## Theory of Nonlinear Meissner Effect in High- $T_c$ Superconductors

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We investigate the nonlinear Meissner effect microscopically. Previous studies did not consider a certain type of interaction effect on the nonlinear phenomena. The scattering amplitude barely appears without being renormalized into the Fermi-liquid parameter. With this effect we can solve the outstanding issues (the quantitative problem, the temperature and angle dependences). The quantitative calculation is performed with use of the fluctuationexchange approximation on the Hubbard model. It is also shown that the perturbation expansion on the supercurrent by the vector potential converges owing to the nonlocal effect.

KEYWORDS: nonlinear Meissner effect, unconventional superconductor, electron correlation, spin fluctuation, vertex correction, nonlocal effect

Many unconventional superconductors have recently been discovered. The evidence that these superconductors are non-s-wave is obtained by thermodynamic measurements in most cases. These types of measurement provide only information averaged over the Fermi surface and then other measurements are needed to determine the position of nodes. Detailed information about the node is useful for judging the accuracy of various theories. One of these, the nonlinear Meissner effect (NLME), which provides a measurement of the magnetic field (H) dependence of the magnetic field penetration depth ( $\lambda + \delta\lambda(H)$ ), was proposed by Yip and coworkers (YS).<sup>1,2)</sup> Their proposal is based on the Doppler-shifted energy spectrum and its predictions are summarized as follows. (i) The supercurrent has a nonanalytical form (written as A|A|, A is the vector potential) and  $\delta\lambda(H)$  is proportional to the magnetic field |H| as a result. (ii)  $\delta\lambda(H)$  varies with the direction of the applied magnetic field and therefore  $\delta\lambda(H)$ provides information on the position of nodes.

To date, the experiments have not provided decisive results because the effect is small and tends to be masked by many extrinsic effects. The first investigation of NLME was carried out by Maeda *et al.*,<sup>3)</sup> however this experiment was performed with the magnetic field perpendicular to the CuO plane. The precision was also poor (order of 10Å) and, at present, the observed quantity is considered to reflect extrinsic effects. Experiments with high precision (order of 0.1Å) were carried out by Bidinosti *et al.*<sup>4)</sup> and Carrington *et al.*<sup>5)</sup> The most reliable results in ref.4 are summarized as  $\delta\lambda(H) \propto H^2$ , the temperature dependence is weak and the

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angle dependence is not observed. These contradict YS's theory. YS also predict that  $\delta\lambda(H) \propto H^2$  below the crossover field, however, the temperature dependence is strong. Two groups have attempted to detect the transverse magnetization, which is the other prediction made by YS.<sup>6,7</sup> The recent experiment with higher precision (two orders of magnitude)<sup>6</sup> showed that the amplitude of this quantity is at most one third of the predicted one and it is almost at the measurable limit. Extensions of YS's theory were made by several researchers.<sup>8–11</sup> The common results obtained by them are that the theoretical predictions are inconsistent with the experimental results. Therefore, some papers suggest that the experiments observed extrinsic effects. (In ref.5 the possible extrinsic effects are listed (the vortex contribution, the weak links, the interlayer contribution).) The important point is that the values predicted by YS and other researchers are at least of the same order of magnitude, compared with the values obtained in the experiments. Therefore, it is not possible that the intrinsic theoretical value is masked by the extrinsic effects which are suppressed up to 0.2Å.

Then, the question arises as to whether the existing theories are correct for judging experimental results. Here, we discuss the NLME effect on the basis of the perturbation theory and show that the previous theories have some defects. The perturbation expansion by the vector potential on the supercurrent and the magnetic field penetration depth converges owing to the nonlocal effect. The intermediate-states interaction<sup>12</sup> (electron-electron) which is not included in the conventional quasiclassical approximation<sup>13</sup>) exists and makes a dominant contribution. This effect solves the inconsistency between the theory and the experiments on the value of  $\delta\lambda$  and its angle and temperature dependences. We adopt the fluctuationexchange (FLEX) approximation for the quantitative calculation. The many-body effect on the response function is included on the basis of the conserving approximation.

