IOPscience iopscience.iop.org

Home Search Collections Journals About Contact us My IOPscience

Quantum reciprocity conjecture for the non-equilibrium steady state

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2004 J. Phys.: Condens. Matter 16 L263

(http://iopscience.iop.org/0953-8984/16/20/L02)

View the table of contents for this issue, or go to the journal homepage for more

Download details:
IP Address: 129.252.86.83
The article was downloaded on 27/05/2010 at 14:37

Please note that terms and conditions apply.

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **16** (2004) L263–L269 PII: S0953-8984(04)76853-8

LETTER TO THE EDITOR

Quantum reciprocity conjecture for the non-equilibrium steady state

P Coleman and W Mao

Center for Materials Theory, Rutgers University, Piscataway, NJ 08854, USA

Received 25 February 2004
Published 7 May 2004
Online at stacks.iop.org/JPhysCM/16/L263
DOI: 10.1088/0953-8984/16/20/L02

Abstract
A consideration of the lack of history dependence in the non-equilibrium steady state of a quantum system leads us to conjecture that in such a system there is a set of quantum mechanical observables whose retarded response functions are insensitive to the arrow of time, and which consequently satisfy a quantum analogue of the Onsager reciprocity relations. Systems which satisfy this conjecture can be described by an effective free energy functional. We demonstrate that the conjecture holds in a resonant level model of a multi-lead quantum dot.

Although the fundamental principles of thermal equilibrium were established by Boltzmann more than a century ago, their generalization to the non-equilibrium steady state has proved elusive. The non-equilibrium steady state is thought to be defined by a set of characteristic variables such as the current and the thermal and chemical potential gradients, and as such it is expected to be independent of the history of how it was prepared. This has led to the notion that general principles should govern the instantaneous properties of the steady state. One recurring idea is that a generalized free energy functional might apply to the non-equilibrium steady state [1–6]. This was first speculated by Rayleigh in the late 19th century [1]. Onsager [2, 3] later used his reciprocity relations to support this conjecture, but the idea has remained controversial to the present day.

Non-equilibrium steady state behaviour plays an important role in electronic transport theory, and becomes particularly important in driven nano-devices, such as a DC biased quantum dot [7]. Variants on Rayleigh's approach would be invaluable in this new context, and might provide an important first step along the road to Boltzmann's approach the non-equilibrium steady state [4–6].

Recent work on non-equilibrium hydrodynamics has shown how Onsager's reciprocity relations can be generalized to the non-equilibrium steady state [8, 9]. This motivates us to re-examine Onsager's reciprocity relations in the context of non-equilibrium quantum physics. By considering the history independence of the non-equilibrium steady state, we are led to

 © 2004 IOP Publishing Ltd Printed in the UK

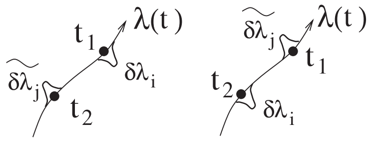

Figure 1. Two variations in the path P where the increments in λ_j at times t_2 and $t_1 > t_2$ are interchanged.

conjecture that Onsager's reciprocity theorem continues within a limited class of quantum variables, in the non-equilibrium steady state. Within this restricted class of variables, the concept of a free energy can be used to describe the steady state of non-equilibrium quantum systems.

The lack of history dependence of the equilibrium steady state means that the work done on the system by coupling various internal degrees of freedom $\hat{A}_i$ $(i = 1, n)$ to corresponding external 'forces' $\lambda_i(t)$,

$$W = \sum_i \int_P \langle A_i(t) \rangle \, \mathrm{d}\lambda_i(t),$$

does not depend on the path P over which the λ_i are adiabatically incremented to their final value. If we increment $\lambda_j(t)$ at two different times t_2 and $t_1 > t_2$, we may do it two ways, illustrated in figure 1.

