

MAGNETIC PROPERTIES OF DISORDERED SYSTEMS NEAR A METAL-INSULATOR TRANSITION

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Abstract. – Using doped semiconductors as a paradigm, the low-temperature magnetic and thermodynamic behaviour of a disordered system undergoing a metal-insulator transition is described, and compared with various theoretical approaches from the metallic and insulating phases. Issues addressed include universal behaviour in the insulating phase, local moments in the metallic phase, and effects of compensation.

1. Introduction

Progress in the understanding of the properties of disordered systems undergoing a metal insulator (MI) transition during the past decade [1] has hinged on the scaling theory of localization. Following the seminal papers for non-interacting electrons in a disordered medium [2, 3], the role of electron interactions has been incorporated into the scaling description [4-7]. These studies suggest that the critical behaviour at the MI transition in disordered systems depends on spin-dependent effects (e.g. spin-flip or spin-orbit scattering, magnetic field) which appear as cut-offs in the scaling equations. This leads to various universality classes for the critical exponents of the transport coefficients (conductivity, Hall coefficient, etc.). Thus the magnetic properties of disordered systems near the MI transition could yield valuable clues towards unraveling the transport behaviour.

Experiments have been performed on a variety of systems such as (i) doped semiconductors, (e.g. uncompensated Si:P (Silicon doped with phosphorus) and compensated Si:P; B); (ii) metal-semiconductor mixtures (e.g., Au-Ge, Nb-Si) and (iii) magnetic semiconductors.

Except for uncompensated silicon [8], the exponent μ characterizing the $T = 0$, the conductivity onset $\sigma \sim |x - x_c|^\mu$ is $\mu \approx 1$. (Here x is the concentration, pressure, uniaxial stress, or magnetic field, and x_c is the value at the MI transition). In contrast, uncompensated doped silicon shows a much sharper onset, with $\mu \approx 0.5 - 0.6$. Recent work [9-15] has focussed towards the resolution of this controversy, through magnetic and thermodynamic measurements at low temperatures, as well as magnetoresistance data in doped silicon. Similar measurements on the other systems would be invaluable in furthering our understanding of the MI transition and the applicability of scaling ideas for thermodynamic quantities.

We limit ourselves to doped semiconductors, comparing various theoretical scenarios with experiments on Si:P and Si:P; B. Doped semiconductors are in some sense the most basic disordered system, because the impurity electron is in a shallow hydrogenic 1s state, with a Bohr radius a_B ($\sim 20 \text{ \AA}$ in Si) that is much larger than lattice spacing because of the large dielectric constant of the host semiconductor. Consequently, it is well modelled as an ensemble of hydrogen atoms, *randomly distributed in three-dimensional space* (i.e. the discreteness of the underlying lattice is unimportant at the densities of interest $n \sim n_c \approx (1/4a_B)^3$). Additional complications due to mass anisotropy, many conduction band minima etc., necessary for quantitative calculations, are of little concern for the qualitative physics, and so will not be discussed here.

At low n , each donor impurity electron is bound to the impurity, in a 1s state; the system behaves magnetically as an ensemble of spins with $s = 1/2$, with very weak exchange interactions, leading essentially to a Curie susceptibility $\chi \sim 1/T$. At high densities, on the other hand, the electrons delocalize, and if we were to take the analogy with lattice systems, we would expect a T -independent Pauli spin-susceptibility. Thus the low T magnetic behaviour considerably with n . It is this change, as well as the associated effect on the low temperature thermodynamics, and its relation with the MI transition and the electronic transport properties that we wish to discuss in the following sections. Section 2 is devoted to the insulating phase, where we believe we have an approximate theory and a good physical understanding of the magnetic behaviour. Section 3 discusses recent developments in our understanding of the behaviour in the vicinity of the MI transition, implications for the disordered metal, and the effect of compensation. Finally, in section 4, we summarize our conclusions.

