Response to Comments from Reviewer 3

We would like to thank the editor and referee for their comments and suggestions on the manuscript. We would also like to answer the questions raised by the report from the reviewer as following.

[Q1] The focus of the paper is unclear: are we studying impurities, or boundary effects? It would be informative to compare the results on mesoscopic domains with no impurities and varying sizes and shapes, and on large domains with impurities. Restricting the study to the simplest TDGL model is justified I think: the parameter space of the full model is too large to be surveyed. (Quoted from the report of Reviewer 3)

[A1] **First of all, we would like to investigate the L-κ_1 phase diagram of the $L \times L$ two-band superconductor with L the sample size and κ_1 the Ginzburg-Landau (GL) parameter in the absence of impurity.** We perform the corresponding numerical calculations based on the time-dependent GL theory (3)-(5) in the revised manuscript. In the procedure of simulations, we set the coefficients in the GL equations as $\Gamma_1 = \Gamma_2 = 5$, $m_1 = 2m_2$, $\alpha_{10} = \alpha_{20}$ and $\beta_1 = \beta_2$. The obtained $L-\kappa_1$ phase diagram is plotted in Fig. 1. Here the sample size L is measured in units of the first-band coherence length $\xi_1 = \hbar / \sqrt{2m_1\alpha_{10}}$, and $\kappa_1 = \lambda_1/\xi_1$ with λ_1 the London penetration depth of the first condensate. We also take the magnetic field in units of $H_0 = \Phi_0/\left(2\pi\xi_1^2\right)$ with the flux quantum $\Phi_0 = \pi\hbar c/e$ and the vector potential $\mathbf{A}$ in units of $A_0 = H_0\xi_1$. It can be seen from Fig. 1 that with the decrease of L, the vortex cluster phase produced by the long-range attractive interaction between vortices gradually vanishes. Meanwhile, we also notice the critical sample size L_c for the disappearance of this cluster state is $32\xi_1$. **Thus, the superconducting system will stay in the type-1.5 regime above L_c and the type-II regime below L_c in the absence of impurity.**

As we know, the type-1.5 superconductor originates from a peculiar vortex interaction that exhibits short-range repulsion and long-range attraction characteristics. The obtained critical size L_c is consistent with the characteristic length scale (about $30\xi_1$) of the crossover from the attractive to repulsive intervortex interaction [J. Carlström, E. Babaev and M. Speight, Type-1.5 superconductivity in multiband systems, Phys. Rev. B 83, 174509

(2011)]. For the sample size $L > L_c$, the long-range attractive potential between vortices will dominate at the external magnetic field $H_e = 0.8H_0$ and the system is allowed to spontaneously form the stable vortex cluster. However for $L < L_c$, the repulsive intervortex interaction will prevail in the mesoscopic superconductor and the vortex cluster phase can only be induced by other effects such as impurities.

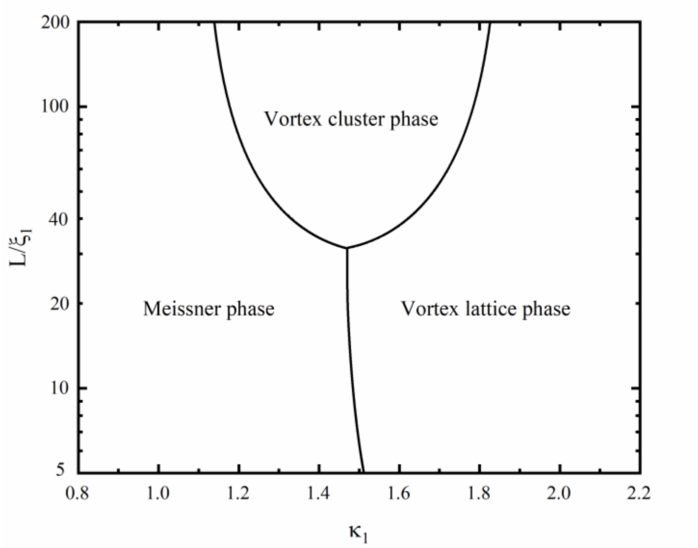

Figure 1: The $L-\kappa_1$ phase diagram of the $L \times L$ two-band superconductor in the absence of impurity. We set the external magnetic field $H_e = 0.8H_0$ in the numerical simulations, and plot the sample size L on a logarithmic scale.

