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Coexistence of Ferromagnetism and Superconductivity?

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Abstract. Some aspects of the problem of coexistence of superconductivity and ferromagnetism are analyzed, in particular the suggestion of JACCARINO and PETER [2] that it might be possible to compensate an exchange field by an external magnetic field, such making superconductivity possible in a range of very high magnetic fields.

The Meissner effect will however in principle impede very high magnetic fields from penetrating the material. We examine in this paper if there are interactions, acting on the orbits of the electrons, that could play the role of effective magnetic fields, and therefore «compensate» the Meissner effect. It is shown that, within our assumptions, this is not possible.

Introduction

CLOGSTON [1] has shown that in the limit of complete field penetration in a superconductor, there is an upper limit imposed on that field due to its effect on the spins of the electrons. JACCARINO and PETER [2] pointed out that magnetic ions, such as in the rare earths, impress upon the spins of the conduction electrons an effective (exchange) field, which in some cases point in the opposite direction as the magnetization. They suggested that this effective field could be cancelled by an external field, such making superconductivity possible in a certain interval of magnetic field. The upper bound of this interval would then be given by the sum of the effective field plus the field given by the Clogston criterion.

The purpose of this paper is to investigate more closely the possibility of this effect in bulk superconductors of type I or of type II below H_{c1} , where the effect of a magnetic field on the orbits of the electrons is of crucial importance.

In part I, we analyze the problems in general terms.

In part II, we analyze microscopically the spatial distribution of currents and fields in a hypothetical superconducting ferromagnet.

Part I

The formulation of the problem is simple: if an effective field, acting on the spins of the conduction electrons, must be compensated by an external field, then this external field must penetrate the superconductor. In a bulk superconductor of type I, or type II below H_{c1} , this is in principle not possible, because the superconductor expels the magnetic field (Meissner effect). In a type II superconductor above H_{c1} , the field in the material is at most H_{c2} . Our purpose is to investigate whether the magnetic field acting on the orbits of the conduction electrons can be compensated in some way by an effective internal field. Note that the equivalence of superconductivity and Meissner effect, which has been proven [3], concerns the response of a conduction

electron system to an external electromagnetic field. Here we have in addition the response of the electron system to a ferromagnetic lattice.

Let us first recall briefly how the Meissner effect comes about. A microscopic treatment [4] shows that to first order in the vector potential A , the total current in an electron gas is given by two contributions:

a) a paramagnetic current

$$\mathbf{J}_p(\mathbf{r}, t) = -\frac{i}{\hbar c} \int d^3r' \int_{-\infty}^t dt' \langle \phi_0 | [\mathbf{j}^p(\mathbf{r}', t'), \mathbf{j}^p(\mathbf{r}, t)] | \phi_0 \rangle \mathbf{A}(\mathbf{r}', t') \quad (1)$$

where $\mathbf{j}^p(\mathbf{r}, t)$ is the usual paramagnetic current operator and $|\phi_0\rangle$ is the ground state of the system.

b) a diamagnetic current

$$\mathbf{J}_d(\mathbf{r}) = \frac{e^2}{m c} \rho_s(\mathbf{r}) \mathbf{A}(\mathbf{r}) \quad (2)$$

where $\rho_s(\mathbf{r})$ is the conduction electron density.

In the normal phase, and for $q = 0$, the two currents exactly cancel, whereas in the superconducting phase $J_p(q)$ disappears for small q so that the total slowly varying current is given by the diamagnetic current (2) which produces a field which exactly cancels the external field in the interior of the specimen.

Our system consists of magnetic ions, where the localized ' d -electrons' are responsible for the magnetic properties of the ferromagnet, and a gas of conduction electrons. In this system there will be a certain spatial distribution of magnetic fields, given by the sum of the external field $H_e(\mathbf{r})$, the internal dipole-field $H_{id}(\mathbf{r})$ due to the magnetic ions, and the magnetic field $H_{is}(\mathbf{r})$ due to whatever currents are present in the conduction-electron system. In addition a part of the exchange interaction between the d -electrons and the conduction electrons will depend on the momentum \mathbf{p} of the conduction electrons when the total angular momentum of the d -electrons is not zero.

If we suppose that this interaction can be written as $\mathbf{p} \cdot \mathbf{A}_{eff}$ and that the system we study is superconducting with the usual singlet pairing, then the slowly varying part of the current will be given by:

$$\mathbf{J}_l(\mathbf{r}) = \frac{e^2}{m c} \rho_s(\mathbf{r}) \{ \mathbf{A}_e(\mathbf{r}) + \mathbf{A}_{id}(\mathbf{r}) + \mathbf{A}_{is}(\mathbf{r}) + \mathbf{A}_{eff}(\mathbf{r}) \}. \quad (3)$$

Using the Maxwell equation:

$$\mathbf{J}_l(\mathbf{r}) = \frac{c}{4\pi} \text{curl } \mathbf{H}_{is}(\mathbf{r}) \quad (4)$$

we get, performing a standard calculation [5] and putting $H_{eff} = \text{curl } A_{eff}$

$$\begin{aligned} -\Delta^2 \mathbf{H}_{is}(\mathbf{r}) &= \frac{4\pi e^2}{m c^2} \rho_s(\mathbf{r}) \{ \mathbf{H}_e(\mathbf{r}) + \mathbf{H}_{is}(\mathbf{r}) + \mathbf{H}_{id}(\mathbf{r}) + H_{eff}(\mathbf{r}) \} \\ &+ \frac{4\pi e^2}{m c^2} \nabla \rho_s(\mathbf{r}) \{ \mathbf{A}_e(\mathbf{r}) + \mathbf{A}_{is}(\mathbf{r}) + \mathbf{A}_{id}(\mathbf{r}) + \mathbf{A}_{eff}(\mathbf{r}) \}. \end{aligned} \quad (5)$$

Supposing that ρ_s is constant (the more general case $\nabla \rho_s \neq 0$ will be discussed later) we have

$$-\nabla^2 \mathbf{H}_{is}(\mathbf{r}) = \frac{4\pi e^2}{m c^2} \rho_s \{ \mathbf{H}_e(\mathbf{r}) + \mathbf{H}_{is}(\mathbf{r}) + \mathbf{H}_{id}(\mathbf{r}) + \mathbf{H}_{eff}(\mathbf{r}) \}. \quad (6)$$

In this the total magnetic field is given by

$$\mathbf{H}(\mathbf{r}) = \mathbf{H}_e(\mathbf{r}) + \mathbf{H}_{is}(\mathbf{r}) + \mathbf{H}_{id}(\mathbf{r}).$$

It is easy to see on a simple geometry that $\mathbf{H}(\mathbf{r})$ inside the superconductor will be reduced to $-\mathbf{H}_{eff}(\mathbf{r})$:

$$\mathbf{H}(\mathbf{r}) \simeq -\mathbf{H}_{eff}(\mathbf{r}) \quad (7)$$

in the interior.

