Time-dependent Ginzburg-Landau theory with floating nucleation kernel; FIR conductivity in the Abrikosov vortex lattice

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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 (FIR) conductivity in the Abrikosov vortex lattice. It yields better agreement with recent experimental data [PRB 79, 174525 (2009)] than the customary time-dependent Ginzburg-Landau theory.

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The time-dependent Ginzburg-Landau (TDGL) equation is a useful extension of the equilibrium Ginzburg-Landau theory. Unfortunately, microscopic derivations\textsuperscript{[1, 2, 3, 4, 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 non-equilibrium 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 introduce 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; 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 \textit{et al} \textsuperscript{[6]}. The optical gap $2\Delta = 5.3$ meV implies the maximal sub-gap 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 sub-gap 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 $eE/\omega m^*$, while a normal electron is accelerated to $eE\tau_n/m$. At the measurement temperature, $T = 3$ K and $T_e = 15$ 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 sub-gap FIR frequencies the 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 $\sigma$ with $\text{Im}\sigma$ giving the off-phase current and $\text{Re}\sigma$ for the in-phase current. Ikebe \textit{et al} \textsuperscript{[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 $\sigma$ with theoretical predictions based on the TDGL theory in the laboratory and the floating coordinate system.

We will use the electric field $E(\tau) = \text{Re}[E e^{-i\omega \tau}]$ and
current $\mathbf{J}(\tau) = \text{Re} \left[ \mathbf{J} e^{-i\omega \tau} \right]$. 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 \cite{2},

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

describes the evolution of the condensate including a relaxation of the GL function $\psi$ towards 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 $\mathbf{E} = -(1/c) \partial_t \mathbf{A}$. The electric current

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

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: $\mathbf{j}_s = \mathbf{j}_s = (B/\Phi_0) \int_{\text{cell}} dxdy j_x$. The supercurrent, $\mathbf{J}_s = \sigma_s \mathbf{E}$, gives the condensate conductivity

$$\sigma_s = \frac{3\sigma_0}{\beta} \frac{1 - t - b}{b - i\omega \tau_s}, \quad (3)$$

where $t = T/T_c$, $b = B/H_{c2}$ are the dimensionless temperature and magnetic field, $\sigma_0$ is the normal state conductivity, $\beta = 1.16$ is the Abrikosov constant for hexagonal vortex lattice, and $\tau_s = \Gamma(1-t)/\alpha$. Deriving Eq. (3) we have used the GL parameter \cite{7}

$$\Gamma = \frac{12\pi\sigma_0 \alpha \kappa^2 \xi^2}{e^2(1-t)^2}, \quad (4)$$

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 $H_{c2} = H_{c2}(1-t)$ from experimental data in Fig 3 of \cite{6}. The normal-state conductivity $\sigma_0 = 2 \times 10^4/O\Omega cm$, experimentally established at 20 K \cite{6}, has weak temperature dependence and can be used at 3 K.

In Fig. 1 one can see that the imaginary part of $\sigma_s$ from formula (3) reproduces recent experimental data of Ikebe et al \cite{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 $\text{Im} \sigma$ slightly depart from the experimental data only at the lowest magnetic field $B = 1$ T.

Due to the relaxation term $\Gamma \partial_t \psi$, the TDGL equation (1) includes a damping and generates Joule heat \cite{4}, $Q = 4k_B T(\omega/2\pi) \langle |\partial_t \psi|^2 \rangle$, where the brackets denote the time average: $\langle \phi \rangle \equiv (\omega/2\pi) \int^\infty_0 2\pi \omega d\omega \phi$. 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 $W = (\mathbf{j}_s \cdot \mathbf{E})$. 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 non-local.

The fraction of Joule heat due to the condensate is small. In Fig. 3 we compare the real part of the condensate conductivity \cite{8} with experiment. Indeed, the discrepancy between experimental data and $\text{Re} \sigma_s$ indicates that the supercurrent produces only a minor part of the Joule heat; the normal current cannot be neglected.

