally applied strain, the form of Ginzburg-Landau free energy (14) remains exactly identical. The result in this paper equally applies to experiments which uses strain instead of stress.

B. Limit on the Magnitude of Stress

The explicit form of coefficient \( b \) in \( F \) is

\[
b = \beta - g_1 \varepsilon \cos(2\theta - 2\Phi) - g_2 \varepsilon \cos(4\theta + 2\Phi).
\]

If we take this expression at its face value, for \( \varepsilon \) large enough, the minimum value of \( b \) turns negative, and the transition from normal to superconducting state becomes first order.

While not implausible, this stress-induced first order transition has not been observed in any material to our knowledge. We therefore limit the range of \( \varepsilon \) in our theoretical investigation to avoid this regime. The appropriate condition is:

\[
\varepsilon \ll \frac{\beta}{|g_1|}, \frac{\beta}{|g_2|}.
\]

C. Sign of \( \gamma_2 \)

In the absence of an SBF, up to order \( D^2 \) the free energy \( F_0 \) is symmetric under arbitrary rotation around the \( z \)-axis. That is, \( \eta_x \) and \( \eta_y \) are completely degenerate. The six-fold symmetric \( \gamma_2 \) term breaks the degeneracy between \( \eta_x \) and \( \eta_y \). When \( \gamma_2 < 0 \), the ground state has non-zero \( \eta_x \), while \( \gamma_2 > 0 \) results in non-zero \( \eta_y \).

Using a two-band lattice model, Fu argued that \( \eta_y \) is the correct superconducting order parameter for \( \text{Cu}_x\text{Bi}_2\text{Se}_3 \), which corresponds to \( \gamma_2 > 0 \). We will thus assume the positive sign for the remainder of the paper. However, we point out that all the result obtained subsequently can be easily mapped to the case where \( \gamma_2 < 0 \), should it turn out that way in experiment.

Let \( \Delta \theta = \theta - \Phi \), and eliminate all occurrences of \( \theta \) in favor of \( \Delta \theta \) in (4) and (13). By shifting \( \Phi \to \Phi + \pi/6 \), one effectively reverses the signs of both \( \gamma_2 \) and \( g_2 \).

III. PHASE DIAGRAMS

In this section we will describe the three possible phase diagrams, assuming the Ginzburg-Landau free energy (14). The reduced temperature \( a \), the magnitude of the stress \( \varepsilon \), and the orientation of the stress \( \Phi \) will span the three axes of the phase diagrams. The derivation and analysis of the features on these phase diagrams will be given in later sections.

First let us consider the stress-free case \( \varepsilon = 0 \). The superconducting transition takes place at \( \alpha = 0 \), and the order parameter is six-fold rotationally degenerate: \( \theta = -\pi/6, \pi/6, \text{ or } \pi/2 \). We will refers to these directions as the “natural minima” of the free energy.

Let’s turn on the stress. By assumption, the size of \( \varepsilon \) is such that \( b \) remain positive for all values of \( \theta \) and \( \phi \). The normal-to-superconducting transition is then solely controlled by the coefficient \( a \) in (14):

\[
a = \alpha - g_0 \varepsilon \cos(2\theta - 2\Phi).
\]

At finite stress, the transition occurs at

\[
\alpha = \alpha_1(\varepsilon) \equiv g_0 \varepsilon,
\]

and the order parameter is directed at \( \theta = \Phi \). We will refer to this as the upper transition.

A. \( \Phi = 0 \)

We first examine the case when the direction of stress is fixed at \( \Phi = 0 \). This is a plane in the full three-dimensional parameter space \((\Phi, \varepsilon, \alpha)\).

We first orient the stress along \( \Phi = 0 \). Below \( T_c \), the order parameter is locked to \( \theta = 0 \) by symmetry. This is labeled as phase A. As discussed earlier, the \( \gamma_2 \) term favors \( \theta \) along one of the natural minima. The competition between \( g_0 \) and \( \gamma_2 \), eventually leads to a second order phase transition from \( \theta = 0 \) to \( \theta \neq 0 \) at a lower temperature \( \alpha_2(\varepsilon) \). This will be referred to as the middle transition. This will be referred to as phase B.

