Nonlinear Responses in Type-II Superconductors. I. Dirty Limit*

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A theoretical study is made of nonlinear responses of a dirty type-II superconductor to microwaves. The generation of a large third-harmonic current is found, arising from the nonlinearity of the dynamics governing the motion of the order parameter in the vortex state. Furthermore, the harmonic currents reveal interesting anisotropy, viz., a dependence on the angle between the direction of the microwave current and that of the dc magnetic field.

I. INTRODUCTION

Recently, there has been an increasing amount of theoretical work[1-10] on the dynamical behavior of the order parameter in superconductors. Abrahams and Tsuneto,[1] among others, have shown that in the vicinity of the transition temperature, where the order parameter is small, one can write a set of equations governing the behavior of the order parameter (i. e, the time-dependent Ginzburg-Landau equations). In this temperature range, the order parameter obeys a diffusionlike equation. More generally, in a gapless superconductor, where excitation of quasiparticles does not require any threshold energy, one can construct a more general formalism to deal with the dynamical behavior of the order parameter, as discussed by Caroli and Maki[4,5] and by Gor'kov and Éliashberg.[10] In a series of papers, Caroli and the present author[4,5] studied the fluctuation of the order parameter in the vortex state in type-II superconductors and found a class of collective modes with different helicity along the dc magnetic field. Furthermore, these modes are strongly coupled to the electromagnetic wave, resulting in a large anisotropy in the electromagnetic surface impedance of the vortex state.

In these articles,[4,5] we limit our discussions to the linear response to the electromagnetic wave. Since the basic equations which describe the motion of the order parameter and the electromagnetic wave are essentially nonlinear,[11] we expect large nonlinear effects in the vortex state.

In the present series of works, we shall study systematically the nonlinear response of a type-II superconductor in the high-field region. In this work we further limit our consideration to a dirty type-II superconductor, where the electronic mean free path is much shorter than the coherence distance. We find that, owing to the nonlinearity of the equation, a large third-harmonic current, easily accessible to experiment, is generated. We would like to stress here that these nonlinear responses are intrinsic to the vortex state and reflect directly the existence of the collective fluctuation of the order parameter. This is in sharp contrast to the nonlinear response in a normal metal, which is mainly due to dislocations in the crystal.[12] In fact, we neglect in the following consideration any effect which may arise from the existence of dislocations or pinning centers in actual crystals.

In Sec. II, we treat the nonlinear response in the framework of the time-dependent Ginzburg-Landau (TDGL) equations. In spite of their limitation (i. e., TDGL applies only in the temperature region close to T_c, the transition temperature), the author believes that TDGL is quite useful in clarifying the underlying physics. Furthermore, we can easily extend the results obtained in the framework of TDGL to all temperatures, at least for a dirty superconductor; this is done in Sec. III.

II. PRELIMINARY CONSIDERATIONS

In order to give a general insight into the problem of the nonlinear response in the vortex state, we start with the following set of equations, found by Abrahams and Tsuneto[1] and by Schmid.[2] In standard notation, we have

$$\left(\frac{\partial}{\partial t} - D(\vec{\nabla} - 2ie\vec{A})^2 - \epsilon_0(T)\right)\Delta^\dagger(\vec{r}, t) + R\left|\Delta(r, t)\right|^2 \Delta^\dagger(\vec{r}, t) = 0 , \qquad (1)$$

$$J(r, t) = \sigma\left(\frac{\partial\vec{A}}{\partial t}\right) - \frac{C}{i}(\vec{\nabla} - \vec{\nabla}' - 4ie\vec{A}) \times \Delta(\vec{r}, t)\,\Delta^\dagger(\vec{r}'t)\big|_{\vec{r}=\vec{r}'} , \qquad (2)$$

where $D = (lv/3)$, the diffusion constant, $\epsilon_0(T) = 2DH_{c2}(T)$, $R = 7\zeta(3)/2\pi^3 T_c$, $C = (e\pi/4T_c)D$, $\zeta(3) = 1.303\cdots$ is the Riemann's ζ function, and $H_{c2}(T)$ is the upper critical field.[13]

The above set of equations [i.e., the so-called TDGL equations describe the motion of the superconducting order parameter $\Delta(\vec{r}, t)$ in the presence of the time-dependent vector potential $\vec{A}(\vec{r}, t)$]. We do not enter into the discussion of the validity of TDGL,[14] but note that in the vortex state the above set of equations hold in the vicinity of the transition temperature.

