## FLUXOID QUANTIZATION AND PHASE TRANSITION

IN HOLLOW SUPERCONDUCTORS CARRYING TRANSPORT CURRENT*

Mario Rabinowitz and Edward L. Garwin Stanford Linear Accelerator Center Stanford University, Stanford, California 94305


#### Abstract

A theory is presented to account for all the experimental observations of fluxoid quantization and phase transition in superconducting cylinders, without invoking the large, unlikely, misalignment between field and cylinder required in previous theory. Correct values are obtained for the ratio of periodic to background quadratic coefficients in the resistance vs. field plot, and for the $0^{\circ} \mathrm{K}$ penetration depth. The new theory predicts for the first time actual penetration depth and superconducting area.


From the earliest observation of periodicity in the transition temperature in units of the flux quantum $h / 2 e$ by Little and Parks $^{1}$ ( $\mathrm{L}-\mathrm{P}$ ) through to the most recent by Meyers and Meservey ${ }^{2}$ (M-M), a quadratic background has been observed upon which the periodicity lies. Tinkham ${ }^{3}$ derived an expression to explain the background seen by L-P. Yet, as he pointed out, unless their hollow cylinder were misaligned with the magnetic field, H , by as much as $9^{\circ}$, his theory differs by a factor of 100 from their result, and "a physical misalignment of this magnitude is unlikely, ...". P-L ${ }^{4}$ observed "the nonperiodic quadratic background which appeared in all of the samples and which varied in magnitude depending upon the diameter of the cylindrical sample, the wall thickness, and the orientation of the sample in the magnetic field." Yet Tinkham's equation predicts no dependence on the cylinder diameter for an aligned cylinder.

On the other side of the coin, the recent experiments of $\mathrm{M}-\mathrm{M}^{2}$ show excellent agreement with Tinkham's theory. Therefore, the theory developed here will attempt to explain these two apparently disparate sets of results. Our effort here is in no way meant to lessen the value of Tinkham's basic analysis underlying the specifics of his theory, as we follow the same basic approach.

[^0]Consider a thin superconducting cylindrical shell of length $D$, thickness $d$, and mean radius $R>d$, in an axial magnotic field $\overrightarrow{I I}$. The penetration depih $\lambda>d$, so the field is approximately uniform across the wall. Since the wave function of the superconducting elcctrons is single-valued, we may apply the BohrSommerfeld quantum condition to the electron pairs.

$$
\begin{equation*}
\int \overrightarrow{\mathrm{P}} \cdot \overrightarrow{\mathrm{~d} \ell}=q h \tag{I}
\end{equation*}
$$

where $q$ is the quantum integer.
The canonical momentum, $\vec{P}=2 m \vec{v}+2 e \vec{A}$, where $2 m$ is the mass, $2 e$ is the charge, and $v$ is the average center-of-mass velocity of the pairs. $\vec{A}$ is the magnetic vector potential so that the magnetic flux density $\vec{B}=\vec{\nabla} \times \vec{A}=\mu_{0} \vec{H}$. Substituting into Eq. (I) and using Stokes's theorem, we have

$$
\begin{equation*}
\int 2 m \vec{v} \cdot d \vec{\ell}+\int_{S} \int(\vec{\nabla} \times 2 c \vec{A}) \cdot d \vec{S}=q h \tag{2}
\end{equation*}
$$

Equation (2) yields

$$
\begin{equation*}
v=\left(\frac{e}{2 \pi m R}\right)\left(\frac{q h}{2 e}-\pi R^{2} B\right) \tag{3}
\end{equation*}
$$

The kinetic energy density associated with trapped flux quanta is

$$
\begin{align*}
E & =\frac{1}{2}\left(\frac{1}{2} n\right)(2 m) v^{2}=\frac{1}{2} n m\left(\frac{e}{2 \pi m R}\right)^{2}\left(q \phi-\pi R^{2} B\right)^{2} \\
& =\frac{1}{2}\left(\frac{n e^{2}}{m}\right)\left(\frac{1}{2 \pi R}\right)^{2}\left(q \phi-\pi R^{2} B\right)^{2}=\frac{1}{2 \Lambda}\left(\frac{1}{2 \pi R}\right)^{2}\left(q \phi-\pi R^{2} B\right)^{2} \tag{4}
\end{align*}
$$

where $\Lambda=m / n e^{2}=\mu_{0} \lambda^{2}$, $n$ is the number density of superconducting electrons, and $\dot{\phi}=h / 2 e$ is the flux quantum for pairs.