So if $\mathbf{H}_{eff}(\mathbf{r})$ has approximately the value of the spin-polarisation field a compensation of the exchange field is possible.

Equation (5) is valid for the slowly varying part of the fields, and this is indeed the part which we want to calculate. Since the right hand side of equation (5) is curl J_i , we have to calculate the Fourier transform of $J_i(\mathbf{r})$ for small wave vectors q . This will be done in the next section.

Part II

We consider first a regular ferromagnet. Our system consists of a regular array of magnetic ions, with the magnetic moment of each ion pointing in the $+z$ -direction, and the gas of conducting-electrons. In the interaction energy between the localized ' d -electrons' and the conduction electrons, we take the expectation value over the d -electrons, thus neglecting dynamic effects.

The one particle Hamiltonian for the conduction electrons is written:

$$\begin{aligned} \mathcal{K} = & \frac{1}{2m} \left(\mathbf{p} - \frac{e}{c} (\mathbf{A}_e(\mathbf{r}) + \mathbf{A}_{id}(\mathbf{r})) \right)^2 + \frac{e}{mc} \left(\mathbf{p} - \frac{e}{c} (\mathbf{A}_e(\mathbf{r}) + \mathbf{A}_{id}(\mathbf{r})) \right) \cdot \mathbf{A}_{eff} \\ & + \sum V(\mathbf{r} - \mathbf{R}_i) + \sum J(\mathbf{r} - \mathbf{R}_i) (\boldsymbol{\sigma} \cdot \mathbf{S}) + g\mu \boldsymbol{\sigma} \cdot (\mathbf{H}_e(\mathbf{r}) + \mathbf{H}_{id}(\mathbf{r})). \end{aligned} \quad (8)$$

In the two first terms, $\mathbf{A}_e(\mathbf{r})$ is the vector potential of the external field and $\mathbf{A}_{id}(\mathbf{r})$ is the vector potential of the internal dipolefield, due to the magnetic ions, and is defined by:

$$\mathbf{A}_{id}(\mathbf{r}) = \sum_i \int d^3r' \frac{\boldsymbol{\mu}(\mathbf{r}' - \mathbf{R}_i) \times (\mathbf{r} - \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^3} \quad (9)$$

$\boldsymbol{\mu}(\mathbf{r}' - \mathbf{R}_i)$ is the spin-density of the localized d -electrons. The sum runs over all lattice points \mathbf{R}_i .

$\mathbf{A}_{eff}(\mathbf{r})$ represents the orbital part of the exchange interaction. The exchange interaction can be written in the form:

$$\mathcal{K}_{ex} = \sum_i J(\mathbf{r} - \mathbf{R}_i) \boldsymbol{\sigma} \cdot \mathbf{S}_i + \sum_i D(\mathbf{r} - \mathbf{R}_i) \mathbf{l}_i \cdot \mathbf{L}_i + \dots$$

where we disregard higher terms. The first term corresponds to the fourth term in (8) $\boldsymbol{\sigma}$ is the spin of the conduction electron and \mathbf{S}_i is the spin of the ion at the i 'th lattice site. In the absence of magnetic fields the second term is

$$\frac{e}{mc} \mathbf{p} \cdot \mathbf{A}_{eff}$$

where

$$\mathbf{A}_{eff} = \frac{mc}{e} \sum_i D(\mathbf{r} - \mathbf{R}_i) (\mathbf{L}_i \times (\mathbf{r} - \mathbf{R}_i)). \quad (10)$$

\mathbf{L}_i is the orbital momentum of the i 'th ion and \mathbf{l}_i is the orbital momentum of the conduction electron with respect to the i 'th lattice point. The third term in (8) is the

ordinary periodic potential and the last term is the Zeeman energy of the conduction electron.

The Hamiltonian (8) can be separated in two parts: a part which has the periodicity of the lattice, \mathcal{K}_{per} , and the rest, \mathcal{K}_{ap} . As we will see, the whole problem depends on \mathcal{K}_{ap} . This aperiodic part contains:

- a) the orbital interaction with the external field.
- b) surface terms.

Surface terms have an important influence on the properties of the whole system only if they have a long range. This is the case for the dipole field A_{id} : as KITTEL [6] has shown in a simple geometry, the non periodic part of the dipole field is just equal to the uniform magnetization.

$$\text{rot}(\mathbf{A}_{id})_{ap} = 4\pi \mathbf{M}.$$

It is clear however that A_{eff} has a short range compared to A_{id} : the range of A_{eff} is approximately equal to the range of the wave functions of the d -electrons, which are well localized.

From equation (7) one expects therefore that the interaction $\phi \cdot A_{eff}$ cannot 'compensate' the Meissner effect.

To prove this, we make our argument self-consistent: we suppose that the total magnetic field, including the induced field arising from the super currents, is small in most of the material. We build therefore a BCS wave function with the eigenfunctions of \mathcal{K}_{per}

$$\mathcal{K}_{per} \varphi_{k_s} = \varepsilon_k \varphi_{k_s} \quad (11)$$

and treat then \mathcal{K}_{ap} as a perturbation. The main point will then show that $\phi \cdot A_{eff}$ does not induce any slowly varying currents in the system. In equation (11) we have restricted ourselves to one conduction band, and dropped the exchange field acting on the spins since this is not essential to the present argument.

