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Time-Dependent Ginzburg–Landau: From Single Particle to Collective Behavior

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The Ginzburg–Landau theory of phase transitions is one of the most successful and elegant theories that have a very general applicability to a large variety of phenomena. Moreover, its extension into time domain has produced a wealth of interesting new predictions and applications all the way from materials science to cosmology. Here, we describe applications of time-dependent Ginzburg–Landau theory to conventional superconductors and show that there is a clear-cut transition in the time dependence from single particle to collective behavior as a function temperature.

KEY WORDS: critical slowing down; Ginzburg-Landau; relaxation times; superconductors.

1. INTRODUCTION

The dynamical response of a system to external perturbations is one of its most fundamental properties. Landau and Khalatnikov (LK) [1,2] developed a theory for the temperature dependence of the relaxation time of the order parameter, τ, that was based on the Ginzburg–Landau theory of phase transitions. The result of this mean-field approach was a divergence of $\tau \propto 1/(T_c - T)$ near a second-order phase transition point, T_c, and this theory correctly described the experimental temperature dependence [3] of the anomalous absorption of sound just below the superfluid λ-point of liquid helium-4. The predicted divergence of τ at any second-order phase transition point is now referred to as "critical slowing down" and it has been studied in many systems, too numerous to mention. Currently, researchers characterize this behavior by the dynamical critical exponent, z, such that $\tau \propto \varepsilon^{-\nu z}$ where $\varepsilon = (T - T_c)/T_c$ (above T_c) and ν is the critical exponent associated with the diverging correlation length. Renormalization-group techniques are used to predict z.

Schmid [4] modified the LK theory essentially by postulating a gauge invariant form of the Landau–Khalatnikov equation that provides a fundamentally important extension of the theory appropriate for systems with a gap in the excitation spectrum, e.g., superconductors. Several microscopic theories [5–8] were developed, using different assumptions or starting points, which, however, disagree on the temperature dependence of the relaxation time of the superconducting order parameter. In this paper, we begin by describing in greater detail the macroscopic phenomenological theory [4]. Then, we will briefly delineate the several microscopic theories. A detailed description of these theories is beyond the scope of this work, and because of this we will only outline the main ideas. Finally we describe the first direct measurement of the divergent relaxation time for the superconducting order parameter near T_c. A clear transition from single particle behavior at low temperatures to collective behavior very near to T_c is found. Simultaneous measurements of the equilibrium energy gap, Δ, further demonstrate that $\tau \propto 1/\Delta$ very near T_c in agreement with the theoretical predictions of Schmid and Schön [8]. It should be noted that the

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fluctuation regime above T_c, where the material is gapless, obeys [9] the LK result, i.e., $\tau \propto 1/(T - T_c)$.

2. PHENOMENOLOGICAL GAUGE INVARIANT LANDAU–KHALATNIKOV THEORY

Since the Ginzburg–Landau equation has proven to be very successful when applied to equilibrium superconductivity, it is reasonable to try to formulate a time-dependent theory based on this equation. Schmid [4] advanced the following gauge invariant modification of the theory [1,2].

Let us assume that in a nonequilibrium situation the free energy of a superconductor can be written as

$$F(P, T, \psi, t) = F_0(P, T) + A(P, T)\psi^2(t) + C(P, T)\psi^4(t) \tag{1}$$

where the time dependence enters through the time dependence of the order parameter ψ and the assumptions are the same as in the Ginzburg–Landau theory. For completeness we will rewrite the assumptions. Below T_c, $C > 0$ and $A < 0$, and above T_c, $A > 0$; the transition point is determined by the condition $A(P, T) = 0$. In the vicinity of the transition temperature T_c we expand $A(P, T)$ in series in the difference $T - T_c$, and neglecting higher order terms we have

$$A(P, T) = a(P)(T - T_c). \tag{2}$$

The temperature dependence of the order parameter ψ is determined from the condition that the free energy has to be a minimum in equilibrium. This gives

$$\psi_{eq}^2 = -\frac{A}{2C} = \frac{a}{2C}(T_c - T) \tag{3}$$

which replicates the BCS temperature dependence of the equilibrium energy gap, close to T_c.