This middle transition only exists for \( \Phi = 0 \). Otherwise, \( \theta \) will simply be pulled toward the nearer of \( \pm \pi/6 \) due to the reduced symmetry of the setup. At \( \varepsilon = 0 \), the upper and middle transition merge into a single superconducting transition.

Below \( \alpha_2(\varepsilon) \), the \( \Phi = 0 \) plane is a first order coexistent surface, separating the \( \theta > 0 \) and \( \theta < 0 \) phases on either side. The middle transition is the critical end line of this surface.

Going still lower in temperature, another first order phase transition may occur at some \( \alpha_3(\varepsilon) \), and forms the lower bound of the coexistence surface. It will be referred to as the lower transition. Below this lower transition, the orientation \( \theta \) is fixed at \( \pi/2 \). The necessary and sufficient condition for the lower transition is

\[
g_1 < g_2.
\]
FIG. 2: The three allowed phases when the stress is oriented at \( \Phi = 0 \) as shown. (a) Phase A: immediately below the upper transition. (b) Phase B: between the middle and the lower transition if applicable. The two orientations depicted in phase B are degenerate and coexisting, and the dotted lines mark \( \theta = \pm \pi/6 \). (c) Phase C below the possible lower transition.

The lower transition occurs at an temperature where \( \bar{\eta} \) is no longer small. The sixth order \( \gamma_2 \) term dominates the free energy, and by the same argument \( g_0 \) is overwhelmed by \( g_1 \) and can be ignored. The \( \gamma_2 \) term has six degenerate minima (see equation (19)) and this degeneracy is lifted by \( g_1 \) and \( g_2 \). It will be shown subsequently that the difference \( (g_1 - g_2) \) determines which of the six minima is most favorable, leading to the criterion (19).

All three lines of phase transition may continue indefinitely into higher stress, or alternatively the middle transition line may bend down and end when it merge with the lower transition at some value of stress \( \varepsilon^* \). Overall, there are three different possible scenarios associated with this given \( \mathcal{F} \) at \( \Phi = 0 \), as shown in FIG 3.

B. \( \Phi = \pi/6 \)

Immediately below the upper transition the order parameter has \( \theta = \pi/6 \), which is again locked by the symmetry. We shall refer to this as phase D. This orientation also minimizes the \( \gamma_2 \) term, however, and there is no middle transition.

On the other hand, the competition between \( g_0 \) and \( g_2 \) may be relevant at low temperature if (19) is satisfied, and then there is a lower transition. Crossing this transition from high to low temperature, the equilibrium state goes from \( \theta = \pi/6 \) to two-fold degenerate \( (\theta - \pi/6) > 0 \) and \( (\theta - \pi/6) < 0 \). Please see FIG 4 for illustration.

Purely from the symmetry standpoint, this transition can be of either first or second order. It will be shown that, for stress \( \varepsilon \) smaller than some critical value \( \varepsilon_c \), the lower transition is of first order; beyond this point it becomes second order. We will let \( \alpha_4(\varepsilon) \) denote the line of this lower transition.

Similar to the middle transition at \( \Phi = 0 \), lower transition here serves as the critical end line of a first order coexistent plane between the \( \theta > \pi/6 \) and \( \theta < \pi/6 \) phases on two sides of \( \Phi = \pi/6 \).

There are two possible scenarios at \( \Phi = \pi/6 \), one with
FIG. 5: The two possible phase diagrams when \( \Phi = \pi/6 \). The dashed lines represent second order transitions, and the dash-dotted line represents first order transition. N denotes the normal phase. (a) the phase diagram in the absence of a lower transition; (b) the phase diagram in the presence of a lower transition. The lower transition change from first to second order at \( \varepsilon = \varepsilon_c \).

and the other without a lower transition. The phase diagrams are as shown in FIG 6.

