Hidden First Order Transition in Strongly Pauli Limited Multiband Superconductors — Application to CeCu$_2$Si$_2$ —

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Motivated by recent experiments on heavy fermion materials CeCu$_2$Si$_2$ and UBe$_{13}$, we develop a framework to capture generic properties of multiband superconductors with strong Pauli paramagnetic effect (PPE). In contrast to the single band case, the upper critical field $H_{c2}$ can remain second order transition even for strong PPE cases. The expected first order transition is hidden inside $H_{c2}$ and becomes a crossover due to the interplay of multibandness. The present theory based on full self-consistent solutions of the microscopic Eilenberger theory explains several mysterious anomalies associated with the crossover and the "empty" vortex core state which is observed by recent STM experiment on CeCu$_2$Si$_2$.

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There has been much attention focused on multiband superconductors that were triggered by discoveries of several typical such systems; MgB$_2$ and iron pnictides in recent years. This leads us to take a fresh look not only on new compounds but also on older systems from this multiband perspective. This new view is particularly fruitful for the oldest heavy fermion superconductors CeCu$_2$Si$_2$ and UBe$_{13}$, which are key driving materials of this heavy fermion community over 30 years [1, 2]. In fact these materials have been regarded as representative examples of unconventional order parameter. Low $T$ thermodynamics are apparently difficult to understand within the single band full gap picture and explained in terms of the nodal gap structure with some power law behaviors. However, it is known that there is no unique solution for the nodal gap structure so far because the power law for each thermodynamic quantity is internally conflicting. The recent studies on CeCu$_2$Si$_2$ [3] and UBe$_{13}$ [4] clearly show that they belong to multiband superconductors with full gaps, which better explains low $T$ thermodynamics than the unconventional nodal gap model within the single band does.

Pauli paramagnetically limited superconductors in the clean limit are characterized by the so-called Maki parameter $\alpha_M = \sqrt{2}H_{c2}^{orb}/H_P$, where $H_{c2}^{orb}$ is the orbital depairing upper critical field and $H_P = \Delta_0/\sqrt{2}\mu_B$ is the Pauli limited field with the order parameter amplitude $\Delta_0$ at $T = 0$ [5, 6]. If $\alpha_M \geq 1.0$, $H_{c2}$ becomes first order transition (FOT) from second order transition. Upon further increasing $\alpha_M$ above $\alpha_M \geq 1.8$ [5, 6], the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [8, 10] should appear in the $HT$ phase diagram. In the strong Pauli limiting $\alpha_M \to \infty$, the upper limit temperature of the FOT $T_{1st}$ along $H_{c2}$ is given by $T_{1st}/T_c \to 0.56$. These important limiting criteria [5, 6, 8] often fail badly in some of the heavy fermion superconductors. For example, CeCu$_2$Si$_2$ and UBe$_{13}$ exhibit a strong rise of $H_{c2}$ at $T_c$ (|$dH_{c2}/dT$| = 23, 34 T/K) while $H_{c2}(T = 0)$ is strongly suppressed (2 T, 9 T) respectively. These numbers give $\alpha_M = 3.0$ and 2.3 through a formula for the orbital $H_{c2}$ reduction by PPE given in Fig. 1 of Ref. [11], yet the two superconductors show neither FOT nor FFLO phase. Thus the single band picture is fundamentally violated.

Another interesting system is KFe$_2$As$_2$ [12, 16] that is known to consist of four bands with widely different gap values and that belongs to iron pnictides family. (Some of the iron pnictides reach to $\alpha_M \geq 6$, see Fig. 19 in Ref. [17]). According to recent studies, moderately large $\alpha_M = 1.7$–1.9 is assigned for KFe$_2$As$_2$ for $H \parallel ab$. The first order transition is indeed reported [15]. However, there exist several outstanding deviations from the single band picture, some of which are common with CeCu$_2$Si$_2$ [3] and UBe$_{13}$ [4, 18].

We summarize the anomalies from the single band picture in the following:

(1) The Sommerfeld coefficient $\gamma(H) = C/T$ in low $T$ shows a kink behavior at $H^*$ above which $\gamma(H)$ starts growing rapidly toward $H_{c2}$ for all three compounds [3, 4, 16]. Simultaneously at $H^*$ the magnetization curve $M(H)$ in low $T$ gives a minimum both for CeCu$_2$Si$_2$ and UBe$_{13}$.