2. The insulating phase

The magnetic character of the insulating phase is by and large understood by now for the "fruit-fly" of disordered systems, the random ensemble of hydrogenic atoms, which is realized in n-doped uncompensated semiconductors. We will only summarize the results here and the reader is referred to a previous review [16] for details. At low densities, deep in the insulating phase, where charge fluctuations are unimportant (at low energy scales) the low energy behaviour can be described in terms of the Heisenberg Hamiltonian:

$$H = \sum_{ij} J(\mathbf{r}_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j \quad (1)$$

$J(r)$, the direct antiferromagnetic exchange interaction between pairs of hydrogen atoms varies roughly as $J(r) \sim J_0 \exp(-2r/a_B)$. Though the microscopic $J(r)$ can be calculated only in the low density limit, we will argue that a similar exchange Hamiltonian with $J(r)$ varying exponentially with distance describes the low-energy behaviour throughout the insulating phase, and experiment suggests that it may also persist somewhat into the metallic phase.

Despite the superficial similarity of equation (1) to a lattice Heisenberg Hamiltonian, the magnetic character of the random antiferromagnet is quite different from that of a lattice of hydrogen atoms (e.g., simple cubic or bcc). For the lattice, the dominant nearest neighbour interactions would lead to simple Neel ordering at a temperature $T_N \sim J(r_0)$ where r_0 is the nearest neighbour distance, implying an ordered ground state for arbitrarily small density, n ($\sim r_0^{-3}$). The random antiferromagnet, on the other hand, is best described as a "valence-bond" insulator, i.e. pairs of sites coupled strongly to each other form an inert singlet ground state (valence-bond) because of the large energy gain due to quantum fluctuations in this spin-1/2 system. That this happens to strongly coupled pairs is not so surprising, because of the wide distribution of initial (bare) couplings covering many orders of magnitude in this highly disordered system; in fact, the necessity of a hierarchical scheme has been pointed out by a number of workers [17]. What is more significant is that this wide distribution and hierarchical scheme works down to extremely low energy scales, as shown by the numerical renormalization group calculations [18] of Bhatt and Lee (BL). As a result, the low temperature magnetic and thermodynamic properties are well described by an ensemble of coupled pairs of spins with a *renormalized* distribution $P_R(J)$ of pair couplings J , analogous to that used in the context of the quasi-1d organic systems [19] which are the one dimensional counterpart of this system. Both experiment and the numerical calculation suggest that the susceptibility at low temperatures $T < 10$ K for densities $0.2n_c < n < n_c$ behaves *approximately* as

$\chi(T) \sim T^{-\alpha}$ where $\alpha \approx 0.6$. In the numerical calculation, α is weakly dependent on T (logarithmically); however, the experiments are consistent with a constant α over almost three decades in T . (The same situation is true in the quasi-1d systems). Further, the experimental $\chi(T)$ at low T is almost independent of n as well, strongly suggesting that some sort of fixed point behaviour has been reached in the entire range of densities.

Within the hierarchical pair approach, the singular behaviour of $\chi(T)$ is viewed as a consequence of an infrared singularity which develops in the *renormalized* pair distribution $P_R(J) \sim J^{-\alpha}$. Because of this singularity the low- T thermodynamics in the insulating phase is dominated by spin excitations, even when charge (electron-hole) excitations are added, as for example, in the Hubbard model with randomly positioned sites. Indeed, as long as one is in the insulating state, an effective spin Hamiltonian with exchanges varying exponentially with distance at long distances is likely to emerge as the low energy description, though $J(r)$ is not the bare hydrogenic exchange.

In the non-interacting valence bond picture, all thermodynamic properties can be calculated explicitly as integrals over the distribution $P_R(J)$, and compared with experiment. The resulting fit in terms of a single parameter α (which is motivated by the numerical studies), and *no further adjustable parameters* to the susceptibility [20], specific heat [10], as well as the scaling behaviour of the non-linear magnetization [21] is very good. Further, by including the interactions between the spin pairs and hyperfine interactions [22], a satisfactory agreement can be obtained for the dramatic temperature dependence of the ESR line width and position [23] as well, suggesting that this gives a good starting point for the spin dynamics as well.