The approach of the order parameter ψ to its equilibrium value is determined from the transport equation

$$\frac{d\psi}{dt} = \gamma \frac{\delta F}{\delta \psi} \tag{4}$$

where the transport coefficient γ was assumed, by Landau and Khalatnikov, not to have any singularity at T_c. Schmid [4], using the Gorkov [10] formulation, showed that $\gamma = -\frac{4k}{ah\pi}$ for a superconductor. Let us assume that we have a small deviation from equilibrium, that is to say $\psi(t) = \psi_{eq} + \delta\psi(t)$ with $|\delta\psi/\psi_{eq}| \ll 1$. Then using Eqs. (4) and (1)

$$\frac{d\delta\psi}{dt} = 2\gamma A\left(\psi_{eq} + \delta\psi\right) + 4\gamma C\left(\psi_{eq} + \delta\psi\right)^3$$

and, since the perturbation is small,

$$\frac{d\delta\psi}{dt} \cong 2\gamma A\left(\psi_{eq} + \delta\psi\right) + 4\gamma C\psi_{eq}^3\left(1 + 3\frac{\delta\psi}{\psi_{eq}}\right)$$

But, from Eq. (3), $C = -(A/2\psi_{eq}^2)$ and $d\delta\psi/dt = -4a\,\gamma\delta\psi$, implying that the order parameter relaxes exponentially with a relaxation time,

$$\tau = \frac{1}{4A\gamma} = \frac{1}{4a\gamma}\frac{1}{T - T_c} \tag{5}$$

It should be pointed out that the temperature dependence of τ is independent of the temperature dependence of ψ_{eq}. If, in addition to the time dependence, the order parameter has a spatial dependence and, allowing for a complex order parameter, the Ginzburg–Landau equation is [9, Eq. (4–1)],

$$F = F_0 + A\psi^2 + C\psi^2 + C\psi^4 + \frac{1}{2m}\left|\left(\frac{\hbar}{i}\nabla - \frac{2e}{C}A\right)\psi\right|^2 + \frac{H^2}{8\pi}$$

where $H = \nabla \times A$ is the magnetic field, A is the vector potential, and ψ is now a complex order parameter or the "superconducting wavefunction." If this form of the free energy is introduced in the Landau–Khalatnikov equation (Eq. (4)), we obtain

$$\frac{\partial\psi}{\partial t} = 2\gamma\left\{\frac{1}{2m}\left(\frac{\hbar\nabla}{i} - \frac{2e}{C}\right)^2\psi + A\psi + 2C|\psi|^2\psi\right\} \tag{6}$$

We notice that a quantum mechanical gauge transformation consists of replacing simultaneously [11, p. 399]

$$A \to A + \nabla\Xi$$
$$\tilde{\phi} \to \tilde{\phi} - \frac{1}{c}\frac{\partial\Xi}{\partial t}$$
$$\psi \to \psi e^{ie\Xi/hc}$$

where ϕ is the effective potential and Ξ is an arbitrary function of r and t. In order to make Eq. (6) gauge invariant, a term has to be added so as to obtain

$$\left(\frac{\partial}{\partial t} - 2ie\tilde{\phi}\right)\psi = 2\gamma\left[\frac{1}{2m}\left(\frac{\hbar\nabla}{i} - \frac{2eA}{c}\right)^2\psi + A\psi + 2C|\psi|^2\psi\right] \tag{7}$$

It was proposed by Schmid [4] that the effective potential $\tilde{\phi}$ should be written as $\tilde{\phi} = \phi - \mu/e$ where

ϕ is the electric potential and μ is the chemical potential. Physically, μ is the energy necessary to add an electron in thermal equilibrium when the fields are kept constant. With this assumption, in the case where there are no electro-magnetic fields present and ψ does not vary spatially, Eq. (7) reduces to

$$\left(\frac{\partial}{\partial t} + 2i\mu\right)\psi = 2\gamma(A\psi + 2/C|\psi|^2\psi) \qquad (8)$$