C. The Full Phase Diagram

A generic value of \( \Phi \) breaks the six-fold rotational symmetry, and there cannot be a middle transition. If the (19) is satisfied, the resulting lower transition indeed span a first order coexistent surface, interpolating between the lines of lower transition we have identified at \( \Phi = 0 \) and \( \Phi = \pi/6 \). Overall there are three possible scenarios, as shown in FIG 6.

As discussed earlier, the magnitude of stress in our discussion is limited by (16). Beyond the small-stress regime, the possible phase diagrams display very rich and complicated behaviors. But as noted above, this regime is unlikely to be physically relevant.

The middle transition at \( \Phi = 0 \) marks the temperature below which \( \gamma_2 \) dominates over \( g_0 \). The lower transition is due to the competition between \( g_2 \) and \( \gamma_2 \). This already exhausts the list of possible competitions. Consequently we do not anticipate any other transition on the phase diagrams.

We have numerically verified the assertions made in this section. In the following sections, we will present analytical derivations the phase diagrams, as well as analyze their experimental implications.

IV. THE SPLIT SUPERCONDUCTING TRANSITION

A. Upper Transition

The normal-to-superconducting transition occurs at \( \alpha_1(\varepsilon) \), as given in (18). Equivalently, one may revert to using the physical temperature:

\[
T_1(\varepsilon) = \kappa \alpha_1(\varepsilon) + T_0
\]  

is the corresponding critical temperature of the upper transition. This critical temperature is isotropic and independent of \( \Phi \).

However, the fact that \( \alpha_1 \) depends linearly in \( \varepsilon \) is itself an experimental signature for the nematic superconducting order. A single-component order parameter cannot couple linearly to \( \varepsilon \), as there is no way to form a combination that is invariant under three-fold rotation.

The specific heat jump will be anisotropic. Following standard analysis, one recover

\[
\Delta c_{v,1} = \frac{\kappa^2}{2T_1(\varepsilon)} \frac{1}{\beta - \varepsilon [g_1 + g_2 \cos(6\Phi)]} 
\]  

This six-fold anisotropy is another unique signature of the nematic superconducting state. It shares similar physical origin with the predicted six-fold anisotropy of \( H_{c2} \): the external field sees only the underlying lattice as the source of anisotropy. By mapping out the \( \varepsilon \) and \( \Phi \) dependence of \( \Delta c_{v,1} \), one can in principle experimentally determine the Ginzberg-Landau coefficients involved. In particular, the the relative size of \( g_1 \) and \( g_2 \).
B. Middle Transition

The existence of a middle transition is another unique signature of the nematic superconducting state. It separates the $\theta = 0$ phase from $\theta \neq 0$. Therefore $\theta$ itself is an appropriate order parameter to describe this transition. To this end, it is useful to first minimizing $\mathcal{F}$ with respect to variation in $D$. Thus we define $\tilde{D}$ such that

$$
\left( \frac{\partial \mathcal{F}}{\partial \tilde{D}} \right)_{D=\tilde{D}} = 0.
$$

(22)

$\tilde{D}$ is an implicit function of $\theta$, $\alpha$, $\varepsilon$, and $\Phi$. We will assume that the stress orientation is tuned to $\Phi = 0$ for the remainder of this part.

One can now derive a Ginzburg-Landau expansion for $\mathcal{F}$ by replacing $D$ with $\tilde{D}$:

$$
\tilde{\mathcal{F}} \equiv a \tilde{D} + b \tilde{D}^2 + c \tilde{D}^3
= k_0 + k_2 \theta^2 + k_4 \theta^4 + \ldots
$$

(23)

Then the standard analysis of second order phase transition applies.

As noted in the previous section, the middle transition is due to the competition between $g_0$ and $\gamma_2$. It can be shown that, for asymptotically small $\varepsilon$, the leading expressions of coefficients $k_2$ and $k_4$ are independent of $g_1$ and $g_2$. Here we present these asymptotic formulas, though the exact analytic result can be obtained by carefully retaining all terms.