We now define the current operator by the relation [7]

$$\delta\mathcal{K} = -\frac{1}{c} \int \mathbf{j}(\mathbf{r}) \delta\mathbf{A}_e(\mathbf{r}) d^3r. \quad (12)$$

Performing the functional derivation we find as usual a paramagnetic and a diamagnetic current:

$$\mathbf{j}(\mathbf{r}) = \mathbf{j}_p(\mathbf{r}) + \mathbf{j}_d(\mathbf{r}). \quad (13)$$

Using the formalism of second quantization and writing $c_{k_s}^+$ (c_{k_s}) for the creation (annihilation) operator for the state φ_{k_s} we get:

$$\mathbf{j}_p(\mathbf{r}) = -\frac{ie\hbar}{2m} \sum_{k_1 k_2} [\varphi_{k_1 s}^*(\mathbf{r}) \nabla \varphi_{k_2 s}(\mathbf{r}) - (\nabla \varphi_{k_1 s}^*(\mathbf{r})) \varphi_{k_2 s}(\mathbf{r})] c_{k_1 s}^+ c_{k_2 s} \quad (14)$$

$$\mathbf{j}_d(\mathbf{r}) = -\frac{e^2}{mc} \sum_{k_1 k_2} \varphi_{k_1 s}^*(\mathbf{r}) \varphi_{k_2 s}(\mathbf{r}) (\mathbf{A}_e(\mathbf{r}) + \mathbf{A}_{id}(\mathbf{r}) + \mathbf{A}_{eff}(\mathbf{r})) c_{k_1 s}^+ c_{k_2 s}. \quad (15)$$

We now split the current operator in two parts:

$$\mathbf{j}(\mathbf{r}) = \mathbf{j}^{per}(\mathbf{r}) + \mathbf{j}^{ap}(\mathbf{r}) \quad (16)$$

where

$$\mathbf{j}^{per}(\mathbf{r}) = \mathbf{j}_p(\mathbf{r}) + \mathbf{j}_d^{per}(\mathbf{r}) \quad \mathbf{j}^{ap}(\mathbf{r}) = \mathbf{j}_d^{ap}(\mathbf{r}) \quad (17)$$

$j_d^{per}(\mathbf{r})$ is defined as in (15) but with the periodic part of the total vector potential, $A_{id}^{per} = A_{eff}^{per}$, similarly $j_d^{ap}(\mathbf{r})$ is defined as the rest, with the aperiodic parts of the vector potentials, $A_e + A_{id}^{ap} + A_{eff}^{ap}$.

In the same way the BCS wave function will have a periodic and a non periodic part

$$|\phi\rangle = |\phi^p\rangle + |\phi^{ap}\rangle \quad (18)$$

where

$$|\phi^p\rangle = \prod_k (u_k + v_k c_{k\uparrow}^+ c_{-k\downarrow}^+) |0\rangle \quad (19)$$

and $|0\rangle$ is the vacuum

$$|\phi^{ap}\rangle = T \exp \left[-\frac{1}{\hbar} \int \mathcal{K}^{ap}(t') dt' \right] |\phi^p\rangle. \quad (20)$$

Correspondingly, the expectation value of the current can be written:

$$\mathbf{J} = \langle \phi | \mathbf{j}(\mathbf{r}) | \phi \rangle = \mathbf{J}^{per} + \mathbf{J}^{ap} \quad (21)$$

where

$$\mathbf{J}^{per} = \langle \phi^p | \mathbf{j}^{per} | \phi^p \rangle. \quad (22)$$

We first discuss \mathbf{J}^{per} . It is easy to see that it has the periodicity of the lattice, and we can therefore expand it as in equation (1)

$$\mathbf{J}^{per}(\mathbf{r}) = \sum_G \tilde{\mathbf{J}}^{per}(G) e^{iG\mathbf{r}} \quad (23)$$

where G are the reciprocal lattice vectors.

As discussed in part I, we are only interested in the small q_- components of the current, and this means here the $G = 0$ component. This term is explicitly discussed in Appendix I, and it is there shown that it is zero:

$$\langle \phi^p | \mathbf{j}^{per}(\mathbf{G} = 0) | \phi^p \rangle = 0. \quad (24)$$

To discuss \mathbf{J}^{ap} , we restrict ourselves to linear terms in the aperiodic part, and write:

$$\mathbf{J}^{ap} \simeq -\frac{i}{\hbar} \int_{-\infty}^0 \langle \phi^p | [\mathcal{K}^{ap}(t'), \mathbf{j}^{per}(\mathbf{r})] | \phi^p \rangle + \langle \phi^p | \mathbf{j}^{ap}(\mathbf{r}) | \phi^p \rangle. \quad (25)$$

The first term in (25) contains among other terms the paramagnetic current. This current goes to zero for $q \rightarrow 0$. In Appendix II we discuss all these terms, and we show explicitly that all terms go to zero for $q \rightarrow 0$ in the same way as the paramagnetic current does.

So the only current which survives in the long wavelength-limit is the second term in (25). This can be written:

$$\mathbf{J}^{ap}(\mathbf{r}) = \frac{e^2}{m c} \varrho(\mathbf{r}) \{ \mathbf{A}_e(\mathbf{r}) + \mathbf{A}_{id}^{ap}(\mathbf{r}) + \mathbf{A}_{eff}^{ap}(\mathbf{r}) \}. \quad (26)$$

If we add $e^2/m c \varrho(\mathbf{r}) \mathbf{A}_{is}(\mathbf{r})$, to make the magnetic field selfconsistent, to the right hand side of (26) we get the equation (3).

Now, from Part I we know that if our system is superconducting, the slowly varying part of the total magnetic field in the interior of the specimen will be reduced

to $-A_{eff}^{ap}$. But because of the short range of the exchange interaction A_{eff}^{ap} is different from zero only on the surface of the specimen, and therefore the total magnetic field is zero in the interior of the specimen.

One could argue that the other momentum-dependent terms in the exchange interaction are of the same order as the one we have used, and might give different results. However the main point is that our result comes from the short range nature of the exchange interaction, and any short range interaction will lead to the same conclusion. The same argument applies also to the ordinary spin-orbit interaction, which we also neglected in (8).

In discussing equations (3) and (26) we supposed that $\nabla\varrho(\mathbf{r}) = 0$. In real metals this is not the case and normally $\varrho(\mathbf{r})$ varies in such a way that the mean field seen by the spin of the electron is larger than the magnetization. One may think that this also produces an effect on the orbits of the electrons, that means that high- q -components of $\varrho(\mathbf{r})$ and $A(\mathbf{r})$ together produce low- q -components of the current. This problem is analyzed in Appendix III, using equation (5) for $\nabla\varrho \neq 0$. It turns out that the slowly varying contribution that one finds, has exactly the range of the Meissner current, so that in the interior it has no effect. Note that in discussing equation (22) and the first part of equation (25) we did not suppose $\nabla\varrho = 0$.

We thus come to the conclusion that h_{eff} is essentially zero and that in ferromagnetic superconductors of type I or type II below H_{c1} , the mean magnetic field, inclusive the magnetization due to the dipoles, will be expelled from the interior of the specimen.