In the first variation $\lambda_i(t_1) \to \lambda_i(t_1) + \delta\lambda_i$ and $\lambda_j(t_2) \to \lambda_j(t_2) + \widetilde{\delta\lambda_j}$, whereas in the second the variations are reversed, $\delta\lambda_j \leftrightarrow \widetilde{\delta\lambda_j}$. The second-order change in the work done along both paths must be equal, i.e.

$$\delta^2 W = \delta\lambda_i \widetilde{\delta\lambda_j} \left(\frac{\delta\langle A_i(t_1)\rangle}{\delta\lambda_j(t_2)} \right) = \delta\lambda_i \widetilde{\delta\lambda_j} \left(\frac{\delta\langle A_j(t_1)\rangle}{\delta\lambda_i(t_2)} \right) \tag{1}$$

from which if follows that

$$\frac{\delta\langle A_j(t_1)\rangle}{\delta\lambda_i(t_2)} - \frac{\delta\langle A_i(t_1)\rangle}{\delta\lambda_j(t_2)} = 0. \tag{2}$$

We can relate these functional derivatives to the corresponding response functions,

$$\frac{\delta\langle A_j(t)\rangle}{\delta\lambda_i(t')} = -\mathrm{i}\langle [A_j(t), A_i(t')]\rangle \theta(t - t') \tag{3}$$

from which it follows that

$$-\mathrm{i}\langle [A_j(1), A_i(2)]\rangle \theta(1-2) = -\mathrm{i}\langle [A_i(1), A_j(2)]\rangle \theta(1-2). \tag{4}$$

These are the quantum generalization of Onsager's reciprocity relations [2, 3]. The relations are understood to hold only in the long-time limit corresponding to a slow adiabatic variation of the source terms. Onsager identified relations with the microscopic reversibility of the equations of motion and the absence of any 'arrow of time' in thermal equilibrium. This derivation shows how reciprocity is directly related to a lack of history dependence. Since our proof makes no reference to thermal equilibrium, it offers the intriguing prospect of an extension to the non-equilibrium steady state.

To extend the discussion away from thermal equilibrium, we consider a tiny system 'S', which may be a quantum dot [7, 10, 11], a quantum wire [12], or another small system that is coupled to two very large baths of electrons ('leads') at different chemical potentials μ_{L} and μ_{R} where $\mu_{\mathrm{L}} > \mu_{\mathrm{R}}$. The entire coupled system is completely isolated from the outside world.

If we connect S to the leads at time $t = 0$, then after an equilibration time τ_1 the system will arrive at a steady state where a current flows from the left- to the right-hand lead (figure 2).

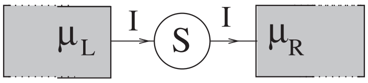

Figure 2. The non-equilibrium steady state is obtained by adiabatically connecting system S to two heat baths at chemical potentials $\mu_{\mathrm{L,R}}$.

This state persists for a long time $\tau_2(L)$ until a substantial fraction of the additional electrons on the left-hand lead have flowed into the right-hand lead. The time $\tau_2(L)$ will diverge rapidly as $L \to \infty$, which permits us to define the steady state value of some variable $\hat{A}$ as

$$\langle A \rangle = \lim_{L\to\infty} \langle A(t) \rangle$$

with the understanding that $\tau_2(L) \gg t \gg \tau_1$.

Suppose the steady state is arrived at by adiabatically turning on an interaction $H_{\mathrm{I}} = gh_{\mathrm{I}}$ between the leads, and by coupling source terms λ_j to various quantities A_j which are localized within S. Since the combined system is closed, when we adiabatically change these variables the amount of work done in reaching the steady state is simply the change in the total energy of the system

$$W_{NE} = \int \langle h_{\mathrm{I}}(t) \rangle \, \mathrm{d}g(t) + \langle A_i(t) \rangle \, \mathrm{d}\lambda_i.$$

If the work done W_{NE} is independent of the path by which g and the λ_j reach their final values, then we can use the previous proof to show that the corresponding variables satisfy a quantum reciprocity relation. The converse will also hold true. This motivates the 'quantum reciprocity conjecture'.

In the non equilibrium steady state, the set of quantum mechanical observables contains a non-trivial subset $\mathcal{P}$ of 'protected' quantum observables $\mathcal{P} = \{a_1, a_2, \ldots, a_n\}$ whose correlation functions in the steady state are insensitive to the arrow of time, and which consequently satisfy a quantum mechanical analogue of the Onsager reciprocity relations

$$\langle [a(1), b(2)] \rangle = \langle [b(1), a(2)] \rangle, \qquad (a, b \in \mathcal{P}).$$

Of course we do not expect the reciprocity relation to extend to *all* variables, as it does in thermal equilibrium, because this would mean that the arrow of time is completely invisible.