The critical temperature $T_2$ is

$$
T_2 = T_0 \left[ 1 - \frac{2 \beta}{3 \kappa} \sqrt{\frac{g_0}{\gamma_2 \varepsilon}} + \frac{g_0}{\kappa} \left( \frac{2 \gamma_2 - \gamma_1}{3 \gamma_2} \right) \varepsilon \right],
$$

(24)

and the specific heat jump is

$$
\Delta c_v = \frac{k^2 T_0 (\varepsilon)}{T_0^2} \left[ 2 \frac{g_0 \varepsilon (4 \gamma_1 - 5 \gamma_2 + 4 \beta \sqrt{\gamma_2 g_0 \varepsilon})}{(\beta + (\gamma_1 + \gamma_2) \sqrt{g_0 \varepsilon / \gamma_2})} \right].
$$

(25)

The formulas (24) and (25) are accurate in the limit where $(g_1 \varepsilon / \beta)$, $(g_2 \varepsilon / \beta)$, $g_1 \sqrt{\varepsilon / g_0 \gamma_2}$ and $g_2 \sqrt{\varepsilon / g_0 \gamma_2}$ are small.

We also note that $\Delta c_{v,2}$ vanishes in the limit $\varepsilon \to 0$. This is consistent with the sum rule

$$
\lim_{\varepsilon \to 0} (\Delta c_{v,1} + \Delta c_{v,2}) = \Delta c_v,
$$

(26)

where $\Delta c_v$ is the specific heat jump of the superconducting transition at zero stress.

V. LOWER TRANSITION

The existence of a lower transition can be inferred by looking at the extremely low temperature $\alpha \to -\infty$ limit:

$$
\tilde{\mathcal{F}} \approx |\alpha|^{3/2} \left[ 3 \gamma_1 + 3 \gamma_2 \cos(6\theta) \right]
+ \left[ |\alpha| \left\{ \beta - \varepsilon \left[ g_1 \cos(2\theta - 2\Phi) + g_2 \cos(4\theta + 2\Phi) \right] \right\} / 3 \right] \left[ \gamma_1 + \gamma_2 \cos(6\theta) \right]
+ O \left( |\alpha|^1 \right).
$$

(27)

The leading term has the six-fold rotational symmetry, favoring all natural minima equally. The sub-leading term lifts this degeneracy.

The sign of $(g_1 - g_2)$ controls the qualitative behavior of (27). First assuming a generic value of $\Phi$ that breaks the six-fold rotational symmetry. If $g_1 > g_2$, out of the three minima, the one closest to $\Phi$ wins out. Conversely if $g_1 < g_2$, the minimum furthest away from $\Phi$ becomes the lowest. On the other hand, the order parameter has $\theta = \Phi$ at the upper transition, and then initially drifts toward the nearest of the natural minima as the temperature is lowered. For $g_1 < g_2$, there must be a first order transition separating the low-temperature asymptotic behavior from that just below the upper transition. This justifies (10) as the criterion for a lower transition.

The cases of $\Phi = 0$ and $\pi/6$ require special attention. For the sake of clarity, we introduce $\sigma = (\theta - \Phi)$, and consider the range $-\pi/2 < \sigma \leq \pi/2$. It can be shown that $\tilde{\mathcal{F}}$ exhibits the following properties at $\Phi = 0$ or $\pi/6$:

1. $\tilde{\mathcal{F}}$ is symmetric under $\sigma \to -\sigma$
2. $\tilde{\mathcal{F}}$ is always stationary at $\sigma = \pi/2$ and 0.
3. $\tilde{\mathcal{F}}$ admits at most five stationary points within the range $-\pi/2 < \sigma < \pi/2$.
4. Just below the upper transition, $\sigma = 0$ is the global minimum of $\tilde{\mathcal{F}}$, and $\sigma = \pi/2$ is the global maximum.