At this point it should be emphasized that we have constructed one solution, in analogy with the BCS solution for the nonmagnetic case, but we have not shown that this is the ground state of the system. This depends on the effective electron-phonon interaction. One could for instance suppose that the magnetic field penetrates the superconductor, and then construct a BCS-like-wavefunction with Landau-functions. One would probably find a state which would not show a Meissner-effect. Such a state has not been observed, and the reason is probably that the effective electron-electron interaction between Landau-states is very small, due to their small overlap [8]. The

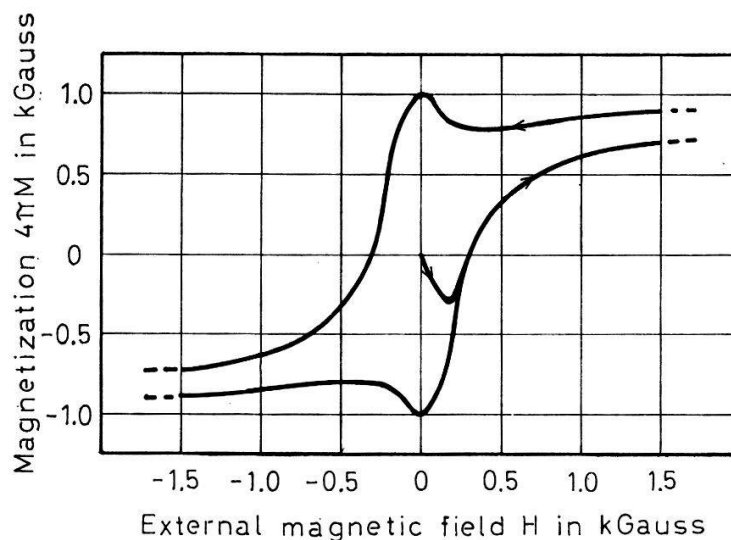


Figure 1

Magnetization versus field for $\text{Ce}_{0.918}\text{Gd}_{0.082}\text{Ru}_2$ at 1.3°K as measured by Bozorth et al. Note that the magnetization is in the same units as the external field.

same argument applies to the ferromagnetic superconductor, and therefore we don't expect such a state to be the ground state of our system.

From the anomalous Hall-effect in ferromagnetics one knows that in normal metals the conduction electrons can 'see' internal fields which are much higher than the magnetization. This seems to be in contradiction with our result. However the anomalous Hall effect is connected in an essential way to scattering processes [9] which produces also the electrical resistance. These scattering processes will be reduced in a superconductor in a similar scale as the resistance and cannot therefore give an effective field which could be of interest in our case.

Practically our results mean the following. In a type I superconductor, or a type II below H_{c2} , it is only possible to make superconductivity and ferromagnetism compatible if the exchange field acting on the spins is negligible. Superconductivity and ferromagnetism will be compatible in an external field that compensates the uniform magnetization. In a type II superconductor above H_{c1} , the exchange field acting on the spins may be at most of the order of H_{c2} .

Some years ago, BOZORTH et al. [10] published magnetization curves for alloys of the form $Ce_{1-x}Gd_xRu_2$ (Fig. 1); these alloys, as shown by MATTHIAS et al. [11] are both superconducting and ferromagnetic, but the question arises whether the specimen consists of an homogeneous phase, or if superconductivity and ferromagnetism exist in different domains.

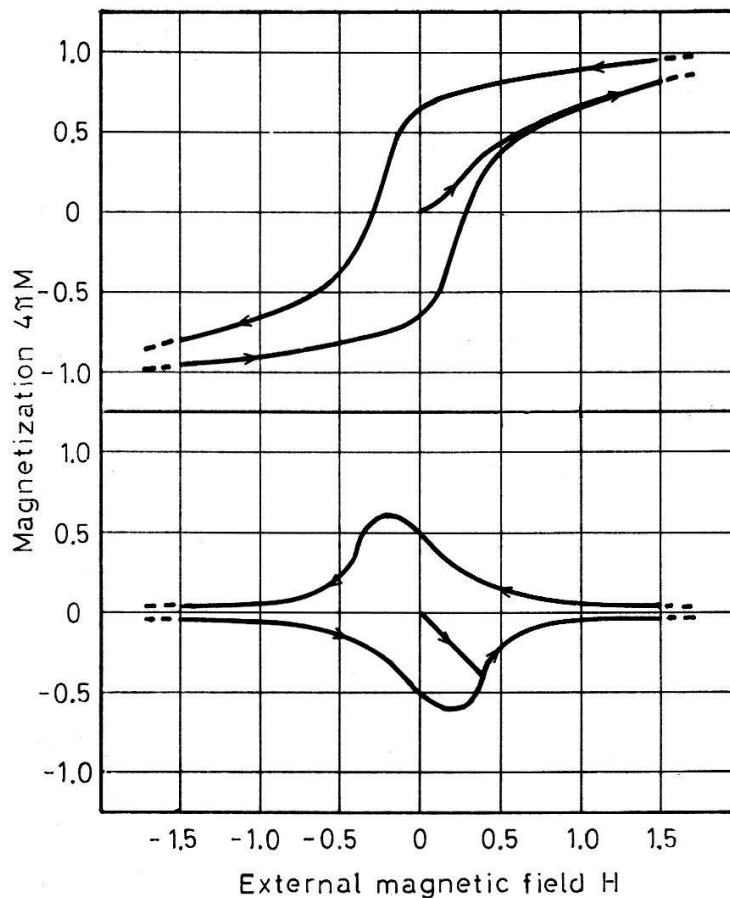


Figure 2

a) Typical ferromagnetic hysteresis curve. b) Typical superconducting hysteresis curve. In both curves the magnetization and the external field are in the same (arbitrary) units.

In Figure 3 we show the magnetization curve for a ferromagnetic superconductor, starting from a hypothetical substance which, if only ferromagnetic (superconducting), would have the magnetization curve shown in Figure 2a (Fig. 2b). The resulting magnetization curve depends on the choice of the curves 2a and 2b, but if the ferromagnetic and superconducting magnetizations are of the same order of magnitude, some important features are independent of the details of curves 2a and 2b: in particular the characteristic peak is always well on the left of the origin, and it is followed by a sharp decrease of $M(H)$. The perfect diamagnetic behaviour of the initial curve is also independent of the details of 2a and 2b.

