Nonequilibrium magnetic and superconducting phases in the correlated electron system coupled to electrodes

Takashi Oka$^{1,2}$ and Hideo Aoki$^1$

$^1$Department of Physics, University of Tokyo, Hongo, Tokyo 113-0033, Japan,
$^2$Theoretische Physik, ETH Zurich, 8093 Zurich, Switzerland

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A theory is presented for a nonequilibrium phase transition in the two-dimensional Hubbard model coupled to electrodes. Nonequilibrium magnetic and superconducting phase diagram is determined by the Keldysh method, where the electron correlation is treated in the fluctuation exchange approximation. The nonequilibrium distribution function in the presence of electron correlation is evoked to capture a general feature in the phase diagram.

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Introduction — While our understanding of the physics of electron correlation has matured, there are still intriguing avenues that are yet to be fully explored. One such avenue is, in our view, strongly correlated electron systems in nonequilibrium situations. While there are a body of intense studies on nonequilibrium states in strong AC fields such as strong light sources that can trigger photo-induced insulator-to-metal transitions (see [1] and refs therein), or nonequilibrium states in strong DC electric fields that can introduce pair-creation of electron and holes in dielectric breakdown [2, 3], here we pursue yet another situation, where nonequilibrium states are conceived for an open, correlated electron system coupled to electrodes (Fig.2 (a) inset). Two effects are expected to arise from the bias voltage $V$ across the electrodes. One is bi-carrier doping, i.e., electrons and holes are simultaneously doped, since two Fermi energies exist due to the two electrodes. Naively one might guess that this can make the system superconducting with Cooper pairs formed by electrons or holes at half-filling, but this has to be tested. There is in fact the second effect of the electron-electron scattering in nonequilibrium that makes the originally sharp Fermi edges to be smeared. The smearing is expected to degrade magnetic orders [4], which in our case implies that the smearing should act to reduce antiferromagnetic order. The natural question then is: will this also destroy the $d$-wave superconducting state?

So we study this problem, which is motivated by two recent experimental developments. One is the fabrication of functional structures with oxides [5, 6, 7]. In refs. [5, 6], properties such as superconducting transition in a clean electron gas formed at an interface of two insulating oxides was studied, while Ueno et al. have succeeded in controlling the superconducting transition in an electrolyte-$\text{SrTiO}_3$ system by changing the applied voltage. Nonlinear transport properties near the Mott transition at interfaces have also been theoretically studied in [8, 9, 10]. The second is an experimental observation by Pothier et al. of the nonequilibrium electron distribution — double-step Fermi distribution — in a mesoscopic copper wire attached to two electrodes [11]. They showed that the step in the distribution is rounded due to electron scattering. The smearing effect is expected to be even stronger in correlated electron systems, so that in order to examine the nature of the nonequilibrium phase transitions in correlated systems, it is theoretically essential to develop a method to deal with the nonequilibrium distribution of quasi-particles in a self-consistent manner. Here we perform this by using the Keldysh method, while the interaction is treated within the fluctuation exchange approximation (FLEX) [12]. The transition to a superconducting state is studied with the linearized Eliashberg equation.

We briefly comment on the past studies on superconductivity transition out of equilibrium. In a pioneering work by Chang and Scalapino [13] who have solved the electron-phonon model self-consistently, it was pointed out that nonequilibrium conditions such as irradiation of light can cause the quasiparticle distribution function to deform, and, under certain conditions, can lead to higher $T_c$ as observed in conventional $s$-wave superconductors [14, 15]. In more recent attempts, the critical properties near an insulator-superconductor transition was studied in [16] followed by several authors [17, 18].

Here we adopt the Hubbard model, a prototype in the study of magnetism, superconductivity and other phase transitions in correlated electron systems. In the two-dimensional square lattice near half-filling, the ground state is the Mott insulator with an antiferromagnetic order [19]. When chemically doped with carriers (electrons or holes), it is believed that Cooper pairs are formed with $d$-wave symmetry and the system turns superconducting, as also discussed phenomenologically in [20, 21, 22]. So the question here is the effect of nonequilibrium on these.