According to Abrikosov,[15] in a magnetic field slightly smaller than $H_{c2}(T)$(we assume that a uniform field H_0 is applied along the z axis), the order parameter [i.e., the static solution of Eq. (1)] is given by

$$\Delta_0(\vec{r}) = \sum_{n=-\infty}^{\infty} c_n e^{inky} \exp\left[-eB\left(x - \frac{nk}{2eB}\right)^2\right], \quad (3)$$

which represents the two-dimensional lattice structure of vortex lines in the x-y plane. Here C_n, k are constants, n is an integer, and B is the induction. Abrikosov[15] showed further that the magnetization is given by

$$\vec{M} = -C\left|\Delta_0(\vec{r})\right|^2 , \quad (4)$$

which can be easily checked by substituting Eq. (3) in Eq. (2) and by solving the Maxwell equation for the magnetic fields. From the equilibrium condition,[15] the amplitude of $\Delta_0(\vec{r})$ is also determined by

$$\langle \left|\Delta_0(\vec{r})\right|^2 \rangle = \frac{2eD(H_{c2} - B)}{R\{\beta_A[1-(2\kappa^2)^{-1}] + (2\kappa^2)^{-1}\}} , \quad (5)$$

where κ is the Ginzburg-Landau parameter, $\beta_A = 1.16$, and $\langle a(\vec{r})\rangle$ is the space average of $a(r)$.

Now let us consider the electromagnetic response of the vortex state. For this purpose we introduce into Eq. (1) the microwave field through the vector potential $\vec{A}(\vec{r}, t)$ and then calculate the resulting current by making use of Eq. (2). It is very convenient to distinguish two situations for the following consideration.

A. Parallel Geometry

First let us consider the situation where the microwave electronic field E_ω is applied parallel to the dc magnetic field. We can take then the vector potential

$$\vec{A}(\vec{r}, t) = (0, A_0(\vec{r}), A_z(t)) , \quad (6)$$

where $A_0(\vec{r})$ describes the dc magnetic field:

$$A_0(\vec{r}) = Hx \quad A_z(t) = A_\omega e^{-i\omega t} . \quad (7)$$

Substituting $\vec{A}(\vec{r}, t)$ given in Eq. (5) into Eq. (1) we have

$$\left(\frac{\partial}{\partial t} - D(\vec{\nabla} - 2ieA_0)^2 + 4e^2DA_z^2(t) - \epsilon_0(T)\right) \times \Delta^+(\vec{r}, t) + R\left|\Delta(\vec{r}, t)\right|^2 \Delta^+(\vec{r}, t) = 0 . \quad (8)$$

Here we have already taken into account the fact that $\Delta(\vec{r}, t)$ does not depend on z, owing to the symmetry of the vortex structure. We note that the lowest-order modification of the order parameter is the second order in $A_z(t)$. Furthermore, since the term $A_z^2(t)$ does not mix the equilibrium state with the collective fluctuations of higher helicity (i.e., with those of higher magnetic quantum number along the z axis), we can assume without loss of generality that the solution of Eq. (8) is given by

$$\Delta(\vec{r}, t) = \Delta_0(\vec{r})\,\phi(t), \quad (9)$$

$$\left(\frac{\partial}{\partial t} - D(\vec{\nabla} - 2ie\vec{A}_0)^2 + 4e^2DA_z^2(t) - \epsilon_0(t) + R\left|\Delta_0(\vec{r})\right|^2\phi^2\right)\Delta_0^\dagger(\vec{r})\,\phi = 0 \quad (10)$$

Multiplying Eq. (10) by $\Delta_0(\vec{r})$ and taking the space average, we obtain

$$\left(\frac{\partial}{\partial t} + \frac{2eD(H_{c2} - H_0)}{1.16[1-(2\kappa^2)^{-1}]}[\phi(t)^2 - 1] + 4e^2DA_z^2(t)\right)\phi(t) = 0, \quad (11)$$

which is also yielded by Eq. (4) and the relation[15]

$$-\langle \Delta_0(r)[D(\vec{\nabla} - 2ie\vec{A}_0)^2 + \epsilon_0(T)]\,\Delta_0^\dagger(\vec{r})\rangle = R\langle \left|\Delta(\vec{r})\right|^4\rangle . \quad (12)$$

Making use of the fact that $A_z^2(t) \propto e^{-2i\omega t}$, we can solve Eq. (10) by iteration

$$\phi(t) = 1 - \frac{4e^2DA_\omega^2 e^{-2i\omega t}}{-2i\omega + 2k(H_0)} + \frac{(4e^2DA_\omega^2 e^{-2i\omega t})^2}{[-4i\omega + 2k(H_0)][-2i\omega + 2k(H_0)]} \times\left(1 - \frac{3k(H_0)}{-2i\omega + 2k(H_0)}\right) , \quad (13)$$

where

$$k(H_0) = \frac{2eD(H_{c2} - H_0)}{1.16[1-(2\kappa^2)^{-1}]} . \quad (14)$$

Thus, if we retain only the lowest-order correction, we will have

$$\Delta(\vec{r}, t) = \Delta_0(\vec{r})\left(1 - \frac{2e^2DA_z^2(t)}{-i\omega + k(H_0)}\right) . \quad (15)$$

Now substituting this expression of $\Delta(\vec{r}, t)$ in Eq. (2), we get the linear as well as nonlinear current

$$\vec{J}_z = i\omega\sigma\vec{A}_z(t) - 4eC\langle\left|\Delta_0(\vec{r})\right|^2\rangle \times\vec{A}_z(t)\,(1 - \{4eDA_z^2(t)/2[-i\omega + k(H_0)]\})^2 . \quad (16)$$

Here we replaced $\left|\Delta_0(\vec{r})\right|^2$ by its space average $\langle\left|\Delta_0(\vec{r})\right|^2\rangle$, since the penetration depth of the electromagnetic wave is much longer than the coherence distance $\xi(T)$ associated with the spatial variation of $|\Delta_0(\vec{r})|^2$.

