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Phys. Rev. B **95**, 121106 — Published 23 March 2017

DOI: 10.1103/PhysRevB.95.121106

Thermodynamic constraints on the amplitude of quantum oscillations.

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(Dated: December 20, 2016)

Quantum oscillation experiments in high temperature superconductors show a strong thermally–induced suppression of the quantum oscillation amplitude approaching the critical dopings [1–3]—in support of a quantum critical origin of their phase diagrams. We suggest that, in addition to a thermodynamic mass enhancement, these experiments may directly indicate the increasing role of quantum fluctuations that suppress the quantum oscillation amplitude through inelastic scattering. We show that the traditional theoretical analysis of the quantum oscillation amplitude in correlated metals results in a contradiction with the third law of thermodynamics and suggest a way to rectify this problem.

Recent advances in high-magnetic field measurements have amassed a body of information about metallic quantum criticality in high-temperature superconductors.[4–7] In particular, quantum oscillation measurements in cuprate, $YBa_2Cu_3O_{6+x}$[1], and pnictide, $BaFe_2(As_{1-x}P_x)_2$[2, 3], systems suggest a strong enhancement of the quasiparticle mass approaching a critical doping—a locus for thermodynamic anomalies in other measurements as well [3, 8, 9].

The quasiparticle mass, $m^\star$, is inferred in these measurements from analysis of the temperature dependence of the quantum oscillation amplitude $A(T)$.[10] In conventional metals, $A(T)$ decays over a temperature range that is inversely proportional to the quasiparticle mass with no parameters other than the magnetic field and temperature entering the functional form, $A_0(T) = X/\sinh X$, where $X = 2\pi^2(k_BT)/(\hbar\omega_c)$ and $\omega_c = eB/m^\star$ is the cyclotron frequency.[11] This form of the temperature dependence of $A(T)$ originates in the temperature smearing (over an energy interval k_BT) of the occupation number of Landau levels (spaced at $\hbar\omega_c$) near the Fermi surface. Importantly, it relies on the presence of well-defined quasiparticles near the Fermi surface, justified by the Fermi liquid theory of conventional metals.[12, 13]

Unlike the renormalizations of $m^\star$, which describe changes in electron velocity without changes in lifetime, electron interactions in correlated metals lead to anomalous quasiparticle relaxation dynamics, observed via the temperature and energy dependence of the quasiparticle relaxation rate $1/\tau(T,\epsilon)$.[4–7] Such departure from Fermi liquid behavior must change the character of quantum oscillations, or at the very least add to the temperature dependence of $A(T)$ and change its interpretation in terms of the quasiparticle mass.

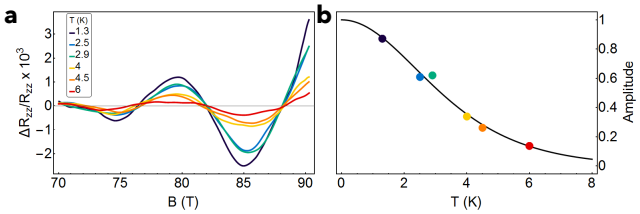

FIG. 1. The temperature dependence of quantum oscillations in $YBa_2Cu_3O_{6+x}$ near optimal doping. **a** Quantum oscillations in the c-axis resistivity, ρ_{zz}, up to 90T for $YBa_2Cu_3O_{6.86}$ ($p = 0.152$, $T_c = 92K$). [1] **b** The temperature-dependent amplitude of the oscillations in **a** closely follows the standard Lifshitz-Kosevich behavior $A(T)$ (black line).

It is therefore puzzling that the observed temperature dependence of $A(T)$ in high-temperature superconductors (Figure 1) appears, within experimental resolution, to be identical to its Lifshitz-Kosevich form $A_0(T)$, even for chemical compositions near the critical doping[1–3] where strong correlation effects are well established.[4–7] Herein we discuss the thermodynamic constraints on the form of $A(T)$ that derive directly from the third law of thermodynamics (Nernst's theorem) and are independent of the nature of the metallic state. Importantly, the form of $A(T)$ required by these constraints suggests that the observed $A(T)$ is universal in its broad features and is not specific to Lifshitz-Kosevich form $A_0(T)$.

