Time-dependent Ginzburg-Landau theory with floating nucleation kernel: Far-infrared conductivity in the Abrikosov vortex lattice state of a type-II superconductor

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(Received 24 September 2009; revised manuscript received 25 November 2009; published 31 December 2009)

We formulate the time-dependent Ginzburg-Landau theory, with the assumption of local equilibrium made in the reference frame floating with normal electrons. This theory with floating nucleation kernel is applied to the far infrared conductivity in the Abrikosov vortex lattice. It yields better agreement with recent experimental data [Phys. Rev. B **79**, 174525 (2009)] than the customary time-dependent Ginzburg-Landau theory.

DOI: 10.1103/PhysRevB.80.212506 PACS number(s): 74.40.+k, 74.25.Ha, 74.25.Nf, 74.25.Qt

The time-dependent Ginzburg-Landau (TDGL) equation is a useful extension of the equilibrium Ginzburg-Landau theory. Unfortunately, microscopic derivations[1–5] guarantee its validity under such restrictive conditions that it seems more difficult to find justified nontrivial applications than to solve it. The TDGL equation is thus most often applied beyond its nominal range of validity.

As one leaves the familiar vicinity of the superconducting phase transition and asymptotically slow processes, the intuitive foundation of the theory becomes shaky. The TDGL theory contains an assumption of local equilibrium, which is dependent on reference frame; when we adapt the equilibrium-based equation to nonequilibrium problems, we should at least work in the reference frame in which electrons are as close to local equilibrium as possible. This is the frame floating with the normal current in the background of a superconducting condensate. To this end, in this paper we present what we refer to as a *floating nucleation kernel*.

The standard TDGL theory is formulated using a kernel static in the laboratory system. We will show that compared to the TDGL theory in the floating system, the laboratory formulation lacks a term which is particularly important at high frequencies of the driving field. We will demonstrate the effects of this term on the conductivity in the subgap far-infrared (FIR) region. Comparing our results with recent FIR magnetotransmission measurements of Ikebe *et al.*,[6] we will show that use of the floating nucleation kernel improves agreement between the theory and experimental data.

Let us first describe the magnetotransmission measurement. It is performed on a thin layer perpendicularly penetrated by the magnetic field in the form of vortices. The incident FIR light is perpendicular to the surface and its electric field drives currents, which determine the amplitude and phase of the transmitted light, which is measured.

Both the normal and the superconducting electrons are accelerated by the electric field and experience a friction with the lattice. The friction of the condensate is much weaker since Joule heat develops only in vortex cores moving perpendicularly to the electric field. The relative contribution of these components to the current depends on the frequency of the driving field; the higher the frequency the higher will be the fraction of the normal current.

It is useful to inspect characteristic times for NbN, the material used by Ikebe *et al.*[6] The optical gap $2\Delta = 5.3$ meV implies the maximal subgap frequency $\omega < 10$ THz. The mean time between two collisions of the normal electron is $\tau_n \sim 5$ fs, therefore, during a single period of the subgap FIR field, the electron loses momentum more than a hundred times. At zero magnetic field, the condensate suffers no friction. The field of amplitude E accelerates the condensate to velocity $e^*E/\omega m^*$, while a normal electron is accelerated to $eE\tau_n/m$. At the measurement temperature, $T = 3$ K and $T_c = 15$ K, the density of condensed electrons exceeds the normal density, therefore, the condensate clearly dominates the total current. A different situation obtains, however, for the Joule heat. The condensate current is out of phase with the driving electric field and generates no heat. The normal current is in-phase, producing heat. If the magnetic field penetrates the sample, the condensate generates the Joule heat due to motion of vortices. We will see that for the subgap FIR frequencies, the condensate Joule heat value is much smaller than the amount of heat generated by normal electrons.

To identify the Joule heat, it is necessary to measure the transmission coefficient, including its phase. This allows one to determine the complex conductivity σ with $\mathrm{Im}\,\sigma$ giving the off-phase current and $\mathrm{Re}\,\sigma$ for the in-phase current. Ikebe *et al.*[6] achieved this task by splitting short pulses and mixing them again after one of branches passed through the sample. As mentioned, we will compare their experimentally established σ with theoretical predictions based on the TDGL theory in the laboratory and the floating coordinate system.

