Universality in Voltage-driven Nonequilibrium Phase Transitions$

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We consider the non-equilibrium ferromagnetic transition of a mesoscopic sample of a resistive Stoner ferromagnet coupled to two paramagnetic leads. The transition is controlled by either the lead temperature $T$ or the transport voltage $V$ applied between the leads. We calculate the $T$ and $V$ dependence of the magnetization. For systems with a flat density of states we find within mean-field theory that even at finite bias the magnetization does not depend on the position along the sample axis, although the charge density and other quantities do vary. This may be relevant for possible spintronics applications. In addition, we establish a generalized control parameter in terms of $T$ and $V$ which allows for a universal description of the temperature- and voltage-driven transition.

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1. INTRODUCTION

Phase transitions in interacting electron systems far from thermodynamic equilibrium are an exciting, relatively young field of research, both for fundamental theoretical reasons and for application purposes. While the theory of equilibrium phase transitions is well developed, a theory for phase transitions out of equilibrium is much more difficult to formulate, largely because in the most general case the thermodynamic states of the system are not characterized by a minimum of the free energy and because often the temperature $T$ as a state variable driving the transition is not even

$\ast$Dedicated to Prof. Hilbert von Löhneysen on the occasion of his 60th birthday.

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defined. On the other hand, nonequilibrium phase transitions are of high technological potential, since a mesoscopic electron system can be driven out of equilibrium in a highly controlled way, and controlling a nonequilibrium phase transition by externally applied fields can be envisaged. It opens up the prospect of fast switching between different magnetic or conducting states of a mesoscopic system, e.g. by applying a transport bias voltage or by an applied laser field, with possible spintronics applications.

A thermodynamic phase transition in equilibrium is in general controlled by the minimum of the free energy

\[ F = U - TS, \]

i.e. by the balance of the internal energy \( U \) and the entropy \( S \). The dependence on the control parameter temperature favors an ordered phase at low \( T \) and a disordered phase at high \( T \). In a stationary nonequilibrium situation internal energy and entropy are statistically still well-defined quantities. One may, therefore, conjecture that a relation analogous to Eq. (1) holds even in this case. The different phases of a stationary non-equilibrium system would then again be determined by the minimum of the free energy. However, in the above relation (1) the entropy coefficient \( T \) would have to be generalized, since temperature \( T \) is in general not defined in nonequilibrium. Based on general thermodynamic arguments we propose in the present article a definition of such a generalized control parameter \( \tau \) which allows to describe electron systems driven out of equilibrium by an applied stationary bias voltage \( V \) in a unified way.

To be specific, we consider the voltage-driven magnetic phase transition of a Stoner ferromagnet. Such a system can be realized as a mesoscopic sample of a magnetic metal, contacted by two paramagnetic electron reservoirs or leads at chemical potentials \( \mu_L \) and \( \mu_R \), respectively, as seen in Fig. 1a). It has recently been demonstrated that the fabrication of such devices, which sustain a high nonequilibrium bias, is technologically feasible using metallic nanobridges with high lead-to-bridge aspect ratio \( 1 \) or using resistive nanowires \( 2 \). As will be seen below, the ferromagnetic transition in the mesoscopic sample can be controlled both by the reservoir temperature \( T \) and by the voltage \( V = \mu_L - \mu_R \) applied between the reservoirs. The magnetic state of the sample may be probed by a third, ferromagnetic electrode, attached to the sample via a tunnel junction. This electrode stays in equilibrium with itself, independent of the transport voltage \( V \), so that it remains in the ferromagnetically ordered state throughout the transition of the sample, and its tunneling conductance depends strongly on the magnetization in the sample. A related proposal of controlling magnetism by a nonequilibrium transport bias has recently been put forward in Ref. [3]. However, in that
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proposal the magnetization was induced in a paramagnetic metal sample by ferromagnetism in the leads, and the sample was assumed to be zero-dimensional in the sense that it was smaller than the electrons’ elastic mean free path $\ell$, implying ballistic transport and a position independent electron distribution function in the metal sample. In the present work we consider the more realistic case of a resistive, ferromagnetic sample whose lateral size $L$ is greater than the elastic mean free path, $L > \ell$, but still smaller than the inelastic and spin relaxation lengths, $L < \ell_{\text{in}}, \ell_{\text{s}}$. In this case the transport is diffusive, leading to a nonequilibrium electron distribution function which depends on the location along the sample. We develop a mean field theory for the temperature- or voltage-driven Stoner transition.