This valence-bond insulator is the disordered counterpart of the RVB picture put forth for the high- T_c superconductors by Anderson *et al* [24]; however, it has no "resonance" in the valence bonds. Further, our result is crucially dependent on the existence of a high degree of disorder, so that the hierarchy is well-defined, and it appears not to depend crucially on dimension, at least for $d = 1-3$ [18]. Finally, it should be emphasized that while this gives a good description, it is a zeroth order model, from which perturbative expansions *may* be necessary, depending upon the question asked; as for example, in the case of spin diffusion [22].

3. Beyond the critical density

The traditional picture of the metallic phase beyond n_c has been in terms of a Fermi liquid, much like systems with translational symmetry (fluids and crystalline solids), especially because at large density, interaction effects are less important than the one electron kinetic energy. This would suggest that in the ab-

sence of magnetic transitions in the insulating phase, the thermodynamic properties across the MI transition would be well described by the Brinkman-Rice [25] result for the MI transition in the half-filled Hubbard model. Namely, the divergent susceptibility would be quenched in the metallic phase at low temperatures at a value $\chi(0) \sim m^*/m$, with an effective mass, diverging as $m^* \sim (n - n_c)^{-1}$, leading to a divergent χ at n_c at $T = 0$. The same divergence is found for the specific heat $\gamma \equiv C/T$, and the ratio (χ/γ) is four times the free electron value. Unfortunately, such a scenario is not borne out by the experimental results on Si:P (Fig. 1), which shows $\chi(T)$ vs. T on a double logarithmic plot for concentrations on both sides.

It is quite conceivable that properties at low-temperatures where Fermi liquid theory is supposed to apply are drastically modified in the presence of disorder, and calculations based on perturbative scaling approaches [5-7] suggest that this is indeed so, particularly in the absence of spin scattering and magnetic fields. The RG equations, involve a dimensionless conductance t , an action parameter γ_2 , and a quasiparticle density of states renormalization z , which vary on the length scale l as:

$$\frac{dt}{dl} = -\frac{\epsilon}{2}t + t^2 \left[4 - 3 \left(\frac{1 - \gamma_2}{\gamma_2} \right) \ln(1 + \gamma_2) \right] \quad (2a)$$

$$\frac{d\gamma_2}{dl} = \frac{t}{2}(\gamma_2 + 1)^2 \quad (2b)$$

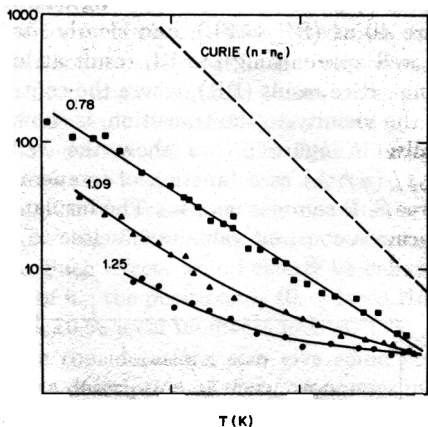


Fig. 1. - Spin susceptibility of Si:P normalized to the Pauli value as a function of temperature for three values of the reduced density n/n_c indicated on a double logarithmic plot. Dashed line is the Curie susceptibility corresponding to the MI transition density n_c .

$$\frac{dz}{dl} = \frac{t}{2}z(3\gamma_2 - 1). \quad (2c)$$

Here $t = \Lambda^{d-2} / 4\pi^2 N_1 D$, where Λ is the cut off in momentum space, N_1 the bare density of states at the Fermi level, and D the charge diffusion constant. Equations (2) are to lowest order in $\epsilon \equiv d - 2$, the

dimensionality above two dimensions, and are appropriate for the non-degenerate band case, though similar equations with slightly different coefficients can be written down for the degenerate (multi-valley) case. Identifying the physical conductivity σ , specific heat γ and magnetic susceptibility χ as:

$$\sigma \sim \Lambda^{d-2} / t \quad (3a)$$

$$\gamma = N_1 z \quad (3b)$$

$$\chi = N_1 z (1 + \gamma_2), \quad (3c)$$

one may show that these equations imply a strong T -dependent enhancement of both χ and γ , $\chi \sim T^{-4\epsilon/(d+3\epsilon)}$ and $\gamma \sim T^{-3\epsilon/(d+3\epsilon)}$ as T is reduced. However, at low T the parameters flow to strong coupling, where equations (2) are no longer applicable.