From Eq. (15) we find easily the third-harmonic current

$$(J_{3\omega})_z = \frac{16CD(eA_\omega)^3\langle|\Delta_0(\vec{r})|\rangle^2}{-i\omega + k(H_0)}$$

$$= \{16(eA_\omega)^3/[-i\omega + k(H_0)]\}D|M| , \tag{17}$$

while the fundamental response current is given by the usual expression[4,16]

$$(J_\omega)_z = (i\omega\sigma - 4eC\langle|\Delta_0(\vec{r})|^2\rangle)A_\omega$$
$$= (i\omega\sigma - 4e|M|)A_\omega , \tag{18}$$

where M is the magnetization, already defined in Eq. (4). The third-harmonic field which will be emitted from the specimen is derived by solving the Maxwell equation

$$-\nabla^2(\vec{A}_{3\omega}) = 4\pi\sigma(3i\omega\vec{A}_{3\omega}) + 4\pi(\vec{J}_{3\omega}) . \tag{19}$$

When the penetration depth of the electromagnetic wave is much longer than the coherence distance, as is usually the case of a dirty superconductor, Eq. (19) is easily solved because of the simple spatial dependence of $\vec{J}_{3\omega}$

$$\vec{J}_{3\omega}(x) \propto |A_\omega(x)|^3 \propto \exp\{-3[(1+i)/\delta]x\} ,$$

where x is the distance from the surface and δ is the normal skin depth of the microwave with the frequency ω [i.e., $\delta = (2\pi\omega\sigma)^{-1/2}$]. We have, in fact,

$$A_{3\omega}(x) = \frac{1}{6i\omega\sigma}[J_{3\omega}(x)]_z = \frac{16[eA_\omega(x)]^3D|M|}{6i\omega\sigma[-i\omega + k(H_0)]} \tag{20}$$

and the corresponding electric field $E_{3\omega}$ is given by

$$E_{3\omega} = -3i\omega A_{3\omega} .$$

In order to estimate the relative importance of the third-harmonic field, it is convenient to compare it with the incident field E_ω

$$\frac{E_{3\omega}}{E_\omega} = -\frac{3i\times 16e^3A_\omega^2D|M|}{6i\omega\sigma[-i\omega + k(H_0)]} = \frac{8e^3A_\omega^2D|M|}{\omega\sigma|-i\omega + k(H_0)|} . \tag{21}$$

Furthermore, making use of the fact $H_\omega \cong \delta A_\omega$, we have finally

$$\left|\frac{E_{3\omega}}{E_\omega}\right| \sim \left|\frac{\epsilon_0(T)}{\omega}\right| \left|\frac{k(H_0)}{-i\omega + k(H_0)}\right|$$
$$\times\kappa^{-2}\left(\frac{\delta}{\xi(T)}\right)^4 \left(\frac{H_\omega}{H_{c2}(T)}\right)^2 , \tag{22}$$

where H_ω is the incident microwave magnetic field. We have here an appreciable third-harmonic field, because of the presence of large factors such as $\epsilon_0(T)/\omega$ and $[\delta/\xi(T)]^4$, where $\xi(T) = [2eH_{c2}(T)]^{-1/2}$.

We also point out that $E_{3\omega}/E_\omega$ has interesting frequency dependences due to the denominator $-i\omega + k(H_0)$.

In fact, it is always possible to choose the experimental conditions so that ω is in the vicinity of $k(H_0)$, where we can observe these dependences easily. $k(H_0)$ in the denominator is exactly the reciprocal of the characteristic damping term associated with fluctuations in the amplitude of the order parameter with a fixed lattice structure. We will see in Sec. III that the more general treatment, which is valid at all temperatures, results in the same expression for the third-harmonic current as given in Eq. (17). This fact implies *a posteriori* that TDGL is useful even in handling the phenomena at low temperatures, at least in the case of the dirty type-II superconductor