Our theory is based on the evaluation of the response function in the supercurrent which is exactly expanded by A up to the third order. The expression for the supercurrent is written as

$$J_{\mu}(q) = -K^{(1)}_{\mu\nu}(q)A_{\nu}(q) - \int_{q'} K^{(2)}_{\mu\nu\alpha}(q,q')A_{\alpha}(q')A_{\nu}(q-q') - \int_{q',q''} K^{(3)}_{\mu\nu\alpha\beta}(q,q',q'')A_{\beta}(q'')A_{\alpha}(q'-q'')A_{\nu}(q-q'),$$
(1)

where  $K^{(1,2,3)}$  are the response functions in the perturbation expansion,  $\mu, \nu, ...$  are the spatial dimensions and the summation of the repeated indices is taken.  $K^{(1)}$  appears in the usual linear response theory. The  $K^{(3)}$  term is dominant in the magnetic field dependence of  $\lambda$ because  $K^{(2)}$  vanishes.

First, we show the convergence of the perturbation expansion. By analyzing various terms it is shown that the most divergent term in the local limit is the type (a) term in Fig. 1 and

is written as

$$K^{(3a)}(q,q',q'') = \frac{T}{V} \sum_{\boldsymbol{k},n} v_{\boldsymbol{k}}^{4} \operatorname{Tr}[\hat{G}_{k+q/2}\hat{G}_{k+q'-q/2}\hat{G}_{k+q''-q/2}\hat{G}_{k-q/2}], \qquad (2)$$

where T,  $v_k$  and  $\hat{G}_k$  are the temperature, the velocity and Green's function in the superconducting state, respectively, and  $k = (\mathbf{k}, i\epsilon_n)$  ( $\epsilon_n = \pi T(2n + 1)$ , n is integer) and  $q = (\mathbf{q}, 0)$ . The uniform component is written as  $K^{(3a)}(0,0,0) \propto -\frac{v}{\Delta_0}\frac{1}{T}$  ( $\Delta_0$  is the maximum of the superconducting gap). This is the result of YS's theory.  $K^{(3)}(q,q',q'')$  diverges for  $T \to 0$  at q = q' = q'' = 0, however, this term is integrated by q' and q'' in the expression of the supercurrent and by q, q' and q'' in the case of  $\delta\lambda$ . Then, the behavior of  $K^{(3)}(q,q',q'')$  in q-space comes into question. The q-dependence of  $K^{(3a)}(q,0,0)$  for small q and at T = 0 is written as  $K^{(3a)}(q,0,0) \propto -\frac{v}{\Delta_0}\frac{1}{v_{\perp}|q|}$  ( $v_{\perp}$  is the mean value of the the interlayer velocity with magnetic field parallel to the ab-plane.) It is difficult to determine analytically the dependence of  $K^{(3)}(q,q',q'')$  on q,q' and q'', however the nonzero values of q' and q'' do not make this term more divergent than 1/q and if we consider the symmetry relation,  $K^{(3)}(q,q,q) = K^{(3)}(q,0,0)$ , the form for small q, q', q'' is considered to be

$$K^{(3)}(q,q',q'') \propto \frac{1}{\sqrt{(\frac{q}{2})^2 + (q'-\frac{q}{2})^2 + (q''-\frac{q}{2})^2 + (\frac{q''}{2})^2}}.$$
(3)

The  $q^{-1}$  divergence of  $K^{(3)}(q, q', q'')$  guarantees the convergence of the perturbation expansion on the physical quantities. Therefore, the nonanalytical behavior does not appear contrary to the prediction of YS.<sup>14</sup>

Next, we estimate the various terms in  $K^{(3)}$ . (The diagrammatic representation of  $K^{(3)}$  is given in Fig. 1.) There are many cumbersome terms unlike the linear response case.<sup>15)</sup> The approximation used is as follows. The three-point vertex correction connected with the odd order of the external field is omitted. This is because the velocity is an odd function in wave-number space and the integral is small. The same approximation holds in some types of the six- and eight-point irreducible vertices. If we consider a system with a strong momentum dependence such as the underdoped region and its doping dependence, this type of vertex is necessary.<sup>16</sup>) We consider, however, mainly the temperature and the angle dependence, and then the above terms have a slight influence.

The formalism for deriving  $\delta\lambda(H)$  consists of the Maxwell equation with the specular boundary condition (here, we consider the situation where the external magnetic field is applied parallel to the *y*-axis and the superconductor occupies z > 0)  $\frac{d^2A_x(z)}{dz^2} = 2H\delta(z) - \frac{4\pi}{c}J_x(z)$ , (*H* is the applied external field) and the nonlinear Pippard equation (eq. (1)) From these two equations the nonlinear equation for *A* is obtained and is solved by the perturbation method (not self-consistently) because we use the perturbation method to obtain the response