(2) The $T$-dependence of $C/T$ under high fields shows an increasing behavior upon lowering $T$ for CeCu$_2$Si$_2$ [3] and KFe$_2$As$_2$ [16].

(3) The empty vortex core state is recently found in CeCu$_2$Si$_2$ by STM [19, 20] where the zero energy density of states (ZDOS) at the core site is suppressed.

The purposes of this Letter are to elucidate generic features of the multiband superconductors with strong Pauli limiting and to place a foundation to explore these intriguing phenomena. We are going to show that FOT is hidden by covering $H_{c2}^{orb}$ in the other band by referring...
to CeCu₂Si₂.

We consider a simplified model of a two-band system with a larger superconducting gap band (band-1) and a smaller gap band (band-2). For simplicity, the superconducting gap is assumed to open isotropically on each three-dimensional spherical Fermi surface.

The electronic state is calculated by the quasiclassical Eilenberger theory in the clean limit [21, 22], including the Pauli paramagnetic effect (PPE) due to the Zeeman term $\mu B(r)$ [23], where $B(r)$ is the flux density of the internal field and $\mu = \mu_0 B_0 / \pi k_B T_c$ is a renormalized Bohr magneton related to $\alpha_M = 1.76 \mu$. The quasiclassical Green’s functions $g_j \equiv g(k_j, r, \omega_n + i \mu B)$, $f_j \equiv f(k_j, r, \omega_n + i \mu B)$, and $\phi_j \equiv \phi(k_j, r, \omega_n + i \mu B)$ with band index $j$ depend on the direction of the Fermi momentum $k_j$ for each band, the center-of-mass coordinate $r$ for the Cooper pair, and Matsubara frequency $\omega_n = (2n + 1) \pi k_B T$ with $n \in \mathbb{Z}$. They are calculated in a unit cell of the triangle vortex lattice by solving the Eilenberger equation

$$\begin{align*}
\{\omega_n + i \mu B(r) + v_j \cdot [\nabla + i A(r)]\} f_j &= \Delta_j(r) g_j, \\
\{\omega_n + i \mu B(r) - v_j \cdot [\nabla - i A(r)]\} g_j &= \Delta_j^*(r) f_j,
\end{align*}$$

(1)

where $g_j = (1 - f_j \phi_j)^{1/2}$, $\text{Re}[g_j] > 0$, and $v_j = (v_{Fj}/v_{F0}) k_j$.

The unit of Fermi velocity $v_{F0}$ is defined by $N_{F0} v_{F0}^2 \equiv N_{F1} v_{F1}^2 + N_{F2} v_{F2}^2$, where the density of states (DOS) in the normal state at each Fermi surface is defined by $N_{F0} \equiv N_{F1} + N_{F2}$. Throughout this Letter, temperatures, energies, lengths, and magnetic fields are, respectively, measured in units of the transition temperature $T_c$, $\pi k_B T_c$, $\xi_0 = h v_{F0}/2 \pi k_B T_c$, and $B_0 = \phi_0/2 \pi \xi_0^2$ ($\phi_0$ is the flux quantum).

The gap value is self-consistently determined by

$$\Delta_j(r) = T \sum_{0 < \omega_n < \omega_C} \sum_{j', j = 1, 2} V_{jj'} N_{Fj'} \left(f_j + f_{j'}^*\right)_{k_j},$$

(2)

where $\langle \cdots \rangle_{k_j}$ indicates the Fermi surface average on each band. We use the energy cutoff $\omega_C = 20 \pi k_B T_c$. The vector potential is also self-consistently determined by

$$\nabla \times \nabla \times A = \nabla \times M_{\text{para}} - \frac{T}{\kappa^2} \sum_{|\omega_n| \leq \omega_C} \sum_{j = 1, 2} N_{Fj} \langle v_j \text{Im}[g_j] \rangle_{k_j},$$

(3)

which includes the contribution of the paramagnetic moment $M_{\text{para}} = \langle 0, 0, M_{\text{para}} \rangle$ with

$$M_{\text{para}} = M_0 \left( \frac{B(r)}{B} - \frac{T}{\mu B} \sum_{|\omega_n| < \omega_C} \sum_{j = 1, 2} N_{Fj} \langle \text{Im}[g_j] \rangle_{k_j} \right).$$

