# Evaluation of Some Nonlocal Theories for a Thin Superconducting Film\*

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The magnetic susceptibility of a thin superconducting film has been evaluated as a function of film thickness on the basis of nonlocal theories proposed by Pippard, Bardeen, and Schafroth and Blatt. Two limiting boundary conditions corresponding to specular reflection and random scattering of the electrons at the surface have been applied, and the susceptibility appears to be insensitive to the boundary condition. Deviations from the London theory occur for a thickness less than twice the penetration depth and give a susceptibility smaller than the London value. Largest deviation is found for the Pippard theory and least for the Schafroth-Blatt version. The experimental data of Lock are inconclusive with regard to the validity of the nonlocal theories.

## 1. INTRODUCTION

 $R^{\rm ECENT}$  experimental and theoretical investigations  $^{1-5}$  have indicated that a generalization of the London phenomenological theory of the electromagnetic behavior of a superconductor should be carried out. On the basis of experimental studies of highfrequency surface impedance of superconductors, Pippard<sup>3</sup> has proposed a specific form of nonlocal theory which accounts for many of the observed effects. Using an energy gap model, Bardeen<sup>4</sup> has derived a theory for T=0 very similar to Pippard's proposal. Schafroth and Blatt<sup>5</sup> have suggested another form of nonlocal theory based on a theorem dealing with the behavior of the nonlocal kernel in momentum space. In order to determine the validity of the nonlocal theories, the magnetic susceptibility of a thin superconducting film has been calculated on the basis of these theories as a function of film thickness. It is well known that the predictions of the local theory of London agree well with the experimental results of Lock<sup>6</sup> on tin films; however, it was suggested by Ginsburg that a nonlocal theory would give a susceptibility considerably smaller than the London value for film thickness of the order of the penetration depth.

Lock's measurements were carried out for a fixed film thickness. By varying the temperature he was able to change penetration depth so as to cover a large range of effective thicknesses, since the quantity which enters the London theory is the ratio of film thickness to penetration depth. The nonlocal theories, however, contain another physical length whose temperature dependence is not well known and for this reason direct comparison with experiment is difficult. If one assumes that this new physical length varies with temperature in a manner similar to that of the penetration depth, then these calculations show that the resulting susceptibility is indeed smaller than the London value.

The calculations are carried out for two limiting boundary conditions corresponding to (1) specular reflection of the superelectrons at the surface and (2) random scattering at the surface. The first condition admits an exact solution as a rapidly convergent series while the second condition is handled by a variational technique. The results are relatively insensitive to the boundary condition.

### 2. TIME-INDEPENDENT SOLUTIONS OF MAXWELL'S EOUATIONS

We shall discuss only time-independent solutions of Maxwell's equations because a complete nonlocal electrodynamics for a superconductor has not been worked out at this time.

The relation of primary interest is that between the vector potential **A** and the supercurrent density **j**. The relations are given both in momentum space and configuration space for convenience. We choose to work in a gauge with  $\nabla \cdot \mathbf{A} = 0$ .

London:

$$\mathbf{j}(\mathbf{r}) = -\frac{1}{\Lambda c} \mathbf{A}(\mathbf{r}), \quad \mathbf{j}(\mathbf{k}) = -\frac{1}{\Lambda c} \mathbf{A}(\mathbf{k}), \quad (2.1)$$

Pippard:

$$\mathbf{j}(\mathbf{r}) = -\frac{3}{4\pi\xi_0\Lambda c} \int \frac{\mathbf{R}[\mathbf{R}\cdot\mathbf{A}(\mathbf{r}')]e^{-R/\xi}}{R^4} d\tau', \quad \mathbf{R} \equiv \mathbf{r} - \mathbf{r}',$$
$$\mathbf{j}(\mathbf{k}) = -\left[\frac{3\pi^2}{\xi_0\Lambda ck}\right] \frac{2}{\pi(\xi k)^2} \{[1 + (\xi k)^2] \tan^{-1}\xi k \qquad (2.2)$$
$$-\xi k\} \mathbf{A}(\mathbf{k}) \simeq -\frac{3\pi^2}{\xi_0\Lambda ck} \mathbf{A}(\mathbf{k}) \quad \text{for} \quad \xi k \gg 1.$$