In addition to the superconducting square discussed above, we further explore the transition behaviors of mesoscopic samples with the aspect ratio different from 1 in the absence of impurity. As a simple example, we choose the $15\xi_1 \times 20\xi_1$ superconducting sample with each side length below L_c. For $H_e = 0.8H_0$, we plot the magnetic field intensity B_z and the order parameter of the first condensate $|\Psi_1|$ at $t = 10^4 t_0$ in Fig. 2. With the increase of the GL parameter κ_1, we can see the direct transition of this system from the perfect diamagnetic state to the Abrikosov lattice phase as shown in Fig. 2. All of these numerical results thus suggest that the vortex cluster phase will be excluded for arbitrary mesoscopic sample with the characteristic scale less than L_c in the absence of impurity.

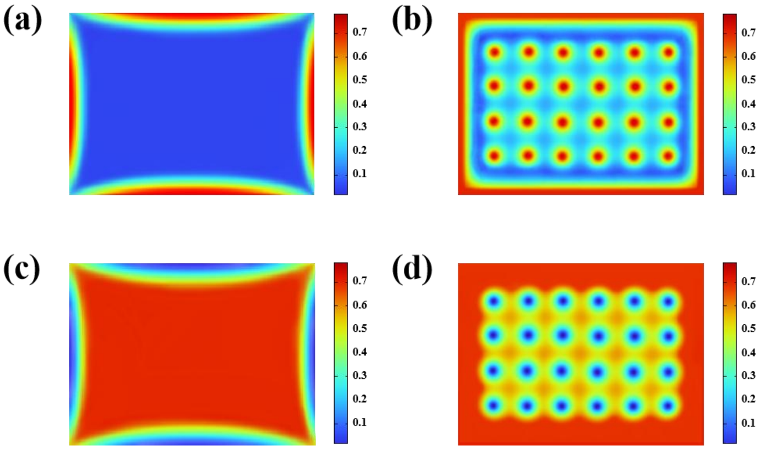

Figure 2: Transition of the magnetic field intensity B_z (a,b) and the order parameter of the first condensate $|\Psi_1|$ (c,d) for the $15\xi_1 \times 20\xi_1$ type-II superconductor. The snapshots show the Meissner phase (a,c) and vortex lattice phase (b,d) at the GL parameter $\kappa_1 = 0.70$ and 2.10 respectively. The magnetization only has the component perpendicular to the superconducting plane.

In this work, we mainly focus on the possible generation of the vortex cluster phase in the mesoscopic superconducting system with $L < L_c$ due to the impurity effect. In the presence of impurity, the phenomenological parameter α_i $(i = 1,2)$ in the GL equations can be expressed as $\alpha_i = \alpha_{i0} g(\mathbf{r})$, and the function $g(\mathbf{r})$ is used to model the defect potential which will deplete the superconducting state at specific positions. As an example, we introduce an isotropic impurity with the radius $0.5\xi_1$ at the center of the $15\xi_1 \times 15\xi_1$ superconducting sample here. Then, the defect function $g(\mathbf{r})$ will be characterized by the disorder strength g inside the impurity. The obtained $g - \kappa_1$ phase diagram is shown in Fig. 3. It can be seen from Fig. 3 that with the increase of the absolute value of g, the vortex cluster phase induced by the attractive interaction from the impurity will gradually appear in the system. Meanwhile, we also see that there exists a critical impurity strength $g_c \approx -0.22$ for the generation of the vortex cluster state in this $15\xi_1 \times 15\xi_1$ sample. **Thus, this mesoscopic superconductor will stay in the type-1.5 regime for $|g| > |g_c|$ in the presence of an isotropic impurity.**

Also see Subsection 4.1 "$L - \kappa_1$ phase diagram in the clean limit" from page 6 to page 8, the first paragraph and Figure 3 on page 8 in the revised version of manuscript.