(4)

The normal state paramagnetic moment $M_0 = (\mu/k)^2 \tilde{B}$ and $\tilde{k} \equiv B_0/(\pi k_B T_c \sqrt{8 \pi N_{F0}}) = [7 \zeta(3)/18]^{1/2} \kappa_{GL}$ with a large Ginzburg-Landau parameter $\kappa_{GL} = 89$. Using Doria-Gubernatis-Rainer scaling [24], we obtain the relation of $B$ and the external field $H$ [23]. Then, the total magnetization $M_{\text{total}} = B - H$ including both the diamagnetic and the paramagnetic contributions is derived.

When we calculate the electronic state, we solve Eq. (1) with $\omega_n \to E + i \eta$. The local density of states (LDOS) is given by $N_j(r, E) = N_{j, \uparrow}(r, E) + N_{j, \downarrow}(r, E)$, where

$$N_{j, \sigma}(r, E) = N_{Fj} \left( \text{Re} \left[ g(k_j, r, \omega_n + i \mu B) \right] |_{\text{Im}[E + i \eta]} \right),$$

(5)

with $\sigma = 1$ (−1) for up (down) spin component. We typically use the smearing factor $\eta = 0.01$. The DOS is obtained by the spatial average of the LDOS as $N(E) = \sum_j N_j(r, E) = \sum_j (\text{Im}[N_{j, \uparrow}(r, E) + N_{j, \downarrow}(r, E)])$.

We set the DOS in the normal state at each Fermi surface to $N_{F1} = \frac{4}{3} N_{F0}$ and $N_{F2} = \frac{1}{3} N_{F0}$. We assume that Cooper pair transfer $V_{12} = V_{21}$ is small. Then, we set the pairing interaction to $V_{22} = 1.5 V_{11}$ and $V_{12} = V_{21} = 0.05 V_{11}$ so that $\Delta_j/\Delta_2 \sim 2$ at zero field. These two parameters, namely the normal DOS and gap ratios are consistent with the fitting parameters of the specific heat for CeCu₂Si₂ by the two-gap model [3]. For the Fermi velocity on each band, we choose $v_{F1} = 4 v_{F2}$ so that $H_{c2}^{\text{orb}(2)} / H_{c2}^{\text{orb}(1)} \sim 4$, where $H_{c2}^{\text{orb}(j)} \propto \xi_j^{-2} \sim (\Delta_j/h v_{Fj})^2$. When the two bands are independent ($V_{12} = V_{21} = 0$), the orbital limits $H_{c2}^{\text{orb}(j)}$ with $j = 1, 2$ are shown schematically by dashed curves in Fig. 1(a), provided that the transition temperatures are equal which is realized by even slight inter-band interaction. We design that the slope of the band-2 at $T_c$ is steeper than that of the band-1, intending to simulate the sharp rise of $H_{c2}$ at $T_c$ as observed in CeCu₂Si₂.

In Fig. 2, we show spatial averaged physical quantities which are probed by thermodynamics. First, we start off with interacting two band superconductivity without PPE. As shown in Fig. 2(a-1) the two order parameters...
$\Delta_1$ and $\Delta_2$ are coupled and vanish at the same $H_{c2}$ when $V_{12} = V_{21}$ is finite. Figure 2(a-2) shows the field dependence of the zero energy ZDOS, $N(E=0)$. At lower fields $N(E=0)$ grows linearly by $B$, which is characteristic to full gap superconductors. The linear slopes of $N(E=0)$ for band-1 and band-2 are different reflecting orbital limits $H_{c2}^{\text{orb}(1)} \approx 0.5$ and $H_{c2}^{\text{orb}(2)} \approx 1.3$. Note that the ratio of the orbital limits changes from the setting parameter $H_{c2}^{\text{orb}(2)}/H_{c2}^{\text{orb}(1)} \approx 4$ owing to the inter-band interaction. The magnetization curve $M(B) = M_{\text{total}} - M_0$ is shown in Fig. 2(a-3) which is not much different from the usual $M(B)$ curve expected for single band systems. Those results confirm the naively expected behaviors for two band superconductors.

