

Griffiths Phase in Diluted Magnetic Semiconductors

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We study the effects of disorder in the vicinity of the ferromagnetic transition in a diluted magnetic semiconductor in the strongly localized regime. We derive an effective polaron Hamiltonian, which leads to the Griffiths phase above the ferromagnetic transition point. The Griffiths-McCoy effects yield nonperturbative contributions to the dynamic susceptibility. We explicitly derive the long-time susceptibility, which has a pseudoscaling form, with the dynamic critical exponent being expressed through the percolation indices.

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Introduction.—There has been increasing interest recently in studying diluted magnetic semiconductors both experimentally and theoretically [1,2]. This interest is motivated not only by the possibility of potential technological applications of the materials but also by a very rich physics, which follows from the unique combination of magnetic and semiconductor properties coexisting in diluted magnetic semiconductors. There is a large number of different materials currently being studied such as $\text{Ga}_{1-x}\text{Mn}_x\text{As}$, $\text{In}_{1-x}\text{Mn}_x\text{As}$, $\text{Ga}_{1-x}\text{Mn}_x\text{P}$, $\text{Ga}_{1-x}\text{Mn}_x\text{N}$, $\text{Ge}_{1-x}\text{Mn}_x$, etc. One of the common important features of these materials is the invariable presence of strong disorder, which plays an essential role in both magnetic and transport properties of the systems with the most prominent effect being the localization of carriers. Disorder arises here from the random locations of magnetic impurities in the host lattice of the semiconductor and also from other impurities and defects in the system. It is important to emphasize that disorder effects are completely neglected in the continuum virtual crystal approximation often used in the theoretical studies of these systems.

It is now widely accepted that ferromagnetism in diluted magnetic semiconductors is due to an indirect interaction between magnetic impurities, which is mediated by holes. The effective magnetic coupling strongly depends upon the ratio $n_i^{1/3}L_{\text{loc}}$ of the localization radius and the mean distance between the magnetic ions. In the strongly localized regime, i.e., when $n_i^{1/3}L_{\text{loc}} \ll 1$, the system may be viewed as a combination of two static magnetic subsystems (magnetic impurities and holes) with the corresponding Hamiltonian having the following form:

$$\mathcal{H} = \sum_{ia} J(|\mathbf{r}_a - \mathbf{r}_i|) \mathbf{S}_a \boldsymbol{\sigma}_i + \sum_{ab} J_{\text{AF}}(|\mathbf{r}_a - \mathbf{r}_b|) \mathbf{S}_a \mathbf{S}_b, \quad (1)$$

where \mathbf{S}_a are impurity spins and $\boldsymbol{\sigma}_i$ are hole spins, $J(r)$ is the manganese-hole interaction, which is given by

$$J(r) = J_0 \exp\left(-\frac{r}{L_{\text{loc}}}\right), \quad (2)$$

and $J_{\text{AF}}(r)$ is the direct antiferromagnetic coupling originating from the exchange interaction of the deep-lying electron states of magnetic impurities, which decays exponentially with distance much more rapidly than the manganese-hole coupling (2). The Hamiltonian (1) leads to a polaron percolation picture [3], which qualitatively explains the mechanism of the ferromagnetic transition in the system: In diluted magnetic semiconductors, the concentration of holes is much smaller than the one of manganese atoms n_i . If $T \ll |J_0|$, the magnetic impurities around each localized hole get polarized forming a bound magnetic polaron. As we lower the temperature, the polaron size $R_p(T)$ increases and polarons overlap forming finite clusters. At some threshold temperature T_c , an infinite cluster appears signaling the ferromagnetic transition. The direct antiferromagnetic coupling cuts off the polaron growth at the distance $R_* = L_{\text{loc}} \ln|J_0/J_{\text{AF}}(n_i^{-1/3})|$, when the manganese impurities prefer the antiferromagnetic alignment to the coupling to a distant hole. If this distance exceeds the polaron size at the transition point $R_* \gg R_p(T_c)$, the antiferromagnetic interaction does not affect the ferromagnetic transition physics and can be neglected at $T \sim T_c$ (we accept this assumption in the present Letter).

