Charge and Magnetization Inhomogeneities in Diluted Magnetic Semiconductors

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It is predicted that III-V diluted magnetic semiconductors can exhibit stripe-like modulations of magnetization and carrier concentration. This inhomogeneity results from the strong dependence of the magnetization on the carrier concentration. Within Landau theory, a characteristic temperature \( T^* \) below the Curie temperature is found so that below \( T^* \) the equilibrium magnetization shows modulations, which are strongly anharmonic. Wavelength and amplitude of the modulation rise for decreasing temperature, starting from zero at \( T^* \). Above \( T^* \) the equilibrium state is homogeneous, but the coupling between charge and magnetization leads to the appearance of an electrically charged layer in domain walls.

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Introduction.—Diluted magnetic semiconductors (DMS) are investigated extensively as promising materials for spintronics applications \([1,2]\) and because of their unique physical properties \([3,4]\). Since the magnetic interaction is mediated by the carriers, the magnetization and the Curie temperature increase for increasing carrier concentration \([5,6,7,8,9,10]\). The magnetic part has the usual form

\[
H_m = \int d^3r \left\{ \frac{\alpha}{2} m^2 + \frac{\beta}{4} m^4 + \frac{\gamma}{2} \nabla \cdot \mathbf{m} \cdot \nabla \mathbf{m} \right\},
\]

where \( \partial_i \equiv \partial / \partial r_i \) and summation over \( i \) is implied. The mean-field Curie temperature is determined by \( \alpha = 0 \). Since experimentally the Curie temperature depends approximately linearly on carrier concentration, we expand \( \alpha = \alpha' (T - T_c - \eta \delta n) \), where \( T_c \) is the Curie temperature for \( \delta n = 0 \). This dependence of \( \alpha \) provides the coupling between magnetism and carrier concentration in our model and is responsible for the physics discussed in the following. Since the equilibrium magnetization for constant \( \alpha \) is \( m_0 \equiv \sqrt{-\alpha/\beta} \), larger magnetization is favored in regions with higher carrier concentration (note \( \alpha < 0 \) in the ferromagnetic phase).

The second ingredient for our model is the screened Coulomb energy due to the charge inhomogeneity,

\[
H_{\delta n} = \frac{1}{2} \int d^3r d^3r' \frac{e^2}{4\pi \epsilon_0 \epsilon} \delta n(r) \delta n(r') e^{-|r-r'|/\xi_0}.
\]

The total Hamiltonian is \( H = H_m + H_{\delta n} \).

We first discuss qualitatively what kind of equilibrium states we expect from \( H \). Any inhomogeneous charge distribution increases \( H_{\delta n} \). On the other hand, the contribution from \( H_m \) is not obvious, since the first term is negative for \( T < T_c + \eta \delta n \). We will see that the magnetic energy decrease in regions of higher carrier concentration and magnetization can outweigh the increase in regions of lower \( \delta n \) and \( m \) and even the increase in electrostatic energy. In that case, the equilibrium state is indeed inhomogeneous. We consider stripe-like, one-dimensional modulations. Two- and three-dimensional patterns seem less likely, because they contain more regions with large magnetization gradients for a given inhomogeneity length scale, which increase the energy due to the gradient term in \( H_m \). One could expect the inhomogeneity to take the form of stripe domains \([16]\) with alternating magnetization. However, we will see that the equilibrium solution shows a magnetization modulation without sign change.

We now turn to the formal derivation of equilibrium states. It is convenient to express \( H_{\delta n} \) in terms of the electrostatic potential \( \phi \). With \( (\Delta - \eta^{-2}) \phi(r) = -e \delta n(r)/\epsilon \epsilon_0 \) (for p-type DMS) we obtain the total Hamiltonian

\[
H = \int d^3r \left\{ \alpha' (T - T_c) m_0^2 + \frac{\beta}{4} m^4 + \frac{\gamma}{2} \partial_i m_i \cdot \partial_j m_j \right\} + \frac{e \epsilon_0}{2\xi_0^2} \phi^2 + \frac{e \epsilon_0}{2} \partial_i \phi \partial_j \phi - \frac{\alpha' \epsilon \epsilon_0}{2e \epsilon_0^2} m^2 \phi - \frac{\alpha' \epsilon \epsilon_0}{e} m_i \cdot (\partial_i m) \partial_j \phi \right\}.
\]

Equilibrium configurations are given by minima of \( H \) subject to the constraint of charge neutrality, \( \int d^3r (\Delta - \eta^{-2}) \phi = 0 \),
This is an integro-differential equation due to the term for only depend on to rescaling. We choose is density since retention (4) retains information about the coupling to the carrier magnetization in terms of .

Periodic solutions exist for for and type 3 solutions for for type 2 and 3 solutions. The homogeneous solutions are given by . Eliminating from the first we find

The homogeneous solutions for type 1 are obtained by setting . This expression satisfies . The homogeneous solutions for type 2 and 3 solutions.