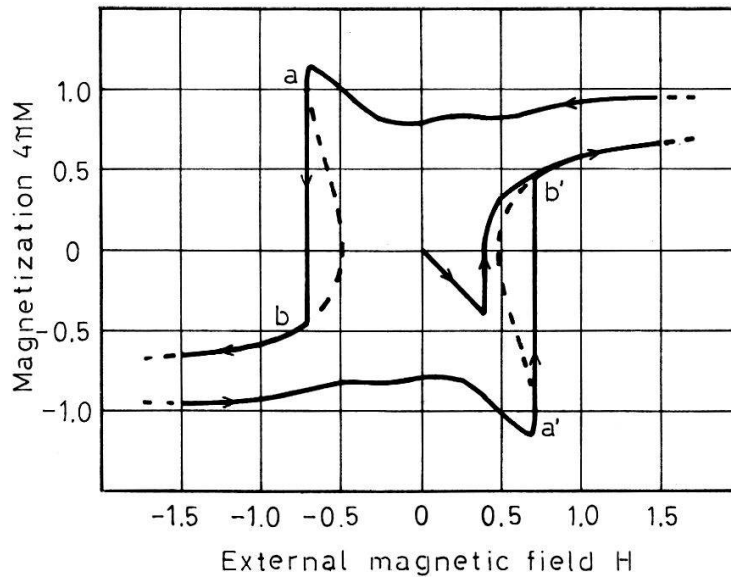


Figure 3

Magnetization versus field for a hypothetical ferromagnetic superconductor, as calculated from the two curves in Figure 2 using the selfconsistent equations

$$h_F = H + 4 \pi M_s(h_s)$$

$$h_s = H + 4 \pi M_F(h_F)$$

$h_F(h_s)$ is the magnetic field acting on the magnetic ions (superconducting electrons) and $M_F(M_s)$ is the magnetization of the ferromagnetic (superconducting) system as given in Figure 2a (2b). H is the external field. At the points a , a' the magnetization becomes unstable, and it changes abruptly to the magnetization at b , b' . The calculated curves $M(H)$ between these points are the dashed curves.

The units are the same as in Figure 2. Demagnetization effects were disregarded.

In Figure 4 we draw the curve which is just the sum of 2a and 2b. This corresponds roughly to the case where two phases are present. Comparison of these two curves with the experimental curve of BOZORTH et al. points clearly towards the second possibility: his samples consisted probably of ferromagnetic and superconducting domains. This view is supported by recent measurements by WILHELM and HILLENBRAND [12] on the same alloys, who checked carefully the homogeneity of the specimens by metallographic methods. They did not find superconductivity in alloys which had already become ferromagnetic.

To conclude this section, we discuss briefly the case of a dilute ferromagnetic alloy, when the magnetic ions are distributed at random.

The potential $A_{id}(r)$ is written as above

$$A_{id}(r) = \sum_i \int d^3r' \frac{\boldsymbol{\mu}(r' - \mathbf{R}_i) \times (r - r')}{|r - r'|^3}$$

where the sum goes over the magnetic sites. As usual one treats this system by averaging over the impurity-sites i.e. by replacing the sum by an integral. Physically this means that the $q \neq 0$ components of the $\boldsymbol{\mu}(r)$ -distribution average out because of the random distribution. It is clear then that this system is equivalent to a regular ferromagnet with a moment per ion reduced proportionally to the concentration.

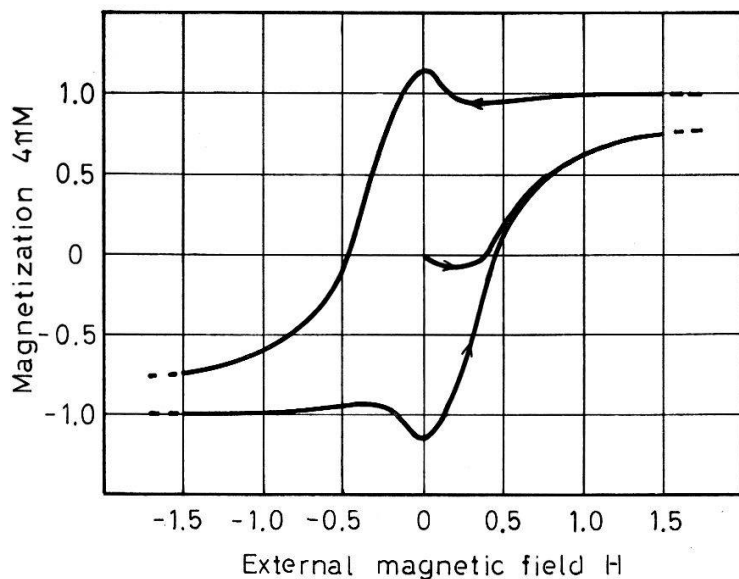


Figure 4

Magnetization versus field obtained by adding the two curves in Figure 2 (units as in Figure 2). This corresponds approximately to the situation where one has cylindric superconducting and ferromagnetic domains which are mainly parallel to the external field.

One can apply more sophisticated averaging procedures, which take into account random distribution of clusters of various sizes. We believe however that these will not give larger contributions to the low q components than would the magnetization of a similar system in a regular dense ferromagnet. In other words, for low q -components:

$$J(q) < \frac{e^2}{m c} \varrho_0 |A_{id}^{ap}(q)|$$

where $A_{id}^{ap}(q)$ is the Fourier transformed of the aperiodic part of the internal field in a dense regular ferromagnet. This is because each of the clusters produces low q -components corresponding at most to their magnetization.

Conclusion

We have examined the possibility of a bulk material being both superconducting and ferromagnetic if placed in a suitable external magnetic field. Our analysis showed that there is no possibility of 'compensating' the Meissner effect; in other words $h_{eff} = 0$ (see equation (5)) in most of the material. This result is due essentially to the

short range nature of all s - d interactions except the dipole field. Practically this means that one cannot compensate an exchange field acting on the spins in a superconductor of type I or type II below H_{c1} . In a superconductor of type II above H_{c1} , or in thin films, the system can become ferromagnetic and superconductor if the exchange field is smaller than the upper critical field.