Whether the doping evolution of $A(T)$ observed in Refs. 1–3 approaching critical doping is a result of quasiparticle mass evolution alone is now an open question. More insight into the dynamic and static effects near a quantum critical point can be gained from comparison between masses obtained in quantum oscillation measurements with those from other experimental probes, such as heat capacity, cyclotron resonance, and Landau level spectroscopy.

At low temperatures, metals in a magnetic field exhibit quantum oscillations—an oscillatory variation of magnetization, resistivity, and other properties with field intensity.[10] The frequency, F, of the oscillations, which are periodic in inverse magnetic field, $1/B$, has a direct geometric interpretation as the extremal area (perpendicular to the field) of the Fermi surface in momentum space.[14] Quasiparticle properties near the Fermi surface, such as the effective mass $m^\star$ and the relaxation time τ, can be obtained from the analysis of the field and temperature dependences of the oscillation amplitude $A(T,B)$. The amplitude $A(T,B)$ can be defined via the oscillatory part of the thermodynamic potential, $\Omega_{\rm osc}(T,B) \propto A(T,B)\cos(2\pi F/B+\gamma)$. Note that $A(T,B)$ contains all of the temperature dependence in this expression, therefore, it determines the oscillatory part of entropy, $S_{\rm osc}(T,B) = -\partial\Omega_{\rm osc}(T,B)/\partial T$.

Nernst's theorem requires that the total entropy vanishes in the zero temperature limit, $[S_{\rm osc}(T,B) + S_{\rm bg}(T,B)]_{T\to 0} = 0$ where $S_{\rm bg}(T,B)$ is the non-oscillatory part.[15, 16] This requirement can only be satisfied at all fields if $S_{\rm osc}(T,B)$ itself vanishes in the zero temperature limit because of its distinct magnetic field dependence, $S_{\rm osc}(T\to 0,B) = 0$. The temperature derivative of the amplitude of quantum oscillations must therefore vanish in the zero-temperature limit,

$$\left(\frac{\partial A(T,B)}{\partial T}\right)_{T\to 0} = 0\,. \qquad (1)$$

Similarly, the oscillating part of the heat capacity, $c_{\rm osc}(T,B) = T(\partial S_{\rm osc}(T,B)/\partial T)$, must vanish at zero temperature. This requires that the curvature of $A(T,B)$ is less singular than $1/T$ in the limit of zero temperature,

$$\left(T\frac{\partial^2 A(T,B)}{\partial T^2}\right)_{T\to 0} = 0\,. \qquad (2)$$

We emphasize that this line of reasoning does not provide justification for the presence of quantum oscillations in any metal, but rather it sets tight bounds on the behavior of $A(T,B)$ if the metal does exhibit quantum oscillations in the zero-temperature limit, i.e., if $A(T,B)$ can be defined at all. It is in this sense that these thermodynamic identities are independent of the character of the metallic state.

Being a result of a well-controlled microscopic calculation, the Lifshitz-Kosevich $A_0(T)$ satisfies identically both Eqs. (1) and (2): $A_0(T)$ approaches zero temperature with zero slope and finite (non-singular) curvature. The temperature dependence of $A_0(T)$ in a broad temperature range is tightly constrained by requirements of vanishing entropy at zero temperature, $dA_0(T)/dT|_{T\to 0} = 0$, and fast decay at high temperatures. Furthermore, its temperature dependence is set by a single energy scale, $\hbar\omega_c$, which requires $A_0(T) = f_0(X)$ where the function $f_0(X)$ approaches zero with zero slope and decays for $X \gtrsim 1$.