Type 2 solutions exist if numerator and denominator must have another zero at the next extremum of this point normally cannot be reached and there is no solution.

Periodic solutions.—For periodic, collinear solutions that must be either larger or smaller than for . The lines A, B, C show the shift of the solutions in terms of for the solutions continue periodically.

A special role is played by the magnetization value for , where . The equations support the homogeneous mean-field solution for for . While Eqs. contain five parameters, it is sufficient to vary only two to obtain all possible solutions up to rescaling. We choose for periodic solutions up to .

Periodic solutions.—For periodic, collinear solutions that only depend on , Eq. becomes

This is an integro-differential equation due to the term for and are imposed, where will be obtained by minimizing the energy.

Using standard methods, we obtain the explicit integral for the inverse function on the interval .

This expression satisfies since and diverges at . To obtain a periodic function, the denominator must have another zero at the next extremum of . However, a solution (here called type 1) crossing is possible if numerator and denominator vanish simultaneously. This solution is oscillating between and is the minimum of the denominator.

For all other periodic solutions must be either larger or smaller than everywhere. For oscillating between and with zero average exist if . For oscillating around for exist for and .

Next, the phase diagram is mapped back onto the parameters of the Euler equation , determining . The results are shown in Fig. 2(a) for . The phase diagram for other values has the same topology. In the distorted triangle with corners A,B,0 two solutions with different wavelength coexist. (b) Phase diagram for periodic magnetization solutions in terms of and for . The various solutions exist to the left of the respectively lines.
crossing points marked A, B, C in Fig. 2(a) with $dr_0^2/a$. Type 3 solutions exist to the left of point A, i.e., for $m_0 > m_{\text{sing}}$.

From Eqs. (3)–(5) we find the average energy density

$$\tau = \frac{1}{\lambda} \int_0^\lambda dx \left\{ -\frac{d}{4} m^4 + \frac{a}{2} m^2 (\partial_x m)^2 \right\} - \frac{a}{8r_0^3} (m^2)^2. \tag{8}$$

For the homogeneous solution we obtain the standard result $\tau = \epsilon_{\text{hom}} \equiv -c^2/4d$. Numerical evaluation shows that type 1 and 2 (type 3) solutions always have higher (lower) energy than the homogeneous solution. Among type 3 solutions the energy is minimized by the maximum amplitude, where $m$ comes arbitrarily close to $m_{\text{sing}}$.

The mean-field magnetization of our DMS model is thus zero for $T \geq T_c$, homogeneous for $T^* \leq T < T_c$, where

$$T^* \equiv T_c - \frac{e^2}{\epsilon_0 \alpha' \eta^2} \tag{9}$$

corresponds to line A in Fig. 2(b), and a periodic spin-density and charge-density wave for $T < T^*$. The dipolar interaction omitted here favors $m$ lying in the $yz$ plane. The magnetization and potential show sharp cusps at the minima of $m$, see the inset in Fig. 3. The cusps lead to negative peaks in the carrier density, which become $\delta$-functions for $m^* \to m_{\text{sing}}$. This divergence is cut off by the condition of non-negative hole concentration. Since the amplitude, wavelength, and energy approach finite values for $m^* \to m_{\text{sing}}$, the Landau theory gives a good impression of the profile, except for some broadening of the cusps.

The optimum solution can be written down explicitly,

$$m(x) = \sqrt{2 m_{\text{min}}^2 - m_{\text{sing}}^2} \sin \left( \sqrt{\frac{2}{a}} x + \theta \right) \tag{10}$$

for $0 \leq x \leq \lambda$ and periodically repeated. Here, $(2m_{\text{min}} - m_{\text{sing}})^1/2 \sin \theta = m_{\text{sing}}$. From $m(x)$ one can obtain expressions for the wavelength $\lambda = 2(2a/d)^1/2 (\pi/2 - \theta)$, the average magnetization $\bar{m}$, the peak-to-peak amplitude $\delta m_{pp} = (2m_{\text{min}} - m_{\text{sing}})^1/2 - m_{\text{sing}}$, and the energy, see Fig. 3. Note that $\bar{m}$ is nonzero for all $T < T_c$. Close to $T^*$ the wavelength becomes small. In this regime, the continuum theory breaks down, since $\lambda$ is not large compared to the disorder length scale. Figure 3 also shows that the fundamental length scale is the screening length $r_0$. The inset shows a typical solution.

It is important to check whether the periodic solution can occur in real DMS. For that, $T_c - T^*$ should be small. Equation 10 shows that this is the case for high dielectric constant, small spin stiffness, strong dependence of $T_c$ on carrier concentration, and rapid onset of magnetization below $T_c$. For (Ga,Mn)As we estimate $T^*$ by comparing experiments and mean-field theory for homogeneous magnetization and to spin-wave theory. We find $T_c - T^*$ of the order of 10 K. The properties of (Ga,Mn)As vary strongly with Mn concentration and growth procedures. In particular, $T_c - T^*$ is inversely proportional to the square of the shift of $T_c$ with carrier concentration, $\eta^2$.