One might think that this transition, driven by the transport voltage at reservoir temperature $T = 0$ might be a quantum phase transition, since it is controlled by a parameter different from the temperature. However, at any finite voltage $V \neq 0$ the nonequilibrium quasiparticle distribution is such that there exists a finite phase space for inelastic scattering (see, e.g., Section 2.1), so that the quantum dephasing rate in the sample, $\gamma_{\varphi}$, remains finite even for reservoir temperature $T = 0$. Thus, critical divergencies of correlation functions at the phase transition are dominated by classical, statistical fluctuations rather than quantum fluctuations, i.e. the transition belongs to the universality class of classical rather than quantum phase transitions, in contrast to the assertion of Ref. [4]. Remarkably, the authors of Ref. [4] do, however, find that the transition is governed by the classical critical exponents. In fact, as will be seen in Section 3, the roles of $V$ and $T$ are completely analogous, as far as thermodynamic properties are concerned.

This paper is organized as follows. In Section 2 we define the Stoner model in a steady-state nonequilibrium situation with finite bias. The quantum Boltzmann equation for a resistive wire with a Stoner mean-field interaction is derived in Section 2.1, and the Stoner mean-field equations in the presence of finite bias are presented in Section 2.2, along with their numerical solution for various temperatures and voltages. In Section 3 we provide general thermodynamic arguments which allow us to introduce a control parameter $\tau(T, V)$, depending on the reservoir temperature $T$ and the bias voltage $V$, as a generalization of the temperature to the voltage-biased non-equilibrium situation, as far as thermodynamic properties are concerned. We demonstrate explicitly that this provides a universal description of the voltage-driven Stoner transition. Some concluding remarks are given in Section 4.
Fig. 1. a) Schematic view of a magnetic transistor. The ferromagnetic mesoscopic sample is shown in light grey. It is attached to paramagnetic leads kept at chemical potentials $\mu_L$ and $\mu_R$, respectively. A magneto-tunneling current depending on the sample magnetization can be extracted through the third electrode FM. The position coordinate along the sample of length $L$ is denoted by $s$, as shown in the figure. b) The Keldysh Hartee-Fock diagrams defining the Stoner mean-field theory in stationary nonequilibrium. Solid lines denote the electron propagators, wavy lines the screened Coulomb interaction $U$.

2. VOLTAGE-DRIVEN STONER FERROMAGNETISM

As a simple model for a ferromagnetic transition in an itinerant, diffusive electron system we consider electrons on a lattice with dispersion $\varepsilon_k$, with a random potential $V_j$ induced by static impurities on lattice sites $j$ and a strongly screened, local Coulomb repulsion $U$ between electrons on the same site,

$$H = \sum_{k\sigma} \varepsilon_k c_{k\sigma}^+ c_{k\sigma} + \sum_{j\sigma} V_j c_{j\sigma}^+ c_{j\sigma} + U \sum_{j} c_{j\uparrow}^+ c_{j\downarrow}^+ c_{j\downarrow} c_{j\uparrow}.$$

Here $c_{k\sigma}^+$, $c_{k\sigma}$ denote the electron creation and destruction operators with momentum $k$ and spin $\sigma = \pm 1/2$, and $c_{j\sigma}^+ = \sum_k \exp(ikx_j)c_{k\sigma}^+$ is the creation operator at site $j$. We assume that any antiferromagnetic instabilities that could be induced by nearest neighbor hopping on a cubic lattice are frustrated, e.g., by an appropriate lattice structure or by a next-nearest neighbor hopping included in the band structure $\varepsilon_k$.