Consider the retarded and advanced Green functions between protected variables,

$$G_{ab}^{(R,A)} = \mp \mathrm{i} \langle [a(1), b(2)] \rangle \theta_{\pm}(t_2 - t_1) \tag{5}$$

where $\theta_{\pm}(t) = \theta(\pm t)$. Since a and b are Hermitian, these are real functions ($G^{R,A}(t) = [G^{R,A}(t)]^*$). The conjectured Onsager relations mean that in the steady state they also satisfy

$$\begin{aligned} G_{ab}^{R}(t_2 - t_1) &= G_{ab}^{A}(t_1 - t_2), \\ G_{ab}^{(R,A)}(t_2 - t_1) &= G_{ba}^{(R,A)}(t_2 - t_1), \end{aligned} \tag{6}$$

where the order of the subscripts and time variables is important. If we write $G^R(t_1 - t_2) = \left[G^R(t_1 - t_2)\right]^*$ in the first relation, and then Fourier transform, we obtain the more familiar result

$$G_{ab}^{A}(\omega) = G_{ab}^{R}(\omega)^*$$

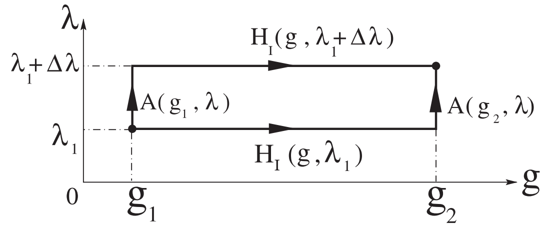

Figure 3. Two paths for turning on the interaction and source terms.

which means that the retarded and advanced Green functions of protected variables share the same spectral decomposition

$$G^{(R,A)}_{ab}(\omega) = \int \frac{\mathrm{d}E}{\pi} \frac{1}{\omega - E \pm \mathrm{i}\delta} A_{ab}(E)$$

where $A_{ab}(E) = \pm\,\mathrm{Im}[G^{(A,R)}_{ab}(E)]$.

Provided that the set of protected quantum variables includes the interaction $H_\mathrm{I} = gh_\mathrm{I}$, then we can define an effective free energy from the virtual work done W_{NE} in reaching the steady state. Suppose we evaluate W_{NE} along the two paths shown in figure 3. Since W_{NE} is the same along both paths, for small $\Delta\lambda$ we have

$$A(g_1, \lambda)\Delta\lambda + \int_{g_1}^{g_2} \frac{\mathrm{d}g'}{g'} H_\mathrm{I}(g', \lambda + \Delta\lambda)\,\mathrm{d}g' = A(g_2, \lambda)\Delta\lambda + \int_{g_1}^{g_2} \frac{\mathrm{d}g'}{g'} H_\mathrm{I}(g', \lambda)\,\mathrm{d}g', \tag{7}$$

so that

$$\Delta A = A(g_2, \lambda) - A(g_1, \lambda) = \frac{\partial}{\partial\lambda}\Delta F \tag{8}$$

where

$$\Delta F = \int_{g_1}^{g_2} \frac{\mathrm{d}g'}{g'} H_\mathrm{I}(g', \lambda). \tag{9}$$

Thus if reciprocity holds, the change in the variables $\{A_j\}$ associated with a change in the coupling constant g can be evaluated as derivatives of a *single* free energy variable ΔF.

We now illustrate the correctness of this conjecture in a simple non-interacting model. We consider a single resonant level in a quantum dot carrying a DC current between two or more leads, where the Hamiltonian $H = H_0 + H_\mathrm{I}$ and

$$H_0 = \sum_{\alpha,\mathbf{k}\sigma} \epsilon(\mathbf{k}) c^\dagger_{\alpha,\mathbf{k}\sigma} c_{\alpha,\mathbf{k}\sigma} + \sum_\sigma \epsilon_{\mathrm{d}\sigma} d^\dagger_\sigma d_\sigma,$$

$$H_\mathrm{I} = J \sum_{\alpha,\mathbf{k}} \big[\gamma_\alpha c^\dagger_{\alpha,\mathbf{k}\sigma} d_\sigma + \mathrm{H.c.}\big].$$

Here $\alpha = 1, N$ labels the leads, each one characterized by a distinct chemical potential μ_α, $\epsilon_{\mathrm{d}\sigma} = \epsilon_\mathrm{d} - \sigma B$ is the energy of the localized state in the dot in a magnetic field B, J is the overall coupling constant and γ_α is a parameter which sets the relative strength of hybridization with the α lead. This is an exactly solvable problem, and has well known results [13] found by the Keldysh method.