B. Perpendicular Geometry ($\vec{H}_0 \perp \vec{E}_\omega$)

Next we shall consider the perpendicular geometry where the microwave field E_ω is applied in a direction (which we will take as the x direction) perpendicular to the dc magnetic field. In the present case, the microwave exerts a Lorentz force on each vortex line and sets them in motion.[8] Because of this possibility, some special care is required in solving the equation for the order parameter [i.e., Eq. (1)]. In the present geometry we choose the vector potential

$$\vec{A}(\vec{r}, t) = (A_x(t), A_0(\vec{r}), 0). \tag{23}$$

Then Eq. (1) may be written as

$$\left(\frac{\partial}{\partial t} - D(\vec{\nabla} - 2ie\vec{A}_0)^2 + 4ieD(\vec{\nabla} - 2ie\vec{A}_0)A_x(t) + 4e^2DA_x^2(t) - \epsilon_0(T)\right)\Delta^\dagger(\vec{r}, t)$$
$$+ R|\Delta(\vec{r}, t)|^2\Delta^\dagger(\vec{r}, t) = 0 \tag{24}$$

and $A_x(t) = A_\omega e^{-i\omega t}$. Here we assume that the penetration depth of the electromagnetic wave δ is much larger than $\xi(T)$, the coherence length of the superconductor [i.e., $|\vec{\nabla}A_x(t)| = 0$] for simplicity.

As in the case of the linear response discussed in the Ref. 8, we can eliminate the linear term in $A_x(t)$ in Eq. (24) by making use of the moving solution, which satisfies the following equation:

$$\left(\frac{\partial}{\partial t} - D(\vec{\nabla} - 2ieA_0)^2 + 4ieD(\vec{\nabla} - 2ieA_0)A_x(t) + 4e^2DA_x^2(t) - 2eDB + 4e^2B^2Df^2(t)\right)$$
$$\times\Delta_f^\dagger(\vec{r}, t) = 0 , \tag{25}$$

where

$$\Delta_f^\dagger(\vec{r}, t) = \sum_n C_n e^{ik_n y} \exp\left[-eB\left(x - \frac{k_n}{2eB} - if(t)\right)^2\right] \tag{26}$$

and
$$f(t) = \frac{2\epsilon_0(T)}{2\epsilon_0(T) - i\omega}\frac{A_x(t)}{B} . \tag{27}$$

This solution represents a helical oscillation of the vortex structure. In the limit $\omega \to 0$, the above solution reduces essentially to that in the flux-flow regime[2,5] (i.e., the uniform motion of the vortex structure in the x direction). Now assuming that the complete solution in the present geometry is given by

$$\Delta^\dagger(\vec{r}, t) = \Delta_f^\dagger(\vec{r}, t)\phi_f(t) , \tag{28}$$

and substituting this in Eq. (24), we have

$$\Delta_f^\dagger(\vec{r}, t)\left(\frac{\partial}{\partial t}\;\phi_f(t) + 4e^2DA_x^2(t)[1-\eta^2(\omega)]\phi_f(t) + [2eDB - \epsilon_0(T)]\phi_f(t) + R\,|\Delta_f(r, t)|^2\phi_f^3(t)\right) = 0, \tag{29}$$

where $$\eta(\omega) = \frac{Bf(t)}{A_x(t)} = \frac{2\epsilon_0(T)}{2\epsilon_0(T) - i\omega} . \tag{30}$$

Here we have made use of Eq. (25). Finally, multiplying Eq. (29) by $\Delta_f(\vec{r}, t)$ and taking the space average, we have

$$\left(\frac{\partial}{\partial t} + 4e^2DA_x^2(t)[1-\eta^2(\omega)] + k(H_0)[\phi_f^2(t) - 1]\right)\phi_f(t) = 0 . \tag{31}$$

Equation (31) is of the same structure as Eq. (11) and we can solve it by iteration. Keeping only the lowest-order term in $A_x^2(t)$ we have

$$\phi_f(t) = 1 - \frac{4e^2D[1-\eta^2(\omega)]}{2[-i\omega + k(H_0)]}\,A_x^2(t) + O(A_x^4(t)) \tag{32}$$

or

$$\Delta^\dagger(\vec{r}, t) = \Delta_f^\dagger(r, t)\left(1 - \frac{4e^2D[1-\eta^2(\omega)]}{2[-i\omega + k(H_0)]}\,A_x^2(t)\right) . \tag{33}$$

As in the case of the parallel geometry, $\phi_f(t)$ describes the fluctuation of the amplitude of $\Delta_f^\dagger(r, t)$.