First let us look at the $\Phi = 0$ case. Coming down from high temperature, $\sigma$ is initially fixed at 0, but drifts away from this high-symmetry direction below the middle transition. The previous argument for generic value of $\Phi$ therefore applies equally. However, below the lower transition, the location of global minimum in this case is pinned exactly at $\sigma = \pi/2$ by the enhanced symmetry.
Now we turn to the case of $\Phi = \pi/6$. As noted in the previous section, the lower transition here may be either first or second order.

If the lower transition is first order, $\tilde{F}$ must develop a maximum-minimum pair on each side of $\sigma = 0$ as the temperature is lowered. This already accounts for five stationary points within $-\pi/2 < \sigma < \pi/2$, and $\sigma = 0$ must always remain a local minimum. On the other hand, a second order lower transition implies the stationary point at $\sigma = 0$ must revert its character as the temperature is lowered. Therefore the criterion separating first and second order transition is whether $(\partial^2 \tilde{F}/\partial \sigma^2)_{\sigma=0}$ changes sign when the temperature is lowered. This gives the critical stress:

$$\varepsilon_c = \frac{36 \, g_0 \gamma_3}{(g_1 - 4g_2)^2}.$$  \hspace{1cm} (28)

If $\varepsilon > \varepsilon_c$, the lower transition becomes second order.

Finally, we will address the question of if and when the middle transition at $\Phi = 0$ ends on the surface of lower transition. While a closed-form analytic solution is possible, the full expression is extremely long and unwieldy. We instead supply a recipe here.

The line of middle transition $\alpha_2(\varepsilon)$ is implicitly defined by

$$\left(\frac{\partial^2}{\partial \sigma^2} \tilde{F}(\alpha = \alpha_2, \varepsilon, \sigma, \Phi = 0)\right)_{\sigma=0} = 0.$$  \hspace{1cm} (29)

Since the phase below the lower transition has exactly $\sigma = \pi/2$ at $\Phi = 0$, the equation

$$\tilde{F}(\alpha_2(\varepsilon_*), \varepsilon, \sigma = 0, \Phi = 0) = \tilde{F}(\alpha_2(\varepsilon_*), \varepsilon, \sigma = \pi/2, \Phi = 0)$$  \hspace{1cm} (30)

determines the stress $\varepsilon_*$ at which the middle and lower transitions meet if a solution exists. If there is no solution, then the line of middle transition extends indefinitly into large $\varepsilon$. Our numerical results indicate that, depending on the actual values of Ginzburg-Landau coefficients, $\varepsilon_*$ can be well within the limit (16), and the ending of the middle transition can be physically relevant.

VI. PRE-EXISTING SBF AND SIGNATURES OF NEMATIC SUPERCONDUCTIVITY

Two-fold anisotropy of upper critical field $H_{c2}$ has been consistently observed in experiment\textsuperscript{14,16-18}. In the work of Pan et al.\textsuperscript{16}, part of the experiment was done with the sample repeatedly cycled between a high temperature (about 5K, or 1.7 times the $T_c$) and 0.3K where the measurements were taken. Yet the same preferred orientation persisted.\textsuperscript{16} This points to a fairly robust SBF that can survive the cycle where sample is taken far into the normal phase and back. We therefore consider in this section a pre-existing SBF that is essentially a given constant background.

To our knowledge, the reported two-fold anisotropy always aligns with a lattice direction in all cases\textsuperscript{15,16-18}. We therefore conjecture that the SBF, whatever it may be, breaks the lattice symmetry by favoring one out of the three two-fold axes in the Bi$_2$Se$_3$ structure. One does not need to know the precise nature of the SBF: all that matters is that it must be a nematic director, and phenomenologically it must couple to the order parameter in a manner similar to that of the stress. In the same notation developed for stress, the SBF $\vec{p}$ can be expressed as

$$\vec{p} = p \begin{pmatrix} \cos 2\Phi_p \\ \sin 2\Phi_p \end{pmatrix} \equiv \begin{pmatrix} p_1 \\ p_2 \end{pmatrix},$$  \hspace{1cm} (31)

where $p$ is positive, and $\Phi_p$ is either 0 or $\pi/6$.

