Spin dynamics across the metal-insulator transition

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A theoretical study of the dynamics of electron spins in disordered insulators and metals is performed. Spin diffusion is found to slow down at low temperatures, making the system unusually sensitive to spin-dependent perturbations. This sensitivity shows up in an unusual frequency and temperature dependence of the linewidth and resonance field of the electron spin resonance (ESR) signal. These results are used to interpret recent ESR measurements in phosphorus doped silicon in both the insulating and metallic phases, and good agreement is obtained with experiment.

I. INTRODUCTION

Doped semiconductors, and especially Si:P, have been used extensively in recent years to understand the properties of disordered interacting electrons.[1] With increasing doping the system is known to undergo a transition from an insulator to a metal at a critical dopant density n_c. In this paper we focus upon two recent ESR experiments in Si:P, one on the insulating side[2] ($n \approx n_c/2$) and the other on the metallic side[3] just above n_c. We show how these experiments can yield useful information on the nature of the spin excitations on both sides of n_c. A unified picture of the slowing down of spin diffusion with falling temperatures emerges from our analysis. This behavior is believed to be an important ingredient in determining the universality class within the scaling picture of the metal-insulator transition. In Sec. II we describe the spin dynamics in the insulating phase in detail (part of this study is discussed elsewhere[4]). We present new results which show how traditional methods fail for spin excitations localized on small clusters, and discuss how to correctly deal with quantum fluctuations in such a case. Section III summarizes results for the metallic phase,[3,5] and conclusions are drawn in Sec. IV.

II. INSULATOR

The ESR experiment[2] was done in a transverse field with a low ac frequency while scanning the longitudinal dc magnetic field. The ESR absorption spectrum was a Lorentzian as a function of the dc magnetic field. The experiment showed not only a large temperature (T) dependence to the resonance field and linewidth, but found that these quantities also depend strongly on the frequency. As we show, these effects are a consequence of the slowing down of spin diffusion, which makes the electron resonance absorption quite sensitive to their coupling to the phosphorus nuclear spins. At low doping densities, the electrons are bound in hydrogenic states about the phosphorus nuclei and interact with each other via an exchange interaction. The system can therefore be modeled as a disordered Heisenberg antiferromagnet.[6] Because of the wide distribution of bond strengths, varying over several orders of magnitude, pairs of strongly coupled spins tend to lock into singlet states hierarchically, as temperature T becomes less than the exchange constant J. The small polarizability of the singlets leads to a slight renormalization of the interactions of the remaining weakly coupled spins. Thus to understand the nature of the low lying spin excitations at any given T we need only look at the spins which have not yet been locked into singlets.

Because of the dilution of the bond strengths with decreasing T we may assume that over the time scales relevant for an ESR experiment (T_2), the spin excitation is localized over a cluster of size N. We show that when N becomes small at the low temperatures where interesting experimental effects occur, well-known classical methods for analyzing the linewidth break down. Consider, to start with, an exactly soluble problem: two electron spins $\mathbf{S}_i$, in an external magnetic field $\mathbf{h}$ interacting with each other via an exchange constant J and with their respective nuclear spins I_i with a hyperfine constant A. The Hamiltonian H for the system is

$$H = J\mathbf{S}_1\cdot\mathbf{S}_2 + A\,\mathbf{S}_1\cdot\mathbf{I}_1 + A\,\mathbf{S}_2\cdot\mathbf{I}_2 + \mathbf{h}\cdot(\mathbf{S}_1+\mathbf{S}_2). \quad (1)$$

In the limit $J,h \gg A$, by solving the electron Hamiltonian exactly and treating the nuclear coupling in first-order perturbation theory, we obtain the eigenvalues:

$$E_{00} = -3J/4,$$
$$E_{1m} = J/4 + m[h + A(i_1+i_2)/2], \quad m = 0, \pm 1, \quad (2)$$

where i_1 and i_2 are the eigenvalues of the nuclear spins. The hyperfine coupling gives a series of lines in the ESR spectrum. The mean square moment of the absorption spectrum can easily be evaluated from the above energies:

$$\langle(\Delta\omega)^2\rangle = T_2^{-2} = A^2/8, \quad (3)$$

i.e., a line whose width is reduced from the single electron value of $A/2$ by a factor $\sqrt{2}$. This reduction can be interpreted as an effect of motional narrowing and is independent of J.