$\text{FLEX+Keldysh method}$ — We consider a thin layer of strongly correlated material described by the two-dimensional Hubbard model which is coupled to electrodes. Here we have assumed for simplicity the top and bottom electrodes (Fig.2 (a) inset), since we want to single out the effect of nonequilibrium situation caused by the bias voltages, while a lateral attachment of the electrodes would cause a change in the spatial symmetry of the phases. The total Hamiltonian is then given by $H = H_{\text{sys}} + H_{\text{sys-lead}} + H_{\text{lead}}$, where $H_{\text{sys}}$ is the Hubbard Hamiltonian with the hopping integral $t$
through the Dyson equation, \( \Sigma^\alpha = \Sigma^\alpha_{\text{lead}} + \Sigma^\alpha_{\text{int}} \) of contributions from the electrodes and from the interaction, where \( \alpha = r, a, <, >, K \) denote, respectively, the retarded, advanced, lesser, greater, and Keldysh components (see e.g., [27]). If we label the top and bottom electrodes by \( \gamma = 1, 2 \) the electrode self-energy becomes \( \Sigma^\alpha_{\gamma} = 2i \sum_{\gamma=1,2} \frac{\Gamma_\gamma}{\pi} \tanh \frac{\gamma \mu}{2T} \), \( \Sigma^\alpha_{\text{lead}} = -i \sum_{\gamma=1,2} \frac{\Gamma_\gamma}{\pi} \) where \( \Gamma_\gamma \) is the coupling strength between the system and the electrodes, \( \mu \) and the energy dependence in the density of states is neglected. Here, \( \mu \) represents the respective chemical potential of the electrodes while their temperature \( T \) is kept to be the same, and we fix \( \Gamma_\gamma = 0.001 \). We note that if the coupling is too strong \( (\Gamma_\gamma \approx 0.1) \), no ordering takes place. Within the FLEX, the self-energy arising from the electron interaction is given by

\[
\Sigma_{\text{int}}^{>,<}(p, \omega) = -i \int \frac{d\omega'}{2\pi N} \sum_k P^{>,<}_\text{eff}(k, \omega') G^{>,<}(p - k, \omega - \omega'),
\]

where \( p, k \) are momenta, \( \omega \) the frequency, \( N \) the number of \( k \)-points considered. The retarded component is determined from \( \text{Im} \Sigma^\alpha_r = \frac{1}{2} (\Sigma^\alpha_+ - \Sigma^\alpha_-) \), to which the real part is related via Kramers-Kronig’s relation. This relation between the lesser, greater components and the retarded component is valid for other quantities as well. The fluctuation interaction is given by \( P^{>,<}_\text{eff} = U^2 \text{Im} \left( \frac{1}{2} \chi^0_{\alpha} < + \frac{1}{2} \chi^0_{\alpha} < - \chi^0_{\alpha} < \right) \), where \( \chi^0_{\alpha} \) represent the spin (charge) susceptibilities, whose retarded components are \( \chi^0_{\alpha} = \chi^0_{\alpha}/(1 - U \chi^0_{\alpha}) \). Here \( \chi^0 \) is the irreducible susceptibility,

\[
\chi^{>,<}_0(q, \omega) = -i \int \frac{d\omega'}{2\pi N} \sum_k G^{<,>}(k, \omega') G^{>,<}(k + q, \omega + \omega').
\]

The lesser and greater components of \( \chi^0_{\alpha} \) are determined with the help of the Langreth rules [27]. Finally, the Green’s function is determined from the self-energy through the Dyson equation, \( (G^{\alpha, -1}) = (G^{\alpha, 0})^{-1} - \Sigma^\alpha \) for the retarded and advanced, and \( G^<, > = G^+ \Sigma^<, > < G^0, > \) for the Keldysh component [26] with \( (G^{\alpha, 0})^{-1} = \omega - \varepsilon_k + i\delta \). The process is repeated until a self-consistent solution is obtained. The nonequilibrium distribution function \( f_{\text{eff}} \) can be extracted from the relation,

\[
G^K = (1 - 2 f_{\text{eff}}) (G^+ - G^-).
\]

We seek for a self-consistent solution of the above equations with iteration until the self-energy converges. In the calculation we take a \( 64 \times 64 \) grid for the square Brillouin zone, while an almost logarithmic mesh [22, 29] with 301 points for the \( \omega \)-axis is used. We note that the distribution function \( f_{\text{eff}} \) deviates significantly from its non-interacting form (double step Fermi function),

\[
f_{\text{eff}}^0 = [\Gamma_1 f_{\text{FD}}(\omega - \mu_1) + \Gamma_2 f_{\text{FD}}(\omega - \mu_2)]/(\Gamma_1 + \Gamma_2),
\]

(with \( f_{\text{FD}} \) being the Fermi-Dirac distribution) as an effect of strong interaction as discussed below.