Now we consider how an external stress can be applied to cancel the effect of this SBF. We will focus on the split superconducting transition here, i.e. the pair of upper and middle transitions. To this end, one only needs to retain the coupling terms quadratic in SBFs. The modified Ginzburg-Landau coupling terms reads:

$$F_p = - (g_0 \varepsilon_g + g_0 \vec{p}) \cdot \vec{S},$$  \hspace{1cm} (32)

where $S$ is given by (8), and $\varepsilon$ by (5) with $\Phi_0 = 0$. The $x$-direction in this case is instead decided by the SBF, so the stress orientation $\Phi$ takes value in the broader range ($-\pi/2, \pi/2$). The Ginzburg-Landau free energy under consideration is:

$$F = F_0 + F_p.$$  \hspace{1cm} (33)

The SBF orientation can be found by looking at the two-fold anisotropy at zero external stress. One may then align the stress $\vec{v}$ with $\vec{p}$. Once the stress is aligned, following (15) and (20), the critical temperature of the upper (normal-to-superconducting) transition will show a kink when one continuously varies the applied stress:

$$(T_1 - T_0) \propto |g_0 \vec{p} - g_0 \varepsilon|.$$  \hspace{1cm} (34)

This behavior is a unique signature of a nematic superconducting state. If the order parameter is single-component, it cannot couple linearly to either stress or the SBF. One would expect instead a quadratic dependence:

$$(T_1 - T_0) \propto (g_0 \vec{p} - g_0 \varepsilon)^2.$$  \hspace{1cm} (35)

In addition, if the kink in (34) can be identified, the predicted six-fold anisotropy purely due to the lattice should be restored at that point. This effect can be seen from the specific heat jump, following (21). The upper critical field $H_{c2}$ should also exhibit the same six-fold anisotropy. This provides further verification for the nematic state.

The existence of the middle transition is another unique signature of the nematic superconducting state.
Adopting to the current case with both SBF and stress, one needs to tune the orientation
\[
(g_0 \vec{p} - g_0 \vec{\varepsilon}) \propto \begin{pmatrix} 0 \\ 1 \end{pmatrix}
\]  
(36)
to see the second order transition. Calorimetry experiments can detect the specific heat jump across the second order transition.

If the SBF itself is already along the desired orientation, the condition (36) can be trivially satisfied by aligning the applied stress with the SBF. The other possibility is that it is off by $\pi/2$. In this case, one instead applies a stress at an arbitrary angle relative to the SBF and continuously changes its magnitude. Although the condition (36) will be exactly satisfied by only one value of stress, at stress slightly off that value the sharp phase transition is smeared into a crossover of some finite width, and one can still observe a steep increase in specific heat as an experimental signature.

**VII. JOSEPHSON JUNCTION WITH AN S-WAVE SUPERCONDUCTOR**

In this section, we change our direction slightly and discuss the phenomenology of tunneling current between a nematic superconductor and an s-wave superconductor. As pointed out by Yip et al., one may write down the effective Hamiltonian of a junction purely based on symmetry considerations, in the same spirit of the Ginzburg-Landau theory. The behavior of the critical current then reflects the symmetry of the nematic order parameter. Here the implicit assumption is that the interface itself has spin-orbit coupling so as to allows a tunneling supercurrent.

The theory for Josephson tunneling between two s-wave superconductors was worked out in the classic paper by Ambegaokar and Baratoff. For a junction between two different s-wave superconductors, right below the lower of the two critical temperatures, the critical current is proportional to $\sqrt{T_c - T}$, and shows no strong stress dependence. In this section we explore how the proposed nematic superconducting state gives rise to qualitative differences.

We consider the scenario where an s-wave superconductor (with a much higher $T_c$) is attached to the $M_2\text{Bi}_3\text{Se}_3$ sample being tested. The surface contact of the sample is cut perpendicular to the $z$-axis. We assume that the junction is in the tunneling limit.