The standard method of moments evaluates the linewidth from the mean square and fourth moments:

$$\langle(\Delta\omega)^2\rangle = -\frac{\mathrm{Tr}([H,S_x])^2}{\mathrm{Tr}\,S_x^2} = \frac{A^2}{4},$$
$$\langle(\Delta\omega)^4\rangle = \frac{\mathrm{Tr}([H,[H,S_x]])^2}{\mathrm{Tr}\,S_x^2} = \frac{A^2J^2}{8} \quad (4)$$

where in the second equation we have assumed $A \ll J$. Assuming that the absorption spectrum falls off sufficiently rapidly in the tails (argument due to van Vleck[7]) we obtain a linewidth,

$$T_2^{-1} = A^2/2J, \quad (5)$$

which suggests that the line continues to narrow with increasing J, unlike the correct result Eq. (3). This discrepan-

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cy can be traced to a slight mixing of the electron spin singlet and triplet manifolds from the coupling to the nuclear spins. The mixing leads to a weak absorption in the ESR spectrum of strength $(A/2J)^2$ at the frequency J away from the center of the line. The weak absorption in the tail, however, doubles the second moment and completely dominates the fourth moment. Physically, the correctness of the initial calculation is clear: these weak absorptions deep in tail are really not relevant in the linewidth considerations.

Using this insight we generalize to the N spin cluster. The total Hamiltonian in this case would be

$$H = \sum_{i<j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + A\sum_i \mathbf{S}_i\cdot\mathbf{I}_i + \sum_i \mathbf{S}_i\cdot\mathbf{h}. \tag{6}$$

The electron part of the Hamiltonian yields a set of manifolds spaced apart by an energy of order J and degeneracy $(2S+1)$. By the Clebsch–Gordon decomposition theorem, within each manifold the individual electron spins can be written in terms of the total spin $\mathbf{S}_i = \alpha_i\mathbf{S}$ where α_i are constants which depend upon the distribution of exchange constants and obey the constraint $\Sigma_i\alpha_i = 1$. The projected Hamiltonian in this manifold is

$$H = \mathbf{S}\cdot\left(\sum_i \alpha_i\mathbf{I}_i + \mathbf{h}\right), \tag{7}$$

which is the Hamiltonian we work with. We consider different limits.

(i) Large h, arbitrary N:

We may apply the method of moments to obtain

$$\langle(\Delta\omega)^2\rangle = \frac{1}{T_2^2} = \frac{A^2}{4}\sum_i \alpha_i^2 \approx \frac{A^2}{4N}. \tag{8}$$

In this limit, motional narrowing on a cluster of size N narrows the line by a factor of $\sqrt{N}$ regardless of the overall magnitude of the exchange constants provided they are much larger than A.

(ii) Large N, arbitrary h:

The total magnetic field acting on the electron with spin $\mathbf{S}$ is $(\mathbf{h}+\mathbf{g})$ where $\mathbf{g} = A\Sigma_i\alpha_i\mathbf{I}_i$ is the internal field satisfying the commutation relation

$$[\,g_\mu, g_\nu\,] = i\epsilon_{\nu\mu\rho}A^2\sum\alpha_i^2 I_{i\rho}. \tag{9}$$

Defining

$$\Delta^2 \quad \langle\mathbf{g}^2\rangle/3 = (A^2/4)\sum\alpha_i^2 \approx A^2/(4N),$$

the RMS value of the right-hand side of Eq. (9) is $2\Delta^2/\sqrt{N}$ whereas the left-hand side has a value of order Δ^2. In the large N limit, therefore, the commutator of $\mathbf{g}$ can be neglected and $\mathbf{g}$ behaves classically. We may also show in this limit $\langle\mathbf{g}^{2n}\rangle = (2n-1)!!\Delta^{2n}$ implying that $\mathbf{g}$ obeys gaussian statistics. Thus we may calculate the ESR dynamics in a field $(\mathbf{g}+\mathbf{h})$ and then average over the gaussian distribution of $\mathbf{g}$. In the large h limit the answer agrees with (i), as could be expected.

(iii) Small N and h:

In this case analytic methods are not available and exact diagonalization has to be performed. The diagonalization is tractable for N up to 7, but we may get information for larger sizes by choosing special values of the α_i. We find that the

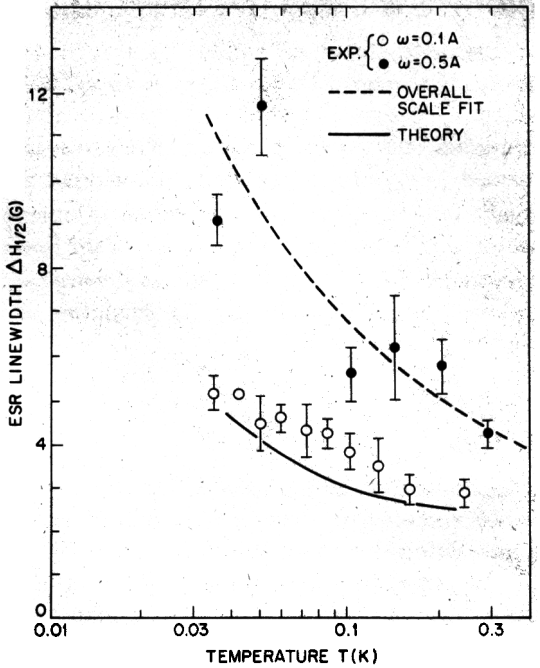

FIG. 1. Linewidth as a function of temperature for two different frequencies. The dashed line is the theoretical fit for determining $N(T)$ while the full line is the theoretical prediction. The constant A is 42 G.