Let us now switch on the PPE. Before going into the numerical results, we explain an intuitive physical picture. Since $H_{c2}^{(1)} > H_{c2}^{(2)}$, because of the Pauli limited field $H_{c2}^{(j)} \propto \Delta_j$ for the band-$j$, it is expected that the $H_{c2}$ curve for the band-2 is suppressed much larger than that for the band-1 as schematically shown in Fig. 1(a). The actual $H_{c2}$ for the two band system is realized by $H_{c2}^{(1)}$ because $H_{c2}^{(2)}$ is less than $H_{c2}^{(1)}$ in low temperatures. Thus, the $H_{c2}^{(2)}$ curve originally characterized by FOT because of $H_{c2}^{(2)} < H_{c2}^{\text{orb}(2)}$ is covered by $H_{c2}^{(1)}$ from above and now hidden under it. This expectation is indeed confirmed by our calculation shown in Figs. 2(b) ($\mu = 1$) and 2(c) ($\mu = 3$).

In the $\mu = 1$ ($\alpha_M = 1.76$) case the resulting $H_{c2}$ is characterized by second order transition. However, inside $H_{c2}$ there exists a crossover field $H^*$ that corresponds to a kink of $\Delta_2$ upon increasing $B$ (Fig. 2(b-1)) and a maximum of ZDOS $N_2(E=0)$ (Fig. 2(b-2)). At $H^*$, $N_2(E=0)$ exceeds the corresponding normal state value, that is, $N_2(E=0) > N_{F2}$ whose origin will be explained later. Because this enhanced DOS recovers to $N_{F2}$ toward $H_{c2}$, the total DOS $N(E=0)$ is also enhanced at $H^*$ as seen from Fig. 2(b-2). This feature is indeed observed in CeCu$_2$Si$_2$ (see Fig. 2 in Ref. 2 where $C_1/T$ data at $T=0.06$ K show an enhancement just below $H_{c2}$). The magnetization curve shown in Fig. 2(b-3) exhibits a concave curvature near $H_{c2}$ which is characteristic to the PPE.

It is seen that $H_{c2}$ is largely suppressed because of the PPE. Judging by the suppressed $H_{c2} \approx 0.3$ from $H_{c2}^{\text{orb}} \approx 1.0$, the effective value of the $\mu$ ($\alpha_M$) amounts to $\mu_{\text{eff}} \approx 2.1$ ($\alpha_{\text{eff}} \approx 3.6$) that is enhanced from the setting $\mu = 1$. The large disparity is due to the difference between gaps on each band, which are $\Delta_1 = 0.58$ and $\Delta_2 = 0.29$ at $B = 0$ and $T = 0.2T_c$. The transition temperature $T_c$ proportional to a gap in the BCS theory is subject to the main band-1. Since the unit of energy is $\pi k_B T_c$, the effective Zeeman energy for band-2 is twice owing to half a gap for band-1. The resulting effective value $\mu_{\text{eff}} \approx 2.1$ implies that the Pauli limited field $H_{c2}^{(2)}$ strongly influences $H_{c2}$ although the FOT is hidden.

Upon further large $\mu = 3$, $H_{c2}$ becomes eventually FOT from second order transition in low $T$ because $H_{c2}^{(1)} < H_{c2}^{\text{orb}(1)}$. As seen from Fig. 2(c-1) in addition to the hidden FOT at $H^*$, also FOT is shown at $H_{c2} = 0.12$ where the two order parameters vanish suddenly. The ZDOS in Fig. 2(c-2) exhibits successively sharp variations at $H^*$ and $H_{c2}$. At low $B$ region the total $N(E=0)$ is strongly suppressed and suddenly grows just below $H^*$ as seen from Fig. 2(c-2). The magnetization curve in Fig. 2(c-3) also exhibits a minimum just below $H^*$, which is caused by the competition between the orbital diamagnetic negative contribution and the paramagnetic positive contribution due to the PPE. This minimum is observed both in CeCu$_2$Si$_2$ (see Fig. 4(b) in Ref. 3) and UBe$_{13}$ (see Fig. 2 in Ref. 15). Note that the minimum of the magnetization curve never occurs in the single band case [23].