The two-fold rotation $C'_{2d}$ in the original $D_{3d}$ group is no longer a valid symmetry on the surface of contact. The junction coupling term in the Hamiltonian respects only $C_3$ and $M$, and of course the $U(1)$ symmetry. The ingredients entering the coupling terms are the s-wave order parameter $\Psi$, the nematic order parameter $\vec{\eta}$, and the total SBF $\vec{p}$ as defined in (31). We make the same gauge choice where $\vec{\eta}$ is entirely real, and then the phase $\chi$ of $\Psi = e^{i\chi}|\Psi|$ is the phase difference across the junction.

We will first restrict ourselves to the case where $\vec{\eta}$ is in the $E_u$ representation. The leading order terms that couple $\vec{\eta}$ and $\Psi$ and respect the symmetry of the junction are
\[
\begin{align*}
F_{J0} &\propto |\Psi|^2 \left( \eta_x p_2 - \eta_y p_1 \right) + \text{h.c.} \\
F_{J1} &\propto |\Psi|^2 \left( \eta_x S_2 - \eta_y S_1 \right) + \text{h.c.},
\end{align*}
\]  
(37)
where $S_1$, $S_2$ are defined in (30). Here $F_{J1}$ is the effective Hamiltonian in the absence of an SBF, and $F_{J0}$ is the additional term allowed when $\vec{\varepsilon}$ is non-zero, so labeled because $F_{J0}$ is of a lower order in $\vec{\eta}$.

Assume that the junction is but a small perturbation to the bulk, and that the system is sufficiently close to the upper transition. We may therefore use the unperturbed solution for the nematic order parameter $\vec{\eta}$, and assume that it aligns with SBF $\vec{p}$, or $\theta = \Phi$. The coupling (37) then reduces to
\[
\begin{align*}
F_{J0} &\propto |\Psi|^2 |p| \sqrt{D} \sin (3\Phi) \cos(\chi) \\
F_{J1} &\propto |\Psi|^2 D^{3/2} \sin (3\Phi) \cos(\chi).
\end{align*}
\]  
(38)

The critical current across the junction can be identified as $2 \left( \frac{\partial}{\partial \theta} \right) (F_{J0} + F_{J1})$. The factor of two is because this describes the tunneling of Cooper pairs. In the presence of an SBF, $F_{J0}$ should dominate when the temperature is close to the upper transition. Using
\[
D \propto (-\alpha + g_0 |p|)
\]  
(39)
as per standard Ginzburg-Landau theory, the dependence of critical current on external parameters $\alpha$, $|p|$ and $\Phi_p$ can be read off:
\[
I_{c0} \propto |p| \sqrt{-\alpha + g_0 |p|} |\sin 3\Phi_p|.
\]  
(40)

The critical current shows three-fold anisotropy with respect to the orientation $\Phi_p$ of the SBF, as well as a non-linear dependence on the strength $|p|$.

Away from the immediate vicinity of the upper transition, the relation $\theta = \Phi_p$ only holds along the high symmetry directions. The spatial angular dependence of $I_{c0}$ will in general contain higher harmonics, but is still three-fold anisotropic.

When the SBF vanishes, $F_{J1}$ is the non-vanishing term, and the nematic order parameter naturally aligns to either $\theta = 0$ or $\theta = \pi/2$. Considerations similar to (40) lead to the critical current:
\[
I_{c1} \propto |\alpha|^{3/2} |\sin 3\theta|.
\]  
(41)
Depending on the equilibrium orientation $\theta$, the tunneling current may be identically zero, or it may show a unique temperature dependence of $|\alpha|^{3/2}$, as opposed to $|\alpha|^{1/2}$ between two s-wave superconductors.