The form of A(T) in the quantum-critical regime need not be identical to that of LK because the underlying metallic state is not the same as the one assumed in calculate the Lifshitz-Kosevich form of temperature dependence. However, for the *scale invariant* dynamics[7] in the vicinity of quantum critical point the temperature dependence of $A(T)$ can only be set by an energy scale associated with external magnetic field, $\alpha \times \hbar\omega_c$. Thus, $A(T) = f_q(X/\alpha)$ where α is a numeric factor. The function $f_q(X)$ is similar in its form to $f_0(X)$: it must approach zero temperature with zero slope (Nernst's theorem) and it must decay for $X \gtrsim 1$. Further refinement in microscopic modeling and experimental sensitivity may point to more subtle differences in the functional form of the two temperature dependences. Unlike the electron renormalization in $m^\star$ which describe changes in electron velocity without changes its lifetime, α originates in quasiparticle relaxation dynamics, via the temperature and energy dependence of the quasiparticle relaxation rate $1/\tau(T,\epsilon)$. We note that fitting $A(T) = f_q(X/\alpha)$ with $A_0(T) = f_0(X)$ yields $m^\star/\alpha$ rather then the quasiparticle mass $m^\star$.[17]

Thus, the observed Lifshitz-Kosevich functional form of the temperature dependence of the amplitude of quantum oscillations in high-temperature superconductors[1–3] should not be taken as conclusive evidence of Fermi liquid behavior, where all electron scattering is elastic and an interaction effects are captured by mass renormalization. This observation leads one to ask whether and how quantum oscillation can arise near quantum critical point where quasiparticles are poorly defined.

First, we note that the *existing* theoretical discussions of the amplitude of quantum oscillations in correlated metals all lead to finite entropy at zero temperature, $dA(T)/dT|_{T\to 0} \neq 0$ [20–28], violating Nernst's theorem. In the remainder of this note we attempt to identify the source of the problem and point to a way to resolve it. In particular, we suggest that rather than signaling inadequacy of their common starting point (Luttinger's functional representation of the Free energy [13]), the violation of Nernst's theorem in these discussions simply indicates that the approximation scheme chosen is inconsistent, following too closely the approximation scheme used for the Fermi liquid metal.

The standard starting point for the existing theoretical analysis of quantum oscillations in a strongly interacting electron liquid[20–27] is the Luttinger's functional

representation for the thermodynamic potential [13, 29],

$$\Omega = T\sum_{i\varepsilon_n,p}\ln(G) + Y\{G\} - T\sum_{i\varepsilon_n,p} G(G_0^{-1}-G^{-1})\,, \quad (3)$$

where $G(i\varepsilon_n,p) = 1/[i\varepsilon_n - \epsilon(p) + \mu - \Sigma(i\varepsilon_n,p)]$ is the exact Green's function for interacting electrons, and $G_0(i\varepsilon_n,p) = 1/[i\varepsilon_n - \epsilon(p) + \mu]$ is the Green's function for free electron propagation.[30] $Y\{G\}$ stands for an infinite set of diagrams in which electron quasiparticle propagation is represented by $G(i\varepsilon_n,p)$[13].

The first term in Eq. (3) evaluates to

$$\Omega_1 = -\int \frac{d\varepsilon}{2\pi}\tanh\frac{\varepsilon}{2T}\sum_p \mathrm{Im}\,\overline{\ln}\left[\varepsilon - \epsilon_p + \mu - \Sigma^R(\varepsilon,T)\right], \quad (4)$$

where ϵ_p are the energies of quasiparticle states. This term is similar in structure to the Green's function representation of the thermodynamic potential for the non-interacting Fermi gas, $\Omega_0 = T\sum_{i\varepsilon_n,p}\ln(G_0) = -T\sum_p \ln\left(1+e^{\frac{\mu-E_p}{T}}\right)$ (see SM for details). In Fermi liquid metals, the oscillatory part of the thermodynamic potential is obtained by analysis of this term, Ω_1, alone.[12, 31]