It is possible to formalize the polaron picture developed in the an earlier Letter [3] by deriving the effective Hamiltonian for interacting polarons. This can be achieved (see below) by integrating out the manganese spins. The resulting Hamiltonian (8) is the ferromagnetic Heisenberg-type Hamiltonian with small logarithmic “non-Heisenberg” corrections, which can be neglected. The corresponding interaction constant depends upon the interpolaron distance, which is the distance between the localization centers of the corresponding holes. Since the spatial distribution of the holes is the random Poisson distribution, the ferromagnetic polaron coupling becomes a random variable with a well-defined distribution function. This maps the initial problem onto a strongly disordered ferromagnetic Heisenberg model with randomly placed sites. This system naturally leads to the well-known Griffiths-McCoy effects [4,5] in the disordered phase near the phase transition. The

Griffiths-McCoy singularities are the central idea of the present Letter.

The original Letter of Griffiths [4] addressed the nature of the transition in a random Ising ferromagnet, although the main conclusions are relevant to much more general systems. The central idea of the Griffiths theory is that above the ferromagnetic transition point in a disordered system there is always a finite probability of finding an arbitrary large ferromagnetic cluster. These rare-fluctuation clusters give singular contributions to the magnetization, which can be proven to be a nonanalytic function of the external magnetic field. The corresponding phase is called the Griffiths phase. The statement of the nonanalyticity of the magnetization is a purely mathematical statement. The corresponding corrections, although singular, are extremely (exponentially) small, and it is not clear whether such effects in thermodynamic quantities are observable in real experiments. However, the Griffiths effects are much more pronounced in dynamic quantities because the disorder in the model is quenched (does not depend on time) and thus the rare-fluctuation induced clusters are infinitely extended in the time direction. Thus, in the Griffiths phase region, the dynamic response is governed by the Griffiths-McCoy effects as was shown in Ref. [6]. Currently there is a great deal of interest in Griffiths physics related to quantum phase transitions [7,8] (a quantum d -dimensional system is equivalent to a $(d + 1)$ -dimensional classical systems at $T = 0$; thus, a quantum phase transition in a disordered system is governed by Griffiths-McCoy effects due to disorder being perfectly correlated in time).

Effective polaron Hamiltonian.—Let us consider two localized holes embedded into the system of impurity spins. Let the interhole distance be large: $r \gg n_i^{-1/3}$. The partition function for the system reads

$$Z = \text{Tr}_{\sigma_1, \sigma_2} \text{Tr}_{\{S_a\}} \prod_a \exp \left\{ -\frac{1}{T} \hat{\mathbf{S}}_a [J(|\mathbf{r}_a - \mathbf{r}_1|) \hat{\boldsymbol{\sigma}}_1 + J(|\mathbf{r}_a - \mathbf{r}_2|) \hat{\boldsymbol{\sigma}}_2] \right\}. \quad (3)$$

It is important to realize that we are building an effective description in terms of bound magnetic polarons, which are very “heavy” classical objects, and the impurity spins are generally large ($5/2$ for manganese impurities). Thus, we can ignore quantum effects and replace the traces in Eq. (3) by the corresponding classical integrals. We rewrite Eq. (3) as

$$Z = \oint \frac{d\Omega_1}{4\pi} \frac{d\Omega_2}{4\pi} \exp \left\{ -\frac{\mathcal{E}_{\text{eff}}(\cos\theta, T)}{T} \right\}, \quad (4)$$

where integration over Ω_i implies averaging over the orientation of the i th polaron’s spin, θ is the angle between the spins of the two polarons, and the effective interaction is defined as [see Eq. (3)]

$$\mathcal{E}_{\text{eff}}(\cos\theta, T) = -T \sum_a \ln \left\{ \frac{\sinh[\tilde{J}(\mathbf{r}_a)/T]}{\tilde{J}(\mathbf{r}_a)/T} \right\}, \quad (5)$$