In writing the "wavefunction" as $\psi = |\psi|e^{i\theta}$ and separating the real and imaginary parts of Eq. (8), two modes are obtained:

$$\frac{\partial|\psi|}{\partial t} = 2\gamma|A|\psi| + 2C|\psi|^3| \qquad \text{(Real part)} \qquad (9)$$

$$\frac{\partial\theta}{\partial t} + 2\mu = 0 \qquad \text{(Imaginary part)} \quad (10)$$

Equation (9) is the same as Eq. (4) except that now ψ is a complex order parameter. The earlier calculation for the relaxation time is still valid, and so the relaxation time for the magnitude of ψ is

$$\tau_{|\psi|} = \frac{1}{4A\gamma} = \frac{1}{4a\gamma}\frac{1}{T - T_c} \qquad (11)$$

The relationship between the superconducting order parameter (wavefunction) and the energy gap was determined by Gorkov [12] to be

$$\psi(\vec{r}, t) = \left[\frac{7\zeta(3)n_0\chi(\rho)}{16\pi^2k^2T_c^2}\right]^{1/2}\Delta(\vec{r}, t) \qquad (12)$$

where $\zeta(3) = 1.202$, n_0 is the density of electrons, $\rho = \frac{1}{2}\pi T_c\tau_{tr}$ with τ_{tr} the mean time between collisions for an electron, and

$$\chi(\rho) = \frac{8}{7\zeta(3)}\frac{1}{\rho}\left|\frac{\pi^2}{8} + \frac{1}{2\rho}\left(G\left(\frac{1}{2}\right) - G\left(\frac{1}{2} + \rho\right)\right)\right|$$

with G the logarithmic derivative of the Γ function.

Since the order parameter and the energy gap of superconductors differ only by a constant, the relaxation time of the energy gap might be expected to be given by Eq. (11), but see below.

The imaginary part of Eq. (8) describes the evolution of the phase of the order parameter. Equation (10) implies that the phase of the order parameter will change in time at a constant rate. The relaxation time associated with Eq. (10) is related to the branch imbalance relaxation [13,14] (determined by the difference between the chemical potential of quasiparticle and pairs) since the evolution of the phase θ is determined by the chemical potential μ of the "superelectrons."

3. THE MICROSCOPIC THEORIES FOR THE RELAXATION OF THE ORDER PARAMETER IN A SUPERCONDUCTOR

The first attempt to determine the time variation of the Ginzburg–Landau order parameter from a microscopic theory was done by Abraham and Tsuneto [15]. They extended the Gorkov [12] formulation to the nonequilibrium situation and found for small deviations from equilibrium, a time-dependent Ginzburg–Landau equation exists near the transition temperature and near absolute zero temperature. Close to the transition temperature they found a differential equation, which is diffusive-like in character, and at zero temperature they found an equation which is wave-like. One of the main limitations of the theory [15] is that they assumed that the thermal excitations in the superconductor are at rest and in equilibrium with the local values of the energy-gap and the external fields. This occurs only if the characteristic interaction time between phonons and thermal excitations is faster than the characteristic time for the "normal to superfluid" conversion.

Later, Lucas and Stephen [5] studied the relaxation of the order parameter in a superconductor and also found that the main mechanism for this relaxation is through the interaction of the quasiparticles with the phonon field. To calculate the relaxation time τ they started from the Hamiltonian for the electron–phonon interaction written in the conventional form [16, p. 32] and used a Boltzmann equation [17] for the quasiparticle distribution. The relaxation time for the order parameter derived as described has the form

$$\tau \propto \ell n\left[\frac{T_c - T}{T_c}\right]^2 \quad \text{for } \frac{T}{T_c} \sim 1 \qquad (13)$$

Woo and Abrahams [6] pointed out that the calculation of [5] is not correct since they used the Boltzmann equation [17], which assumes local equilibrium between the quasiparticles and the energy gap. In the case, where the energy gap is time dependent, there is no local equilibrium between the quasiparticles and the energy gap at a time scale shorter than the electron–phonon inelastic collision time. Woo and Abrahams [6] further derive a transport equation for the superconductor in the presence of electron–phonon interaction. This transport equation is obtained by a technique of thermodynamic Green's functions developed by Kadanoff and Baym [18]. Their conclusion is that for weak coupling materials, for $0.9 \leq T/T_c \leq 0.99$ the relaxation time of the energy gap (10^{-9} to 10^{-8} s) is about an order of magnitude faster than the quasiparticle recombination rate, but very close to T_c it diverges as

$$\tau \propto \frac{1}{T - T_c} \tag{14}$$

This temperature dependence agrees with the prediction of the phenomenological Landau–Khalatnikov theory.