This dependence of $\chi \sim T^{-2/3}$ and $C = \gamma T \sim T^{1/2}$ (for $\epsilon = 1$) are qualitatively in agreement with experimental results (Figs. 1 and 2). However, from equa-

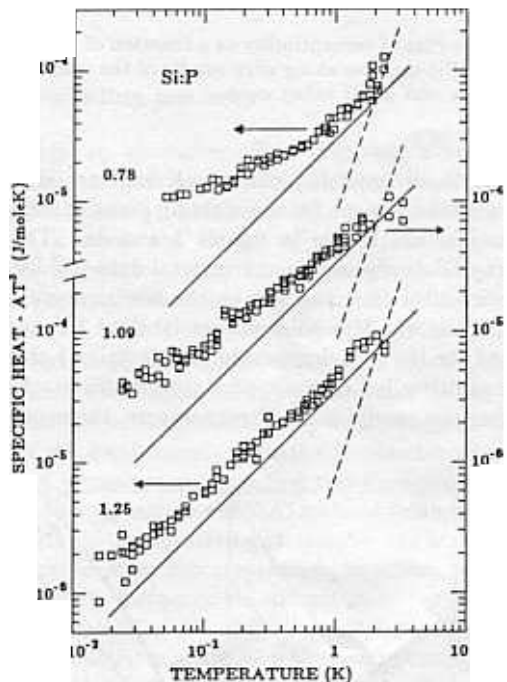


Fig. 2. - Electronic specific heat of the same three samples versus temperature. Also shown is the "free electron" (solid line) and phonon (dashed line) specific heat. Below 1 K, the specific heat for the insulator (top curve) is consistent with $C \sim T^{1-\alpha}$ with $\alpha \approx 0.6$.

tions (2) and (3), we can also write down equations for the variation in susceptibility χ/χ_0 , specific heat γ/γ_0 , and conductivity σ/σ_0 (where symbols with the subscripts 0 refer to the bare, high temperature values), in terms of each other, which are independent of cutoff to lowest order in ϵ and can be integrated up to (down to) any desired length scale (tempera-

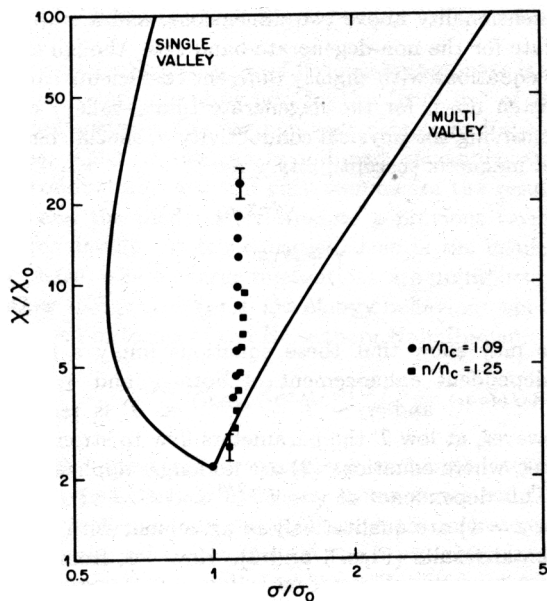


Fig. 3. - Plot of susceptibility as a function of conductivity for metallic samples along with results of the scaling theory for single and multi valley case.

ture). Such *implicit* plots, which have no adjustable parameters, except for the starting point at high temperature, are shown in figures 3 and 4a. The various symbols represent experimental data [10] from the two metallic Si:P samples as the temperature is varied, while the two solid curves labelled SV and MV stand for the non-degenerate (single valley) case, and the multi-valley case appropriate for silicon. Despite fitting the results at high temperature, the experimen-