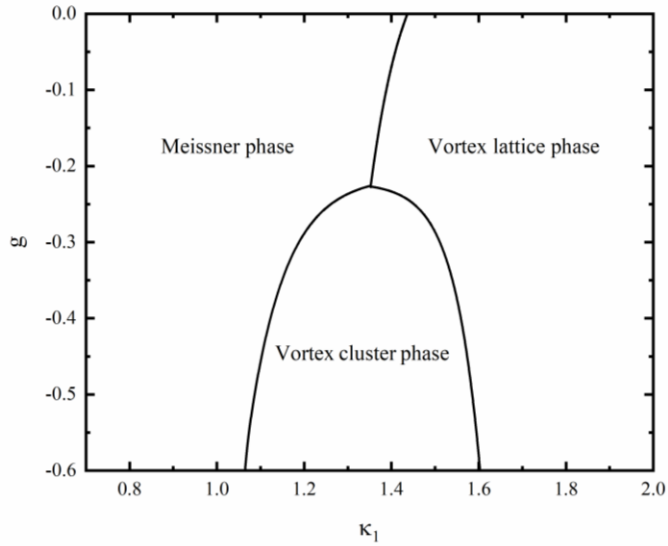

Figure 3: The $g-\kappa_1$ phase diagram of the $15\xi_1 \times 15\xi_1$ two-band superconductor in the presence of an isotropic impurity. We set the external magnetic field $H_e = 0.8H_0$ in the numerical simulations.

[Q2] Since boundary effects are important for the results here, I'm a bit troubled by the authors' choice of boundary condition. It's true that the conditions assumed ensure that no supercurrent passes through the boundary, but they are much stronger than is required by that condition. They are not gauge invariant, and they impose that each "component" of the supercurrent (associated with each condensate) is confined separately. For the simple model studied here, which has $\mathrm{U}(1)\times\mathrm{U}(1)$ symmetry, and hence separately conserved supercurrents, this may well be justified, but for two component GL models in general this strikes me as a very artificial assumption. The authors justify their choice by citation to the literature, but having followed the thread back 3 links I still haven't found a derivation of them. Given the importance (presumably) of boundary effects, I think a derivation of the boundary conditions from physical/mathematical principles is needed. (Quoted from the report of Reviewer 3)

[A2] **Firstly, we would like to show that in the zero electric potential gauge, the dimensionless boundary conditions will take the form**

$$\nabla\Psi_i \cdot \mathbf{n} = 0, \qquad \mathbf{A}\cdot\mathbf{n} = 0 \qquad \text{and} \qquad \nabla\times\mathbf{A} = \mathbf{H}_e \tag{1}$$

as adopted in our work. Here Ψ_i $(i=1,2)$ represents the superconducting order parameter. $\mathbf{n}$ is the outward unit vector normal to the boundary. $\mathbf{H}_e$ is the external magnetic field and $\mathbf{A}$ is the vector potential.

We start from the following gauge invariant boundary conditions between a two-band superconductor and an insulator (or vacuum)

$$(\nabla - \mathrm{i}\mathbf{A})\Psi_i \cdot \mathbf{n} = 0, \qquad \left(\frac{\partial \mathbf{A}}{\partial t} + \nabla\varphi\right)\cdot \mathbf{n} = 0 \qquad \text{and} \qquad \nabla \times \mathbf{A} = \mathbf{H}_e. \tag{2}$$

Here φ is defined as the electric potential. Given an arbitrary function χ, the gauge transformation takes the form as

$$\Psi_i \to \Psi_i e^{\mathrm{i}\chi}, \qquad \mathbf{A} \to \mathbf{A} + \nabla\chi \qquad \text{and} \qquad \varphi \to \varphi - \frac{\partial \chi}{\partial t}. \tag{3}$$

It is easy to show that the boundary conditions in Eq. (2) maintain the gauge invariance. Then with the zero electric potential gauge, we can get from the transformation in Eq. (3)

$$\frac{\partial \chi}{\partial t} = \varphi. \tag{4}$$

Plugging this condition into the second equation of the boundary conditions (2) , it leads to

$$\frac{\partial \mathbf{A}}{\partial t} \cdot \mathbf{n} = 0 \tag{5}$$

in this new gauge. This equation can be integrated to give $\mathbf{A}\cdot\mathbf{n} = 0$, which transforms the boundary condition $(\nabla - \mathrm{i}\mathbf{A})\Psi_i \cdot \mathbf{n} = 0$ into the form $\nabla\Psi_i \cdot \mathbf{n} = 0$. Based on the analysis above, we can see that the boundary conditions in Eq. (1) are simply the result of a particular gauge choice.