In either cases of (40) or (41), the critical current shows a markedly different behavior compared with that between two s-wave superconductors.
If the order parameter $\eta$ transforms instead in the parity-even $E_g$ representation, the junction coupling terms \cite{37} are slightly modified:

\begin{align}
(\mathcal{F}_{J0})_{E_g} & \propto \Psi^\ast (\eta_x p_1 + \eta_y p_2) + \text{h.c.} \\
(\mathcal{F}_{J1})_{E_g} & \propto \Psi^\ast (\eta_x S_1 + \eta_y S_2) + \text{h.c.} \\
\end{align}

The upshot is that both occurrences of $\sin$ are replaced by $\cos$ in \ref{10} and \ref{44}. All the qualitative conclusions for the $E_g$ case stand unaffected for $E_g$.

The above treatment amounts to the direct coupling of the two superconducting bulks. This is admittedly an over-simplification: for unconventional superconductors, surface depairing may occur depending on the exact detail of the gap function\cite{17}. The extent of this surface effect is dictated by the coherence length. Therefore, near $T_c$ when the coherence length is large, surface depairing may substantially suppress the tunneling current; the overall temperature dependence will then have a higher power\cite{15,48} than our simple $|A|^3/2$ prediction. Nonetheless, the temperature dependence is clearly distinct from the s-wave-to-s-wave case.

\section{VIII. CONCLUSION}

In this paper, we explore the coupling between the two-component superconducting order parameter $\eta$ and the (traceless part of) stress $\varepsilon$ in the basal plane, and map out all possible phase diagrams allowed by symmetry constraints. Indeed the analysis is not restricted to mechanical stress: any charge-neutral SBF must couple on the exact detail of the gap function\cite{17}. The extent of this surface effect is dictated by the coherence length. Therefore, near $T_c$ when the coherence length is large, surface depairing may substantially suppress the tunneling current; the overall temperature dependence will then have a higher power\cite{15,48} than our simple $|A|^3/2$ prediction. Nonetheless, the temperature dependence is clearly distinct from the s-wave-to-s-wave case.

The same cannot be said for the lower transition, which is not connected to the superconducting transition without a SBF. The assumption that Ginzburg-Landau coefficients are temperature-independent may no longer be a quantitatively accurate approximation, and we focus our effort on the qualitative results. The existence of a lower transition hinges on the criterion \cite{16}. It is first order almost everywhere, except when $\Phi = \pi/6$ and $\varepsilon > \varepsilon_c$ it becomes second order. The critical value $\varepsilon_c$ is given in \ref{29}.

The observed two-fold anisotropy of $H_{c2}$ suggested that a pre-existing SBF explicitly breaks the rotational symmetry in the sample. At the phenomenological level, the total symmetry breaking field is the sum of the SBF and an externally applied stress, and we discuss how a stress can counteract and cancel this SBF to unmask the true anisotropy, if any, of the superconducting state itself.

We point out two thermodynamics experimental signatures unique to the nematic state. First, the superconducting critical temperature is linearly proportional to the strength of the total SBF, as given in \ref{44}, as opposed to the quadratic relation if the order parameter is single-component. Second, the existence of the middle transition, and the associated finite crossover even if the total SBF isn’t exactly tuned to the right orientation, can be observed through calorimetry experiments.

When linked to another s-wave superconductor, the Josephson tunneling current also offers hints to the superconducting pairing symmetry. The nematic scenario results in a critical current that depends anisotropically on the SSB, as given in \ref{44}. In the absence of SSB, the temperature dependence of the critical current is also markedly different from the tunneling between two s-wave superconductors, as shown in \ref{44}. The Josephson tunneling current may provide yet another “smoking gun” test for the nematic superconductivity.

We hope our findings here will guide future experimental effort in discerning the pairing symmetry of the $\mathrm{Bi}_2\mathrm{Se}_3$ family of superconductor, thereby helping to settle the debate on the topological nature of the superconductivity.

\section{ACKNOWLEDGMENTS}

The authors would like to thank Anne de Visser for sharing the details of experimental procedures carried out in Pan et. al\cite{69}. This work is supported by Ministry of Science and Technology, Taiwan under grant number MOST 107-2112-M-001-035-MY3, and PTH is supported under grant number MOST 107-2811-M-001-045.

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