We will use the electric field $\mathbf{E}(\tau) = \mathrm{Re}[\mathbf{E}e^{-i\omega\tau}]$ and current $\mathbf{J}(\tau) = \mathrm{Re}[\mathbf{J}e^{-i\omega\tau}]$. The complex conductivity is defined via $\mathbf{J} = \sigma\mathbf{E}$. The current has a small Hall component, which we neglect in our discussion for convenience.

The TDGL equation derived using the static kernel,[7]

$$\frac{1}{2m^*}\left(-i\hbar\nabla - \frac{e^*}{c}\mathbf{A}\right)^2\psi + \alpha\psi + \beta|\psi|^2\psi = -\Gamma\partial_\tau\psi, \qquad (1)$$

describes the evolution of the condensate including a relaxation of the GL function ψ toward its equilibrium value. The vector potential is that of the internal magnetic field as well as the electric field of the FIR light; $\mathbf{B} = \nabla\times\mathbf{A}$ and $\mathbf{E} = -(1/c)\partial_\tau\mathbf{A}$. The electric current

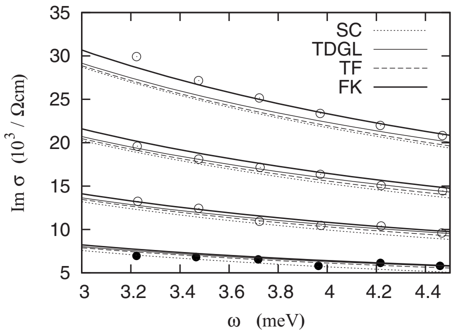

FIG. 1. Imaginary part of the conductivity giving nondissipative currents: thin lines are the superconducting condensate conductivity Im σ_s (dotted), the TDGL conductivity Im $\sigma_{\rm GL}$ (full), and the two-fluid modification of the TDGL conductivity Im $\sigma_{\rm tf}$ (dashed). The heavy line is the conductivity Im $\sigma_{\rm fk}$ evaluated in the floating system. Experimental data of Ikebe *et al.* (Ref. 6) at 7 T (•) are in the nominal validity range of the TDGL theory, while the lower magnetic fields 5 T, 3 T, and 1 T (○) are not.

$$\mathbf{j}_s = \frac{e^*}{m^*}\mathrm{Re}\left[\bar{\psi}\left(-i\hbar\nabla - \frac{e^*}{c}\mathbf{A}\right)\psi\right] \tag{2}$$

is composed of circulating diamagnetic currents and oscillating response to the light. We solve Eq. (1) to linear order in $\mathbf{E}$ and eliminate the diamagnetic currents by averaging over the elementary cell of the Abrikosov vortex lattice; $\mathbf{J}_s = \langle \mathbf{j}_s \rangle = (B/\Phi_0)\int_{\rm cell} dxdy\,\mathbf{j}_s$. The supercurrent, $\mathbf{J}_s = \sigma_s \mathbf{E}$, gives the condensate conductivity

$$\sigma_s = \frac{3\sigma_0}{\beta_{\rm A}}\frac{1-t-b}{b-i\omega\tau_s}, \tag{3}$$

where $t=T/T_c$, $b=B/H_{c2}$ are the dimensionless temperature and magnetic field, σ_0 is the normal state conductivity, $\beta_{\rm A} = 1.16$ is the Abrikosov constant for hexagonal vortex lattice, and $\tau_s = \Gamma(1-t)/\alpha$. Deriving Eq. (3), we have used the GL parameter[8]

$$\Gamma = \frac{12\pi\sigma_0\alpha\kappa^2\xi^2}{c^2(1-t)^2}. \tag{4}$$

The zero-temperature coherence length is determined by the upper critical field; $\xi^2 = \Phi_0/(2\pi H_{c2}^0)$. Here, $H_{c2}^0 = 15$ T is obtained via the linear extrapolation $H_{c2} = H_{c2}^0(1-t)$ from experimental data in Fig. 3 of Ref. 6. The normal-state conductivity $\sigma_0 = 2\cdot 10^4/\Omega$ cm, experimentally established[6] at 20 K, has weak temperature dependence and can be used at 3 K.