Secondly, we would like to give a microscopic derivation of the dimensionless boundary condition $(\nabla - \mathrm{i}\mathbf{A})\Psi_i \cdot \mathbf{n} = 0$ which is presented in Eq. (2). We try to show that at the interface of two-band superconductor and insulator (or vacuum), this boundary condition is applicable not only for the simple $\mathrm{U}(1)\times\mathrm{U}(1)$ symmetric model studied here but also the two-component GL models in general. In this process, we will follow the procedure in the single-band case suggested by de Gennes [P.G. de Gennes, Superconductivity of metals and alloys, Westview Press, New York, 1966, pp. 229-232].

Based on the work of Zhitomirsky and Dao [M.E. Zhitomirsky and V.H. Dao, Ginzburg-Landau theory of vortices in a multigap superconductor, Phys. Rev. B 69, 054508 (2004)], we write the Hamiltonian of a two-band superconductor as

$$H = \sum_{i\sigma} c_{i\sigma}^{\dagger}(\mathbf{r})\hat{h}(\mathbf{r})c_{i\sigma}(\mathbf{r}) - \sum_{ii'} g_{ii'} c_{i\uparrow}^{\dagger}(\mathbf{r})c_{i\downarrow}^{\dagger}(\mathbf{r})c_{i'\downarrow}(\mathbf{r})c_{i'\uparrow}(\mathbf{r}). \tag{6}$$

Here, $i,i'=1,2$ are the band indices and $\sigma=\uparrow,\downarrow$ is the spin index. $\hat{h}(\mathbf{r})$ is the single particle Hamiltonian of the normal metal, and $g_{ii'}$ are the effective electron-electron interaction constants with $g_{12}=g_{21}$.

We can introduce the gap functions

$$\Delta_i(\mathbf{r})=-\sum_{i'} g_{ii'}\left\langle c_{i'\downarrow}(\mathbf{r})c_{i'\uparrow}(\mathbf{r})\right\rangle \tag{7}$$

and transform the Hamiltonian into the mean field form

$$H_{\text{eff}}=\sum_{i\sigma} c_{i\sigma}^{\dagger}(\mathbf{r})\hat{h}(\mathbf{r})c_{i\sigma}(\mathbf{r})+\sum_i\left[\Delta_i(\mathbf{r})c_{i\uparrow}^{\dagger}(\mathbf{r})c_{i\downarrow}^{\dagger}(\mathbf{r})+\text{H.c.}\right]. \tag{8}$$

This effective Hamiltonian can be diagonalized by means of the Bogoliubov transformation with b and $b^{\dagger}$ the annihilation and creation operators of quasi-particle excitations

$$c_{i\uparrow}(\mathbf{r})=\sum_{\mathbf{k}}\left[u_{i\mathbf{k}}(\mathbf{r})b_{i\mathbf{k}\uparrow}-v_{i\mathbf{k}}^{*}(\mathbf{r})b_{i\mathbf{k}\downarrow}^{\dagger}\right] \tag{9}$$

and

$$c_{i\downarrow}(\mathbf{r})=\sum_{\mathbf{k}}\left[u_{i\mathbf{k}}(\mathbf{r})b_{i\mathbf{k}\downarrow}+v_{i\mathbf{k}}^{*}(\mathbf{r})b_{i\mathbf{k}\uparrow}^{\dagger}\right] \tag{10}$$

where $\mathbf{k}$ is the wave vector. With the anti-commutation relations between the fermion operators and the equation of motion for $c_{i\sigma}(\mathbf{r})$, we can obtain the Bogoliubov-de Gennes equations for a two-band superconductor

$$\begin{pmatrix}\hat{h} & \Delta_i(\mathbf{r})\\ \Delta_i^{*}(\mathbf{r}) & -\hat{h}^{*}\end{pmatrix}\begin{pmatrix}u_{i\mathbf{k}}(\mathbf{r})\\ v_{i\mathbf{k}}(\mathbf{r})\end{pmatrix}=E_{i\mathbf{k}}\begin{pmatrix}u_{i\mathbf{k}}(\mathbf{r})\\ v_{i\mathbf{k}}(\mathbf{r})\end{pmatrix} \tag{11}$$

where $E_{i\mathbf{k}}$ is the energy of the excitation. Then with Eq. (7), we can transform the self-consistent gap equations into

$$\Delta_i(\mathbf{r})=\sum_{i'\mathbf{k}} g_{ii'}v_{i'\mathbf{k}}^{*}(\mathbf{r})u_{i'\mathbf{k}}(\mathbf{r})\left[1-2f(E_{i'\mathbf{k}})\right] \tag{12}$$

with $f\left(E_{i\mathbf{k}}\right)=\left[1+\exp\left(E_{i\mathbf{k}}/k_BT\right)\right]^{-1}$ and T the temperature.