2.1. Nonequilibrium Distribution in a Diffusive Ferromagnet

Any theory of an interacting system out of equilibrium requires knowledge of the quasiparticle distribution $f_{\sigma x}(\omega)$ as a function of the particle energy $\omega$. In a paramagnetic, diffusive system at bias $V$, if inelastic relax-
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ations can be neglected, \( f_{\sigma x}(\omega) \) is known to have a double-step form according to the two Fermi edges in the reservoirs, and to depend linearly on the position \( x = s/L \) along the sample axis (see Fig. 1a). In a ferromagnet, however, the Stoner splitting may in general depend on \( x \) as well, inducing nontrivial position and spin dependences to \( f_{\sigma x}(\omega) \) even in the absence of inelastic or dynamical spin flip processes. In the following we derive the distribution function for the case of a mesoscopic ferromagnet, where inelastic and spin flip processes can be neglected.

The kinetic equation (quantum Boltzmann equation) for \( f_{\sigma x}(\omega) \) is obtained as the off-diagonal part of the Keldysh Dyson equation for the lesser Green’s function \( G_{\sigma x}^<(\omega, x, x') \). After transforming to center-off-mass and relative coordinates, \( r = (x + x')/2, \Delta x = x - x' \), taking \( \Delta x \) to be confined in a small volume compared to the scale on which the external fields vary, and Fourier transforming to momentum space with respect to \( \Delta x \) within that volume, the resulting quantum Boltzmann equation acquires the form of a continuity equation in phase space,

\[
\left[ \frac{\partial}{\partial t} + v_k \cdot \nabla r + \frac{-e}{\hbar} \mathbf{E}(r) \cdot \nabla k \right] G_{\sigma x}^<(k, \omega) = \mathcal{I} \{ f_{\sigma x}(k) \} \quad (3)
\]

where \( G_{\sigma x}^<(k, \omega) = 2\pi i f_{\sigma x}(k) A_{\sigma x}(k, \omega) \) is expressed in terms of the distribution \( f_{\sigma x}(k) \) and the spectral density \( A_{\sigma x}(k, \omega) \) of momentum \( k \) at position \( r \) in the sample. \( v_k = \partial r/\partial t = \partial \varepsilon_k/\partial k \) is the electron group velocity and \(-e\mathbf{E}(r) = \hbar \partial k/\partial t \) the force acting on the electrons due to an external field \( \mathbf{E}(r) \). The latter includes both, the external bias voltage and the electrostatic potential generated by the random impurities. Since we consider a stationary situation and assume that the sample is smaller than the inelastic and the spin relaxation lengths, the explicit time derivative and the collision integral \( \mathcal{I} \{ f_{\sigma x}(k) \} \) vanish in Eq. (3). After summation over \( k \) the force term vanishes as well. This is because of the relation

\[
\sum_{k, \sigma} \nabla_k \cdot \mathbf{E}(r) f_{\sigma x}(k) A_{\sigma x}(k, \omega) = \oint_{S_k} \mathbf{E}(r) f_{\sigma x}(k) A_{\sigma x}(k, \omega) \cdot dS_k = 0 \quad (4)
\]

Here the second term implies an integration over the surface of the 1st Brillouin zone, where \( f_{\sigma x}(k) \) is either 0 or 1, i.e. \( f_{\sigma x}(-k) = f_{\sigma x}(k) \) and \( dS_{-k} = -dS_k \). The same \( k \)-summation also introduces the particle and the current density in Eq. (3),

\[
\rho_{\sigma x}(\omega) = \sum_k f_{\sigma x}(k) A_{\sigma x}(k, \omega) \quad (5)
\]

\[
j_{\sigma x}(\omega) = \sum_k v_k f_{\sigma x}(k) A_{\sigma x}(k, \omega) \quad (6)
\]
which in a diffusive system with diffusion coefficient $D$ are related by Fick’s law,

\[ j_{\sigma r}(\omega) = -D \nabla_{r} \rho_{r}(\omega) \, . \] (7)