As pointed out by Tinkham, ${ }^{3}$ near $T_{c}$ "due to inhomogeneity, one is dealing with isolated threads girdling the flux." However, he does not pursue this aspect of the theory near $T_{c}$ in terms of the kinetic energy of the shielding currents of these filaments. It is likely that the regions of the grain boundaries become normal first, leaving superconducting regions of thickness $d$ and average width $w$, each being a singly-connected surface with circulating current density ' $J_{c}$, as illustrated in Fig. 1. Parks and Littie ${ }^{4}$ pointed out that even with superconducting and normal regions, pairs can still traverse the cylinder circumference, and $E$ is preserved. The London equation determines $J_{c}$,

$$
\begin{equation*}
\int \Lambda \vec{J}_{\mathrm{c}} \cdot \overrightarrow{\mathrm{~d} l}=-\iint \overrightarrow{\mathrm{B}} \cdot \overrightarrow{\mathrm{~d}} \overrightarrow{\mathrm{~S}}, \tag{5}
\end{equation*}
$$

giving the approximate solution

$$
\begin{equation*}
J_{c} \doteq \frac{-1}{2 \Lambda}\left(\frac{d w}{d+w}\right) B \tag{6}
\end{equation*}
$$

Since $J_{c}=\left(\frac{1}{2} n\right)(2 e) v_{c}$, the shielding contribution to the kinetic energy density is

$$
\begin{equation*}
E_{c}=\frac{1}{2} A J_{c}^{2}=\frac{1}{8 \Lambda}\left(\frac{d w}{d \div W}\right)^{2} B^{2} \tag{7}
\end{equation*}
$$

A transport current, $I_{p}$, parallel to the axis, is impressed on the cylinder (as is done experimentally to measure the resistance change of the cylinder as His varied) by means of leads at each end. These electrons have total velocity, $v_{t}$, with a component of velocity $\mathrm{v}_{\mathrm{s}}$, due to H , around the cylinder in addition to the component of velocity $v_{p}$ they have parallel to the cylinder's axis. The effect of the transport current has been neglected in the previous analyses. ${ }^{1-6}$ Consider. the power supply and normal leads to be equivalent to a superconducting wire attached to the ends of the cylindex in a plane parallel to $\vec{B}$. In this case, as before, Eq. (1) holds. ${ }^{7}$

Before proceeding, we point out that in all the experiments to date, $1,2,4,5$ connection has been made to the cylindex by means of the thin film on the plate unon which the cylinder rests, i.e., at the bottom edge of the cylinder. Thus for smali $B$, the spiralling electrons will not necossarily be collected in their first traversal of the cylinder, as they may reach the cylinder end at a point distant from the anode. In this case, we assume they will be specularly reflected at each end, and that for collection there are only discrete angles $\sigma=t n^{-1} \frac{v_{s}}{v_{p}}$ as long as $\sigma$ is small. Consideration that an electron must make an odd number of traversals for collection leads to the condition

$$
\begin{equation*}
\operatorname{tn} \sigma=\frac{2 \pi \mathrm{Rk}}{\mathrm{D}(2 \mathrm{i}+1)} \tag{8}
\end{equation*}
$$

where k and $\mathrm{i}=1,2,3, \ldots \mathrm{k}$ is the number of times the electron has gone around the cylinder, and 2 i is the number of reflections. When $\operatorname{tn} \sigma$ is large, one may relax the capture condition, as tunnelling, space charge effects, and small perturbations allow collection with an essentially continuously variable $\operatorname{tn} \sigma=\frac{v_{S}}{v_{p}}$. For small $\sigma$ (essentially small $B$ for the experiments performed):