with

$$\tilde{J}(\mathbf{r}) = J_0 \sqrt{2} \exp \left(-\frac{r_1 + r_2}{2L_{\text{loc}}} \right) \sqrt{\cosh \left(\frac{r_1 - r_2}{L_{\text{loc}}} \right) + \cos\theta}, \quad (6)$$

where $r_{1,2}$ are the distances between a magnetic impurity and polaron centers (localized holes). Taking into account the fact that the density of magnetic impurities is high, we can replace the sum in Eq. (5) by the integral

$$\frac{\partial \mathcal{E}_{\text{eff}}(\cos\theta, T)}{\partial \cos\theta} = -n_i \int d^3\mathbf{r} \tanh \left[\frac{\tilde{J}(\mathbf{r}_a)}{T} \right] \frac{\partial \tilde{J}(\mathbf{r})}{\partial \cos\theta}. \quad (7)$$

In the limit $L_{\text{loc}} \ll r$, we have

$$\begin{aligned} \mathcal{E}_{\text{eff}}(\cos\theta, T) = & -\frac{J_0^2}{T} \left(\frac{\pi}{6} L_{\text{loc}}^2 n_i \right) \exp \left(-\frac{r}{L_{\text{loc}}} \right) \\ & \times \ln \left[f(\theta) \frac{T}{J} e^{r/2L_{\text{loc}}} \right] \cos\theta, \end{aligned} \quad (8)$$

where $f(\theta) \sim 1$ is a function of θ , which is always non-zero and can be neglected within the logarithmic accuracy. We conclude that the effective polaron Hamiltonian is the ferromagnetic exchange Hamiltonian with randomly placed sites and the couplings between them exponentially decaying with distance.

A similar random exchange Hamiltonian was considered by Korenblit *et al.* in Ref. [9], where it was found that the ferromagnetic transition in the system can be conveniently described with the help of the percolation theory. Within the percolation approach, two interacting polarons are considered either locked in the same direction (if the effective interaction is larger than temperature: $J_{\text{eff}} > T$) or completely uncorrelated in the opposite limit, $J_{\text{eff}} < T$ (which essentially means that the weak couplings can be set to zero). This means that we replace the initial system of randomly interacting polarons by a system in which the weaker couplings are cut. When decreasing temperature, the number of strongly coupled polarons increases and at some temperature, an infinite cluster appears. This temperature is identified with the ferromagnetic transition temperature of the system. We emphasize that the percolation theory is able to establish T_c only up to a numeric factor of the order of unity. In the immediate vicinity of the ferromagnetic transition point, the percolation theory may fail since it is not able to account for critical fluctuations, which become increasingly important at $T \rightarrow T_c$. However, there is a small parameter in our theory, namely, $n_h^{1/3} L_{\text{loc}} \ll 1$. The parameter p of the percolation theory is related to temperature by

$$p(T) \equiv n_h^{1/3} L_{\text{loc}} \ln \left(\frac{J_0}{T} \right), \quad (9)$$

with $p_c \approx 0.86$. Thus there exists a domain $1 \gg |p - p_c|/p_c \gg n_h^{1/3} L_{\text{loc}}$, which is outside the thermodynamic critical region $|T - T_c|/T_c \ll 1$ but still close to the percolation transition, where we can use the asymptotic results of the percolation theory.

Griffiths theorem.—Let us prove the existence of the Griffiths phase in the framework of the percolation theory. In the percolation picture, we have a number of finite clusters above the transition point. The concentration of clusters containing N polarons for $N \gg \xi^D$ is [8,10]

$$P(N, T) \propto N^{-\tau} \left(\frac{N}{\xi^D} \right)^{\tau-\theta} \exp \left[-A \left(\frac{N}{\xi^D} \right)^\zeta \right], \quad (10)$$

where N is the number of polarons in a cluster and $\xi \propto [|p(T) - p_c|/p_c]^{-\nu}$ is the dimensionless correlation length. Percolation exponents are as follows:

$$\tau \approx 2.18, \quad \nu \approx 0.88, \quad (11a)$$

$$T > T_c : \zeta = 1, \theta = 3/2, D = 2, \quad (11b)$$

$$T < T_c : \zeta = 2/3, \theta = -1/9, D = 3 \quad (11c)$$

for the three-dimensional percolation. Constant A in Eq. (10) is a number of the order of unity.