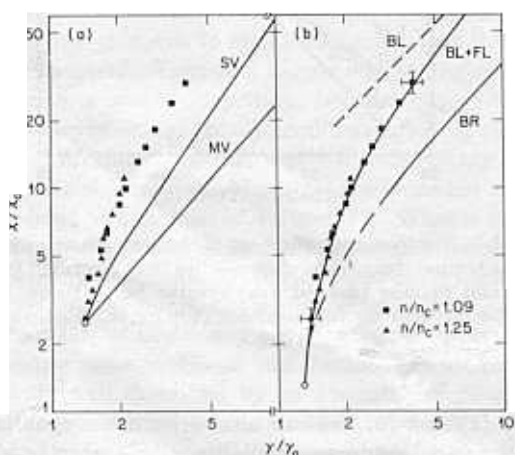


Fig. 4. - Plot of the susceptibility versus specific heat γ for metallic samples. In (a) the experimental data are compared with the single and multi valley scaling theory, while (b) compares it with the Brinkman-Rice and the two-fluid (BL + FL) model. Dashed line is the asymptotic low temperature result where BL pairs dominate.

tal data do not fit the theory well, and the fits do not improve much by changing the starting point for the RG equation. We mention in passing that with spin scattering or finite magnetic field, the divergences in χ and γ at low T are quenched in the RG equations, so these would not fit the data even qualitatively.

The large enhancement of χ relative to γ suggests a magnetic instability of the Fermi liquid, namely the formation of local moment (or long-lived quasi localized spin fluctuations). This is found in studies of the disordered Hubbard model [26], which suggests a continuity with the thermodynamics of the insulating phase, where it was dominated by localized spins interacting with each other. Following this line of thought, we may write a generalization of the results of the insulating phase to include a Fermi-liquid contribution to γ and χ in a two fluid model:

$$\gamma / \gamma_0 = m^* / m + (T / T_0)^{-\alpha} \quad (4a)$$

$$\chi / \chi_0 = m^* / m + \beta (T / T_0)^{-\alpha} \quad (4b)$$

We assume an $\alpha \approx 0.62$, taken from the susceptibility and specific heat data for the $n / n_c = 0.78$ sample (Figs. 1 and 2), independent of n , $\beta = 10.5$ from the spin pair model for $\alpha = 0.62$, and a mass enhancement $m^* / m = 1.3$ from earlier measurements [29]. Then equations (4) contain only one adjustable parameter T_0 (which measures the fraction of localized spins), which may be eliminated by considering the plot of χ / χ_0 vs. γ / γ_0 (as for the scaling theory) where (T / T_0) is an implicit parameter. The resulting curve is shown in figure 4b as (BL + FL), and clearly fits the data rather well approaching the BL result at low T . The Brinkman-Rice result (BR), where the control parameter is the vicinity to the transition, is shown as well.

Finally, in figure 5, we show the Wilson ratio $(\chi / \chi_0) / (\gamma / \gamma_0)$ as a function of temperature T for the three Si:P samples near n_c . The insulating sample approaches a constant value ≈ 9.3 close to the BL re-

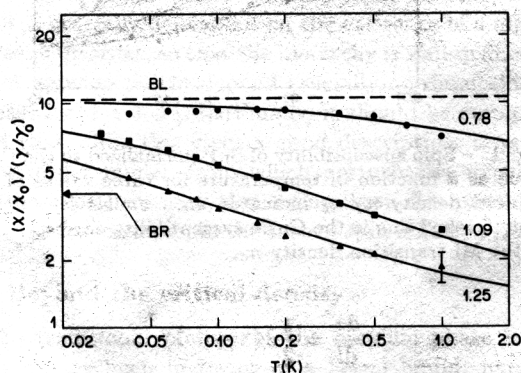


Fig. 5. - Wilson ratio as a function of temperature for the three samples and fits using the two-fluid model. Note the constant value for the insulating sample below $T \sim 0.2$ K, close to the Bhatt-Lee model value (dashed line).

sult of 10.5, and much above the BR value of 4. By adjusting T_0 for each sample the solid curves are obtained from equations (4), implying that the ratio of spins for the three samples in ascending density, is 20:5:2. (Assuming nearly 100 % localized in the insulating sample gives 25 % and 10 % localized spins in the two metallic samples).