In the analogy with the single-band case, for small gap functions Δ_i, we can obtain the linearized form of self-consistency conditions from Eqs. (11) and (12) as

$$\Delta_i(\mathbf{r})=\sum_{i'}\int K_{ii'}(\mathbf{r},\mathbf{r}')\Delta_{i'}(\mathbf{r}')\mathrm{d}\mathbf{r}' \tag{13}$$

with the kernel

$$K_{ii'}(\mathbf{r},\mathbf{r}') = \frac{g_{ii'}}{2}\sum_{\mathbf{k}\mathbf{k}'}\frac{\tanh\left(\varepsilon_{i'\mathbf{k}}/2k_BT\right)+\tanh\left(\varepsilon_{i'\mathbf{k}'}/2k_BT\right)}{\varepsilon_{i'\mathbf{k}}+\varepsilon_{i'\mathbf{k}'}}\Phi^*_{i'\mathbf{k}}(\mathbf{r}')\Phi^*_{i'\mathbf{k}'}(\mathbf{r}')\Phi_{i'\mathbf{k}}(\mathbf{r})\Phi_{i'\mathbf{k}'}(\mathbf{r}). \quad (14)$$

Here $\Phi_{i'\mathbf{k}}(\mathbf{r})$ and $\varepsilon_{i'\mathbf{k}}$ are defined as the normal-state eigenfunction and eigenvalue of the electron with $\hat{h}\Phi_{i'\mathbf{k}} = \varepsilon_{i'\mathbf{k}}\Phi_{i'\mathbf{k}}$.

We now assume the small spatial variations in the vector potential $\mathbf{A}$. Then the eigenfunctions $\Phi_{i'\mathbf{k}}$ in the normal metal in the presence of $\mathbf{A}$ will differ from the eigenfunctions $w_{i'\mathbf{k}}$ in the absence of $\mathbf{A}$ by only a phase factor, i.e.,

$$\Phi^*_{i'\mathbf{k}}(\mathbf{r}')\Phi_{i'\mathbf{k}}(\mathbf{r}) \to w^*_{i'\mathbf{k}}(\mathbf{r}')w_{i'\mathbf{k}}(\mathbf{r})\exp\left[\frac{\mathrm{i}}{2}\mathbf{A}\cdot(\mathbf{r}-\mathbf{r}')\right]. \quad (15)$$

Plugging into Eq. (13), it will lead to

$$\Delta_i(\mathbf{r}) = \sum_{i'}\int \bar{K}_{ii'}(\mathbf{r},\mathbf{r}')\Delta_{i'}(\mathbf{r}')\exp\left[\mathrm{i}\mathbf{A}\cdot(\mathbf{r}-\mathbf{r}')\right]\mathrm{d}\mathbf{r}' \quad (16)$$

with the kernel in the absence of external magnetic field

$$\bar{K}_{ii'}(\mathbf{r},\mathbf{r}') = \frac{g_{ii'}}{2}\sum_{\mathbf{k}\mathbf{k}'}\frac{\tanh\left(\varepsilon_{i'\mathbf{k}}/2k_BT\right)+\tanh\left(\varepsilon_{i'\mathbf{k}'}/2k_BT\right)}{\varepsilon_{i'\mathbf{k}}+\varepsilon_{i'\mathbf{k}'}}w^*_{i'\mathbf{k}}(\mathbf{r}')w^*_{i'\mathbf{k}'}(\mathbf{r}')w_{i'\mathbf{k}}(\mathbf{r})w_{i'\mathbf{k}'}(\mathbf{r}). \quad (17)$$