In this way the diffusive Boltzmann equation in a ferromagnet takes the form

\[ -D \nabla_{r}^{2} [f_{\sigma r}(\omega)N_{\sigma r}(\omega)] = 0 \, , \] (8)

where $N_{\sigma r}(\omega) = \sum_{k} A_{\sigma r}(k, \omega)$ is the Stoner split electronic density of states (DOS) at position $r$ in the sample. Note that in a paramagnetic system $N_{\sigma r}(\omega)$ is in general position independent and can be dropped from Eq. (8). In a ferromagnet, however, the spectral function contains the Stoner selfenergy $\Sigma_{\sigma r}$ (see Section 2.2), which depends in general on the position $r \equiv s$,

\[ A_{\sigma x}(k, \omega) = \frac{1}{\pi} \text{Im} \frac{1}{\omega + \mu_{0} - \varepsilon_{k} - \Sigma_{\sigma x} + i0} \, , \] (9)

where $s$ is the coordinate along the sample axis (see Fig.1a)) and $x = s/L$. Taking into account only the linear dependence of the DOS on $x$, with $N'_{\sigma}(\omega) \equiv dN_{\sigma x}(\omega)/dx|_{x=0.5} = -[dN_{\sigma x}(\omega)/d\omega \cdot d\Sigma_{\sigma x}/dx]|_{x=0.5}$ and $N_{\sigma}(\omega) \equiv N_{\sigma x}(\omega)|_{x=0.5}$, the quantum Boltzmann equation in a resistive ferromagnet takes the final form

\[ -D \frac{d^{2}f_{\sigma x}(\omega)}{dx^{2}} - 2D \frac{N'_{\sigma}(\omega)}{N_{\sigma}(\omega)} \frac{df_{\sigma x}(\omega)}{dx} = 0 \, . \] (10)

The second term in this equation implies a selfconsistent coupling to the Stoner mean-field equations via the spin and position dependent DOS. However, Eq. (10) can readily be solved analytically, once $N'_{\sigma}(\omega)$ and $N_{\sigma}(\omega)$ are determined. This will be done in a forthcoming publication. In the present paper we wish to focus on universality related issues, which can be discussed as well for the particle-hole symmetric case. Therefore, we will assume in the following that the DOS is flat within the energy window defined by the applied bias voltage $V$. In this case $N'_{\sigma}(\omega) = 0$, and the kinetic equation (10) reduces to the form familiar from noninteracting diffusive systems. In particular, $f_{\sigma x}(\omega)$ is then spin independent, since the boundary conditions on $f_{\sigma x}(\omega)$ are spin symmetric, i.e. both spin species in the sample couple to the same paramagnetic reservoirs.

\[ f_{\sigma x}(\omega) = f_{-\sigma x}(\omega) \equiv f_{x}(\omega) \quad \text{for} \quad N'_{\sigma}(\omega) = 0 \, . \] (11)

The Boltzmann equation (10) is easily solved for this case with the boundary conditions that in the left (right) lead there is an equilibrium distribution with chemical potential $\mu_{L} = +eV/2$ and $\mu_{R} = -eV/2$, respectively,

\[ f_{x}(\omega) = x f^{(0)}(\omega + \frac{eV}{2}) + (1 - x) f^{(0)}(\omega - \frac{eV}{2}) \, , \] (12)
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where \( f^{(0)}(\omega) = 1/[\exp(\omega/k_BT) + 1] \) is the Fermi distribution and \( T \) the reservoir temperature.

### 2.2. Stoner Mean-Field Equations in Stationary Nonequilibrium

For simplicity we assume an elliptic DOS (per spin) of the disordered system without interactions,

\[ N_0(\omega + \mu_0) = \frac{2}{\pi} \sqrt{1 - (\omega + \mu_0)^2}, \]

where the single-particle energy \( \omega \) is measured relative to the equilibrium chemical potential \( \mu_0 \), and all energies are given in units of the half bandwidth \( B \).

To realize a flat DOS for energies within the voltage window, \(-eV/2 \leq \omega \leq eV/2\), \( \mu_0 \) is placed at the band center, \( \mu_0 = 0 \), and \( eV \ll B \). As discussed in Section 2.1, the distribution function is then decoupled from the solution of the Stoner model and is given by Eq. (12).