Since the factor $1-\eta^2(\omega)$ vanishes for $\omega \to 0$, we conclude that in the perpendicular geometry the fluctuation in the amplitude of $\Delta^\dagger(\vec{r}, t)$ is suppressed by a factor $[1-\eta^2(\omega)]$ in comparison with that in the parallel geometry. Substituting then $\Delta^\dagger(\vec{r}, t)$ thus found in Eq. (2), we have

$$J_x(r, t) = i\omega\sigma A_x(t) - 4eC\,\frac{i\omega}{2\epsilon_0(T) - i\omega} \times A_x(t)\langle|\Delta_f|^2\rangle\left(1 - \frac{4e^2D[1-\eta^2(\omega)]}{2[-i\omega + k(H_0)]}\,A_x^2(t)\right)^2 . \tag{34}$$

From this we have the fundamental current[8]

$$(J_\omega)_x = i\omega\left(i\sigma - 4eC\,\frac{1}{2\epsilon_0(T) - i\omega}\,\langle|\Delta_f|^2\rangle\right)A_x(t) = i\omega\,\left(\sigma - \frac{4e\,|M|}{2\epsilon_0(T) - i\omega}\right)A_x(t) \tag{35}$$

and the third-harmonic current

$$J_{3\omega} = 4eC\,\frac{i\omega}{2\epsilon_0(T) - i\omega}\,\frac{4e^2D[1-\eta^2(\omega)]}{-i\omega + k(H_0)}\,\langle|\Delta_f|^2\rangle A_x^3(t) , = -\frac{16[eA_x(t)]^3}{-i\omega + k(H_0)}\left(\frac{i\omega}{2\epsilon_0(T) - i\omega}\right)^2\left(\frac{4\epsilon_0 - i\omega}{2\epsilon_0 - i\omega}\right)\,D|M| . \tag{36}$$

Making use of an analysis similar to that in Sec. II A, we find easily that at the surface of the specimen the third-harmonic electric field radiated from the specimen is given by

$$E_{3\omega} = \frac{8(eA_x)^2}{\sigma}\,\frac{1}{-i\omega + k(H_0)}\left(\frac{i\omega}{2\epsilon_0 - i\omega}\right)^2 \times\left(\frac{4\epsilon_0 - i\omega}{2\epsilon_0 - i\omega}\right)\,D|M| . \tag{37}$$

As is seen in Eq. (34), the third-harmonic response in the present case is strongly reduced by a factor $(\omega/\epsilon_0)^2$ in the low-frequency limit in comparison with that in the parallel geometry. However, in the frequency range $\omega \sim \epsilon_0(T)$, the third harmonic in the present geometry becomes appreciable. Furthermore, the amplitude of the third-harmonic current in the present geometry has a much stronger frequency dependence. This difference comes from the fact that in the parallel geometry the microwave excites the fluctuation of the amplitude of the order parameter, while in the perpendicular geometry the microwave excites the helical oscillation of the vortex structure, reducing the amplitude fluctuation. Since the nonlinear response is essentially related to the amplitude fluctuation of the order parameter (as seen above), we have a smaller nonlinear response in the perpendicular geometry.

C. Arbitrary Geometry

It is not difficult to consider the nonlinear response in the arbitrary geometry, where the microwave field E_ω is applied with an angle to the dc magnetic field. We have then the third-harmonic currents both in the parallel and in the perpendicular direction to the dc field. For this purpose let us consider the case where the vector potential is given by

$$\vec{A}(\vec{r}, t) = (A_x(t), A_0(\vec{r}), A_z(t)),$$

where

$$A_x(t) = A(t)\sin\theta \quad \text{and} \quad A_z(t) = A(t)\cos\theta .$$

It is then easy to see that now $\Delta(\vec{r}, t)$ [the solution of Eq. (11)] is given by

$$\Delta^\dagger(\vec{r},t)=\Delta_f^\dagger(\vec{r},t)\times\left(1-\frac{2e^2D}{-i\omega+k(H_0)}\{A_z^2(t)+[1-\eta^2(\omega)]A_x^2(t)\}\right)=\Delta_f^\dagger(\vec{r},t)\left(1-\frac{2e^2DA^2(t)}{-i\omega+k(H_0)}\times\{\cos^2\theta+[1-\eta^2(\omega)]\sin^2\theta\}\right), \tag{38}$$

where $\Delta_f^\dagger(\vec{r},t)$ has already been defined in Eq. (26).

Making use of this expression for the order parameter, the third-harmonic currents in the parallel direction and in the perpendicular direction to the dc field is given by

$$J_{3\omega}=\frac{16(eA_\omega)^3}{-i\omega+k(H_0)}D|M|\{\cos^2\theta+\sin^2\theta[1-\eta^2(\omega)]\}\cos\theta \tag{39}$$

and

$$J_{3\omega}=\frac{16(eA_\omega)^3}{-i\omega+k(H_0)}\frac{i\omega}{2\epsilon_0(T)-i\omega}\times D|M|\{\cos^2\theta+\sin^2\theta[1-\eta^2(\omega)]\}\sin\theta . \tag{40}$$

Equations (39) and (40) show that the third-harmonic currents have a strong anisotropy, depending on the angle θ between the microwave field $\vec{E}_\omega$ and the dc field $\vec{H}_0$. As already explained, this reflects the fact that the parallel microwave excites only the amplitude fluctuation of the order parameter, while the perpendicular microwave excites both the helical motion of the vortex structure and the amplitude fluctuation. Therefore, the measurement of the nonlinear response provides a useful means of studying the dynamic behavior of the vortex state.