Thus from Eq. (16), we can write

$$\Delta_i(\mathbf{r}) = \bar{\Delta}_i(\mathbf{r})\exp(\mathrm{i}\mathbf{A}\cdot\mathbf{r}) \quad (18)$$

with $\bar{\Delta}_i(\mathbf{r})$ the superconducting gap function in the absence of $\mathbf{A}$. Then we have

$$\bar{\Delta}_i(\mathbf{r}) = \sum_{i'}\int \bar{K}_{ii'}(\mathbf{r},\mathbf{r}')\bar{\Delta}_{i'}(\mathbf{r}')\mathrm{d}\mathbf{r}'. \quad (19)$$

Now, we can examine the behavior of the superconducting gap functions near the superconductor-insulator interface. Following the procedure pioneered by de Gennes, we suppose that the gap functions close to the surface behaves as

$$\bar{\Delta}_i(s) = \bar{\Delta}_{i0} + \left(\sum_{i'}\frac{\xi_1}{b_{ii'}}\bar{\Delta}_{i'0}\right)s. \quad (20)$$

Here s measures the normal distance from the boundary in units of ξ_1 and $s > 0$ is defined in the superconductor. For simplicity, we set the cross section of the boundary as 1. $\bar{\Delta}_{i0}$ represents the gap function at the boundary and $b_{ii'}$ denotes the intraband or interband surface extrapolation length for the two-band superconductor. **From Eq. (20), we can establish the boundary condition between the two-band superconductor and the insulator (or vacuum) at $s = 0$**

$$\frac{\mathrm{d}\bar{\Delta}_i}{\mathrm{d}s} = \sum_{i'} \frac{\xi_1}{b_{ii'}} \bar{\Delta}_{i'} \tag{21}$$

in the absence of external magnetic field.

Meanwhile, with the explicit expressions of the kernels in the bulk system and the addition of nonlinear terms to the gap equations, we can obtain the two-band GL equations from Eq. (19) as [M.E. Zhitomirsky and V.H. Dao, Ginzburg-Landau theory of vortices in a multigap superconductor, Phys. Rev. B 69, 054508 (2004)]

$$-\alpha_1\bar{\Delta}_1 + \beta_1\left|\bar{\Delta}_1\right|^2\bar{\Delta}_1 - \gamma_1\nabla^2\bar{\Delta}_1 - R_{12}\bar{\Delta}_2 = 0 \tag{22}$$

and

$$-\alpha_2\bar{\Delta}_2 + \beta_2\left|\bar{\Delta}_2\right|^2\bar{\Delta}_2 - \gamma_2\nabla^2\bar{\Delta}_2 - R_{12}\bar{\Delta}_1 = 0, \tag{23}$$

with the GL parameters

$$\begin{gathered}\alpha_{1,2} = N_{1,2}\left[\frac{1}{\lambda_{\max}} - \frac{\lambda_{22,11}}{\lambda} + \ln\left(\frac{T_{c0}}{T}\right)\right], \quad \beta_i = \frac{7\zeta(3)N_i}{16\pi^2\left(k_B T_{c0}\right)^2}, \\ \gamma_i = \frac{7\zeta(3)\hbar^2 N_i v_{Fi}^2}{16\pi^2\left(k_B T_{c0}\right)^2} \quad \text{and} \quad R_{12} = \frac{N_1\lambda_{12}}{\lambda} = \frac{N_2\lambda_{21}}{\lambda}.\end{gathered} \tag{24}$$

Here $\lambda_{ii'} = g_{ii'}N_{i'}$ with $N_{i'}$ the density of states at the Fermi level for each band, $\lambda = \lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21}$ and $\lambda_{\max} = \frac{1}{2}\left[\left(\lambda_{11} + \lambda_{22}\right) + \sqrt{\left(\lambda_{11} - \lambda_{22}\right)^2 + 4\lambda_{12}\lambda_{21}}\right]$ the largest eigenvalue of λ-matrix. T_{c0} is the bulk critical temperature and v_{Fi} is the average Fermi velocity for each band.

In the spatially homogeneous case, we can neglect the gradient γ-terms. Eqs. (22) and (23) yield the gap equation at $T = T_{c0}$

$$\begin{pmatrix} \lambda_{11} & \lambda_{12} \\ \lambda_{21} & \lambda_{22} \end{pmatrix}\begin{pmatrix} \bar{\Delta}_1 \\ \bar{\Delta}_2 \end{pmatrix} = \lambda_{\max}\begin{pmatrix} \bar{\Delta}_1 \\ \bar{\Delta}_2 \end{pmatrix}, \tag{25}$$

which obviously gives the consistent result.