As a first step, by comparing the retarded and advanced correlation functions, we are able to explicitly confirm that the interaction, together with the dot magnetization M and occupancy n_d, form a set of protected variables $\{H_\mathrm{I}, M, n_\mathrm{d}\}$ which satisfy reciprocity and for which a free energy functional can be defined.

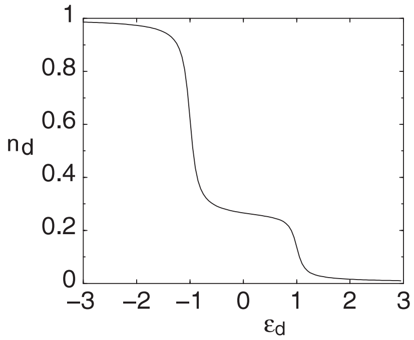

Figure 4. Distribution function of $n_{\rm d}$ as a function of $\epsilon_{\rm d}$. $\mu_1 = 1$, $\mu_2 = -1$, $\lambda_1 = 0.75$, $\lambda_2 = 0.25$, $\Delta = 0.01$ and $T = 0.001$.

For example, to confirm the relation

$$\langle [H_{\rm I}(t_1), n(t_2)]\rangle = \langle [n(t_1), H_{\rm I}(t_2)]\rangle, \tag{10}$$

we compare the retarded and advanced Green functions:

$$G^R_{H_{\rm I}n}(\omega) = {\rm Tr}\sum_\alpha J\gamma_\alpha \int \frac{{\rm d}\epsilon}{2\pi}\left[\mathcal{G}_{{\rm dd}^\dagger}(\epsilon)({\rm i}\tau_1)\mathcal{G}_{{\rm c}_\alpha{\rm d}^\dagger}(\epsilon+\omega) + \mathcal{G}_{{\rm dc}_\alpha^\dagger}(\epsilon)({\rm i}\tau_1)\mathcal{G}_{{\rm dd}^\dagger}(\epsilon+\omega)\right] \tag{11}$$

and

$$G^A_{H_{\rm I}n}(\omega) = {\rm Tr}\sum_\alpha J\gamma_\alpha \int \frac{{\rm d}\epsilon}{2\pi}\left[\mathcal{G}_{{\rm c}_\alpha{\rm d}^\dagger}(\epsilon+\omega)({\rm i}\tau_1)\mathcal{G}_{{\rm dd}^\dagger}(\epsilon) + \mathcal{G}_{{\rm dd}^\dagger}(\epsilon+\omega)({\rm i}\tau_1)\mathcal{G}_{{\rm dc}_\alpha^\dagger}(\epsilon)\right] \tag{12}$$

where the G_{ab} refer to the Larkin–Ovchinikov matrix Green function [14, 15] between electron fields a and b and the trace is over Keldysh indices. By writing these expressions out explicitly, we are able to explicitly confirm that they are related by complex conjugation, $G^R_{H_{\rm I}n}(\omega) = [G^A_{H_{\rm I}n}(\omega)]^*$, from which reciprocity between $n_{\rm d}$ and $H_{\rm I}$ holds. A similar method enables us to check that

$$\langle [H_{\rm I}(t_1), M(t_2)]\rangle = \langle [M(t_1), H_{\rm I}(t_2)]\rangle. \tag{13}$$

The correlation function between M and $n_{\rm d}$ identically vanishes, trivially satisfying reciprocity.