The HT phase diagram is shown in Fig. 1(b) where the second order transition $H_{c2}$ and the hidden FOT $H^*$ are depicted for the $\mu = 1$ case. It is seen from Fig. 1(b) that $H^*$ terminates at a finite $B$ hit on the $H_{c2}$ curve because the PPE becomes effective at a finite $B$. The hidden FOT may be contrasted with the “hidden criticality” examined by Komendova et al. [27] who find a similar critical behavior in two band superconductors along the $T$-axis at $B = 0$. We note that the hidden FOT line $H^*$ relative to $H_{c2}$ shown in Fig. 1(b) looks very similar to $H_{\text{Max}}^{*}$ observed in UBe$_{13}$ [18].

The spatial averaged DOS $N(E)$ calculated for various $B$ in the $\mu = 1$ case is shown in Fig. 3(a). At $B = 0.002$ the gap due to $\Delta_1 = 0.58$ and $\Delta_2 = 0.29$ widely opens and $N(E)$ exhibits sharp edge singularities at $\Delta_2$. Upon increasing $B$ from $B = 0.002$ toward $H_{c2} \approx 0.3$ it is seen that

**FIG. 2:** (Color online) Each column is a series of results for $\mu = 0$ (a), $\mu = 1$ (b), and $\mu = 3$ (c) at $T = 0.2T_c$. The first row: field dependence of the two order parameters $\Delta_1$ and $\Delta_2$. The second row: ZDOS $N_1$, $N_2$, and $N$. The third row: magnetization curve.
from temperatures with an appropriate scale transformation the vortex bound state has a peak exactly at $E = 0$. At $\tilde{B} = 0.002$, $\Delta_1 = 0.58$ and $\Delta_2 = 0.29$. By increasing $\tilde{B}$, the gap edge singularities corresponding to the minor gap $\Delta_2$ move inward to $E = 0$. At $\tilde{B} = 0.225$, total DOS has a maximum at $E = 0$. ZDOS increases gradually due to the depaired quasiparticles in the vortex core region \cite{23}. Finally at $\tilde{B} = 0.225$ below $H_{c2}$ corresponding to the hidden FOT $H^* = N(E)$ exhibits a maximum at $E = 0$ because the gap edge singularity of the minor band gap $\Delta_2$ is shifted to $E = 0$ by the PPE. The Zeeman shifted DOS for up spin component in band-2 is shown in Fig. 3(b) whose energy is shifted by $\mu B$. Note that the DOS for the down spin component has a relation $N_{j,+}(E) = N_{j,-}(E)$. The depaired quasiparticles in vortex cores accumulate inside the gap and the smeared gap edge singularity approaches toward the Fermi level at $E = 0$ according to the magnitude of magnetic fields. The Zeeman shifted gap edge singularity at $E = \Delta_2 - \mu H^* = 0$ explains also the enhanced $N_2(E = 0)$ behaviors at $H^*$ shown in Fig. 2(b-2).

Low energy DOS can be observed as $C(T)/T$ in low temperatures with an appropriate scale transformation from $k_BT$ to $E$ as demonstrated in Supplemental Material \cite{24}. In CeCu$_2$Si$_2$, the observed electronic specific heat $C_v/T$ increases toward low temperature under high fields (Fig. 1 in Ref. \cite{3}), which will reflect the enhancement of the DOS toward $E = 0$ at $H^*$.

So far we have mainly discussed the averaged physical quantities. Now we touch upon the local ones that are also important to characterize the hidden FOT associated with the multiband Pauli limited superconductors. In Fig. 4 we show the local ZDOS around the vortex core, each corresponding to major band-1 (Fig. 4(a)) and minor band-2 (Fig. 4(b)). The ring shaped “crater” like landscapes are clearly seen, which are quite different from that of the ordinary cases without the PPE where the single peaked mountain like landscape is seen \cite{28, 29}.

The physical origin of this structure of ZDOS can be understood as follows. At the ordinary vortex core site the vortex bound state has a peak exactly at $E = 0$. As moving away from the core this peak splits into two peaks (see Fig. 9 in Ref. \cite{28}), which are eventually absorbed and merged into the continuum above the gap edges. In the cases with the PPE, the Zeeman split two peaks at the core site evolve into two peaks each when moving away. These inward two peaks intersect somewhere away from the core. Thus these particular sites situated circularly give rise to the peak, resulting in a ring structure in the ZDOS landscapes. This peak position $r_{\max}$ is roughly estimated by $r_{\max}/\xi_0 \sim E_B/\Delta_0$ with the Zeeman shift $E_B$ from $E = 0$.