Bardeen:

$$\mathbf{j}(\mathbf{r}) = -\frac{c}{4\pi} \int \mathbf{R} [\mathbf{R} \cdot \mathbf{A}(\mathbf{r}')] f_B(R) d\tau',$$

$$\mathbf{j}(\mathbf{k}) = \frac{-3}{4\Lambda c} \left(\frac{\Delta k}{k}\right) \ln \left(1 + \frac{k}{\Delta k}\right) \mathbf{A}(\mathbf{k}).$$
(2.3)

 <sup>\*</sup> Supported by the Office of Ordnance Research, U. S. Army.
 <sup>1</sup> A. B. Pippard, Proc. Roy. Soc. (London) A203, 98 (1950).
 <sup>2</sup> A. B. Pippard, Proc. Roy. Soc. (London) A203, 829 (1950).
 <sup>3</sup> A. B. Pippard, Proc. Roy. Soc. (London) A216, 547 (1953).
 <sup>4</sup> J. Bardeen, Phys. Rev. 97, 1724 (1955).
 <sup>5</sup> M. R. Schafroth and J. M. Blatt, Phys. Rev. 100, 1221 (1955).
 <sup>6</sup> L. M. L. J. Dure, Phys. Rev. 100, 1221 (1955).

<sup>&</sup>lt;sup>6</sup> J. M. Lock, Proc. Roy. Soc. (London) A208, 391 (1951).

Schafroth-Blatt:

$$\mathbf{j}(\mathbf{r}) = -\frac{c}{4\pi} \int \mathbf{R} [\mathbf{R} \cdot \mathbf{A}(\mathbf{r}')] f_{SB}(R) d\tau',$$

$$\mathbf{j}(\mathbf{k}) = -\frac{k}{\Lambda c(k+\mu)} \mathbf{A}(\mathbf{k}).$$
(2.4)

The functions f(R) are obtained from the scalar kernels in momentum space by

$$f(R) = -\frac{c}{32\pi^4} \int \left\{ \int^k k' K(k') dk' \right\} e^{i\mathbf{k}\cdot\mathbf{R}} d^3k, \quad (2.5)$$

where

$$-\frac{4\pi}{c}\mathbf{j}(\mathbf{k}) \equiv K(\mathbf{k})\mathbf{A}(\mathbf{k}).$$
(2.6)

The coherence length,  $\xi_0$ , in the Pippard theory is qualitatively related to the minimum size of the wave packets which can be made from a group of states near the Fermi surface with energy spread  $\sim kT_c$ . The quantity  $\xi$  is an effective coherence length which is determined in part by the electronic mean free path in the normal state. The parameter  $\mu^{-1}$  in the Schafroth-Blatt theory is of the order of Pippard's coherence length. The parameter  $\Delta k$  in the Bardeen theory is related to the energy gap model by  $\hbar^2 k_F \Delta k/m = \epsilon_0$ , where  $k_F$  is the magnitude of the wave vector at the Fermi surface and  $\epsilon_0$  is the width of the energy gap at T=0.

It should be noted that the Pippard theory reduces to the Bardeen theory for  $\xi k \gg 1$  except for a slowly varying logarithmic term. Also, the Schafroth-Blatt theory reduces to the London theory for  $k/\mu \gg 1$ .



FIG. 1. A superconducting film of thickness 2a in a uniform external field  $H_0$  with the supercurrent  $j_x$  shielding the interior.

### 3. MAGNETIC SUSCEPTIBILITY OF A THIN FILM

We consider an infinite superconducting film of thickness 2*a* with the normal to the surface in the *z* direction. A uniform external magnetic field  $H_0$  is applied parallel to the surface in the *y* direction. Diamagnetic supercurrents will be established along the *x* axis which shield the interior of the film as shown in Fig. 1. The magnetic susceptibility of the film is given by

$$\frac{\kappa}{\kappa_0} = \frac{1}{2a} \int_0^{2a} \frac{[H_0 - H_y(z)]}{H_0} dz = 1 - \frac{A_x(2a)}{aH_0}, \quad (3.1)$$

where  $\kappa_0 \equiv -1/4\pi$ , and we have used  $A_x(0) = -A_x(2a)$ , which can be chosen because of symmetry.