Each polaron has a spin $S_p = S n_i v_p$, where S is the spin of a single magnetic impurity and v_p is the polaron volume which is the volume of the sphere of radius $R_p \approx L_{\text{loc}} \ln(J_0/T)$. The magnetization of the system in an external magnetic field \mathbf{H} can be written as follows:

$$M(H) = S_p \sum_N N P(N) \left[\coth \frac{N S_p H}{T} - \frac{T}{N S_p H} \right]. \quad (12)$$

Let us expand this function in Taylor series

$$M(H) = \sum_N a_N H^N. \quad (13)$$

Following Ref. [11], we obtain the radius of convergence R_H of the series

$$R_H = \left(\frac{T}{S_p} \right) \lim_{N \rightarrow +\infty} \left| \frac{a_{N-1}}{a_N} \right| = \zeta^{1/\zeta} \left(\frac{2\pi T}{S_p} \right) \lim_{N \rightarrow \infty} N^{-1/\zeta} = 0.$$

Above the percolation threshold, $\zeta = 1$ and the series (13) has zero radius of convergence, which implies the non-analyticity of the magnetization as a function of the external magnetic field due to rare disorder fluctuations, i.e., the essential Griffiths singularity. The corresponding singularity in the magnetization and susceptibility is, however, very weak:

$$M(H) \propto \exp \left[-\text{const} \left(\frac{T}{H} \right) \right], \quad (14)$$

which is typical for the Griffiths effects on thermodynamic quantities in classical systems.

Dynamic susceptibility.—The Griffiths physics manifests itself much stronger in dynamic quantities. The quantity of interest is the dynamic susceptibility, which is connected with the spin-spin autocorrelation function

$$C(t) = \frac{1}{V n_i} \sum_a \langle \mathbf{S}_a(t) \mathbf{S}_a(0) \rangle. \quad (15)$$

In the Griffiths phase, the disorder-averaged correlator

can be written as follows [6,8]:

$$C(t) = \sum_N P(N) C_N(t), \quad (16)$$

where $C_N(t)$ is a function describing the relaxation of the magnetic moment of a cluster of size N .

The relaxation of the magnetic moment in a cluster is governed by the Gilbert equation:

$$\frac{\partial \mathbf{m}}{\partial t} = \gamma \mathbf{m} \times \left\{ -\frac{\partial \mathcal{F}}{\partial \mathbf{m}} - \eta \frac{\partial \mathbf{m}}{\partial t} + \mathbf{h}(t) \right\}, \quad (17)$$

where γ and η are constants corresponding to the gyro-magnetic ratio and Gilbert damping and $\mathbf{m} = \mathbf{M}/V_N$ is the magnetization density with V_N being the volume of the cluster $V_N = N v_p = N(4\pi/3) L_{\text{loc}}^3 \ln^3(J_0/T)$. Function $\mathcal{F}(\mathbf{m})$ is the free energy of the cluster per volume as a function of the direction of the magnetic moment, and $\mathbf{h}(t)$ is the fluctuating magnetic field. Let us note that the case of $h = 0$ and $\mathcal{F}(\mathbf{m}) = -\mathbf{H} \mathbf{m}$ yields the well-known Landau-Lifshitz equations [12] with the dimensionless damping coefficient $\alpha = \eta \gamma m$. We assume that anisotropy of the cluster is weak enough so that parameter $[V_N(\mathcal{F}_{\text{max}} - \mathcal{F}_{\text{min}})/T]$ is very small, which corresponds to the superparamagnetic limit.