In Fig. 1, one can see that the imaginary part of σ_s from formula (3) reproduces recent experimental data of Ikebe *et al.*[6] Here, we use the GL parameter $\kappa = 40$, the only fitting parameter in the present theory. It is adjusted to fit the imaginary part of the conductivity at 7 T. Our main interest is in the Joule heat given by the real part of the conductivity.

Formula (3) was derived for the dense Abrikosov vortex lattice. Theoretically, the region of nominal validity is $B > 4$ T, at the temperature $T = 3$ K. It is, therefore, somewhat surprising that theoretical curves of Im σ slightly depart from the experimental data only at the lowest magnetic field $B = 1$ T.

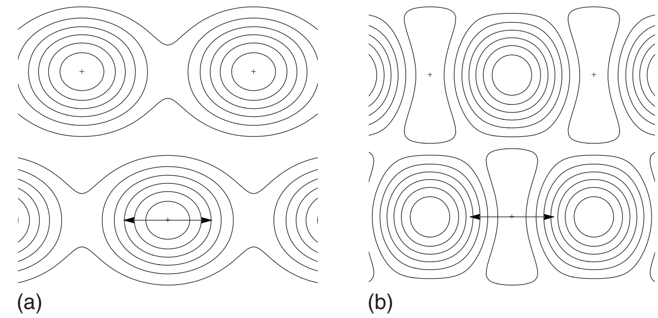

FIG. 2. Heat production (left) and the power absorption (right) in the hexagonal Abrikosov vortex lattice: crosses denote centers of vortices. The electric field is polarized vertically so that vortices oscillate horizontally with amplitude shown by arrows. The Joule heat is produced at vortex cores, their horizontal motion is responsible for elongation of the heated region. Absorption of power is rather delocalized. Its maxima are also around vortex cores but elongated vertically. The rounded minima are between vortices. Difference of these two maps shows that the "rigid" GL function transfers the power to be dissipated in cores.

Due to the relaxation term $\Gamma\partial_t\psi$, the TDGL Eq. (1) includes a damping and generates Joule heat,[9] $\dot{Q} = 4k_{\rm B}T\Gamma(\omega/2\pi)\langle|\partial_\tau\psi|^2\rangle$, where the brackets denote the time average: $\langle\phi\rangle \equiv (\omega/2\pi)\int_0^{2\pi/\omega} d\tau\phi$. The left-hand panel of Fig. 2 shows that the supercurrent produces Joule heat only at vortex cores. The right-hand panel of Fig. 2 presents the spatial distribution of the power absorbed by the condensate from the electric field $W = \langle \mathbf{j}_s \cdot \mathbf{E}\rangle$. The most intensive absorption is around vortices in regions elongated in the vertical direction which is parallel to the electric field. Deep minima of the absorption are between vortices in horizontal rows. Comparing the two panels shows that the relation between absorption and heat production is very nonlocal.

The fraction of Joule heat, due to the condensate, is small. In Fig. 3, we compare the real part of the condensate conductivity (3) with experiment. Indeed, the discrepancy between experimental data and Re σ_s indicates that the supercurrent produces only a minor part of the Joule heat; the normal current cannot be neglected.

From microscopic derivations[1–3,10] of the GL theory, it follows that the normal current and the supercurrent simply add. Adding the current $\mathbf{J}_n = \sigma_0(1+i\tau_n\omega)\mathbf{E}$, which would appear in the normal state one obtains the TDGL conductivity

$$\sigma_{\rm GL} = \sigma_s + \sigma_n, \tag{5}$$

with the normal conductivity $\sigma_n = \sigma_0(1+i\tau_n\omega)$. For experimentally established[6] values $\sigma_0 = 2\cdot 10^4/\Omega$ cm and $\tau_n = 5$ fs, the normal conductivity yields a negligible contribution to Im $\sigma_{\rm GL}$, as seen in Fig. 1, but it provides the dominant contribution to Re $\sigma_{\rm GL}$. One can see in Fig. 3 that Re $\sigma_{\rm GL}$ is much closer to observed values than Re σ_s. It is higher than the observed values, however. This problem becomes more serious at lower magnetic fields, where the observed real part of total conductivity is further reduced well below the level of the normal conductivity, see Fig. 4, while the TDGL conductivity is always larger, Im $\sigma_{\rm GL} >$ Im σ_n.