The single-particle energy levels ϵ_p in a magnetic field consist of a set of quantized Landau levels $\lambda_p = \omega_c(p+1/2)$ with degeneracy BA/Φ_0 where A is the area of the crystal, Φ_0 is the flux quantum, $\omega_c = eB/m^\star$ is the cyclotron frequency, and $m^\star = (\hbar^2/2\pi)\left(\partial A_p/\partial\epsilon\right)$. A_p is the Fermi surface area perpendicular to the magnetic field.[11, 12] Performing the sum over Landau level index p in Eq. (4) (SM) we obtain for the oscillatory (periodic in $2\pi/\omega_c$) part of Ω_1 per unit area,

$$\Omega_{1,\mathrm{osc}} = -2m\varpi_c\int_{-\infty}^{\infty}\frac{d\varepsilon}{2\pi}\tanh\left(\frac{\varepsilon}{2T}\right) \times \mathrm{Im}\,\overline{\ln}\left(1+e^{-i\frac{\varepsilon-\Sigma^R(\varepsilon,T)+\mu}{\varpi_c}}\right), \quad (5)$$

where $\varpi_c = \omega_c/2\pi$. Quasiparticle lifetime effects are introduced in Eq. (5) via the temperature and energy dependence of the imaginary part of the self-energy, $1/\tau = -2\mathrm{Im}\Sigma^R(\varepsilon,T)$. The real part of the self energy, $\mathrm{Re}\Sigma^R(\varepsilon,T)$, is responsible for quasiparticle mass renormalization[30]. We distinguish the "static" and "dynamic" parts of the quasiparticle relaxation rate, $\mathrm{Im}\Sigma^R(\varepsilon,T) = \mathrm{Im}\Sigma^R_{\mathrm{stat}} + \mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon,T)$. The static part, $1/\tau_0 = -2\mathrm{Im}\Sigma^R_{\mathrm{stat}}$, is independent of temperature and energy, equal to $\mathrm{Im}\Sigma^R(\varepsilon\to 0, T\to 0)$. This term typically describes the effects of elastic disorder, and introduces a temperature-independent exponential envelope in field to the oscillation amplitude—the so-called "Dingle" factor.[10] The dynamic part, $1/\tau_{\mathrm{dyn}} = -2\mathrm{Im}\Sigma^R_{\mathrm{dyn}}$, left out of calculations of $A_0(T)$ in the Fermi liquid,[10] contains all the temperature and energy dependence of $\mathrm{Im}\Sigma^R(\varepsilon,T)$ and is constrained by $\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon\to 0, T\to 0) = 0$. For the remainder of this discussion we will explicitly focus on the dynamic effects introduced through $\mathrm{Im}\Sigma^R_{\mathrm{dyn}}$, and all mass-renormalization effects that enter through $\mathrm{Re}\Sigma^R(\varepsilon,T)$ are contained in the renormalized ϖ_c.

The principal harmonic of $\Omega_{1,\mathrm{osc}}$, Eq. (5), defines the temperature dependent amplitude $A_1(T)$

$$\Omega_{1,\mathrm{osc}} \propto R_D A_1(T)\,\cos\left(\mu/\varpi_c\right),$$
$$A_1(T) = \frac{1}{\varpi_c}\int_{-\infty}^{\infty}\frac{d\varepsilon}{2}\sin\left(\frac{\varepsilon}{\varpi_c}\right)\tanh\left(\frac{\varepsilon}{2T}\right)\, e^{-\frac{\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon,T)}{\varpi_c}}, \quad (6)$$