The random magnetic field $\mathbf{h}(t)$ is due to thermal fluctuations. They are supposed to be Gaussian, with the following white-noise correlation functions:

$$\langle \mathbf{h}(t) \rangle = \mathbf{0} \quad \text{and} \quad \langle h_\alpha(t) h_\beta(0) \rangle = \mu \delta_{\alpha\beta} \delta(t), \quad (18)$$

where Greek indices label Cartesian coordinates. It is possible (see Ref. [13]) to connect the constant μ with the properties of the cluster and temperature. To do so, one imposes the natural condition that the stationary solution of the stochastic Gilbert equation (17) subject to the thermal noise (18) reduces to the Boltzmann equilibrium distribution for the magnetization density: $\mathbf{m}(\theta, \phi) \propto \exp[-V_N \mathcal{F}(\theta, \phi)/T]$. This leads to the following expression for the constant in the correlator (18): $\mu = 2\eta T/V_N$.

It is further possible to derive general Fokker-Planck equations for the distribution function of the magnetic moment $P[\mathbf{m}; t]$ in the case of an arbitrary anisotropy $\mathcal{F}(\mathbf{m})$ (this was done in the pioneering paper of Brown [13]). We, however, confine ourselves to studying the isotropic case only when the Fokker-Planck equation reduces to the simple diffusion equation on a sphere (also described phenomenologically by Bray in Ref. [6]):

$$\left[\frac{\partial}{\partial t} - \mathcal{D} \Delta_{\theta, \phi} \right] P[\mathbf{m}, t] = 0, \quad (19)$$

where $\Delta_{\theta, \phi}$ is the angular part of the Laplacian and the effective diffusion coefficient is equal to (see, e.g., Ref. [13])

$$\mathcal{D}(N, T) = \frac{3}{4\pi S} \left(\frac{\alpha \gamma}{1 + \alpha^2} \right) \frac{T}{\ln^3 \frac{J_0}{T}} \frac{1}{n_i L_{\text{loc}}^3 N}, \quad (20)$$

where α is the dimensionless damping coefficient introduced in the previous paragraph, γ is the gyromagnetic ratio, and S is the magnetic impurity spin.

Let us suppose that the initial magnetization $\mathbf{m}(t=0)$ of the cluster is in the z direction, which corresponds to the initial condition $P[\theta, \phi; t=0] = \delta(\theta)$ for the distribution function, defining the Green function for the diffusion equation (19). The corresponding solution reads

$$P[\theta; t] = \sum_{l=0}^{\infty} \left(l + \frac{1}{2} \right) P_l(\cos\theta) \exp[-\mathcal{D}l(l+1)t] \sin\theta, \quad (21)$$

where $P_l(\cdot)$ are Legendre polynomials. The z component of the magnetization averaged over the distribution function (21) is exactly the function $C_N(t)$ introduced in Eq. (16), which describes the relaxation of the magnetic moment in a cluster of size N :

$$C_N(t) = M \exp[-2\mathcal{D}(N, T)t], \quad (22)$$

We now proceed to calculate the spin autocorrelation function by replacing the sum in Eq. (16) by the integral

$$C(t) \propto \int_0^{+\infty} dN \frac{N^{1-\theta}}{\xi^{d(\tau-\theta)}} \exp \left[-A \left(\frac{N}{\xi^d} \right)^{\xi} - 2\mathcal{D}(N, T)t \right], \quad (23)$$

which can be evaluated by the saddle point approximation:

$$C(t) \propto \left(\frac{p(T) - p_c}{p_c} \right)^a t^b \exp \left[-g(T) \left(\frac{p(T) - p_c}{p_c} \right)^c t^{\Delta} \right], \quad (24)$$

where, for the sake of brevity, we have introduced the following indices:

$$\Delta = \frac{\xi}{1 + \xi}, \quad (25a)$$

$$c = \frac{\nu D \xi}{1 + \xi}, \quad (25b)$$

$$b = \frac{2 - \theta - \xi/2}{1 + \xi}, \quad (25c)$$

$$a = \nu D \left[\tau - \theta - \frac{\xi}{1 + \xi} \left(\frac{5}{2} - \theta \right) \right], \quad (25d)$$