In Ref. 9, $\eta \approx 5.4 \times 10^{-3} \text{KÅ}^3$, which was used for the above estimate, whereas in Ref. 7 $\eta \approx 1.5 \times 10^3 \text{KÅ}^3$, which would increase $T_c - T^*$.

Figure 3 suggests that measurements of the average magnetization, which have been performed extensively, are unlikely to find evidence for the inhomogeneous state. For that, probes sensitive to the spatial variation are required. For example, the magnetic modulation should be observable in neutron-scattering experiments. In real space, magnetic scanning-tunneling microscopy (STM) and, for large $\lambda$, scanning Hall probe experiments or magneto-optical techniques are promising. Conversely, the modulation in carrier concentration should be observable in optical reflection or transmission for large enough $\lambda$. It also leads to a modulation of the local density of states which could be probed by STM.

**Domain walls.**—Finally, we study the effect of magnetization-carrier coupling on domain walls. We restrict ourselves to solutions that are homogeneous in the $y$, $z$ directions. Equations 4,5 are solved under the boundary conditions $\lim_{x \to \pm \infty} m(x) = \pm m_0 z$, where $z$ is the unit vector in the $z$ direction. Since $m^2(x)$ only deviates appreciably from $m_0^2$ in a finite interval, we have $m^2 = m_0^2 = -c/d$ in the limit of infinite system size, $L \to \infty$. However, it turns out that charge neutrality can only be satisfied by keeping terms of order $1/L$ in $m^2$. One such term comes from the region far from the wall, where we write $m(L/2) \approx m_0 + m_1/L$. This means that the enhanced carrier density compensating the reduction in the wall is spread out over the bulk.

With the additional condition $m(0) = 0$ we obtain

$$x = \int_0^m d\tilde{m} \sqrt{\frac{b - a\tilde{m}^2}{c\tilde{m}^2 + d\tilde{m}^4/2 - c'm_0^2 - d'm_0^4/2}} \tag{11}$$
FIG. 4: (color online). Magnetization and electrostatic potential for typical domain-wall solutions for \(a/dr_0^2 = 1\). The curves are for \(cr_0^2/b = -0.1, -0.3, -0.5, -0.7, -0.9, -0.99\). The asymptotical values \(m_0 = m(x \to \infty)\) are indicated by triangles. Inset: Energy density \(\Delta \sigma\) of a domain wall as a function of coupling strength for the same values of \(c\).

The area energy density of the domain wall is obtained by integrating the energy density over \(x\), where corrections to \(m(x)\) of order \(1/L\) are again relevant,

\[
\Delta \sigma = \int_{-\infty}^{\infty} dx \left[ -\frac{cd}{2(d+a/2r_0^2)} \Delta m^2 - \frac{1}{4} \left( d - \frac{a}{2r_0^2} \right) \right. \\
\times \left. (\Delta m^2)^2 + \frac{a}{8} (\partial_x \Delta m^2)^2 \right] 
\]  

(12)

with \(\Delta m^2 \equiv m^2(\pm \infty) - m^2(x)\). The dependence of \(\Delta \sigma\) on the coupling \(a \propto \eta^2\) is shown in the inset in Fig. 4. \(\Delta \sigma\) first decreases with increasing coupling and then increases again, finally diverging as \(d' = d - a/2r_0^2\) goes to zero. For larger \(|cr_0^2/b|\) the divergence is not reached, since the condition \(m_0 = m_{\text{sing}}\) (i.e., \(T = T^*\)) is satisfied first. The initial decrease is dominated by the \(1/L\) term in \(\Delta m^2(x)\) far from the wall, i.e., from the redistribution of carriers. In this regime, domain walls are (slightly) less costly than they would be without coupling. The strong increase mostly comes from the increased width due to Coulomb repulsion. Domain walls could be observed in the real-space experiments discussed above. The charged layer should also affect electronic transport through domain walls.

Conclusions.—The carrier-concentration dependence of the magnetization in DMS introduces a characteristic temperature \(T^* < T_c\) such that the mean-field magnetization \(m\) and excess carrier density \(\delta n\) show periodic modulations for \(T < T^*\), whereas \(m\) is homogeneous and \(\delta n = 0\) above \(T^*\). \(T_c - T^*\) can be of the order of 10 K in p-type DMS. The modulation is strongly anharmonic, and amplitude and wavelength increase for decreasing temperature, starting from zero at \(T^*\). For \(T \geq T^*\) the equilibrium state is homogeneous, but the coupling between magnetism and carrier concentration leads to the appearance of a negatively charged layer in the vicinity of a domain wall for p-type DMS.

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[19] If one imposes the same boundary conditions for \(T < T^*\) the solution must cross the singularities at \(\pm m_{\text{sing}}\). Only an oscillatory solution of type 1 is possible.