The Hartree-Fock selfenergy for this model is represented by the Keldysh diagrams shown in Fig. 1b). Note that all vertex points of the instantaneous interaction \( U \) are on the same branch of the Keldysn contour, as indicated by the “−” signs, and that the equal-time operator products are normal-ordered. The internal equal-time propagators are, therefore, the lesser Green’s functions \( G^< (\omega) \). The Hartree-Fock selfenergy then reads,

\[
\Sigma_\sigma(T, V, x) = U \langle n_{-\sigma} \rangle = U \int d\omega f_x(\omega) N_0[\omega + \mu_0 - \Sigma_{-\sigma}(T, V, x)] ,
\]

where \( \langle n_\sigma \rangle \) is the conduction electron number per site with spin \( \sigma \). At \( T = 0 \) the integral can be evaluated, and the \( V \) and \( x \) dependence can be made explicit,

\[
\Sigma_\sigma(0, V, x) = \frac{U}{\pi} \left[ x \left( \Omega_{-\sigma} \sqrt{1 - (\Omega_{-\sigma})^2} + \arcsin(\Omega_{-\sigma}) \right) \right.
+ \left. (1 - x) \left( \Omega_{+\sigma} \sqrt{1 - (\Omega_{+\sigma})^2} + \arcsin(\Omega_{+\sigma}) \right) + \pi \right] ,
\]

with \( \Omega_{\pm,\sigma} = \pm eV/2 + \mu_0 - \Sigma_\sigma(0, V, x) \). The selfenergy can be separated in a spin independent and a spin dependent part, \( \Sigma_\sigma = \Sigma_0 + \sigma \Delta \Sigma \), with

\[
\Delta \Sigma = \Sigma_\uparrow - \Sigma_\downarrow \quad \text{and} \quad \Sigma_0 = \frac{1}{2} \langle n \rangle \Delta \Sigma.
\]

It is now crucial that the definition (12) of the nonequilibrium distribution fixes the zero of the energy to be in the middle between the two Fermi steps for each position \( x \) along the sample. Therefore, in Eqs. (13), (14) the interaction-induced, spin independent shift of the energy \( \Sigma_0 \) must be
Fig. 2. The magnetization per site is shown as a function of $T$ for various values of the transport voltage $V$ as indicated. $V$ is given in units of the equilibrium Curie temperature $T_{c0}$. The screened Coulomb interaction was chosen such that $T_{c0} = 0.01 \, B$ (i.e., $UN_0(0) = 1.0001669$). The small dips in the $m(T)$ curves near $T = 0$ are a consequence of the slightly non-constant DOS for high bias $V$.

The mean-field solutions for the magnetization $m$ as a function of reservoir temperature $T$ are shown for various applied transport voltages $V$ in Fig. 2 and as a function of $V$ for various $T$ in Fig. 3. For these data...
Fig. 3. The magnetization per site is shown as a function of $V$ for various values of $T$, as indicated. The parameter values used are as in Fig. 2.

the strength of the screened Coulomb interaction $U$ has been chosen such that the Stoner criterion, $U N_0(0) > 1$, is satisfied and that the equilibrium Curie temperature is $T_{c0} = 0.01 \, B$, corresponding to $T_{c0} \approx 300 \, K$ for a half bandwidth of $B = 3 \, eV$.

3. UNIVERSALITY

The fact that the Stoner transition can be controlled by both, temperature $T$ and transport voltage $V$, the similarity of the magnetization curves $m(T)$ and $m(V)$ in Figs. 2 and 8 and the independence of the magnetization of the position $x$ in the sample for a flat DOS suggest that there is a universality in this transition in terms of the two control parameters $T$ and $V$. Indeed, the complete thermodynamics of any system is determined, once the internal energy $U$ or the free energy $F$ (with appropriate state variables) is known. At least the internal energy is well-defined also in a stationary nonequilibrium situation. For any stationary electron system in or out of equilibrium, $U$ is uniquely determined by the quasiparticle distribution function $f_x(\omega)$. The latter is, in turn, uniquely defined by the applied transport voltage $V$ (and the concomitant boundary conditions), if the system is coupled to one or more reservoirs to ensure stationarity, and by the reservoir temperature $T$. This suggests that it should be possible to define a single, unified state variable, involving both $T$ and $V$, in terms of the internal energy.
Fig. 4. Collapse of all magnetization data contained in Figs. 2 and 3 onto a single curve, depending on $\tau(T, V)$ (Eq. 18) only. The data for different parameter values of $T$ and $V$ are displayed here using different symbols.