The simple addition of normal current and supercurrent

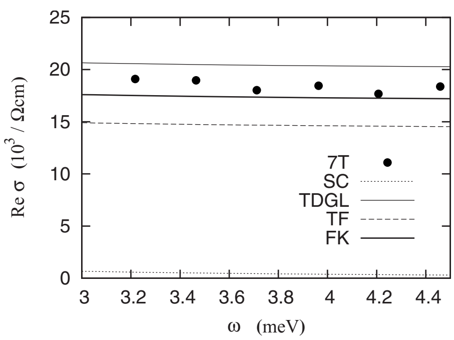

FIG. 3. Real part of the conductivity giving Joule heat: points are experimental data of Ikebe *et al.* (Ref. 6) for 7 T (•). The superconducting condensate contribution (dotted line) given by formula (3) is by an order of magnitude too small. The time-dependent Ginzburg-Landau theory (thin line) adds a contribution of normal electrons, see Eq. (5), arriving at too high values. The two-fluid approach (dashed line) reduces the conductivity subtracting double-counted condensed electrons from the normal conductivity, see Eq. (7). The floating kernel approach (heavy line) given by Eq. (11) removes double-counting from the supercurrents and yields the closest agreement with experiment.

works well close to the phase transition but it badly overestimates conductivity far from it. Apparently, it is insufficient simply to add the supercurrent and the normal current; the electric field accelerates all electrons. Since electrons in the condensate escape frictional effects, this fraction of electrons must be removed in order to obtain the normal conductivity. An intuitive way to avoid double counting of condensed electrons is to introduce a normal current reduced in the spirit of the two-fluid model,

$$\tilde{\mathbf{j}}_n = \left(1 - \frac{2|\psi|^2}{n}\right)\mathbf{J}_n. \tag{6}$$

The total current averaged over the elementary vortex lattice cell, $\mathbf{J}=\mathbf{J}_s+\tilde{\mathbf{J}}_n$, leads to a conductivity

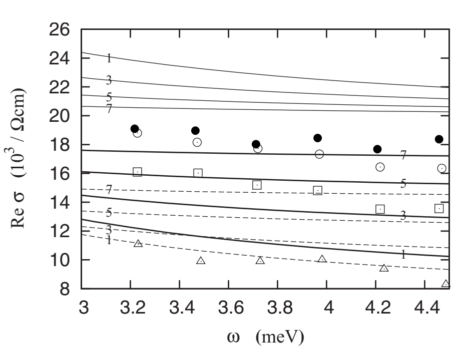

FIG. 4. Real part of the conductivity giving the Joule heat: points are experimental data of Ikebe *et al.* (Ref. 6) for 7 T (•), 5 T (○), 3 T (□), and 1 T (△). The time-dependent Ginzburg-Landau theory (thin line) given by Eq. (5) overestimates the dissipation. The two-fluid approach given by Eq. (7) reduces the dissipation too much leading to the underestimate. The floating kernel approach (heavy line) given by Eq. (11) yields higher values although still smaller than experimental data.

$$\sigma_{\rm tf} = \sigma_s + (t+b)\sigma_n, \tag{7}$$

where we have evaluated the averaged normal fraction, $1-2\langle|\psi|^2\rangle/n=t+b$. One can see in Figs. 1 and 3 that the two-fluid conductivity yields the same nondissipative currents described by Im $\sigma_{\rm tf}$ as the TDGL theory, but that it allows for Re $\sigma_{\rm tf}$ smaller than the normal conductivity. In fact Re $\sigma_{\rm tf}$ is too small, when compared to experimental data.

The reduced normal current (6) contradicts microscopic studies.[1–5] Indeed, the total current is derived from the Nambu-Gor'kov Green's function expanded in the gap, $G \approx G_0+G_0\Delta^*\tilde{G}_0\Delta G_0$, where G_0 gives $\mathbf{j}_n$ and the second term provides the supercurrent. Apparently, the double counting has to be remedied within the supercurrent itself.