ESR spectrum does indeed show a frequency dependence to the resonance field and linewidth. We assume that the sole effect of temperature is to change the effective (average) cluster size N. In the experiment the ESR spectrum is actually a superposition of a distribution of cluster sizes, which, we conjecture, leads to the Lorentzian line shape. The function $N(T)$ is determined by fitting the experimental linewidth at the higher frequency (where the results are almost classical) as shown in Fig. 1. The remaining data on the resonance field and linewidth is now predicted with no further adjustable parameters, and a comparison of theory with experiment is shown in Figs. 1 and 2. Considering the simplicity of our

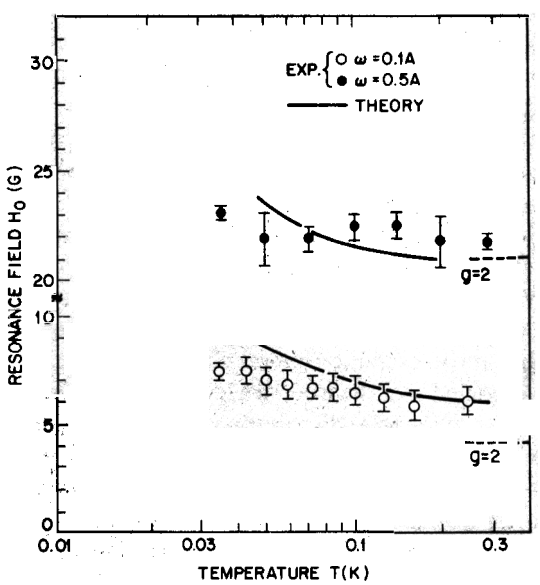

FIG. 2. Resonance field as a function of temperature for two different frequencies with the theoretical predictions. The constant A is 42 G.

model, it is heartening that the calculation captures the correct trends in the unusual temperature and frequency dependences.

The cluster size distribution $N(T)$ can also be obtained independently from a simulation of the disordered antiferromagnet. We impose the requirement that the exchange constants J_{ij} in the cluster are large enough that the spin excitation can propagate across the cluster before it dephases with the rotating magnetic field. The results of this analysis have been published elsewhere[4] and agree well with the above determination of $N(T)$.

III. METAL

The electrons are now delocalized, and travel by the nuclear spins at the Fermi velocity, leading to a motionally narrowed linewidth $(\Delta H_{1/2})_0 \approx A^2/E_F$. In a disordered metal, the diffusive motion of the electron spin leads to a correction to this ballistic linewidth.[3,5] In particular, while the spin is diffusing, repeat visits to the same phosphorus nuclear spin reduces the amount of motional narrowing, leading to a broader line. This effect can be expected to increase as the spin diffusion slows down. Indeed one can show analytically,[3] not too close to the metal-insulator transition, that the renormalized linewidth $\Delta H_{1/2}$ is given by

$$\Delta H_{1/2}(T) \approx (\Delta H_{1/2})_0 \frac{D_0}{D_s(T)},$$

where D_s, is the temperature dependent spin diffusion coefficient and $D_0 \approx v_F l/3$ is the bare Ioffe–Regel spin diffusion constant. The broadening of the line has now been related to the slowing down of spin diffusion. A quantitative understanding of the decrease of D_s, is not as easy as on the insulating side. Perturbation theory in disorder shows that long wavelength spin and density fluctuations enhance the spin susceptibility and suppress D_s by the same temperature dependent factor.[8] The prediction of our analysis here would therefore be that the linewidth and the susceptibility remain directly proportional to each other with falling temperatures. Experimentally this expectation is borne out very well.[3]

IV. CONCLUSION

We have presented a unified picture of the slowing down of spin diffusion in insulating and metallic Si:P. ESR has been shown to be a powerful experimental probe in understanding the nature of spin excitations, and amenable to a theoretical analysis. Such an analysis is an important ingredient in a complete understanding of the nature of the metal-insulator transition in a disordered system such as Si:P.

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