### (A) Specular Reflection

We shall solve for  $A_x(z)$  by introducing image current sheets placed periodically along the z axis, parallel to the film surface as shown in Fig. 2. Due to the anti-



FIG. 2. An array of image current sheets introduced to solve for the susceptibility for the specular reflection boundary condition.

symmetry of the solutions for reflection in the plane at z=2a, for example, one can see that, for each electron in region I approaching the surface with a component of velocity in the positive z direction, an electron in region II is approaching this surface with the same velocity parallel to the surface but the opposite z component of velocity. Hence, we may consider that these electrons exchange roles as they pass through the surface z=2a and thus an apparent specular reflection of the electron in region I has occurred. The magnitude of the image surface current is

$$\sigma = cH_0/2\pi. \tag{3.2}$$

Fourier-analyzing this array of image sources leads to the following expression for the source current density:

$$j_{x}{}^{\sigma}(z) = -\frac{cH_{0}}{2\pi a} \sum_{n=0}^{\infty} \cos k_{n} z, \qquad (3.3)$$

where  $k_n = (2n+1)\pi/a$ .

Introducing this source distribution into Maxwell's equation and using the momentum space relation between the supercurrents,  $j^s$ , and the vector potential, we obtain

$$k_{n}^{2}A_{x}(k_{n}) = -K(k_{n})A_{x}(k_{n}) - 2H_{0}/a$$

$$= \frac{4\pi}{c} [j_{x}^{s}(k_{n}) + j_{x}^{\sigma}(k_{n})], \quad (3.4)$$

and

$$A_{x}(z) = -\frac{2H_{0}}{a} \sum_{n=0}^{\infty} \frac{\cos k_{n} z}{k_{n}^{2} + K(k_{n})}.$$
 (3.5)

Combining (3.1) and (3.5) gives the susceptibility for an arbitrary nonlocal kernel,

$$\frac{\kappa}{\kappa_0} = 1 - \frac{2}{a^2} \sum_{n=0}^{\infty} \frac{1}{k_n^2 + K(k_n)} = \frac{2}{a^2} \sum_{n=0}^{\infty} \frac{K(k_n)}{k_n^4 + k_n^2 K(k_n)}.$$
 (3.6)

Figure 3 shows the ratio  $\kappa/\kappa_0$  plotted as a function of the half-thickness of the film divided by  $\lambda_0$ , the penetration depth at T=0, with

$$\xi_0/\lambda_L=6$$
,  $\Delta k\lambda_L=0.13$ ,  $\lambda_L/\lambda_0=0.7$ , and  $\mu\lambda_L=0.05$ ,

where  $\lambda_L$  is the London penetration depth. These values were computed from the work of Pippard and Faber,<sup>7</sup> Schafroth and Blatt,<sup>5</sup> and Bardeen.<sup>8</sup> Lock's experimental data on tin films are seen to be in agreement with the London theory as well as the Schafroth-Blatt theory. Because of the similarity of these theories for  $k/\mu \ll 1$ , it is not surprising that they give virtually the same susceptibility, since the most important fourier components for the penetration law satisfy this inequality.

The results of the Pippard and Bardeen theories fall below the experimental curve for  $a < 2\lambda_0$ , with the logarithmic term in the Bardeen kernel increasing the susceptibility for small a relative to the Pippard value.

The penetration law follows from (3.5) and is

$$\frac{H_y(z)}{H_0} = \frac{2}{a} \sum_{n=0}^{\infty} \frac{k_n \sin(k_n z)}{k_n^2 + K(k_n)}.$$
(3.7)

In the limit of an infinitely thick film, the penetration law is

$$\frac{H_y(z)}{H_0} = \frac{2}{\pi} \int_0^\infty \frac{k \sin(kz)}{k^2 + K(k)} dk, \qquad (3.8)$$

and the penetration depth is given by

$$\lambda \equiv \int_{0}^{\infty} \frac{H_{y}(z)dz}{H_{0}} = \frac{2}{\pi} \int_{0}^{\infty} \frac{dk}{k^{2} + K(k)}.$$
 (3.9)

<sup>7</sup> A. B. Pippard and T. E. Fabor, Proc. Roy. Soc. (London) A231, 336 (1955). <sup>8</sup> J. Bardeen, *Handbuch der Physik* (Springer-Verlag, Berlin) (to be published), Vol. 15.



FIG. 3. Susceptibility of a film, as a function of the half-thickness of the film, calculated on the basis of several theories. Lock's experimental data agree rather well with curve 1.