Fig. 1. Diagrammatic representation of  $K^{(3)}$ . The solid lines express the propagator of the electron in the Nambu representation. The wavy lines express the electro magnetic fields. The vertex with *n*-wavy lines denotes the *n*-times derivative of the dispersion of electrons and the circle at the vertex means a diagonal matrix  $\hat{\tau}_3$  with  $\text{Tr}\hat{\tau}_3 = 0$ . The vertex with a shaded triangle satisfies the integral equation with a four-point irreducible vertex  $I^{(2)}$ . The shaded rectangle denotes the reducible four-point vertex  $\Gamma^{(2)}$ .  $I^{(3)}$  and  $I^{(4)}$  represent the irreducible six- and eight- point vertices, respectively.

kernel  $K^{(3)}$ . Then, the nonlinear correction for  $\lambda$  is obtained as (the magnetic field is parallel to the intralayer crystal axis)

$$\delta\lambda_{ab} = -8H^2 \frac{4\pi}{c} \int \frac{dq}{2\pi} \int \frac{dq'}{2\pi} \int \frac{dq''}{2\pi} K^{(3)}_{xxxx}(q,q',q'') \times D_{xx}(q'') D_{xx}(q'-q'') D_{xx}(q-q') D_{xx}(q), \qquad (4)$$

where the definition of the magnetic field penetration depth is  $\lambda := \frac{1}{H} \int_0^\infty H_y(z) dz = -\frac{1}{H} \int \frac{dq}{2\pi} A_x(q)$ , and  $D_{xx}(q) := 1/(q^2 + \frac{4\pi}{c} K_{xx}^{(1)}(q))$ .

First, we consider the terms without the intermediate-states interaction. We classify these terms into two groups. One group consists of divergent terms without nonlocality and the other of terms similar to  $K^{(1)}$  (the nonlocality is negligible).  $K^{(3a)}$  and  $K^{(3b,c,d,e)}$  are categorized into the former and the latter groups, respectively. We estimate these terms only with the experimentally observed values. (On the other hand, a specific microscopic model and approximation are needed in the case of the intermediate-states interaction.) It is difficult to analytically calculate the paramagnetic term  $K^{(3a)}$  at a finite temperature, however, by noting that  $E_{k+q} - E_k \simeq v_{\perp}q$  near nodes and the characteristic value of q is  $\lambda^{-1}$  it can be shown that there is a crossover temperature  $T_0 \simeq \frac{\hbar v_{\perp}}{\lambda} \simeq \frac{\xi_c}{\lambda} \Delta_0$  ( $\xi_c$  is the interlayer coherence length). By substituting  $K^{(3a)}(q,q',q'')$  into eq. (4),  $\delta\lambda^{(3a)} = \left(\frac{H}{H_c}\right)^2 \lambda\kappa^{(3a)}(\frac{T_0}{\Delta_0})$ . Here,  $\kappa^{(3a)}(\frac{T_0}{\Delta_0})$  is a dimensionless quantity and its integration is calculated numerically. ( $H_c := \phi_0/2\sqrt{2\pi\xi_{ab}\lambda}$  is the

thermodynamic critical field,  $\phi_0 = 2\pi\hbar c/2e$ ,  $\xi_{ab}$  is the in-plane coherence length).  $\kappa^{(3a)}(\frac{T_0}{\Delta_0})$ is strongly dependent on temperature and shows a maximum around  $T \simeq T_0$ . If we put  $\lambda = 1600\text{\AA}, T_0/\Delta_0 = 0.01$  and  $H_c = 8000\text{G}, \delta\lambda^{(a)} \lesssim 0.2\text{\AA}$  for H = 200G; this is small compared with the experimental result  $\delta\lambda|_{\exp} \simeq 5\text{\AA}. \delta\lambda^{(b,c,d,e)}$  is smaller than one tenth of  $\delta\lambda$  due to the intermediate-states interaction discussed below and is negligible.

Next, we consider the intermediate-states interaction. (The local approximation holds as in the case of (b,c,d,e) and  $K^{(1)}$ .) The terms are calculated after they are transformed into equations which explicitly equal 0 when  $\Delta = 0$ . The term with  $\Gamma^{(2)}$  ((g)-type) is evaluated as follows. The reducible four-point vertex satisfies

$$\breve{\Gamma}^{(2)}(k,k') = \breve{I}^{(2)}(k,k') + \frac{T}{V} \sum_{k''} \breve{I}^{(2)}(k,k'') \breve{g}(k'') \breve{\Gamma}^{(2)}(k'',k')$$
(5)