$$
\begin{align*}
& \int \overrightarrow{\mathrm{P}} \cdot \overrightarrow{\mathrm{~d} \ell}=\int 2 \mathrm{~m} \overrightarrow{\mathrm{v}}_{\mathrm{t}} \cdot \overrightarrow{\mathrm{~d} \ell}+\int_{\mathrm{S}}(2 \mathrm{eB} \cdot \mathrm{~d} S=\mathrm{qh} .  \tag{9}\\
& \int 2 \mathrm{~m} \overrightarrow{\mathrm{v}}_{\mathrm{t}} \cdot \overrightarrow{\mathrm{dl}}=2 \mathrm{~m}\left(\mathrm{v}_{\mathrm{s}}^{2}+\mathrm{v}_{\mathrm{p}}^{2}\right)^{1 / 2}\left[(2 i+1)^{2} \mathrm{D}^{2}+(2 \pi \mathrm{Rk})^{2}\right]^{1 / 2} \tag{10}
\end{align*}
$$

where the path of integration is chosen to lie deep within the connecting wire so that $v_{t}=0$ there.

$$
\begin{equation*}
\iint_{S} 2 \mathrm{e} \overrightarrow{\mathrm{~B}} \cdot \mathrm{~d} \stackrel{\rightharpoonup}{\mathrm{~S}}=2 \mathrm{eBk} \pi \mathrm{R}^{2} \tag{11}
\end{equation*}
$$

Combining Eq. (9), (10), and (11), we obtain the solution for the kinetic energy
density associated with transport current, $E_{t 1}=\frac{1}{2} n m\left(v_{s}^{2}+v_{p}^{2}\right)$,

$$
\begin{equation*}
E_{t 1}=(2 \Lambda)^{-1}\left[(2 i+1)^{2} D^{2}+(2 \pi R k)^{2}\right]^{-1}\left(q \phi-k \pi R^{2} B\right)^{2} \tag{12}
\end{equation*}
$$

for small $\sigma$ (equivalently small $B$ ).
Now let us consider large $\sigma$ (large B).

$$
\begin{align*}
& \int 2 m \vec{v}_{t} \cdot \overrightarrow{\mathrm{~d}}=2 m v_{t}(2 i+1) \mathrm{D}\left(v_{t} / v_{p}\right)=2 m\left(v_{s}^{2}+v_{p}^{2}\right)(2 i+1) \mathrm{D} / \mathrm{v}_{\mathrm{p}}  \tag{13}\\
& \iint_{\mathrm{S}} 2 \mathrm{e} \overrightarrow{\mathrm{~B}} \cdot \overrightarrow{\mathrm{dS}}=\operatorname{eB}(2 i+1) D R v_{s} / v_{p} \tag{14}
\end{align*}
$$

Combining Eq. (9), (13), and (14), we find

$$
\begin{equation*}
\mathrm{v}_{\mathrm{s}}=\left\{-\mathrm{eRLB} \pm\left[(\mathrm{eRLB})^{2}-8 m L \mathrm{v}_{\mathrm{p}}\left(2 \mathrm{~mL} \mathrm{v}_{\mathrm{p}}-\mathrm{q}_{1}\right)\right]^{1 / 2}\right\} /(4 \mathrm{~mL}) \tag{15}
\end{equation*}
$$

where $I=(2 i+1)$. Thus, in this case, the kinetic energy density is

$$
\begin{equation*}
E_{t 2}=\frac{R^{2} B^{2}}{16 \Lambda}+\frac{J_{p}}{2 L}\left(q \phi-\Lambda L J_{p}\right)+\frac{1}{2}\left[\left(\frac{R^{2} B^{2}}{8 \Lambda}\right)^{2}+\frac{R^{2} B^{2} J_{p}}{4 \Lambda L}\left(q \phi-\Lambda L J_{p}\right)\right]^{\frac{1}{2}}+\frac{1}{2} \Lambda J_{p}^{2} \tag{16}
\end{equation*}
$$

where $J_{p}=n e v_{p}=I_{p} /(2 \pi R d)$.