and the function in the exponent reads

$$g(T) = A^{1/(1+\xi)} [\xi^{-\xi/(1+\xi)} + \xi^{1/(1+\xi)}] [2N\mathcal{D}(N, T)]^{\xi/(1+\xi)}, \quad (26)$$

where the diffusion coefficient is defined by Eq. (20) and A is the number of the order of unity introduced in Eq. (10). The explicit values for the parameters above (below) the transition are $\Delta = 1/2$ ($2/5$), $c \approx 0.88$ (1.06), $b \approx 0$ ($14/15$), and $a \approx 0.32$ (3.29).

Conclusion.—We have shown that a diluted magnetic semiconductor in the strongly localized regime (i.e., if

$n_h^{1/3} L_{\text{loc}} \ll 1$) must be in the Griffiths phase in the vicinity of the ferromagnetic transition point. The magnetization is a nonanalytic function of the external magnetic field with the corresponding singularity being exponentially small. The dynamic response in the vicinity of the transition point is determined by rare-fluctuation Griffiths droplets. These droplets correspond to the percolating clusters in the polaron percolation picture. The dynamic susceptibility has the form (24) of the stretched exponential $\ln C(t) \propto -t^{\Delta}$, with Δ being connected to the percolation indices and equal to $1/2$ in the Griffiths phase and $2/5$ in the ferromagnetic phase. An important consequence of our work relates to the role of disorder in ferromagnetic semiconductors. Since disorder is fundamental to the occurrence of the Griffiths phase, an observation of Griffiths physics in diluted magnetic semiconductors will have significance with respect to the role of quenched disorder in these systems. In fact, we believe that insulating diluted magnetic semiconductors (e.g., GaMnAs or InMnAs on the insulating side of the metal-insulator transition) may be an ideal system to look for signatures of evasive Griffiths physics.

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- [1] H. Ohno, *Science* **281**, 951 (1998).
 - [2] R. N. Bhatt, M. Berciu, M. Kennett, and X. Wan, *J. Supercond.* **15**, 71 (2002); S. Sanvito, G. Theurich, and N. A. Hill, *ibid.* **15**, 85 (2002); S. Das Sarma, E. H. Hwang, and A. Kaminski, *Phys. Rev. B* **67**, 155201 (2003).
 - [3] A. Kaminski and S. Das Sarma, *Phys. Rev. Lett.* **88**, 247202 (2002).
 - [4] R. B. Griffiths, *Phys. Rev. Lett.* **23**, 17 (1969).
 - [5] B. M. McCoy and T. T. Wu, *Phys. Rev.* **176**, 631 (1968); **188**, 982 (1969).
 - [6] A. J. Bray, *Phys. Rev. Lett.* **60**, 720 (1988).
 - [7] O. Motrunich, K. Damle, and D. A. Huse, *Phys. Rev. B* **63**, 224204 (2001); B. N. Narozhny, I. L. Aleiner, and A. I. Larkin, *ibid.* **62**, 14898 (2000); B. Spivak and F. Zhou, *Phys. Rev. Lett.* **74**, 2800 (1995).
 - [8] T. Senthil and S. Sachdev, *Phys. Rev. Lett.* **77**, 5292 (1996).
 - [9] I. Y. Korenblit, E. F. Shender, and B. I. Shklovskii, *Phys. Lett.* **46A**, 275 (1973).
 - [10] D. Stauffer and A. Aharony, *Introduction to Percolation Theory* (Taylor & Francis, London, 1994).
 - [11] M. M. Bakri and D. Stauffer, *Phys. Rev. B* **14**, 4215 (1976).
 - [12] L. Landau and E. Lifshitz, *Phys. Z. Sowjetunion* **8**, 153 (1935).
 - [13] W. F. Brown, *Phys. Rev.* **130**, 1677 (1963).