III. THIRD-HARMONIC CURRENT

In Sec. II we studied the nonlinear response of the vortex state in the framework of TDGL and found a large third-harmonic current. We are going to show in this section that most of the results obtained there hold in wider context (i.e., at all temperatures) by making use of the general equations for the time-dependent order parameter as obtained by Caroli and the present author.[5] In fact, in a dirty superconductor and in the relatively low-frequency region [i.e., $\omega/\epsilon_0(0)\ll 1$], we will have exactly the same expression for the third-harmonic current as that found in Sec. II. In a dirty superconductor in a high magnetic field H_0 [slightly smaller than $H_{c2}(T)$], the motion of the order parameter is described by the following set of equations[5]:

$$\{\ln T/T_{c0}+\psi[\tfrac{1}{2}+\Lambda/4\pi T]-\psi(\tfrac{1}{2})\}\Delta^\dagger(\vec{r},t)+[1/16(\pi T)^2]\psi^{(2)}(\tfrac{1}{2}+\rho)|\Delta(\vec{r},t)|^2\Delta(\vec{r},t)=0 \tag{41}$$

and

$$\vec{J}(\vec{r},t)=\sigma\left(\frac{\partial\vec{A}}{\partial t}\right)+\frac{eiN}{2m}(\vec{q}_1-\vec{q}_2)\left\{(2\omega_1+\Lambda_2-\Lambda_1)^{-1}\times\left[\psi\left(\frac{1}{2}+\frac{2\omega_1+\Lambda_2}{4\pi T}\right)-\psi\left(\frac{1}{2}+\frac{\Lambda_1}{4\pi T}\right)\right]+(2\omega_2+\Lambda_1-\Lambda_2)^{-1}\times\left[\psi\left(\frac{1}{2}+\frac{2\omega_2+\Lambda_1}{4\pi T}\right)-\psi\left(\frac{1}{2}+\frac{\Lambda_2}{4\pi T}\right)\right]\right\}\times\Delta(1)\Delta^\dagger(2)\Big|_{1=2=(\vec{r},t)}, \tag{42}$$

where Λ_i are the operators defined by

$$\Lambda_1=\omega_1+Dq_1^2,\qquad \Lambda_2=\omega_2+Dq_2^2,$$
$$\omega_1=i\frac{\partial}{\partial t_1},\qquad \omega_2=i\frac{\partial}{\partial t_2}, \tag{43}$$
$$q_1=-i\vec{\nabla}_1-2e\vec{A}(1),\quad q_2=-i\vec{\nabla}_2+2e\vec{A}(2)$$

and $\psi_{(z)}$, $\psi^{(1)}_{(z)}$ are the digamma and the trigamma function, respectively. Furthermore, ρ is defined by

$$\rho=DeH_{c2}(T)/2\pi T .$$

The above set of equations may be called the generalized TDGL. In particular, in the vicinity of the transition temperature, where $\omega/\pi T$ and $\Lambda/\pi T$ are small operators, Eqs. (41) and (42) reduce to Eqs. (1) and (2), respectively.

In the following, we assume as in Sec. II that a static magnetic field H_0 (slightly smaller than H_{c2}) is applied in the z direction. We shall consider again the same special geometries.

A. Parallel Geometry ($\vec{H}_0\parallel\vec{E}$)

First let us consider the case where the microwave electric field is applied parallel to the dc magnetic field. Then, as previously, we assume

$$\vec{A}(\vec{r},t)=(0,A_0(x),A_z(t)) . \tag{44}$$

Substituting $\vec{A}(\vec{r},t)$ in Eq. (41), we can in principle determine $\Delta(\vec{r},t)$. However, we are only interested here in the second-order corrections to $\Delta(\vec{r},t)$ in $A_z(t)$; thus we first simplify Eq. (41) by expanding $\psi(\frac{1}{2}+\Lambda/4\pi T)$ in the vicinity of $\psi(\frac{1}{2}+\rho)$:

$$\psi\left(\frac{1}{2}+\frac{\Lambda}{4\pi T}\right)=\psi(\tfrac{1}{2}+\rho)+\psi^{(1)}(\tfrac{1}{2}+\rho)\frac{\Lambda-\epsilon_0(T)}{4\pi T}\cdots , \tag{45}$$

where $\epsilon_0(T)=2DeH_{c2}(T)$.