Now, we try to determine the coefficients $b_{ii'}$ in Eq. (21) by solving the linearized gap equation (19) in absence of external magnetic field. If we introduce $\bar{K}^0_{ii'}\left(s, s'\right)$ as the kernel of gap functions in the superconducting bulk system, we can transform Eq. (19) into

$$\bar{\Delta}_i(s)-\sum_{i'}\int \bar{K}^0_{ii'}(s,s')\bar{\Delta}_{i'}(s')\mathrm{d}s' = -\sum_{i'}\int\left[\bar{K}^0_{ii'}(s,s')-\bar{K}_{ii'}(s,s')\right]\bar{\Delta}_{i'}(s')\mathrm{d}s' \equiv -\sum_{i'}H_{ii'}(s)\ . \quad (26)$$

From Eqs. (22) and (23) with the higher order β-terms omitted, also noting that $\bar{K}^0_{ii'}(s,s')=\bar{K}^0_{ii'}(s-s')$ due to the translational symmetry, we can read out the Laplace transformation of $\bar{K}^0_{ii'}$ as

$$\bar{K}^0_{ii'}(p)=\frac{\lambda_{ii'}}{\lambda_{\max}}+\frac{\lambda_{ii'}\gamma_{i'}}{N_{i'}\xi_1^2}p^2. \quad (27)$$

Plugging Eq. (27) into Eq. (26), we can get

$$\bar{\Delta}_i(p)-\sum_{i'}\left(\frac{\lambda_{ii'}}{\lambda_{\max}}\right)\bar{\Delta}_{i'}(p)-\sum_{i'}\left(\frac{\lambda_{ii'}\gamma_{i'}}{N_{i'}\xi_1^2}\right)p^2\bar{\Delta}_{i'}(p)=-\sum_{i'}H_{ii'}(p). \quad (28)$$

Here $\bar{\Delta}_i(p)$ and $H_{ii'}(p)$ are the Laplace transformations of $\bar{\Delta}_i(s)$ and $H_{ii'}(s)$ respectively. Since the first two terms of the left-handed side in Eq. (28) can be cancelled out according to Eq. (25), we then have

$$\sum_{i'}\left(\frac{\lambda_{ii'}\gamma_{i'}}{N_{i'}\xi_1^2}\right)p^2\bar{\Delta}_{i'}(p)=\sum_{i'}H_{ii'}(p). \quad (29)$$

We can see that both sides in Eq. (29) take the main contribution from the boundary region.

Notice that the Laplace transformation of the gap function in Eq. (20) takes the form

$$\bar{\Delta}_i(p)=\frac{\bar{\Delta}_{i0}}{p}+\sum_{i'}\frac{\xi_1\bar{\Delta}_{i'0}}{b_{ii'}p^2}. \quad (30)$$

Then at $p\to 0$, we will obtain from Eq. (29)

$$\sum_{i'i''}\left(\frac{\lambda_{ii'}\gamma_{i'}}{N_{i'}\xi_1 b_{i'i''}}\right)\bar{\Delta}_{i''0}=\sum_{i'}H_{ii'}(p=0). \quad (31)$$

Parallel to de Gennes' analysis, we have the sum rules

$$\int \bar{K}^0_{ii'}(s,s')\mathrm{d}s'=\frac{\lambda_{ii'}}{\lambda_{\max}} \quad \text{and} \quad \int \bar{K}_{ii'}(s,s')\mathrm{d}s'=\frac{\lambda_{ii'}N_{i'}(s)}{\lambda_{\max}N_{i'}} \quad (32)$$

with $N_{i'}(s)$ the local density of states at the Fermi surface. Then, we can write the Laplace transformation of the kernel difference at $p\to 0$

$$H_{ii'}(p=0)=\int H_{ii'}(s)\mathrm{d}s=\frac{\lambda_{ii'}\bar{\Delta}_{i'0}}{\lambda_{\max}}\int\frac{\bar{\Delta}_{i'}(s)}{\bar{\Delta}_{i'0}}\left[1-\frac{N_{i'}(s)}{N_{i'}}\right]\mathrm{d}s. \quad (33)$$