with the irreducible four-point vertex  $\check{I}^{(2)}(k, k')$ . ('denotes the 4×4 matrix in the particle-hole space.) We use eq. (24) in ref.17 as  $\check{g}(k)$ . There are various terms in  $\check{I}^{(2)}(k, k')$ , the particlehole vertex, the particle-particle vertex, the number-nonconserving vertices. These are the functional derivatives of the self-energy by Green's function (the conserving approximation<sup>18</sup>). One of these vertices (the number-conserving particle-hole vertex) is written as follows with the FLEX approximation (the FLEX approximation in the superconducting state, for example, see refs.19 and 20)

$$I_c(k,k') = V_{k-k'}^n - \frac{T}{V} \sum_q G_{k-q} W_q(G_{k'-q} + G_{k'+q}).$$
(6)

Here,

$$V_q^n = U\left[U\chi_q^s + \frac{3}{2}\frac{(U\chi_q^s)^2}{1 - U\chi_q^s} - \frac{1}{2}\frac{(U\chi_q^c)^2}{1 + U\chi_q^c}\right],\tag{7}$$

with  $\chi_q^{s,c} = -\frac{T}{V} \sum_k (G_{k+q}G_k \pm F_{k+q}F_k)$ , ((s,c) correspond to (+,-), and

$$W_q = U^2 \left[ \frac{3}{2} \frac{1}{(1 - U\chi_q^s)^2} + \frac{1}{2} \frac{1}{(1 + U\chi_q^c)^2} - 1 \right].$$
 (8)

The other vertices are derived in the same way. We adopt the Hubbard model with the on-site Coulomb interaction U and take the same dispersion of electrons as in ref.16. The terms with  $I^{(3)}$  and  $I^{(4)}$  are calculated in the same way (e.g.,  $I^{(3)}(k_1, k_2, k_3) = \delta^2 \hat{\Sigma}_{k_1} / \delta \hat{G}_{k_3} \delta \hat{G}_{k_2}$ ). We calculate  $K^{(3)}$  without integrating out the incoherent part to derive a low-energy expression (e.g., ref.16.) Therefore, the effect of the renormalization factor is implicitly included.

To compare the calculation with the experimental results quantitatively, we consider the following quantity

$$\frac{1}{H^2} \frac{\delta\lambda(H)}{\lambda} = -\frac{\pi}{16} \frac{1/\phi_0^2}{(e/\hbar c)^2 K^{(1)} t/a} \frac{a^2 K^{(3)}}{K^{(1)}}.$$
(9)

Here, a is the lattice constant. We numerically calculate  $K^{(3)}$  and  $K^{(1)}$  by putting t = 1 and a = 1 and quantify the values of t and a in units of [eV] and [Å], respectively, and then

 $\frac{1}{H^2} \frac{\delta \lambda(H)}{\lambda} = -0.13 \times 10^{-13} \times \frac{a^3}{t} \frac{K^{(3)}}{(K^{(1)})^2} [\mathrm{G}^{-2}]. \text{ If we put } t = 0.25 [\mathrm{eV}] \text{ and } a = 8 [\mathrm{\AA}], \text{ we get} \frac{1}{H^2} \frac{\delta \lambda(H)}{\lambda} \simeq 0.35 \times 10^{-7} [\mathrm{G}^{-2}] \text{ for } U = 6.0 \text{ and the hole doping } \delta = 0.20. (K^{(3)} \simeq -12.8, K^{(1)} \simeq 0.1 \text{ and the dominant contribution comes from } K^{(3g)}. \text{ This value of } K^{(1)} \text{ yields } \lambda \simeq 2600 \mathrm{\AA}. \text{ This} \text{ is roughly 1.6 times longer than the experimental value.) On the other hand, the experimental result is <math>\frac{1}{H^2} \frac{\delta \lambda(H)}{\lambda} \simeq (0.7 \sim 1.0) \times 10^{-7} [\mathrm{G}^{-2}].^4$  Our calculation is quantitatively consistent with the experimental results in order of magnitude. As for the parameter dependence, the value of  $\frac{1}{H^2} \frac{\delta \lambda(H)}{\lambda}$  is not strongly dependent on the parameters U and  $\delta$  in the FLEX calculation. For example,  $\frac{1}{H^2} \frac{\delta \lambda(H)}{\lambda} \simeq 0.34 \times 10^{-7} [\mathrm{G}^{-2}]$  for U = 7.0 and  $\delta = 0.20$  and  $0.33 \times 10^{-7} [\mathrm{G}^{-2}]$  for U = 6.0 and  $\delta = 0.15$ . This is because the effect of the renormalization factor on  $K^{(3)}$  and  $(K^{(1)})^2$  cancels each other (as for U-dependence) and the integral-equation structure for  $\Gamma^{(2)}$  weakens the variation of the spin-fluctuation effect on  $I^{(2)}$ . If we put  $W_q \to U^2$ , this corresponds to the case of the weak spin fluctuation, for example, the more overdoped region, this results in a smaller value of  $\frac{1}{H^2} \frac{\delta \lambda(H)}{\lambda}$ . Therefore, an experimental study on the doping dependence is expected.