Thus the total kinetic energy donsity of superconducting electrons in the cylinder is

$$
\begin{equation*}
E_{T u}=E+E_{c} \div E_{t u} \tag{17}
\end{equation*}
$$

where $u=1$ for small $\sigma$, and $u=2$ for large $\sigma$.
For small $\sigma$, we have, on substituting the order parameter $\omega=\frac{\Lambda_{0}}{\Lambda}$,

$$
\begin{align*}
E_{T I}= & \left(2 \Lambda_{0}\right)^{-1}(2 \pi R)^{-2}\left(q \phi-\pi R^{2} B\right)^{2} \omega+\left(8 \Lambda_{0}\right)^{-1}\left(\frac{d W}{d+W}\right)^{2} B^{2} \omega+ \\
& +\left(2 \Lambda_{0}\right)^{-1}\left[L^{2}+(2 \pi R k)^{2}\right]^{-1}\left(q \phi-k \pi R^{2} B\right)^{2} \omega \tag{18}
\end{align*}
$$

This must be added to the Ginzburg-Landau total free energy difference, thus

$$
\begin{equation*}
\Delta G(\omega, T)=-a(T) \omega+\frac{1}{2} b(T) \omega^{2}+E_{T 1}(\omega) \tag{19}
\end{equation*}
$$

where the spatial variation of the order parameter $|\nabla \omega|^{2}$ is negligible due to the thinness of the cylinder.

$$
\begin{equation*}
\mathrm{a}(\mathrm{~T})=\mu_{0} \mathrm{H}(\mathrm{~T})^{2}\left[\lambda(\mathrm{~T}) / \lambda_{0}\right]^{2}=\mu_{0} \mathrm{H}_{0}^{2}\left(1-t^{2}\right)^{2}\left(1-t^{4}\right)^{-1}=\mu_{0} \mathrm{H}_{0}^{2}\left(1-\mathrm{t}^{2}\right)\left(1+t^{2}\right)^{-1} \tag{20}
\end{equation*}
$$

where $\mathrm{H}_{0}$ is the thermodynamic critical field at $0^{\circ} \mathrm{K}, \quad \lambda$ is the equilibrium penetration depth, and the reduced temperature $t=T / T$.

$$
\begin{equation*}
\mathrm{b}(\mathrm{~T})=\mathrm{a}(\mathrm{~T})\left[\lambda(\mathrm{T}) / \lambda_{0}\right]^{2} \tag{21}
\end{equation*}
$$

We want $\frac{\partial \Delta G}{\partial \omega}=0$ to find the minimum free energy difference, and set $\omega=0$ for the transition condition of no superconducting electrons. Thus

$$
\begin{equation*}
0=-\mu_{0} H_{0}^{2}\left(1-t^{2}\right)\left(1+t^{2}\right)^{-1}+\frac{\Lambda}{\Lambda_{0}} E+\frac{\Lambda}{\Lambda_{0}} E_{c}+\frac{\Lambda}{\Lambda_{0}} E_{t 1} \tag{22}
\end{equation*}
$$

Expanding to first order in $\Delta t=\left[T_{c}-T_{c}(I)\right] / T_{c} \ll 1$, we have

$$
\begin{equation*}
\frac{T_{c}-T_{c}(H)}{T_{c}}=\frac{1}{8 \lambda_{0}^{2} \mu_{0}^{2} H_{0}^{2}}\left[\frac{\left(q \phi-\pi R^{2} B\right)^{2}}{\pi^{2} R^{2}} \div\left(\frac{\mathrm{a}}{\mathrm{Q}+\mathrm{W}}\right)^{2} \mathrm{~B}^{2}+\frac{4\left(q \phi-k \pi R^{2} B\right)^{2}}{\left[\mathrm{~L}^{2}+(2 \pi R \mathrm{k})^{2}\right]}\right] \tag{23}
\end{equation*}
$$

for small $\sigma$.
When k is small and/or $L$ is large, Eq. (23) yields the results of $M-\mathbb{M} .^{2}$ The other experimental resuits ${ }^{1,4,5}$ also come naturally from Eq. (23) without the necessity of invoking misaligmment. Let us look at the quadratic coefficients $\alpha_{p}$ and $\alpha_{p}$, and include the possibility of a misalignment angle $\theta$.