The superconducting transition is studied in terms of the linearized Eliashberg equation, here extended to nonequilibrium. To this end, we iteratively \( (i = 1, 2, \ldots) \) obtain a series of anomalous self-energy \( (\phi^0_i) \) and anomalous Green’s function \( (F^i_1) \) using \( \Sigma^\alpha, \chi^\alpha_{r,c} \) obtained in the previous step. With a random initial guess for \( \phi^0_1 \), the Green’s function is determined from the linearized Nambu-Gor’kov equation, \( F^i_1 = \phi^i_1/(\langle \omega Z \rangle^2 - \langle \varepsilon_k + X \rangle^2) \), with \( \omega Z = \omega - \langle \Sigma^\alpha (\omega) - (\Sigma^\alpha (\omega))^\dagger \rangle / 2 \) and \( X = \langle \Sigma^\alpha (\omega) + (\Sigma^\alpha (\omega))^\dagger \rangle / 2 \). Then the lesser (greater) components are calculated with the generalized distribution function,

\[
F^i_1 = (1 - 2 f_{\text{eff}}) (F^i_1 - F_1^0).
\]

We assume here that the distribution of the anomalous component is the same as the normal component. Finally, we plug this into the Eliashberg equation,

\[
\phi^{>,<}_{i+1}(p, \omega) = -i \int \frac{d\omega'}{2\pi N} \sum_k P^{>,<}_\text{sing}(k, \omega') F^{>,<}_i(p - k, \omega - \omega'),
\]

where the effective interaction in the singlet channel is \( P^{>,<}_\text{sing} = U^2 \text{Im} \left( \frac{1}{2} \chi^0_{\alpha} < - \frac{1}{2} \chi^0_{\alpha} < \right) \). The eigenvalue of the linearized Eliashberg equation is obtained as \( \lambda = \lim_{\delta \to \infty} \langle \delta \phi^{i+1}_1 || \delta \phi^i_1 || \rangle \), where \( \langle \delta \phi^i_1 || \rangle = (\int d\omega \frac{1}{N} \sum_k \phi^i_1(p, \omega))^2 \) is the norm. The superconducting transition takes place when \( \lambda \) exceeds unity.

**Nonequilibrium phase transition** — We have applied the above formalism to obtain the nonequilibrium phase diagram of the two-dimensional (square lattice) Hubbard model attached to two electrodes by numerically solving the equations self-consistently. In equilibrium the phase diagram within FLEX as obtained in [12] has an antiferromagnetic phase when the doping level \( \delta = 1 - n \) is small, which is taken over by a \( d \)-wave superconductor as \( \delta \) is increased. So the interest is how these are modiﬁed. The result for the nonequilibrium situation in Fig. [1](a), where we plot the spin susceptibility \( \text{Im} \chi_\alpha(q, \omega) \) for \( V = 0.1 \) and a doping level \( \delta = 0.14 \), shows that the antiferromagnetic fluctuation remains strong near half-filling. We have four incommensurate peaks around \( q = (\pi, \pi) \), as in equilibrium. The effect of increased bias is that the peak height is reduced and the peak position on energy axis shifts upwards as displayed in Fig. [1](c), where \( \text{Im} \chi_{\text{peak}}(q, \omega) \) for \( q = (\pi, 1.1\pi) \) is plotted. We notice that no features such as dip or hump appear around \( \omega \approx V \). The dominant superconducting solution in Fig. [1](b) is again similar to the equilibrium case, that is, the \( d \)-wave gap has the largest \( \lambda_d \) for the linearized Eliashberg equation. However, the critical temperature \( T_c \) at which
λ_d = 1 does depend on V, as shown by the temperature dependence of λ_d plotted in Fig. 1(d). So the bias V reduces T_c, until finally the superconducting state no longer exists even at zero temperature when the bias becomes too strong. We define this as the critical bias V_c. For the region of the band filling for which the antiferromagnetic order dominates over the superconducting state, we can similarly define the bias-dependent Néel temperature T_N as the temperature at which the spin susceptibility diverges. The spin susceptibility is reduced as the bias increases, until antiferromagnetic order ceases to exist even at zero temperature beyond the “critical Néel bias” V_N. The doping dependence of the Néel bias and the critical temperatures for a fixed bias is shown in Fig. 2(a). We can see that, while the antiferromagnetic (AF) phase is relatively persistent, the superconducting (SC) region rapidly shrinks with the bias V and disappears at V ≈ 0.1. In Fig. 2(b) we give the zero-temperature phase diagram on the (V, δ) plane. The Néel bias, peaked at the undoped point with V_N ≈ 0.36, decreases with the doping, and the AF phase is replaced with the SC phase around δ ≈ 0.1 with a maximum critical bias for SC V_c ≈ 0.1. As we further increase the doping, the SC phase finally disappears. Figure 2(c) schematically summarizes these phase transitions in the (T, V, δ) space.