To investigate the angle dependence we consider the case where the applied field is parallel to the node direction  $(\delta \lambda_{45^{\circ}}(H))$ . In this case,  $K_{xxxx}^{(3)}$  in eq.(4) is replaced by  $(K_{xxxx}^{(3)} + 3K_{xxyy}^{(3)})/2$ . Then the relationship between  $K_{\mu\mu\mu\mu\mu}^{(3)}$  and  $K_{\mu\mu\alpha\alpha}^{(3)}$  with  $\mu \neq \alpha$  plays an important role in the angle dependence. If we consider a conventional s-wave superconductor, the relation  $K^{(3)}_{\mu\mu\mu\mu\mu} = 3K^{(3)}_{\mu\mu\alpha\alpha}|_{\mu\neq\alpha}$  holds because  $\langle v^4_{\mu} \rangle_{\rm FS} = 3 \langle v^2_{\mu}v^2_{\alpha} \rangle_{\rm FS}$  in the superconductor with no nodes and  $v_{\mu\mu\mu} = 0$  and  $v_{\mu\mu}^2 = v_{\mu\mu}v_{\alpha\alpha}$  hold in the electron gas.  $(< ... >_{\rm FS}$  denotes the average over the Fermi surface and  $v_{\mu\mu} = \partial v_{\mu}/\partial k_{\mu}$ , etc.) Both of these relations do not hold in the unconventional superconductor in the lattice system and therefore the relationship between  $K^{(3)}_{\mu\mu\mu\mu\mu}$  and  $K^{(3)}_{\mu\mu\alpha\alpha}|_{\mu\neq\alpha}$  is not trivial. In fact,  $K^{(3a)}_{xxxx}$  and  $K_{xxyy}^{(3a)}$  give the same contribution to  $\delta\lambda$  because a dominant contribution to the integral over the Fermi surface comes from nodes  $(v_x = v_y)$  at this point) except for  $T >> T_0$ . Then,  $\delta\lambda_{45^{\circ}}(H) = 2\delta\lambda_{ab}(H)$  in the conventional quasi-classical approximation.<sup>21</sup> We made sure above, however, that the intermediate-states interaction term contributes to  $\delta\lambda$  sufficiently and can be dominant. In this case,  $K_{xxyy}^{(3)} = K_{xxxx}^{(3)}/3$  with the approximation noted above and then  $\delta\lambda_{45^{\circ}}(H) = \delta\lambda_{ab}(H)$ . This explains the experimental results. (The reason for this is that the correlation between different vertices is broken by the intermediate-states interaction. For example,  $\int_{q} [\int_{k} G_{k} v_{k\mu} G_{k} v_{k\nu} G_{k} G_{k-q} W_{q} \int_{k'} G_{k'-q} G_{k'} v_{k'\alpha} G_{k'} v_{k'\beta} G_{k'}]$  is negligible for  $\mu \neq \nu$ or  $\alpha \neq \beta$ . The same holds for the case of  $I^{(3)}$  and  $I^{(4)}$ . This discussion also applies to the temperature dependence.)

The temperature dependence of  $\delta\lambda$  is as follows.  $\delta\lambda \propto 1/T$  for  $T > T_0$  in the conventional quasi-classical approximation. On the other hand, the temperature dependence of the intermediate-states interaction term is same as that of  $K^{(1)}$  (*T*-linear) and the decreasing rate compared with the value at T = 0 is almost same. Therefore,  $\delta\lambda$  shows a slight increase. (If  $\lambda$  increases 5Å per 1K as the experimental results indicate, the increasing rate of  $\delta\lambda$  is  $\frac{3\times 5}{\lambda/\delta\lambda} \simeq 0.015$  [Å/K]. This value is no larger than the experimental precision.)