Schmid [19] calculated the relaxation of the order parameter starting from a dynamical version of the BCS theory [20]. He included both the electron–electron and the electron–phonon collision times since, at the temperatures considered, they are comparable in weak coupling materials, especially in aluminum [21]. For small deviations from equilibrium [19], he finds that the relaxation time for the superconducting order parameter is

$$\tau = \begin{cases} \frac{\pi}{16(T_c - T)} & \text{if } \Delta_{eq} < \frac{1}{\tau_e} \\ \frac{\pi^3}{7\zeta(3)} \frac{T}{\Delta_{eq}} \tau_e & \text{if } \Delta_{eq} > \frac{1}{\tau_e} \end{cases} \tag{15}$$

where τ_e is the inelastic electron collision time, Δ_{eq} is the equilibrium energy gap and $\zeta(3) = 1.2$. The condition $\Delta_{eq} < \frac{1}{\tau_e}$ is the condition of gapless superconductivity where the energy gap is smaller than the collision broadening of the energy levels. For the relaxation of the phase of the order parameter [19] he obtains

$$\tau = \begin{cases} \frac{6}{\nu_0^2 k^2} \tau_e \Delta_{eq}^2 & \text{if } \nu_0 k \ll \frac{1}{\tau_e} \text{ and } \Delta_{eq} \gg \frac{1}{\tau_e} \\ \dfrac{12\pi^3 T_c}{\zeta(3)\nu_0^3 k^3} \Delta_{eq} & \text{if } \nu_0 k \gg \frac{1}{\tau_e} \end{cases} \tag{16}$$

where ν_0 is the Fermi velocity and k is the wavevector of the particular mode under consideration.

Schmid and Schön [8] extended this work [19] by using a temperature-dependent Green's function technique introduced by Eliashberg [22] and Eilenberger [23]. The calculation is based on a model of a superconductor where the electrons interact via phonons. The phonons are assumed to be in thermal equilibrium and the deviations from equilibrium of the electronic system are small so that a linearized theory applies. The linearization of the theory also permits the classification of various modes, especially useful when the theory is applied close to the transition temperature T_c. Schmid and Schön [8] obtain an equation for energy gap that is very similar to the Landau–Khalatnikov equation (8)

$$\left(\frac{\partial}{\partial t} + 2iM\right)\Delta = -\frac{8T}{\pi N_0} \text{ linearized } \left\{\frac{\delta F_{GL}}{\delta \Delta}\right\} \tag{17}$$

where N_0 is the density of states at the Fermi energy and M is a complex valued function, unlike in Eq. (8), where it was a real quantity, μ, the chemical potential, and they find

$$M = \frac{4T}{\pi|\Delta|} \int dE' \beta(E') \delta f_{E'} \tag{18}$$

where $\delta f_{E'}$ is the quasiparticle distribution and $\beta(E')$ is related to the details of the theory.

In addition to Eq. (17), the energy gap and the quasiparticle distribution are coupled through a Boltzmann-type equation for the quasiparticle distribution,

$$\frac{d\delta f_E}{dt} - K(\delta f) - P_E - Q_E = \mathring{h}_E \tag{19}$$

where the term $d\delta f_E/dt$ is the time derivative of the quasiparticle distribution, $K(\delta f)$ is the collision integral as in a Boltzmann equation for a gas, P_E is the perturbation term since we are considering nonequilibrium situations, h_E is the term that couples the energy gap and the quasiparticle distribution, and Q_E is an additional correction term that arises from a detailed theory. It should be pointed out that Q_E and h_E are different for different modes that arise.