Then we can recast Eq. (41) in the form

$$\left(\frac{\partial}{\partial t} - D[\vec{\nabla} - 2ei\vec{A}(r,t)]^2 - \epsilon_0(T)\right)\Delta^\dagger(r,t) + \frac{1}{4\pi T}\frac{\psi^{(2)}(\frac{1}{2}+\rho)}{\psi^{(1)}(\frac{1}{2}+\rho)}|\Delta(\vec{r},t)|^2\Delta^\dagger(\vec{r},t) = 0\ , \quad (46)$$

which has exactly the same form as Eq. (1), where R is now given by

$$R = \frac{1}{4\pi T}\frac{\psi^{(2)}(\frac{1}{2}+\rho)}{\psi^{(1)}(\frac{1}{2}+\rho)}\ . \quad (47)$$

Then the solution of Eq. (46) is readily found to be

$$\Delta(\vec{r},t) = \Delta_0(\vec{r})\left(1 - \frac{2e^2DA_z^2(t)}{-i\omega + k(H_0)}\right)\ , \quad (48)$$

with $k(H_0)$ now given by

$$k(H_0) = \{2eD[H_{c2}(T) - H_0]/1.16[1-(2\kappa_2^2(T))^{-1}]\}. \quad (49)$$

Equation (48) is exactly the same that obtained using the TDGL. However, note the appearance[6,7] of the parameter $\kappa_2(T)$ in the definition of $k(H_0)$.[17,18]

Now substituting the order parameter $\Delta(\vec{r},t)$ in Eq. (42) and noting that

$$\Lambda\Delta(\vec{r},t) = \epsilon_0(T)\Delta(\vec{r},t)\ ,$$

we have

$$J_z(\vec{r},t) = \sigma\left(-\frac{\partial}{\partial t}A_z(t)\right) - \frac{\sigma}{\pi T}\langle|\Delta_0(\vec{r})|^2\rangle \times \left(\psi^{(1)}(\tfrac{1}{2}+\rho)A_z(t) - 2\pi T/-2i\omega \times[(\tfrac{1}{2} - i\omega/\pi T + \rho) - \psi(\tfrac{1}{2}+\rho)]\frac{4e^2DA_z^3(t)}{-i\omega + k(H_0)}\right). \quad (50)$$

From this the third-harmonic current is easily found to be

$$(J_{3\omega})_z = 2\sigma\langle|\Delta_0(\vec{r})|^2\rangle(1/-i\omega) \times[\psi(\tfrac{1}{2} - i\omega/\pi T + \rho) - \psi(\tfrac{1}{2}+\rho)]\frac{4e^2DA_z^3(t)}{-i\omega + k(H_0)}$$

$$\simeq \frac{8e^2D\sigma A_z^3(t)}{\pi T[-i\omega + k(H_0)]}\langle|\Delta_0(\vec{r})|^2\rangle\psi^{(1)}(\tfrac{1}{2}+\rho)\ , \quad (51)$$

$$(J_{3\omega})_z = 16[eA_z(t)]^3D|M|/[-i\omega + k(H_0)]\ . \quad (52)$$

Thus we find in the low-frequency limit $\omega \ll \epsilon_0(0)$ exactly the same expression for the third-harmonic current that we have already obtained in Sec. II. We may conclude, therefore, that as long as we are interested in the low-frequency response the TDGL are extremely useful.

Now the calculation of $E_{3\omega}$ can be done exactly as described in Sec. II. Consequently, we shall not pursue this problem further here. We note only that the ratio $E_{3\omega}/E_\omega$ is exactly given by Eq. (21), where $k(H_0)$ is now given in Eq. (49). Therefore, the estimate of this ratio made in Sec. II still holds in the present, more general context.

B. Perpendicular Geometry ($\vec{H}_0 \perp \vec{E}$)

As already mentioned in Sec. II in the present geometry the microwave induces the helical motion of the vortex lattice. However, we can still use the same simplification as described in Sec. III A. We find now that the order parameter is given exactly by Eq. (33) with $k(H_0)$ defined in Eq. (49). Here we assumed the vector potential $\vec{A}(\vec{r},t) = (A_x(t), A_0(r), 0)$ as in Sec. II. Substituting $\Delta^1(\vec{r},t)$ in Eq. (42), we obtain the current

$$J_x(\vec{r},t) = \sigma\left(-\frac{\partial}{\partial t}A_x(t)\right) + \frac{\sigma}{\pi T}\langle|\Delta_f(r)|^2\rangle\left\{\frac{2\pi T}{-i\omega}\times\left[\psi\left(\tfrac{1}{2} - \frac{i\omega}{2\pi T} + \rho\right) - \psi(\tfrac{1}{2}+\rho)\right]\frac{i\omega}{2\epsilon_0(T) - i\omega}A_x(t) - \frac{2\pi T}{-3i\omega}\left[\psi\left(\frac{1}{2} - \frac{3i\omega}{2\pi T} + \rho\right) - \psi\left(\frac{1}{2}+\rho\right)\right] \times\frac{i\omega}{2\epsilon_0(T) - i\omega}\frac{[1-\eta(\omega)^2]4e^2DA_x^3(t)}{-i\omega + k(H_0)}\right\}\ . \quad (53)$$