The existence of local moments in the metallic phase as $n \rightarrow n_c^+$ can also be directly inferred from the phosphorus NMR measurements [14] which see a dramatic loss of signal from nuclei coupled to the itinerant electron gas as n_c is approached. Silicon NMR measurements [13, 15] are also consistent with the two-fluid picture [27, 15]. Bulk magnetization measurements [11] at higher temperatures and susceptibility measurements [28] have also provided evidence in favor of local moments in the metallic phase.

The idea of local moments just above n_c is quite old – it was proposed well before the scaling theory on the basis of susceptibility and specific heat measurements by a number of workers [29]. What is new is that by going to lower temperatures and measuring both χ and C on a series of samples, it has been possible to not only test different theoretical models, but also provide an idea of the interactions between the local moments in the poor metal.

A number of questions are raised by the proposition of local moments in a metal – e.g. why do they not get quenched by a Kondo effect, and what is their effect on transport properties? It has recently been shown [30] that for randomly (Poisson) distributed sites, that the Kondo effect is not capable of quenching the susceptibility even deep in the metallic phase, because of the presence of rare regions consisting of an odd number of sites with low density ($\ll n_c$). The RKKY interaction between these rare regions is more effective, but still does not appear to prevent a divergence in $\chi(T)$ as $T \rightarrow 0$, suggesting a breakdown of a pure Fermi liquid description. Such effects would clearly be enhanced in the vicinity of n_c ; the positionally disordered Hubbard model has ~ 10 % local moments near n_c [26].

The above considerations also give some justification for why a description of *thermodynamics* in terms of a two-fluid model is appropriate – the exchange *between* these moments is much more efficient than that between the moments and the “itinerant part” of the electron gas. Nevertheless, the two non-interacting fluids (Eq. (4)) is clearly a zeroth order description.

The local moments provide a T -dependent spin-flip scattering for transport within a Fermi liquid picture [31]. Analysis of the magnetoresistance data [12] shows that in uncompensated Si:P, the spin-flip rate remains close to $k_B T$ down to millikelvin temperatures, while in compensated Si:P; B and Ge:Sb the rate is actually higher. Recent susceptibility measurements on compensated Si:P; B by ESR show an *enhanced* susceptibility over uncompensated Si:P. This is entirely consis-

tent if the spin-flip scattering is due to local moments – the higher rate corresponds to their larger number, evidenced by the higher χ . (In the one-fluid scaling theory, on the other hand, large spin-flip scattering rate implies no divergence in $\chi(T)$, contrary to the stronger divergence seen in experiment.) The differences in spin-flip scattering may be responsible for the differences in the conductivity exponent, though incorporating the local moments within a scaling theory for transport is not yet complete.

Finally, the ESR line width at small fields was found to be proportional to the susceptibility in uncompensated Si : P; this is easier understood in the disordered Fermi-liquid [9, 32], than in the BL model [22]. However, this proportionality *does not* occur in the compensated samples. In fact the slower divergence in linewidth compared to the uncompensated case correlates very well with the faster divergence of $\chi(T)$ with a BL picture, and is under investigation.

4. Concluding remarks

Through an extensive effort involving a series of measurements *and* theoretical modelling of both transport and thermodynamic properties in doped semiconductors, a consistent picture of the MI transition in disordered systems seems to be emerging. The data favour the appearance of local moments in the disordered metal, and a pure Fermi liquid picture of the disordered metal may break down in a more serious way than perturbative scaling approaches to the MI transition (from the metallic side) would suggest. The effect of the local moments for the thermodynamics is to form a second component in a two-fluid description which is the counterpart of the localized valence bonds (spin pairs) in the insulating phase. For the transport they provide spin flip scattering, and their greater abundance in compensated silicon provides a plausible explanation of the different effective exponents for the conductivity onset seen in uncompensated and compensated systems. Thermodynamic and magnetic experiments on amorphous metal-semiconductor mixtures would be invaluable in furthering our understanding of the MI transition, and the issue of the pervasiveness of local moment formation in disordered metals.

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