There are two modes that can be distinguished by studying Eq. (17), associated with the imaginary and real parts of M. This is similar to the two modes obtained in Section 2, when studying the real and imaginary parts of Eq. (8).

The real part of Eq. (17) gives rise to the relaxation of the magnitude of Δ, the so-called longitudinal mode. According to Schmid and Schön the longitudinal mode can be excited by a superposition of a dc and an ac current [24], or by irradiation of the superconductor by an electromagnetic wave. The relaxation time for this mode was found to be

$$\tau^{(L)} = \frac{\pi^3}{7\zeta(3)} \frac{T}{\Delta_{eq}} \tau_e, \quad \zeta(3) \sim 1.2 \tag{20}$$

The imaginary part of Eq. (17) gives rise to the relaxation of the phase of Δ, the so-called transverse mode. This equation is the same as Eq. (10), obtained from a simple phenomenological theory (μ has to be replaced in Eq. (10) by $\mathrm{Re}(M)$). This mode can be excited by electron tunneling injection at high voltage [13,14,25], or by driving a current in the direction of a spatial change of the order parameter, as at a normal–superconductor interface [26]. The relaxation time associated with this mode was found to be

$$\tau^{(T)} = \frac{4}{\pi}\frac{T}{\Delta_{eq}}\tau_e \tag{21}$$

It should be pointed out that in both modes the relaxation time diverges as $1/\Delta_{eq}$ for $T \lesssim T_c$ and that although the processes involved are quite different, the relaxation times of the two modes are quite close. It is pointed out in [8] that a divergence should occur in any mode that involves the order parameter essentially.

This theoretical result is seen to be reasonable with the following argument. Quasiparticles are scattered to states over an energy interval of $\sim kT$ around the Fermi level at an average rate, $1/\tau_e$. However, in the BCS energy-gap equation, only those states reaching within $\sim\Delta$ participate in relaxing Δ. Then the relaxation of the gap is simply $\sim kT\tau_e/\Delta$, which agrees with Eq. (20) to within a numerical factor of order one. Similar reasoning was applied to the relaxation of charge (or branch) imbalance [27] that corresponds to the transverse mode, i.e., Eq. (21).

Since the details of the calculations are quite involved and beyond the scope of this work, we are going to recapitulate the main ideas and conclusions of this theory. The equations that govern the relaxation of the superconducting order parameter are a pair of coupled differential equations for the energy gap and the quasiparticle distribution. One of the equations has the form of the Landau–Khalatnikov equation for the energy gap, the other equation is a Boltzmann-like equation for the quasiparticle distribution function. In solving these equations one obtains two different modes, one associated with the relaxation of the magnitude of the order parameter, the other associated with the phase of the order parameter. Both these modes have similar relaxation times that diverge as $1/\Delta_{eq}$ for $T \to T_c$. The difference between superconductors and other systems that exhibit second-order phase transitions (liquid He^4, ferromagnets, etc.) can be understood physically (Morrel Cohen, private communication). Superconductors exhibit a forbidden energy gap unlike such systems. When a Cooper pair is broken into quasiparticles, by perturbing the energy gap (with electromagnetic radiation for instance), the energy gap will decrease. Since the energy gap decreases more final states are made available for the quasiparticles and consequently more quasiparticles are thermally excited into these newly available states. Because of this we would expect the behavior of the energy gap to be coupled to that of the quasiparticles, and we expect the relaxation time of the order parameter to have a different dependence in superconductors than in systems that exhibit a second-order phase transition but do not have an energy gap.

To check this hypothesis in other systems besides superconductors it would be very interesting to study the relaxation of the order parameter in liquid He^3 in B phase, which also has a real energy gap that presumably is like BCS. This could be done by studying the low frequency sound attenuation in the B phase of He^3 in a similar way to that done by Chase [3] for the relaxation time of the order parameter in He^4.