The third-harmonic current is then easily found to be

$$(J_{3\omega})_x = -\frac{2\sigma}{3(2\epsilon_0 - i\omega)}\frac{[1-\eta^2(\omega)]4e^2DA_x^3(t)}{-i\omega + k(H_0)} \times\{\psi[\tfrac{1}{2} - 3i\omega/2\pi T + \rho] - \psi(\tfrac{1}{2}+\rho)\}\ \langle|\Delta_f|^2\rangle\ , \quad (54)$$

$$(J_{3\omega})_x \cong -\frac{\sigma}{\pi T}\psi^{(1)}(\tfrac{1}{2}+\rho)\langle|\Delta_f|^2\rangle \times\frac{i\omega}{2\epsilon_0 - i\omega}\ \frac{1-\eta^2(\omega)}{-i\omega + k(H_0)}\ 4e^2DA_x^3(t)\ ,$$

$$(J_{3\omega})_x = -\frac{16[eA_x(t)]^3}{-i\omega + k(H_0)}\left(\frac{i\omega}{2\epsilon_0(T) - i\omega}\right)^2 \times\frac{4\epsilon_0(T) - i\omega}{2\epsilon_0(T) - i\omega}\ D|M|\ , \quad (55)$$

which is equivalent to Eq. (36). In the passage from Eq. (54) to Eq. (55), we have made use of the approximation $\omega \ll \epsilon_0(0)$, which is usually valid for the microwave range. We again find that the full microscopic treatment results in the same expression that we obtained with the help of TDGL if we limit ourselves to low-frequency phenomena.

C. Aribitrary Geometry

It is unnecessary to repeat the calculation here. We can easily extend the validity of Eqs. (39) and (40) to lower temperatures following the same reasoning. We conclude, therefore, that in a dirty type-II superconductor we can use the results obtained in the framework of TDGL at all temperatures, provided the frequency of the microwave ω is sufficiently low [i.e., $\omega \ll \epsilon_0(0)$]. For the Pb-In alloys, for example, with $T_c \cong 6\ °K$, the above condition gives $\omega \ll 10^{12}\ \text{sec}^{-1}$, which is completely satisfied in the usual experimental conditions. On the other hand, for experiments using a frequency in the far-infrared range and specimens with relatively low T_c (say, $T_c \leqq 1\ °K$), we have to use a more general form as given in Eqs. (51) and (54). However, under these conditions we can no longer use the simplification of Eq. (41) as achieved in Eq. (45), and a more delicate treatment of the problem is then required.

IV. CONCLUDING REMARKS

We have here considered the intrinsic nonlinear responses in the vortex state of a dirty type-II superconductor, first in the framework of TDGL and then in the generalized TDGL. We find as long as we are concerned with the low-frequency limit [i.e., $\omega \ll \epsilon_0(0)$], TDGL gives results essentially valid at all temperatures. Substantial generation of third-harmonic current can be observed, which is associated with the amplitude fluctuation of the superconducting order parameter in the vortex state. The emitted third-harmonic field is estimated to be of the order of

$$\left|\frac{E_{3\omega}}{E_\omega}\right| = \left|\frac{\epsilon_0(T)}{\omega}\right| \times \left|\frac{k(H_0)}{-i\omega + k(H_0)}\right| \times \kappa^{-2}\left(\frac{\delta}{\xi(T)}\right)^4 \left(\frac{H_\omega}{H_{c2}(T)}\right)^2 \quad \text{for } \vec{H}_0 \parallel \vec{E}_\omega, \tag{56}$$

$$\left|\frac{E_{3\omega}}{E_\omega}\right| = \left|\frac{\epsilon_0(T)}{\omega}\right| \times \left|\frac{\omega}{2\epsilon_0(T) - i\omega}\right|^2 \times \left|\frac{k(H_0)}{-i\omega + k(H_0)}\right| \kappa^{-2}\left(\frac{\delta}{\xi(T)}\right)^4 \left(\frac{H_\omega}{H_{c2}(T)}\right)^2 . \tag{57}$$

Here E_ω and H_ω are the incident microwave electric and magnetic fields, respectively. Since in the dirty type-II superconductor the factor $[\delta/\xi(t)]^4$ is extremely large, we expect a large third-harmonic response. We note also that the third-harmonic response is reduced by a factor $(\omega/2\epsilon_0)^2$ in perpendicular geometry from that in parallel geometry. This follows from the fact that in perpendicular geometry the microwave excites the helical motion of the order parameter, which cancels a part of the amplitude fluctuation giving rise to the nonlinear response. In the surface-sheath regime of a dirty superconductor, we expect similarly large nonlinear responses. However, the mathematical treatment is somewhat different from what we have done here. A discussion of such a situation will be presented in a future work.

In the pure superconductor also, we have large nonlinear responses as discussed here. However, we cannot simply transcribe the result obtained in Sec. II for this case, because the basic equations describing the changes in the order parameter and the current are much more complicated.[5,7] A detailed discussion of the nonlinear response in a pure type-II superconductor will be given in the second paper in this series.

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