The empty core with the crater like landscape is actually observed by a recent STM experiment on CeCu$_2$Si$_2$ \cite{14, 20}. Observed $r_{\max}/\xi_0 \sim 0.5$ by the experiment at $H = 1.6$ T implies $E_B/\Delta_0 \sim 0.5$. The Zeeman energy can be directly checked by future STM experiment at the core site where we expect that the split two peaks are observed at the half energy of the gap.

In summary, we have constructed a general framework to describe the Pauli paramagnetic effect (PPE) for multiband superconductors within microscopic Eilenberger theory applicable to most type II superconductors. The present theory yields a better and advanced understanding for a superconductor with PPE than those firmly established frameworks \cite{3, 4, 8} based on the single band assumption. We applied it to CeCu$_2$Si$_2$, which is recently found to be multiband superconductor with strong PPE. We showed the hidden FOT as a crossover at $H^*$ deep inside $H_{c2}$ in the HT plane, as evidenced by an increase of low temperature specific heat and a minimum of the magnetization curve. Those features also observed in UBe$_{13}$ and KFe$_2$As$_2$. The empty core vortex is now emerging in the recent STM in CeCu$_2$Si$_2$ \cite{14, 20}. Our picture explains why FOT is avoided along $H_{c2}$ for large $\alpha_M$ superconductors, and furthermore may give a hint why the FFLO is difficult to realize in general.

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Supplementary Material

S1. Transformation from $C(T)/T$ to $N(E)$

Specific heat is given by a temperature derivative of entropy as

$$C = T \left( \frac{dS}{dT} \right) = \sum_k E_k \frac{\partial f_k}{\partial T}$$
$$= \int_{-\infty}^{\infty} \frac{E_k e^{\beta E} e^{\beta E}}{(e^{\beta E} + 1)^2} N(E) dE,$$ (S.1)

where $f_k = (e^{\beta E_k} + 1)^{-1}$ is the Fermi distribution function with $\beta = 1/k_B T$. We transform a variable of the integral from $E$ to $k_B Tx$; then, the specific heat is described by

$$\frac{C}{T} = k_B^2 \int_{-\infty}^{\infty} \frac{x^2 e^x}{(e^x + 1)^2} N(k_B Tx) dx.$$ (S.2)

If low energy DOS can be expanded to $N(E) = N(E = 0) + A|E|^\alpha$ for $\alpha > 0$, the specific heat in low temperature is expanded to

$$\frac{C}{T} = k_B^2 \int_{-\infty}^{\infty} \frac{x^2 e^x}{(e^x + 1)^2} [N(0) + A|k_B Tx|^{\alpha}] dx$$
$$= k_B^2 N(0) \int_{-\infty}^{\infty} \frac{x^2 e^x}{(e^x + 1)^2} dx + k_B^2 A|k_B T|^{\alpha} \int_{-\infty}^{\infty} \frac{|x|^\alpha x^2 e^x}{(e^x + 1)^2} dx$$
$$= B_0 k_B^2 N(0) + B_0 A|k_B T|^{\alpha}$$
$$= B_0 k_B^2 \left[ N(0) + \frac{B_\alpha}{B_0} A|k_B T|^{\alpha} \right]$$
$$= B_0 k_B^2 N(k_B T'),$$ (S.3)

where $T' = \left( \frac{B_\alpha}{B_0} \right)^{1/\alpha} T$ and

$$B_\alpha = 2 \int_0^{\infty} \frac{x^{\alpha+2} e^x}{(e^x + 1)^2} dx$$
$$= (2 - 2^{-\alpha})(\alpha + 2)\Gamma(\alpha + 2)\zeta(\alpha + 2),$$

particularly, $B_0 = \pi^2/3$, $B_1 = 9\zeta(3)$, and $B_2 = 7\pi^4/15$. Therefore, $C/T$ in low temperature gives low energy DOS after a scale transformation from $k_B T$ to $E$. 