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Appendix I

Here we show that $J^{per}(q=0) = 0$:

$$\mathbf{J}^{per}(q=0) = \langle \phi^{per} | \mathbf{j}^{per}(q=0) | \phi^{per} \rangle = \int \mathbf{J}^{per}(\mathbf{r}) d^3r, \quad (\text{I.1})$$

$$\begin{aligned} \mathbf{J}^{per}(\mathbf{r}) &= \frac{i\hbar}{2m} \Sigma [\varphi_k^*(\mathbf{r}) \nabla_r \varphi_k(\mathbf{r}) - (\nabla_r \varphi_k^*(\mathbf{r})) \varphi_k(\mathbf{r})] v_k^2 \\ &+ \frac{e}{mc} \Sigma \varphi_k^*(\mathbf{r}) \varphi_k(\mathbf{r}) (\mathbf{A}_{id}^{per} + \mathbf{A}_{eff}^{per}) v_k^2. \end{aligned} \quad (\text{I.2})$$

Using now that apart from a phase factor

$$\varphi_k(\mathbf{r}) = \varphi_{-k}(-\mathbf{r})$$

and furthermore

$$v_k^2 = v_{-k}^2,$$

$$\mathbf{A}_{id}^{per}(\mathbf{r}) + \mathbf{A}_{eff}^{per}(\mathbf{r}) = -(\mathbf{A}_{id}^{per}(-\mathbf{r}) + \mathbf{A}_{eff}^{per}(-\mathbf{r}))$$

one finds:

$$\begin{aligned} \mathbf{J}(\mathbf{r} + \mathbf{R}_N) &= \mathbf{J}(\mathbf{r}) = \frac{i\hbar}{2m} \Sigma (-\varphi_{-k}^*(-\mathbf{r}) (\nabla \varphi_k)(-\mathbf{r}) + (\nabla_r \varphi_{-k}^*)(-\mathbf{r}) \varphi_{-k}(-\mathbf{r})) v_k^2 \\ &- \frac{e}{mc} \Sigma |\varphi_{-k}(-\mathbf{r})|^2 (\mathbf{A}_{id}^{per}(-\mathbf{r}) + \mathbf{A}_{eff}^{per}(-\mathbf{r})) v_{+k}^2 \\ &= -\mathbf{J}(-\mathbf{r}) = -\mathbf{J}(-\mathbf{r} + \mathbf{R}_N). \end{aligned} \quad (\text{I.3})$$

Therefore the current is antisymmetric with respect to every lattice points, so that the integral in (I.1) is zero.

Appendix II

Here we discuss the first part of the aperiodic current, defined in equation (26). It is given by

$$\mathbf{J}^{ap}(\mathbf{r}, t) = -\frac{i}{\hbar} \int_{-\infty}^t dt' \langle \phi^{per} | [\mathcal{K}^{ap}(t') \mathbf{j}^{per}(\mathbf{r}, t)] | \phi^{per} \rangle. \quad (\text{II.1})$$

The aperiodic Part of the hamiltonian can be written

$$\mathcal{K}^{ap}(t') = -\frac{1}{c} \int d\mathbf{r}' \left(\mathbf{j}_p(\mathbf{r}' t') - \frac{e^2}{mc} \mathbf{a}^{per}(\mathbf{r}' t') \right) \mathbf{A}^{ap}(\mathbf{r}')$$

where $j(\mathbf{r})$ is the paramagnetic current operator, $a^{per}(\mathbf{r}', t')$ is defined by $a^{per}(\mathbf{r}', t') = \psi^+(\mathbf{r}', t') (A_{id}^{per}(\mathbf{r}') + A_{eff}^{per}(\mathbf{r}')) \psi(\mathbf{r}', t')$ (ψ^+ , ψ are field operators), and $A^{ap}(\mathbf{r}) = A_e(\mathbf{r}) + A_{id}^{ap}(\mathbf{r}) + A_{eff}^{ap}(\mathbf{r})$,

Following SCHRIEFFER [4] we then write the aperiodic current as:

$$\begin{aligned} \mathbf{J}^{ap}(\mathbf{r}, t) &= \frac{i}{\hbar c} \iint_{-\infty}^t dt' d^3r' \langle \phi^{per} | \left[\mathbf{j}_p(\mathbf{r}', t') - \frac{e^2}{m c} \mathbf{a}^{per}(\mathbf{r}', t'), \mathbf{j}^{per}(\mathbf{r}, t) \right] | \phi^{per} \rangle A^{ap}(\mathbf{r}' t') \\ &= \iint dt' d^3r' \mathbf{K}(\mathbf{r}, t \mathbf{r}' t') A^{ap}(\mathbf{r}' t') \end{aligned} \quad (\text{II.2})$$

where the Kernel $K(\mathbf{r}, t \mathbf{r}' t')$ is given by

$$\mathbf{K}(\mathbf{r}, t \mathbf{r}' t') = \frac{i}{\hbar c} \langle \phi^{per} | \left[\mathbf{j}_p(\mathbf{r}' t') - \frac{e^2}{m c} \mathbf{a}^{per}(\mathbf{r}' t'), \mathbf{j}^{per}(\mathbf{r}, t) \right] | \phi^{per} \rangle \theta(t - t').$$

Fourier transforming, and using the fact that the Kernel is periodic, we get:

$$\mathbf{J}^{ap}(\mathbf{q}, q_0) = \sum_{\mathbf{G}} \mathbf{K}(\mathbf{q}, -\mathbf{q} - \mathbf{G}, q_0) \mathbf{A}(\mathbf{q} + \mathbf{G}, q_0) \quad (\text{II.3})$$

where \mathbf{G} is a vector of the reciprocal lattice.

As we are interested in the zero frequency response we put $q_0 = 0$. We then find:

$$\begin{aligned} \mathbf{K}(\mathbf{q}, -\mathbf{q} - \mathbf{G}, 0) &= \frac{1}{\hbar c} \int_{-\infty}^0 d\tau \langle \phi^{per} | \\ &\times \left[\mathbf{j}_p(-\mathbf{q} - \mathbf{G}, \tau) - \frac{e^2}{m c} \mathbf{a}^{per}(-\mathbf{q} - \mathbf{G}, \tau), \mathbf{j}^{per}(\mathbf{q}, 0) \right] | \phi^{per} \rangle. \end{aligned}$$

We now write our operators in 2. Quantization.

$$\begin{aligned} \mathbf{j}_p(\mathbf{q}, t) &= \sum_{k_1 k_2 s_1} \mathbf{j}_{k_1 k_2}^{(1)}(\mathbf{q}) c_{k_1 s_1}^+(t) c_{k_2 s_1}(t) \\ \mathbf{a}^{per}(\mathbf{q}, t) &= \sum_{k_1 k_2 s_1} \mathbf{a}_{k_1 k_2}(\mathbf{q}) c_{k_1 s_1}^+(t) c_{k_2 s_1}(t), \\ \mathbf{j}^{per}(\mathbf{q}, t) &= \sum_{k_3 k_4 s_2} \mathbf{j}_{k_3 k_4}^{(2)}(\mathbf{q}) c_{k_3 s_2}^+ c_{k_4 s_2}. \end{aligned}$$