Nonequilibrium distribution function — As was experimentally found in a tunneling measurement in a mesoscopic wire of copper by Pothier et al., the nonequilibrium electron distribution becomes smeared from the simple double-step Fermi distribution f_0 due to electron scattering. In correlated materials, with a strong electron-electron interaction, we expect a greater smearing effect to take place. Indeed, as we shall reveal below, the key feature to understand the nonequilibrium phase diagram for the open Hubbard model may be captured by the way in which the nonequilibrium distribution function is rounded by the interaction effect.

Figure 3(a) plots the effective distribution f_eff defined in eq. (3) obtained self-consistently for V = 0.06 and V = 0.38. The temperature in the electrodes and thus in f_0 is set to zero. We compare the result with the corresponding noninteracting distribution function f_0 (eq. (1)) (dashed lines), f_eff is seen to significantly deviate from f_0. More importantly, we find here that the effective temperature approximation breaks down, that is, we cannot fit f_eff to f_0 with the temperature as a fitting parameter. Instead, the best fit to the data is given by

\[ f_{\text{fit}}(\omega) = \begin{cases} 
1 - \alpha e^{-\omega/V/2/\tau}, & \omega < -V/2 \\
-(1 - 2\alpha \omega/V + 1/2), & -V/2 < \omega < V/2 \\
\alpha e^{-(\omega-V/2)/\tau}, & V/2 < \omega
\end{cases} \] (7)

where α and τ are the fitting parameters. The parameter τ having the dimension of energy represents the extent to which the distribution is smeared from the double-step function. We have found in Fig. 3(b) that the
FIG. 3: (Color online) (a) Nonequilibrium distribution function for two values of the bias \( V \) at half filling \( (\delta = 0) \). Dashed lines are the noninteracting distribution function \( f_{\text{eq}}^0 \). (b) Nonequilibrium distribution function \( (\text{dots}) \) against \( \omega < -V/2 \) region for \( V = 0.08, 0.19, 0.32, 0.47, 0.63, 0.80 \) from the top, where curves represent a fit with eq. (8). (c) The smearing parameter \( \tau \) against the bias \( V \) for various values of \( \delta \) and fixed \( U = 4.5 \) and \( T = 0 \). Fitting errors are smaller than the size of each symbol.

The fitting function eq. (8) is general in the present open Hubbard model in that all the data with different parameters \( (V, \Gamma, U, \delta, \ldots) \) are reproduced within the numerical errors. If we specifically plot the bias-dependence of the smearing parameter in Fig. 3 (c), we can see that they fall upon an approximately linear, universal relation

\[ \tau \propto V. \]  

(8)

The proportionality constant in this relation depends on the interaction strength \( U \) and the coupling \( \Gamma \) to the electrodes, but not on the filling \( \delta \) as seen from the figure. The constant is reduced when the coupling to the electrode becomes stronger.

From the viewpoint of the smeared distribution, we can view the bias-driven phase transitions in the following way. We have seen in Fig. 2 (b) that the AF (SC) orders die out at \( V \approx 0.4 \) \((V \approx 0.1)\) respectively. In terms of the relation (8), these values correspond to the smearing parameters \( \tau \approx 0.1 \) \((\tau \approx 0.02)\). We can then note that these values are similar to the highest Néel (critical) temperatures in the zero bias phase diagram (Fig. 2(a), upper panel). Thus, the transition takes place when the smearing parameter \( \tau \) attains a value (depth of each ordered region in Fig. 2(c) translated to \( \tau \)) similar to the transition temperature (height of the region). AF spin fluctuations are suppressed in finite bias voltages in this manner, which is similar to what happens in itinerant electron magnets [4].

**Discussion** — We have obtained a nonequilibrium phase diagram for the two-dimensional Hubbard model and pointed out the possibility of controlling the phases in strongly correlated heterostructures by external bias. Both of AF and SC regions shrink with the bias \( V \), which we attribute to the smearing of the nonequilibrium distribution function. While the smearing can be reduced if we make the system more strongly coupled to the electrodes (in, e.g., a thinner sample), this will lead to the destruction of order because a larger electrode coupling \( \Gamma \) will make the spin fluctuations weaker. Thus we conclude the smearing of the distribution function is an important property of correlated electron systems out of equilibrium, and an experimental verification of this should be interesting. We have to make a caution that FLEX employed here has limitations in that it ignores the vertex correction, cannot address, due to its weak-coupling nature, the behavior close to the Mott insulator point. Effects of electrodes (on, e.g., the pairing symmetry) when they are attached laterally are also intriguing. A more ambitious future problem is a possibility of bi-carrier induced superconductivity in nonequilibrium, for which the present formalism may serve as a starting point.

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