We now confirm that an effective free energy correctly determines the occupancies and magnetization. The expectation value of the interaction energy is determined by the equal time Keldysh Green functions between the conduction and dot electron, given by

$$\langle H_{\rm I}\rangle = J\sum_{\alpha,\sigma}\gamma_\alpha\int\frac{{\rm d}\omega}{4\pi{\rm i}}\left[G^{\rm K}_{{\rm d}_\sigma{\rm c}_\alpha^\dagger}(\omega) + G^{\rm K}_{{\rm c}_\alpha{\rm d}_\sigma^\dagger}(\omega)\right].$$

After integrating over the coupling constant we obtain

$$\Delta F_{\rm eff} = \int_0^J \frac{{\rm d}J'}{J'}\langle H_{\rm I}\rangle = \sum_{\alpha,\sigma}\frac{2\gamma_\alpha^2}{\pi}{\rm Re}\left[-2\pi T\log\Gamma\left(\frac{1}{2}+\frac{\epsilon_{{\rm d}\sigma}+{\rm i}\Delta-\mu_\alpha}{2\pi{\rm i}T}\right)\right.$$
$$\left. + 2\pi T\log\Gamma\left(\frac{1}{2}+\frac{\epsilon_{{\rm d}\sigma}-\mu_\alpha}{2\pi{\rm i}T}\right) + \Delta\ln\left(\frac{D}{2\pi T}\right)\right], \tag{14}$$

where $\Delta = \sum_\alpha \pi\rho(J\gamma_\alpha)^2$. The expectation values of local state occupancy $n_{\rm d}$ and magnetization M are then

$$\begin{aligned}\langle n_{\rm d}\rangle &= \frac{\partial\Delta F_{\rm eff}}{\partial\epsilon_{\rm d}} + c_1,\\ \langle M\rangle &= -\frac{\partial\Delta F_{\rm eff}}{\partial B} + c_2,\end{aligned} \tag{15}$$

where the constant terms give the limiting value of the occupancy and magnetization when $J \to 0$. We can fix these constants by using the condition that $\langle n_{\rm d}\rangle \to 1$ and $\langle M\rangle \to 0$ as $\Delta \to \infty$, which gives

$$\begin{aligned}\langle n_{\rm d}\rangle &= 1 + \sum_{\alpha,\sigma} \frac{\gamma_\alpha^2}{\pi} \,\mathrm{Im}\left[\psi\left(\frac{1}{2} + \frac{\epsilon_{\rm d\sigma} - \mu_\alpha + \mathrm{i}\Delta}{2\pi \mathrm{i}T}\right)\right],\\ \langle M\rangle &= \sum_{\alpha,\sigma} \frac{\gamma_\alpha^2}{\pi}\sigma \,\mathrm{Im}\left[\psi\left(\frac{1}{2} + \frac{\epsilon_{\rm d} + \sigma B - \mu_\alpha + \mathrm{i}\Delta}{2\mathrm{i}\pi T}\right)\right].\end{aligned} \tag{16}$$

Both results can be independently confirmed by direct calculation from the Keldysh Green functions. It is remarkable that the derivative of a single free energy functional reproduces the results of two separate Keldysh calculations, even though a DC current is flowing through the dot. It is interesting to see that, even at the zero-coupling limit, the occupancy and magnetization of the 'dot' have a non-thermalized form, and depend on the ratios between hybridization γ_α. The non-thermal function $n_{\rm d}(\epsilon_{\rm d})$ (figure 4) is reminiscent of the occupancy observed in quantum wire experiments [12]. Here the parameters λ_i play the similar role of distances between the measured point and leads in the experiment.

It is instructive to examine the magnetization in the two-lead case which for zero temperature is

$$\chi(B,\Delta) = \frac{2\Delta(B^2 + \Delta^2 + V^2)}{\pi((B-V)^2 + \Delta^2)((B+V)^2 + \Delta^2)} \tag{17}$$

whilst for $\Delta \to 0$,

$$\chi(B,T) = \frac{1}{4T}\left[\mathrm{sech}^2\left(\frac{B+V}{2T}\right) + \mathrm{sech}^2\left(\frac{B-V}{2T}\right)\right]. \tag{18}$$

In both limits, the bias voltage dramatically reduces the susceptibility and at a finite voltage the $T = 0$ magnetic susceptibility in the limit of $J \to 0$ is always zero. Non-thermal magnetizations of this kind have recently been obtained in the zeroth order magnetic susceptibility calculation for quantum dot [16–18]. Can we extend the set of 'protected' variables to include other quantities of interest, such as the current or the spin current? The answer appears to be 'no'. When we directly compare the retarded and advanced correlators involving any operator that involves the lead electrons, *other* than $H_{\rm I}$, we find that they are not complex conjugates. This means that we cannot change the ratio of the couplings γ_α as we turn on the interaction, for to do this would be to introduce new variables which do not satisfy the Onsager reciprocity relation with $h_{\rm I}$.