If one expresses the c_{k_s} operators by the γ_{k_s} operators, defined by the Valatin-Bogoliubov-transformations

$$\begin{aligned} c_{k\uparrow}^+ &= u_k \gamma_{k\uparrow}^+ + v_k \gamma_{-k\downarrow} & c_{k\downarrow}^+ &= u_k \gamma_{k\downarrow}^+ - v_k \gamma_{-k\uparrow} \\ c_{k\uparrow} &= u_k \gamma_{k\uparrow} + v_k \gamma_{-k\downarrow}^+ & c_{k\downarrow} &= u_k \gamma_{k\downarrow} - v_k \gamma_{-k\uparrow}^+ \end{aligned}$$

and use that $\gamma_{k_s}(t) = \gamma_{k_s}(0) e^{(i/\hbar)E_k t}$, where E_k is the excitation energy of quasiparticles from the superconducting ground-state, the Kernel $K(\mathbf{q}, -\mathbf{q} - \mathbf{G}, 0)$ can easily be calculated and one obtains:

$$\begin{aligned} \mathbf{K}(\mathbf{q}, -\mathbf{q} - \mathbf{G}, 0) &= \frac{1}{c} \sum_{k_1 k_2} \left[\mathbf{j}_{k_1 k_2}^{(1)}(-\mathbf{q} - \mathbf{G}) - \frac{e^2}{m c} \mathbf{a}_{k_1 k_2}(-\mathbf{q} - \mathbf{G}) \right] \\ &\times \mathbf{j}_{k_2 k_1}^{(2)}(\mathbf{q}) \frac{v_{k_1}^2 u_{k_2}^2 + u_{k_1}^2 v_{k_2}^2}{E_{k_1} + E_{k_2}} + \frac{1}{c} \sum_{k_1 k_2} \left[\mathbf{j}_{-k_2, -k_1}^{(1)}(-\mathbf{q} - \mathbf{G}) \right. \\ &\left. - \frac{e^2}{m c} \mathbf{a}_{-k_2, -k_1}(-\mathbf{q} - \mathbf{G}) \right] \mathbf{j}_{k_2 k_1}^{(2)}(\mathbf{q}) \frac{2 u_{k_1} u_{k_2} v_{k_1} v_{k_2}}{E_{k_1} + E_{k_2}}. \end{aligned} \quad (\text{II.4})$$

We have here used the inversion symmetry of \mathcal{K}_{per} , from which follows that $E_k = E_{-k}$, $v_k = v_{-k}$, $u_k = u_{-k}$.

Furthermore, using the periodicity, we find:

$$\begin{aligned} \mathbf{j}_{k_1 k_2}^{(1,2)}(\mathbf{q}) &= \sum_{G'} \delta(\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{q} + G') \mathbf{j}_{k_1 k_2}^{(1,2)}(\mathbf{q}) \\ \mathbf{a}_{k_1 k_2}(\mathbf{q}) &= \sum_{G'} \delta(\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{q} + G') \mathbf{a}_{k_1 k_2}(\mathbf{q}). \end{aligned}$$

Finally we suppose that our specimen has a center of symmetry and then, using the antisymmetric and hermitian properties of the vectorpotentials and the current-operators one obtains

$$\begin{aligned} \mathbf{j}_{-k_2 - k_1}^{(1,2)}(\mathbf{q}) &= -\mathbf{j}_{k_2 k_1}^{(1,2)}(-\mathbf{q}) = -\mathbf{j}_{k_1 k_2}^{(1,2)}(\mathbf{q})^* \\ \mathbf{a}_{-k_2 - k_1}(\mathbf{q}) &= -\mathbf{a}_{k_2 k_1}(-\mathbf{q}) = -\mathbf{a}_{k_1 k_2}(\mathbf{q})^*. \end{aligned}$$

We thus get for the Kernel:

$$\begin{aligned} K(\mathbf{q}, -\mathbf{q} - G, 0) &= \frac{1}{c} \sum_{k G'} \left[\mathbf{j}_{k, k-q-G-G'}^{(1)}(-\mathbf{q} - G) - \frac{e^2}{m c} \mathbf{a}_{k, k-q-G-G'}(-\mathbf{q} - G) \right] \\ &\times \mathbf{j}_{k-q-G-G', k}^{(2)}(\mathbf{q}) \frac{(u_k v_{k-q-G-G'} - u_{k-q-G-G'} v_k)^2}{E_k + E_{k-q-G-G'}} \\ &+ \frac{1}{c} \sum_{k G'} \left[\mathbf{j}_{k, k-q-G-G'}^{(1)}(-\mathbf{q} - G) - \mathbf{j}_{k, k-q-G-G'}^{(1)*}(-\mathbf{q} - G) \right] \\ &\times \mathbf{j}_{k-q-G-G', k}^{(2)}(\mathbf{q}) \frac{2 u_k u_{k-q-G-G'} v_k v_{k-q-G-G'}}{E_k + E_{k-q-G-G'}} \\ &+ \frac{1}{c} \sum_{k G'} \left[\mathbf{a}_{k, k-q-G-G'}(-\mathbf{q} - G) - \mathbf{a}_{k, k-q-G-G'}^*(-\mathbf{q} - G) \right] \\ &\times \mathbf{j}_{k-q-G-G', k}^{(2)}(\mathbf{q}) \frac{2 u_k u_{k-q-G-G'} v_k v_{k-q-G-G'}}{E_k + E_{k-q-G-G'}}. \end{aligned}$$

We now restrict ourselves to one band, which means that k , and $k - q - G - G'$ must be in the first Brillouin zone. For q tending to zero, this means $G = -G'$, and so it follows $K(0, -G, 0) = 0$ for all G which means that $j(q) \rightarrow 0$ for $q \rightarrow 0$. Now, because our system is not translation invariant, we cannot use the Schafroth argument to prove the Meissner effect. The important thing is, however, that all terms (II.3) behave in the same way for small q . This can be seen from (II.5). In the first line the factor making the expression going to zero for $q \rightarrow 0$ is independent of the different fields and currents contained in $K(q, -q - G, 0)$. In the second and third line the factors which goes to zero are

$$\begin{aligned} &(\mathbf{j}_{k, k-q-G-G'}(-\mathbf{q} - G) - \mathbf{j}_{k, k-q-G-G'}^*(-\mathbf{q} - G)) \text{ and} \\ &(\mathbf{a}_{k, k-q-G-G'}(-\mathbf{q} - G) - \mathbf{a}_{k, k-q-G-G'}^*(-\mathbf{q} - G)) \end{aligned}$$

respectively. The behaviour of these two differences for small q are however both given by the behaviour of $(\varphi_k - \varphi_{k-q}^*)$ and $(\varphi_k^* - \varphi_{k-q}^*)$ where φ_k are the Blochfunctions which belong to the periodic part of our Hamiltonian. On the other hand is the Kernel of the paramagnetic current contained in the two first lines in (II.5). It is therefore not possible to replace the paramagnetic current (which disappears for small q) by

some of the other contributions to (II.3), because all these contributions behave in the same way as the paramagnetic current for small q . Thus none of these current contributions can have an influence on the Meissner effect.