Now we suppose $\bar{\Delta}_{i'}(s)/\bar{\Delta}_{i'0}$ approaches zero in the insulating region and is of the

order of 1 in the metallic region. $N_{i'}(s)/N_{i'}$ also passes from $0 \to 1$ in a few interatomic distances from the boundary. Therefore, the integrand in Eq. (33) is nonvanishing only in a width of order of the lattice constant *a*. We can then estimate $H_{ii'}(p=0)$ as

$$H_{ii'}(p=0) = \frac{\lambda_{ii'} a}{\lambda_{\max} \xi_1} \bar{\Delta}_{i'0}. \tag{34}$$

Comparing Eq. (31) with Eq. (34), we can finally obtain

$$\frac{1}{b_{ii}} = \frac{N_i a}{\gamma_i \lambda_{\max}} \quad \text{and} \quad \frac{1}{b_{12}} = \frac{1}{b_{21}} = 0. \tag{35}$$

At this stage, we would like to point out that $1/b_{ii'} = 0 \ (i \neq i')$ is only an approximation and will become nonzero in the higher-order calculation. **Even for a contact between a superconductor and an insulator, the Cooper pairs can still diffuse into the insulating region with some probability.** Algebraically, this means that the gap function $\bar{\Delta}_{i'}(s)$ will also extend into the $s<0$ region, and we can roughly estimate $\bar{\Delta}_{i'}(s) \sim \sum_{i''} T_{i'i''} \bar{\Delta}_{i''0} e^{\xi_1 s/a} \ (s<0)$ with $T_{i'i''}$ the element of the transmission matrix at the boundary. Including the $s<0$ part in the integration of Eq. (33) and noting $N_{i'}(s)/N_{i'} \approx 0$ in this region, we can get $H_{ii'}(p=0) = (\lambda_{ii'} a/\lambda_{\max}\xi_1)\left(\bar{\Delta}_{i'0} + \sum_{i''} T_{i'i''}\bar{\Delta}_{i''0}\right)$.

Plugging into Eq. (31), the coefficients of boundary terms are given by

$$\frac{1}{b_{ii}} = \frac{N_i a}{\gamma_i \lambda_{\max}}(1+T_{ii}), \quad \frac{1}{b_{12}} = \frac{N_1 a}{\gamma_1 \lambda_{\max}} T_{12} \quad \text{and} \quad \frac{1}{b_{21}} = \frac{N_2 a}{\gamma_2 \lambda_{\max}} T_{21}. \tag{36}$$

With the transmission coefficient from the superconductor to the insulator $T_{ii'} \ll 1$, we can obviously see that Eq. (35) is a good approximation.

For a typical two-band superconductor, we can estimate $\gamma_i \lambda_{\max}/N_i \sim \xi_1^2$ with $\xi_1 \sim 10^{-4}$ cm and the lattice constant $a \sim 10^{-8}$ cm, which will give $b_{ii} \sim 1$ cm. **Therefore for a boundary separating a two-band superconductor from an insulator we can set $\xi_1/b_{ii'} \approx 0$. This leads to the boundary condition $\mathrm{d}\bar{\Delta}_i/\mathrm{d}s = 0$ from Eq. (21). For an arbitrary superconducting domain and in the presence of the magnetic field, we can generalize this result to $(\nabla - \mathrm{i}\mathbf{A})\Delta_i \cdot \mathbf{n} = 0$ according to Eq. (18). With the**

phenomenological superconducting order parameter $\Psi_i \propto \Delta_i$, we can finally write down the boundary condition $(\nabla - \mathrm{i}\mathbf{A})\Psi_i \cdot \mathbf{n} = 0$ for the interface of two-band superconductor and insulator. Also see Appendix A "Zero electric potential gauge and boundary conditions" from page 15 to page 19 in the revised version of manuscript.

[Q3] The figures refer to "evolution" of physical quantities. This is misleading as it is unrelated to the "time evolution" used to generate the solutions. (Quoted from the report of Reviewer 3)

[A3] We thank the referee for pointing out this problem to us. We have replaced the misleading word "evolution" with "transition" in the revised manuscript, to reflect the transformation of the system from the perfect diamagnetism state to the vortex cluster phase, and ultimately to the vortex lattice phase. Also see the captions from Figure 4 on page 9 to Figure 11 on page 14 in the revised version of manuscript.