Periodic parabola: $(\Delta t)_{p, q=0}=\alpha_{p} B^{2}=R^{2} B^{2} \cos ^{2} \theta\left(8 \lambda_{0}^{2} \mu_{0}^{2} H_{0}^{2}\right)^{-1}$.

$$
\begin{equation*}
(\Delta t)_{b}=\alpha_{b} B^{2}=\left(8 \lambda_{0}^{2} \mu_{0}^{2} H_{0}^{2}\right)^{-1}\left[\left(\frac{d w}{d+w}\right)^{2} B^{2} \cos ^{2} \theta+\frac{4\left(\pi^{2}\right)^{2}\left(\frac{q}{x}-k\right)^{2} B^{2} \cos ^{2} \theta}{\left[I^{2}+(2 \pi R k)^{2}\right]}+4 R^{2} B^{2} \sin ^{2} \theta\right] \tag{25}
\end{equation*}
$$

where for small $\theta$ and $B$ the $\sin ^{2} \theta$ misalignment term enters in as for Tinkham, ${ }^{2}$ and $\mathrm{x}=\pi \mathrm{R}^{2} \mathrm{~B} / \phi$. Therefore

$$
\begin{equation*}
\frac{\alpha_{p}}{\alpha_{b}}=R^{2} \cos ^{2} \theta\left[\left(\frac{d w}{d+w}\right)^{2} \cos ^{2} \theta+\frac{4\left(\pi R^{2}\right)^{2}\left(\frac{a}{X}-k\right)^{2} \cos ^{2} \theta}{\left[L^{2}+(2 \pi R L)^{2}\right]}+4 R^{2} \sin ^{2} \theta\right]^{-1} \tag{25}
\end{equation*}
$$

Equation (26) is a simple quadratic in $k$, and can be solved exactly for $k$ for all the different experimental values of $\alpha_{p} / \alpha_{b}$, with $\theta$ either $=0$ or $\neq 0$ as the true case may be. The Sn data taken by $\mathrm{L}-\mathrm{P}^{1,4}$ is shown in Fig. 2 for reference.

Let us consider the $L-P^{1,4}$ data (a lower Iimit for $\frac{\alpha_{p}}{\alpha_{b}}$ ), and get an approximate solution for $k$ which reveals the inherent simplicity between the parameters. In their data, $\frac{\alpha_{p}}{\alpha_{b}} \sim 10$ for $\operatorname{Sn}$ and $\sim 25$ for In. Hence $R^{2} \gg \frac{\alpha_{p}}{\alpha_{b}}\left(\frac{d}{d+w}\right)^{2}$. We may also neglect the $\sin ^{2} \theta$ term if $R^{2} \cos ^{2} \theta \geq 10\left(4 \frac{\alpha}{\alpha_{b}} R^{2} \sin ^{2} \theta\right)$. Therefore, up to a $3^{\circ}$ misalignment, the $\sin ^{2} \theta$ term is negligible. Also for $x \geq 1, k \gg \frac{q}{x} \sim 1$. With these approximations, Eq. (26) reduces to

$$
\begin{equation*}
k \doteq(2 i+1) D(2 \pi R)^{-1}\left(\frac{\alpha p}{\alpha_{b}}-1\right)^{-1 / 2} \quad(\text { for } L-P) \tag{27}
\end{equation*}
$$

The upper limit of $\frac{\alpha_{p}}{\alpha_{b}}$ comes from the $N-M^{2}$ data in which $\theta=0$, and $\alpha_{p} / \alpha_{b} \doteq 3 R^{2} / d^{2}$ so that $\left(\alpha_{p} / \alpha_{b}\right)\left(\frac{d w}{d+w}\right)^{2}=\frac{3}{N} R^{2}$, where $N=d^{2}\left(\frac{d+w}{d w}\right)^{2}$, e.g., $N=4$ when $w=d$. Equation (26) gives

$$
\begin{equation*}
k \doteq(2 i+1) D(2 \pi R)^{-1}\left[\frac{N}{N-3}\left(\frac{\alpha_{p}}{\alpha_{b}}-1\right)\right]^{-1 / 2} \quad(\text { for } M-M, x \geq 1) \tag{28}
\end{equation*}
$$

Thus with the values of $k$ from Eq. (27) or (28), one may obtain values of $\lambda_{0}$ and $a_{p} / \alpha_{b}$ consistent with all the experiments, without invoking misaligmment.