where the temperature-independent Dingle factor $R_D = e^{-1/2\varpi_c\tau_0}$ accounts for the effects of elastic scattering. In the limit where the dynamic part of the self-energy vanishes, $\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon,T) = 0$, $A_1(T)$ reduces to its Fermi liquid form, $A_0(T) = -\left(1/\varpi_c\right)\int d\varepsilon \sin\left(\varepsilon/\varpi_c\right) n_F(\varepsilon) = X/\sinh X$, where $X = \pi T/\varpi_c$ and $n_F(\epsilon) = \left[1-\tanh\varepsilon/2T\right]/2$ is the Fermi-Dirac distribution function. This limit is used to set the normalization factor in $A_1(T)$ in Eq. (6). In this approximation, where we have only considered Ω_1, correlation effects introduced via the self energy change the form of $A_1(T)$. We can now examine whether this modified form of $A_1(T)$ satisfies the thermodynamic constraint imposed by Nernst's theorem, or whether a more complete analysis of Luttinger's functional is required.

Consider the temperature derivative of $A_1(T)$ in Eq. (6),

$$\frac{dA_1(T)}{dT} = \frac{1}{\varpi_c}\int_{-\infty}^{\infty}\frac{d\varepsilon}{2}\sin\frac{\varepsilon}{\varpi_c}e^{-\frac{\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon,T)}{\varpi_c}} \times\left[-\frac{\varepsilon}{T}\left(\frac{\partial\tanh\frac{\varepsilon}{2T}}{\partial\varepsilon}\right) - \frac{1}{\varpi_c}\tanh\frac{\varepsilon}{2T}\left(\frac{\partial\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon,T)}{\partial T}\right)\right]. \quad (7)$$

The frequency integration of the first term on the second line vanishes in the limit of zero temperature because it is confined to an interval $\propto k_BT$ about zero. This term is the only one present in the expression for the temperature derivative of $A_0(T)$. The second term is finite unless $\partial\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon, T\to 0)/\partial T$ decays fast enough with ε. We emphasize that it is the behavior of $\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon,T)$ at energies below as well as above k_BT that determines the slope of $A_1(T\to 0)$. We therefore conclude that the

violation of the Nernst's theorem constraint $dA_1(T \to 0)/dT = 0$ in Ω_1 is a associated with the anomalous energy-dependence of $\partial \mathrm{Im}\Sigma^R(\varepsilon, T \to 0)/\partial T$. For example, in the Fermi liquid, where $\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon, T) = a\varepsilon^2 + bT^2$, the derivative $\partial \mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon, T \to 0)/\partial T$ is zero and therefore the Nernst's theorem constraint is satisfied by the oscillating component $\Omega_{1,\mathrm{osc}}$ alone.[31] For an arbitrary form of self-energy, this constraint is not necessarily satisfied without fine tuning. Therefore, a complete analysis of the effects of electronic correlations on the amplitude of quantum oscillations must include the effects captured by the other two terms in the Luttinger functional, Eq. (3).

It is instructive to compare this situation with a Fermi liquid metal. Analyticity of the Fermi liquid description of a normal metal ensures that oscillating components in the last two terms of Eq. (3) cancel out.[12] At the same time, quasiparticle relaxation near the Fermi surface in Fermi liquids is nearly temperature– and energy–independent.[32, 33] Therefore, analysis of $\Omega_{\mathrm{osc}}(T, B)$ constrained to Ω_1 alone is consistent in the Fermi liquid: $\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon, T)$ has weak temperature and frequency dependence. In contrast, for correlated metals such as cuprates near the critical doping, both of these conditions—that of a weak temperature–and frequency–dependence of $\mathrm{Im}\Sigma^R_{\mathrm{dyn}}(\varepsilon, T)$ and that of analyticity which warrants cancellation of the last two terms in Eq. (3)—break down.[34, 35]

The work at the National High Magnetic Field Laboratory is supported by National Science Foundation Cooperative Agreement No. DMR-1157490 and the State of Florida. A.S. acknowledges hospitality of the Aspen Center for Physics, where part of the work was done. Aspen Center for Physics is supported by National Science Foundation grant PHY-1066293.

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