# (B) Random Scattering

We now impose the boundary condition that each electron approaching the surface has an equal probability of being scattered into any solid angle within the film. An equivalent problem is that of solving the Maxwell equation over all space but requiring that the electrons crossing the surfaces from outside the film come from a field-free region and hence have a random velocity distribution. Thus, we wish to solve

$$\nabla^2 \mathbf{A}(\mathbf{r}) = -\frac{4\pi}{c} \mathbf{j}(\mathbf{r}) = \int \mathbf{R} [\mathbf{R} \cdot \mathbf{A}(\mathbf{r}')] f(\mathbf{R}) d\tau', \quad (3.10)$$

where the integral is to be performed over the film. Using the plane symmetry of the problem, (3.10) can be reduced to a one-dimensional problem, where we have chosen the origin of coordinates at the center of the film.

$$\frac{d^2A_x(z)}{dz^2} = \int_{-a}^{a} G(z-z')A_x(z')dz', \qquad (3.11)$$

$$G(z-z') = \pi \int_{|z-z'|}^{\infty} \eta(\eta^2 - |z-z'|^2) f(\eta) d\eta. \quad (3.12)$$

For Pippard's kernal

$$G_{P}(z-z') = \frac{3\pi}{\xi_{0}\Lambda c} \int_{|z-z'|}^{\infty} \left(\frac{1}{\eta} - \frac{|z-z'|^{2}}{\eta^{3}}\right) e^{-\eta/\xi} d\eta. \quad (3.13)$$

The boundary condition to be imposed is

$$\left. \frac{dA_x}{dz} \right|_{z=a} = \frac{dA_x}{dz} \right|_{z=-a} = H_0.$$
(3.14)

A variational equivalent of (3.11) incorporating the boundary condition (3.14) is

$$\delta \left\{ \frac{1}{2} \int_{-a}^{a} \left[ \frac{dA(x)}{dx} \right]^{2} dx + \frac{1}{2} \int_{-a}^{a} \int_{-a}^{a} A(x) A(y) G(x-y) dx dy - H_{0} \int_{-a}^{a} \frac{dA(x)}{dx} dx \right\} = 0. \quad (3.15)$$

Choosing a trial function of the form

$$A(z) = \sum_{n \text{ odd}} C_n z^n.$$
(3.16)

(3.15) leads to the following set of equations for the  $C_n$ :

$$\sum_{m} C_{m} \left\{ \frac{mna^{m+n-1}}{m+n-1} + G_{mn} \right\} = H_{0}a^{n}, \qquad (3.17)$$

where

$$G_{mn} = \int_{-a}^{a} \int_{-a}^{a} x^m y^n G(x-y) dx dy.$$

The calculation of A has been carried out analytically retaining cubic terms in z for the Pippard kernel. The corresponding susceptibility is shown in Fig. 3. We note that the curve for specular reflection lies very close to that for random scattering over the entire range of  $a/\lambda_0$ . The calculation for random scattering with the Bardeen theory becomes quite involved due to the complex form of the one-dimensional kernel in configuration space and for this reason the calculation has not been carried out explicitly. However, because of the strong similarity between the Bardeen and Pippard theories, it is felt the Bardeen theory should also be insensitive to the nature of the scattering at the surface. A similar argument holds for the Schafroth-Blatt theory with respect to the London theory.

### 4. DISCUSSION

In comparing the theoretical susceptibilities based on the nonlocal theories with Lock's experimental data. it is important to realize that the data for which the deviation is large  $(a/\lambda_0 < 2)$  were taken close to the critical temperature. The temperature dependence of  $\xi_0$  in the Pippard theory is not well known and for this reason there is considerable doubt about drawing conclusions as to the validity of the theory. Direct comparison of theory and experiment in Fig. 3 implicitly assumes that the temperature dependence of  $\xi_0$  is the same as that of  $\lambda_L$ . Thus, it appears that the Pippard theory in its present form with this dependence of  $\xi_0$ on temperature is incorrect. If one assumes, for example, that  $\xi_0$  increases somewhat more slowly with temperature than  $\lambda_L$ , the theoretical curve would come into closer agreement with experiment. The Bardeen theory has only been worked out for T=0 and again direct comparison with Lock's data is inconclusive. Thus, until the theory has been extended to higher temperature, the present experimental data give little insight into the validity of the nonlocal theories. It is hoped that experimental techniques will become available in the near future so that measurements can be made on very thin films and give a critical check of the existing theories.

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