With this issue in mind, we shift to our formulation of the theory, expressing the nucleation of superconductivity using the floating nucleation kernel. The Cooper pairs are created from electrons initially in the normal state, with mean velocity $\mathbf{v}=\mathbf{J}_n/(en)$. The free energy of condensation has to supply the kinetic energy which electrons gain going from the normal component into the condensate, therefore, the stability condition reads

$$\frac{1}{2m^*}\left(-i\hbar\nabla - \frac{e^*}{c}\mathbf{A} - m^*\mathbf{v}\right)^2\varphi + \alpha\varphi + \beta|\varphi|^2\varphi = -\Gamma\partial_\tau\varphi. \tag{8}$$

We note that quantum kinetic energy is in fact a nonlocal contribution of the nucleation kernel. For the floating kernel it depends exclusively on the velocity differences of the normal and superconducting component.[11]

The corresponding supercurrent

$$\tilde{\mathbf{j}}_s = \frac{e^*}{m^*}\mathrm{Re}\,\bar{\varphi}\left(-i\hbar\nabla - \frac{e^*}{c}\mathbf{A} - m^*\mathbf{v}\right)\varphi \tag{9}$$

we can write as $\tilde{\mathbf{j}}_s=\mathbf{j}_s-e^*\mathbf{v}|\varphi|^2=\mathbf{j}_s-(2|\varphi|^2/n)\mathbf{J}_n$, therefore, this approach is free of double-counting.

If an effect of velocity $\mathbf{v}$ on the GL function is negligible, then $\varphi=\psi$ and the total current $\mathbf{j}_{\rm fk}=\tilde{\mathbf{j}}_s+\mathbf{J}_n$ obtained with the floating kernel is not different from the current in the two-fluid approximation $\mathbf{j}_{\rm tf}=\mathbf{j}_s+\tilde{\mathbf{j}}_n$. In the presence of vortices, the kinetic energy is nonzero due to diamagnetic currents and the perturbation enters the TDGL equation in the linear order leading to changes in the GL function. The averaged total current $\tilde{\mathbf{J}}_s+\mathbf{J}_n$ then differs from $\mathbf{J}_s+\tilde{\mathbf{J}}_n$. The magnetotransmission thus allows us to test the TDGL theory formulated with the floating nucleation kernel.

To obtain the conductivity, we do not need to evaluate the modified GL function. The supercurrent modified by the inertial force $m^*\partial_\tau\mathbf{v}$ is readily obtained from the condensate conductivity (3). The driving force in Eq. (9) is $\partial_\tau[-(e^*/c)\mathbf{A}-m^*\mathbf{v}]=e^*\mathbf{E}+i(\omega/e^*n)\sigma_n\mathbf{E}$, therefore,

$$\tilde{\mathbf{J}}_s = \sigma_s\left(1 + i\frac{\omega}{e^{*2}n}\sigma_n\right)\mathbf{E}. \tag{10}$$

The conductivity corresponding to the current $\tilde{\mathbf{J}}_s+\mathbf{J}_n$ is given by

$$\sigma_{\mathrm{fk}} = \sigma_s\left(1 + i\frac{\omega}{e^{*2}n}\sigma_n\right) + \sigma_n. \quad (11)$$

In Fig. 1, we compare Im σ_{fk} with Im σ_s. One can see that both values are very close except for at the smallest magnetic field where Im σ_{fk} is closer to experimental data.

In contrast, the Joule heat obtained within various approximations is rather different. In Fig. 4, we compare the standard TDGL theory with the floating kernel formulation. Although none of the approximations provides satisfactory values, among the tested approaches our floating kernel prescription leads to values closest to experiment.

In summary, we have formulated a version of TDGL theory using a floating nucleation kernel, meaning that the assumption of local equilibrium is applied to electrons in the moving reference frame of the normal current.

When compared with standard TDGL theory in the context of far-infrared spectroscopy, we have found that the floating kernel formulation yields better agreement with experiment. In particular, recent published measurements of conductivity were considered; since we have established the GL parameter κ from the nondissipative response given by the imaginary part of the conductivity, our theory has no fitting parameters with respect to the Joule heat given by the real part of the conductivity.

Finally, since use of this approach does not generally present significant additional complexity, it may be promising in the consideration of systems farther from equilibrium than is usually amenable to analysis via standard TDGL theory.

The authors are grateful to Peter Matlock for valuable comments and help in preparation of the manuscript. This work was supported by research plans MSM under Grants No. 0021620834 and No. AVOZ10100521 and by Grants No. GAČR 202/07/0597 and No. GAAV 100100712.

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