Appendix III

We write equation I in the form

$$-\nabla^2 h(\mathbf{r}) = \varrho(\mathbf{r}) \{h(\mathbf{r}) + \bar{h}(\mathbf{r})\} + \nabla \varrho(\mathbf{r}) \times \{a(\mathbf{r}) + \bar{a}(\mathbf{r})\} \quad (\text{III.1})$$

where

$$h = \text{curl } a, \quad \bar{h} = \text{curl } \bar{a},$$

h represents the self-consistent field h_{i_s} and \bar{h} is the inhomogeneous term $h_{i_d} + h + h_{eff}$.

If ϱ is a constant, the equation is separable in the Fourier space, i.e. the low q -components of $h(q)$ are determined by the low q -components of $\bar{h}(q)$, and it is enough to calculate accurately these low q -components of $\bar{h}(q)$.

The situation must be analyzed more carefully when one allows ϱ to be \mathbf{r} dependent. To discuss this, we write

$$\varrho(\mathbf{r}) = \varrho_0 + \delta\varrho(\mathbf{r}) \quad (\text{III.2})$$

where ϱ_0 is the average of $\varrho(\mathbf{r})$ and $|\delta\varrho(\mathbf{r})| \ll \varrho_0$. Accordingly we write

$$h(\mathbf{r}) = h_0(\mathbf{r}) + \delta h(\mathbf{r}) \quad (\text{III.3})$$

and

$$a(\mathbf{r}) = a_0(\mathbf{r}) + \delta a(\mathbf{r}) \quad (\text{III.4})$$

where $h_0(\mathbf{r})$ is defined by

$$-\nabla^2 h_0(\mathbf{r}) = \varrho_0 \{h_0(\mathbf{r}) + \bar{h}(\mathbf{r})\}. \quad (\text{III.5})$$

We discuss our problem in the simple half space geometry: the y - z plan separates the superconducting body ($x > 0$) from the vacuum ($x < 0$).

Suppose we write for $\bar{h}_z(\mathbf{r})$ (the z -component of $\bar{h}(\mathbf{r})$)

$$\bar{h}_z(\mathbf{r}) = d e^{-\beta x} + \bar{h}$$

where d is some constant. Then one finds

$$h_{0z}(\mathbf{r}) = (\bar{h} - c') e^{-V|\varrho_0|x} + c' e^{-\beta x} - \bar{h} \quad (\text{III.7})$$

where

$$c' = \frac{\varrho_0 d}{-\beta^2 - \varrho_0}. \quad (\text{III.8})$$

Also

$$a_{0y}(\mathbf{r}) = -\frac{\bar{h} - c'}{V|\varrho_0|} e^{-V|\varrho_0|x} - \frac{c'}{\beta} e^{-\beta x} - \bar{h} x. \quad (\text{III.9})$$

The other components are zero

$$a_{0x} = a_{0z} = h_{0x} = h_{0y} = 0.$$

Replacing in (III.1) ϱ , h and a by (III.2), (III.3) and (III.4), and using (III.5) we get

$$-\nabla^2 \delta h(\mathbf{r}) = \varrho_0 \delta h(\mathbf{r}) + \delta\varrho(\mathbf{r}) h_0(\mathbf{r}) + \delta\varrho(\mathbf{r}) \bar{h}(\mathbf{r}) + \nabla \delta\varrho(\mathbf{r}) \times \{a_0(\mathbf{r}) + \bar{a}(\mathbf{r})\}. \quad (\text{III.10})$$

In (III.9) we have neglected the term $\delta\varrho(r) \delta h(r)$ and $\nabla \delta\varrho \times \delta a(r)$. Setting $e i$

$$\delta\varrho(r) = \delta\varrho e^{i(2\pi/a)(x+y+z)} \quad (\text{III.11})$$

and using (III.6), (III.7) and (III.9), we get for the z component of equation (III.10)

$$\begin{aligned} -\nabla^2 dh_z(r) &= \varrho_0 dh_z(r) + \delta\varrho e^{i(2\pi/a)(x+y+z)} \left\{ (\bar{h} - c') e^{-V|\varrho_0|x} \right. \\ &\quad \left. + c' e^{-\beta x} + d e^{-\beta x} - i \frac{2\pi}{a} \frac{\bar{h} + c'}{V|\varrho_0|} e^{-V|\varrho_0|x} - i \frac{2\pi}{a} \frac{c' + d}{\beta} e^{-\beta x} \right\} \\ &= \varrho_0 dh_z(z) + e^{i(2\pi/a)(x+y+z)} (l e^{-V|\varrho_0|x} + m e^{-\beta x}) \end{aligned}$$

with

$$dh_z(r) = e^{i(2\pi/a)(y+z)} f(x)$$

we get

$$-\frac{\partial^2}{\partial x^2} f(x) = \left(\varrho_0 - \frac{8\pi^2}{a^2} \right) f(x) + e^{i(2\pi/a)x} (l e^{-V|\varrho_0|x} + m e^{-\beta x}).$$

The solution can be written

$$f(x) = (q_2 + q_1) e^{-\sqrt{(8\pi^2/a^2) + |\varrho_0|}x} - a_2 e^{(2\pi/a)x - V|\varrho_0|x} - a_1 e^{i(2\pi/a)x - \beta x} \quad (\text{III.12})$$

where a_1 and a_2 are given by

$$a_2 = \frac{\varrho_0 - (8\pi/a^2)^2 + l}{-(i(2\pi/a) - \sqrt{|\varrho_0|})^2} \quad a_1 = \frac{\varrho_0 - (8\pi/a^2)^2 + m}{-(i(2\pi/a) - \beta)^2}.$$

The form (III.12) shows that the penetration depth is again given by the larger of $1/\sqrt{|\varrho_0|}$ (London penetration depth) and $1/\beta$ (range of the aperiodic effective field). If $1/\beta$ is small, the total magnetic field will be zero in the interior of the specimen. Finding that δh and δa are negligible in most of the material, we also justify self-consistently the fact that we neglected $\delta\varrho \times \delta h$ and $\nabla \delta\varrho \times \delta a$ in our solution.

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