Furthermore, values of $\lambda(T)$ and $T$ may be obtained from this theory and the experimiental data, which cannot be obtained from Tinkham's theory. ${ }^{2}$

From Eq. (8), we have

$$
\begin{equation*}
J_{p}=m v_{S}(2 i+1) D\left(\mu_{0} \lambda^{2} e 2 \pi R k\right)^{-1} \tag{29}
\end{equation*}
$$

Substituting for $\mathrm{v}_{\mathrm{S}}$ from Eq. (12), and solving for $\lambda$,

$$
\begin{equation*}
\lambda(T)=\left(\phi / \mu_{0}{ }_{p}\right)^{1 / 2}(\mathrm{kx}-\mathrm{q})^{1 / 2}\left[(2 \mathrm{i}+1)^{2} \mathrm{D}^{2}+2(2 \pi \mathrm{Rk})^{2}+\frac{(2 \pi \mathrm{Pk})^{4}}{(2 \mathrm{i}+1)^{2} \mathrm{D}^{2}}\right]^{-1 / 4} \tag{30}
\end{equation*}
$$

Equations (27) and (28) in (30) yield

$$
\begin{align*}
& \lambda(T) \doteq\left(\phi \times / 2 \pi R \mu_{0} J_{p}\right)^{1 / 2}\left[\frac{\alpha_{p}}{\alpha_{b}}+1\right]^{-1 / 4} \quad(\text { for } L-P), \text { and }  \tag{31}\\
& \left.\lambda(T) \doteq\left(\phi \times / 2 \pi R \mu_{0} J_{p}\right)^{1 / 2}\left[\left(\frac{N}{N-3}\right)^{\alpha_{p}} \frac{\alpha_{b}}{\alpha_{b}}\right]^{-1 / 4} \quad \text { (for } M-M I\right) . \tag{32}
\end{align*}
$$

. Now $\lambda(T)=\lambda_{0}\left(1-t^{4}\right)^{-1 / 2} \doteq \frac{1}{2} \lambda_{0}(1-t)^{-1 / 2}$ for $t \doteq 1$. (This qppears to fit the experimental data as well or better than the BCS expression.) Therefore

$$
\begin{equation*}
T \doteq T_{c}\left[I-\frac{1}{4}\left(\lambda_{0} / \lambda\right)^{2}\right] \tag{33}
\end{equation*}
$$

Values of $\lambda$ from Eq. (31) and (32), when subsututed into (33), give values of $T$ which agree well with experiment. ${ }^{1,2,4}$ Since the cylindrical shell may break up into superconducting and normal regions, $j_{p}$ may be $>I_{p} / 2 \pi R d$. When $T$ is given, then from Eq. (31), (32), and (33), one may calculate the true $J_{p}$ and hence the effective cross sectional area which remains superconducting.

It should now be clear that the transport current, $I_{p}$, plays a vital role in the experimental results, though it was totally neglected in the previous analyses, ${ }^{1-6}$ having been regarded mexcly as a means for sensing the phase transition. $I_{p}$ canses
a spiralling electron trajectory, a change in the path length of which affects $\mathrm{v}_{\mathrm{s}}$ and hence the quadratic background. For a given flux, the number of quantum mechanical wavelengths is conserved, but the electron total path length is a function of $I_{p}$, and therefore so is the electron wavelength, with a concomitant inverse change in electron momentum.

## References

1. W. A. Little and R. D. Parks, Phys. Rev. Letters 9, 9 (1962).
2. L. Meyers and R. Meservey, Phys. Rev. B 4, 824 (1971).
3. M. Tinkham, Phys. Rev. 129, 2413 (1963).
4. R. D. Parks and W. A. Little, Phys. Rev. 133, A97 (1964).
5. R. P. Groff and R. D. Parks, Phys. Rev. 176, 567 (1968).
6. D. H. Douglas, Jr., Phys. Rev. 132, 513 (1963).
7. If only the London Eq. (5) applies, this is equivalent to a 0 angular momentum state with $\mathrm{q}=0$.

## Figure captions

Fig. 1. Thin-walled superconducting cylindrical shell showing shielding current density $J_{c}$.

Fig. 2. Variation of resistance of tin cylinder with magnetic field at its transition temperature showing a periodic parabolic array superimposed upon a quadratic background (from L-P ${ }^{1,4}$ ).


PIG. 1,


FIG. 2


[^0]:    * Work supported by the U. S. Atomic Energy Commission.