From microscopic derivations \cite{1,2,3,10} of the GL theory it follows that the normal current and the supercurrent simply add. Adding the current $\mathbf{J}_n = \sigma_0(1 + i\tau_s \omega)\mathbf{E}$ which would appear in the normal state one obtains the TDGL conductivity

$$\sigma_{GL} = \sigma_s + \sigma_n, \quad (5)$$

with the normal conductivity $\sigma_n = \sigma_0(1 + i\tau_s \omega)$. For experimentally established values $\sigma_0 = 2 \times 10^4/O\Omega cm$ and
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, \( \text{Im} \sigma_{\text{GL}} > \text{Im} \sigma_n \).

The simple addition of normal current and supercurrent 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,

\[
\tilde{j}_n = \left( 1 - \frac{2|\psi|^2}{n} \right) J_n. \tag{6}
\]

The total current averaged over the elementary vortex lattice cell, \( J = J_s + \tilde{j}_n \), leads to a conductivity

\[
\sigma_{tf} = \sigma_n + (t + b) \sigma_n, \tag{7}
\]

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 non-dissipative currents described by \( \text{Im} \sigma_{tf} \) as the TDGL theory, but that it allows for \( \text{Re} \sigma_{tf} \) smaller than the normal conductivity. In fact \( \text{Re} \sigma_{tf} \) is too small, when compared to experimental data.

The reduced normal current (6) contradicts microscopic studies [1, 2, 3, 4, 5]. Indeed, the total current is derived from the Nambu-Gor’kov Green function expanded in the gap, \( G \approx G_0 + G_0 \Delta^* G_0 \Delta G_0 \), where \( G_0 \) gives \( j_n \) 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 new 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 \( v = J_n / (en) \). 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

\[
\frac{1}{2m^*} \left( -i\hbar \nabla - \frac{e^*}{c} A - m^* v \right)^2 \varphi + (\alpha + (t + b) \beta) \varphi = -\Gamma \partial \varphi. \tag{8}
\]

We note that quantum kinetic energy is in fact a non-local 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

\[
\tilde{j}_s = \frac{e^*}{m^*} \text{Re} \varphi \left( -i\hbar \nabla - \frac{e^*}{c} A - m^* v \right) \varphi \tag{9}
\]
we can write as \( \tilde{j}_s = j_s - \epsilon^* v|\varphi|^2 = j_s - (2|\varphi|^2/n)J_n \), therefore this approach is free of double-counting.

If an effect of velocity \( v \) on the GL function is negligible, then \( \varphi = \psi \) and the total current \( j_{tk} = j_s + J_n \) obtained with the floating kernel is not different from the current in the two-fluid approximation \( J_{tf} = j_s + j_n \). In the presence of vortices, the kinetic energy is non-zero due to diamagnetic currents and the perturbation enters the TDGL equation in the linear order leading to changes of the GL function. The averaged total current \( j_s + J_n \) then differs from \( J_s + J_n \). The magneto-transmission 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 \( m^* \partial_t \mathbf{v} \) is readily obtained from the condensate conductivity \( \tilde{\sigma}_n \). The driving force in Eq. (9) is \( \partial_t (- (e^*/c)A - m^* \mathbf{v}) = e^* \mathbf{E} + i(\omega/e^* n)\sigma_n \mathbf{E} \), therefore

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

The conductivity corresponding to the current \( \tilde{j}_s + J_n \) is given by

\[
\sigma_{tk} = \sigma_s \left( 1 + i \frac{\omega}{e^* 2n} \sigma_n \right) + \sigma_n. \tag{11}
\]

In Fig. 1 we compare \( \text{Im} \sigma_{tk} \) with \( \text{Im} \sigma_s \). One can see that both values are very close except for at the smallest magnetic field where \( \text{Im} \sigma_{tk} \) is closer to experimental data.

In contrast, the Joule heat obtained within various approximations is rather different. In Fig. 2 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 \( \kappa \) from the non-dissipative 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 new approach does not generally introduce 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.

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