The validity of our conjecture in more complex systems is an open issue. We cannot prove that reciprocity is stable against the presence of interactions within the dot, but we have circumstantial support for this idea. The above methods can be used in the large-N limit of the infinite-U Anderson model to examine how the mean-field equations evolve away from equilibrium. We have also compared the local susceptibility in the non-equilibrium Kondo problem obtained using the reciprocity conjecture with that obtained using Majorana techniques [19]. An interesting recurring feature of these calculations is the appearance of non-thermal distribution functions in the limit that the coupling with the leads is taken to zero. In interacting systems, these limiting distribution functions will need to be computed self-consistently from the limiting form of the Dyson equation, before the change in free energy can be computed [20][1].

[1] The results of Coleman and Mao [20] did not take this fact into account and are therefore incorrect. A revised calculation to support the idea that the Kondo effect flows to strong coupling at large voltages will shortly be posted.

In conclusion, we have examined the idea that the principle of virtual work can be extended to the non-equilibrium steady state of quantum systems. This has led us to conjecture the existence of a class of steady state variables which satisfy the quantum generalization of Onsager's reciprocity relation out of equilibrium. If the interaction component of the Hamiltonian belongs to the conjectured set of protected variables, then the notion of a free energy can be extended to the quantum non-equilibrium steady state, permitting the expectation values of steady state variables to be computed as derivatives of a free energy functional. This idea works for the simplest possible example, and leaves open the possibility that it will apply to more complex and interesting interacting situations.

We wish to thank Chris Hooley, David Langreth, Joel Lebowitz and Olivier Parcollet, for the many lively discussions at the Rutgers Center for Materials Theory that led to this paper. This work was supported by DOE grant DE-FG02-00ER45790.

References

[1] Lord Rayleigh 1873 *Proc. Math. Soc. Lond.* **4** 357
Lord Rayleigh 1873 *Proc. Math. Soc. Lond.* **4** 363
Lord Rayleigh 1877 *Theory of Sound* 1st edn, vol 1 (London: Macmillan) p 78 (1878 2nd edn, vol 1, p 102)
[2] Onsager L 1931 *Phys. Rev.* **37** 405
[3] Onsager L 1931 *Phys. Rev.* **38** 2265
[4] Hershfield S 1993 *Phys. Rev. Lett.* **70** 2134
[5] Christen T 1997 *Phys. Rev.* B **55** 7606
[6] Derrida B, Lebowitz J L and Speer E R 2001 *Phys. Rev. Lett.* **87** 1506001
[7] Kastner M A 1992 *Rev. Mod. Phys.* **64** 849
Ashoori R C 1996 *Nature* **379** 413
Kouwenhoven L P and Marcus C 1998 *Phys. World* **11** (June) 35
[8] McLennan J A 1974 *Phys. Rev.* A **10** 1272
[9] Dufty J W and Rubí J M 1987 *Phys. Rev.* A **36** 222
[10] Kaminski A, Nazarov Yu V and Glazman L I 2000 *Phys. Rev.* B **62** 8154
[11] Coleman P, Hooley C and Parcollet O 2001 *Phys. Rev. Lett.* **86** 4088
[12] Pothier H *et al* 1997 *Phys. Rev. Lett.* **79** 3490
[13] Wingreen N, Jauho A-P and Meir Y 1993 *Phys. Rev.* B **48** 8488
[14] Larkin A I and Ovchinnikov Yu N 1975 *Zh. Eksp. Teor. Fiz.* **68** 1915
Larkin A I and Ovchinnikov Yu N 1975 *Sov. Phys.—JETP* **41** 960 (Engl. Transl.)
[15] For a review of Keldysh method, see Rammer J and Smith H 1986 *Rev. Mod. Phys.* **58** 323
[16] Kaminski A and Glazman L 2001 private communication
[17] Parcollet O and Hooley C 2002 *Phys. Rev.* B **66** 085315
(Parcollet O and Hooley C 2002 *Preprint* cond-mat/0202425)
[18] Rosch A, Passke J, Kroha J and Wölfle P 2003 *Phys. Rev. Lett.* **90** 076804
[19] Mao W *et al* 2002 to be published
[20] Coleman P and Mao W 2002 *Preprint* cond-mat/0203001 v1