Therefore, we put forward the following procedure. The temperature of an equilibrium system is usually measured by bringing it in heat contact with a reference system whose internal energy is known as a function of $T$, like an ideal gas (ideal gas thermometer). Since the ideal gas is not suitable for applying a transport voltage, we propose to use a diffusive, noninteracting electron gas as a reference system for any voltage-biased, interacting system, like, e.g., a mesoscopic Stoner ferromagnet, Eq. (2). As before, we will restrict ourselves here to the case that the DOS is constant within the voltage window, $N_0(0)$, and defer the general case to a forthcoming paper. The change of the internal energy density of this reference system as a function of $T$ and $V$ is well-defined and is readily calculated as,

$$
\Delta U = U(T, V) - U(0, 0)
= N_0(0) \int_{-\infty}^{\infty} d\omega \omega \left[ f_x(\omega, T, V) - f^{(0)}(\omega, 0, 0) \right]^2 \ .
$$

Remarkably, $\Delta U$ does not depend on the position $x$ where the energy density is considered, as long as the DOS is flat, in accordance with the results of Section 2.2. The expression (17) can now be used to extract a generalized state variable, $\tau(T, V)$, that uniquely determines the internal energy of the reference system, and thus the thermodynamics of any stationary electron
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system controlled by the state variables $T$ and $V$,

$$\tau(T, V) = \sqrt{T^2 + \frac{1}{4L}V^2},$$

(18)

where $L = \frac{\pi^2 k_B^2}{3e^2}$ is the Lorentz number, and $\Delta U = (\pi^2/6)N_0(0)\tau^2$. Note that the form of the expression for $\tau(T, V)$ is equivalent to the effective temperature of a voltage-biased, hot electron system that has equilibrated due to strong inelastic processes. However, in our line of arguments above no assumption has been made about whether or not there are inelastic processes involved. This supports the universality of the definition Eq. (18).

We have tested whether the Stoner transition can be described by $\tau(T, V)$ in a universal way. In Fig. 4 we have plotted all the data contained in Figs. 2 and 3 as a function of $\tau(T, V)$. Indeed there is a perfect collapse of all data onto a single curve, as expected.

4. CONCLUSION

Even the simple Stoner model of a ferromagnetic transition can exhibit interesting behavior when considered in a non-equilibrium situation. We have used this model to define a generalized control parameter $\tau(T, V)$ which allows to describe the voltage and temperature dependence of the Stoner transition in a unified way. The model has been evaluated within mean-field theory where the critical exponent of the magnetization in dependence of both $T$ and $V$ is $\alpha = 1/2$. We emphasize that the generality of our thermodynamic arguments that led to the definition of $\tau(T, V)$ strongly suggest that it not only allows for a universal description of the magnetization but also of the complete thermodynamics of the Stoner model, and that this holds true even beyond mean field theory. In particular, correlations will certainly change the value of $\alpha$, but it will remain the same for the $T$ and the $V$ dependence. Moreover, the thermodynamic arguments suggest that not only Stoner ferromagnets but the thermodynamics of any interacting electron system controlled by the reservoir temperature $T$ and a nonequilibrium transport voltage $V$ can be described by $\tau(T, V)$ in a universal way. Conversely, it is obvious that dynamical and spectral properties are influenced by $T$ and $V$ in quite different ways, which cannot be captured only by the $T$ and $V$ dependence of the internal energy. Therefore, no unified behavior is expected for these quantities.

As mentioned above, the magnetization $m$ will acquire an $x$-dependence, as soon as the density of states in the sample as function of energy is not flat. In this case the interaction-induced correlation length will also influence the spatial dependence of $m$. The thermodynamics of voltage-driven systems,
the influence of correlations in general and the position dependence of the magnetization in particular will be subjects of further research.

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