4. EXPERIMENTAL RELAXATION TIMES IN SUPERCONDUCTING ALUMINUM

The most comprehensive measurements of the relaxation times in superconductors were accomplished in aluminum films and published in two

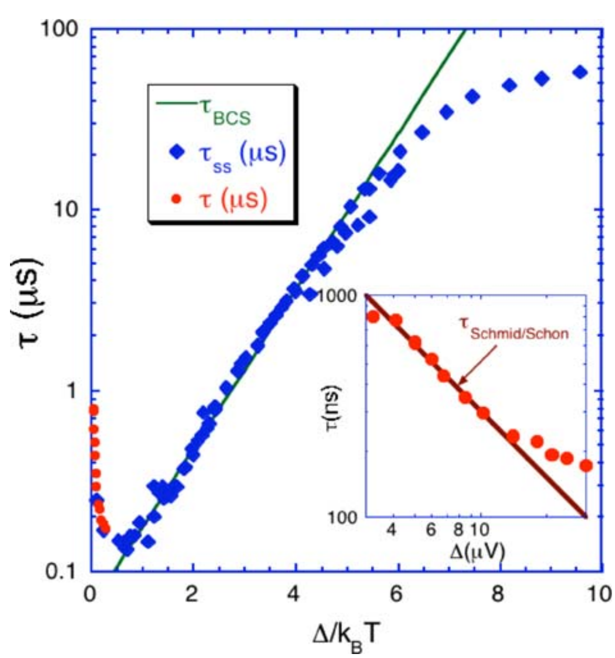

Fig. 1. Relaxation times in superconducting aluminum as a function of the temperature-dependent energy gap, Δ divided by k_BT. At low temperatures ($\Delta/k_BT > 1$) the single particle recombination rate is proportional to the thermal population of other quasiparticles available for recombination. The inset shows the collective behavior of the response close to T_c that follows Ginzburg–Landau critical slowing down for systems with an energy gap in the spectrum.

papers. The first describes the low-temperature relaxation of the quasiparticle density with a steady-state injection/detection method [28] using two superimposed tunnel junctions with a common electrode. The critical region near T_c was reported [29] using real-time response of the current–voltage characteristic of a tunnel junction to fast laser pulses. The results of these are shown in Fig. 1, in a semi-log plot against Δ/kT to emphasize the agreement with theory at low temperatures (i.e., large Δ/kT). Since quasiparticles must recombine in pairs, the recombination *rate* should be proportional to the density of thermally activated quasiparticles which is shown as the solid line that is proportional to $\exp(-\Delta/kT)$. The saturation of τ at the longest times is likely due to defects, such as magnetic vortices with normal cores, where the recombination rate is enhanced. Critical slowing down is observed near T_c (Δ/kT approaching zero) as a sharp increase in the relaxation time. Agreement with the theoretical temperature (or energy-gap) dependence of Schmid and Schön is demonstrated by the inset, in which the solid line represents $\tau \sim 1/\Delta$ i.e. $\Delta \sim \frac{1}{(T-T_c)^{1/2}}$ using the BCS temperature dependence of Δ found in the films studied.

5. SUMMARY

In spite of the richness and complications of real systems, the generic divergence of the order parameter relaxation that was first predicted by the LK extension of the Ginzburg–Landau theory is the conceptual genesis of the phenomena currently called critical slowing down. In addition, the simple physical picture of critical slowing down is transparent in this mean-field formulation.

It is interesting to note that these ideas have been revived and used again in the context of high-temperature superconductivity in the oxide cuprates [30]. Of course, in these materials additional complications or interesting flexibility arises from the fact that the superconductivity can be tuned by doping [31] and the order parameter [32,33] may no longer have the simple s-wave symmetry. In this case, interesting modifications were found in the quasiparticle [34] and collective mode [34–36] dynamics, although the analysis of these measurements were done still within a similar frame work described here. Moreover, the pervasive nature of these ideas based on the original Ginzburg–Landau theory extend even recently from superconductivity [37], to closely related areas such as magnetism [38] all the way to cosmology [39]. The fact that these ideas have been applied to countless physical systems, in equilibrium and way from it, in the last 50 years point to the genius in the original Ginzburg–Landau theory. The Ginzburg–Landau theory is a classic, it does not need a reference.

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