The other phenomenon related to the NLME is the transverse magnetization. The predicted behavior in refs.1 and 2 is that the supercurrent is not perpendicular to H except for the case in which H is parallel to the nodes or the antinodes, and therefore the transverse magnetization has a period of  $\pi/2$  as the direction of H is rotated. Our perturbation theory shows that the supercurrent is written as follows in the arbitrary direction of H.

$$J_{\mu}(q) \simeq \{K_{\mu\mu}^{(1)}(q)A(q) + \int_{q'} \int_{q''} [K_{\mu\mu\mu\mu}^{(3)}(q,q',q'')X_{\theta}^{2} + 3K_{xxyy}^{(3)}(q,q',q'')Y_{\theta}^{2}]A(q'')A(q'-q'')A(q-q')\}X_{\theta}.$$
(10)

Here,  $(X_{\theta}, Y_{\theta}) = (\cos\theta, \sin\theta)$ ,  $(\sin\theta, \cos\theta)$  for  $\mu = x, y$ , respectively.  $\theta$  is the angle between the applied field and the intralayer crystal axis.) If the relation  $K^{(3)}_{\mu\mu\mu\mu\mu} = 3K^{(3)}_{\mu\mu\alpha\alpha}|_{\mu\neq\alpha}$  holds, the transverse magnetization does not appear. Then, we can have the same discussion as in the case of  $\delta\lambda$ .

Finally, we comment on previous studies. The nonlocal effect considered in ref.9 is different from our approach in several points. The behavior  $\delta\lambda \propto H^2$  at low H is seemingly the same as that in the perturbation approach. They predict, however,  $\delta\lambda \propto H$  above the crossover field  $H^*$  and argue that the NLME is unobservable owing to  $H^* > H_{c1}$ . They consider that the nonanalytical current exists above  $H^*$ . Although they do not consider the angle dependence of  $\delta\lambda$  and the transverse magnetization, their theory contradicts the experimental results. The origin of their error is that they consider  $K^{(1)}(q, A_{q=0})$ . They derive  $H^*$  by comparing the effect of q and A, however, it does not make sense to compare the intrinsic spatial variation with the external field. The absence (or very small value) of the transverse magnetization below the first vortex penetration<sup>22)</sup> implies the absence of  $H^*$ .

The quasi-classical approach in ref.23 gives observable values  $(\delta \lambda \simeq 1 \text{\AA} \text{ for} H \simeq 200[G])$  with the experimental parameters  $(H^* \simeq 2[T] \text{ and } \lambda/\xi \simeq 100)$ . Then, this also contradicts the experimental results in the angle and temperature dependences qualitatively. The cause is as follows. The interaction with the external field in the Gor'kov equation  $\frac{1}{2m} \left( \nabla_r + \frac{\nabla_R}{2} - i\frac{e}{c}A(R + r/2) \right)^2 G(r, R)$  is approximated as  $\frac{1}{2m} \left( \nabla_r + \frac{\nabla_R}{2} - i\frac{e}{c}A(R) \right)^2 G(r, R)$  in the quasi-classical approach. (The propagator is transformed as  $G(x, x') \to G(x - x', \frac{x+x'}{2}) = G(r, R)$ .) This means that the external field interacts with the center of mass of the electron propagator and therefore the nonlocal effect is underestimated. The comparison with our (a)-term is as follows. The Green function in the third order of the external field is written as

for the case of the nonlocal effect included correctly, but

in the quasi-classical approximation. This is interpreted as meaning that the magnetic field penetration depth effectively doubles in this approximation and then  $\delta\lambda$  roughly increases eightfold. Therefore, the quasi-classical term ((*a*)-term in our paper) makes less contribution to  $\delta\lambda$  if it is evaluated properly.

Larkin and Ovchinnikov suggest that the quasi-classical approximation does not give correct results in some cases.<sup>24)</sup> Our theory presents a definite example of this proposition.

In this paper, we present the microscopic formulation of the nonlinear Meissner effect. We show that the previous studies on this effect are insufficient and some of them are incorrect. The nonanalytical response is intrinsically absent. The experimental results possibly observe the intrinsic NLME. This is not YS's one, but originates from the intermediate-states interaction. We consider that this effect is interesting because it does not appear in the zeroth order of interactions but it reflects interactions between quasiparticles themselves. The spin fluctuation is quantitatively dominant in our calculation. This is consistent with the properties of the high- $T_c$  cuprates. Experiments on other materials and the theoretical investigations of various scattering mechanisms are expected in the future.

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- 13) The conventional quasi-classical approximation means the approximation which fails to treat the intermediate-states interaction properly, though an extension to include this effect remains an issue. The previous studies based on the quasi-classical approximation fall into this category.
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