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Commensurate and incommensurate phases of epitaxial semiconductor antiferromagnets with built-in strain

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We construct a Landau-Ginzburg theory for a new class of epitaxial semiconductor antiferromagnets with built-in strain. Using present knowledge from experiment we extract the mean-field phase diagram of these systems as a function of the built-in strain field. We find both the commensurate (type-III) and incommensurate (helical) phases seen in experiment as well as a new commensurate phase not yet seen to our knowledge.

Quite recently, a new class of layered magnetic structures has been synthesized by the use of state-of-the-art molecular-beam epitaxy techniques,[1–3] consisting of magnetic layers of MnSe or MnTe sandwiched between nonmagnetic layers of II–VI semiconductors such as ZnSe or ZnTe and grown in a superlattice configuration. Because of the mismatch between the cubic lattice constants of the magnetic and nonmagnetic layers, the magnetic layers are created with an intrinsic built-in strain field whose sign and strength can be tuned via the choice of the nonmagnetic material. The magnetic layers themselves grow in the zinc-blende structure and are found to be type-III Heisenberg antiferromagnets at zero strain.[2] The strain engineering technique described above then allows one the unique opportunity to systematically vary the strain field and thus the magnetic interactions in these systems. This is particularly interesting since zinc-blende Heisenberg antiferromagnets are prototypes of frustrated magnets.

Four systems from this new class of semiconductor antiferromagnets have been grown and characterized, two having built-in compressive strain [ZnSe/MnSe (Ref. 1) and ZnTe/MnTe (Ref. 2)], one with built-in tensile strain [ZnTe/MnSe (Ref. 3)] and a strain-free system [MnTe (Ref. 2)]. The magnetic ordering for the strain-free system consists of the three possible type-III domains, one for each of the cubic axes. For compressive strain, however, only the type-III domain with a doubled unit cell along the growth direction (001) is seen. In the case of tensile strain, the magnetic order consists of a helicoidal spin wave with a wave vector along either of the cubic axes in the growth plane and with a wavelength which is incommensurate with the underlying crystal structure. In contrast, type-III phases are commensurate with a wavelength equal to twice the lattice constant.

Each of the four systems discussed above is characterized by a single and different value of the built-in strain field, and thus one can visualize them as representing four isolated points along a continuous strain axis. Of immediate interest is then the construction of the complete phase diagram as a continuous function of the built-in strain field. Using well established methods, we will show that one can construct a Landau-Ginzburg theory for this purpose which predicts the phases seen in experiment, provides naturally for the observed strain-induced commensurate-incommensurate transition, and predicts the existence of a new commensurate phase not yet seen in experiment.

Our work builds on an earlier Landau-Ginzburg theory developed by Mukamel[4] to study the critical behavior of a type-III antiferromagnet, the ground state of the magnetic multilayers at zero built-in strain. By extending Mukamel's work to include the coupling of the strain field to the magnetic degrees of freedom we arrive at the desired Landau-Ginzburg theory whose mean-field ground states are the strain-induced phases. Without knowledge from experiment, however, one cannot unambiguously determine the ground state and so we discuss the derivation of relations between the Landau-Ginzburg parameters which allows such specification. We begin with the group theory necessary to construct the Landau-Ginzburg theory.

As is well known,[5] the type-III antiferromagnetic structure is described by the wave vector $\mathbf{k}_1 = (2\pi/a)(\frac{1}{2},1,0)$. The magnetic multilayers grow in the zinc-blende structure, which has the space group $F\bar{4}3m$. One then finds that the star of $\mathbf{k}_1$ consists of the six vectors $\mathbf{k}_1 = (2\pi/a)(\frac{1}{2},1,0)$, $\mathbf{k}_{\bar{1}} = (2\pi/a)(-\frac{1}{2},1,0)$, $\mathbf{k}_2 = (2\pi/a)(0,\frac{1}{2},1)$, $\mathbf{k}_{\bar{2}} = (2\pi/a)(0,-\frac{1}{2},1)$, $\mathbf{k}_3 = (2\pi/a)(1,0,\frac{1}{2})$, $\mathbf{k}_{\bar{3}} = (2\pi/a)(1,0,-\frac{1}{2})$. The group of $\mathbf{k}_1$ is S_4 which has both one- and two-dimensional representations, implying that the magnetic order parameter is either six or twelve dimensional. In fact, present experiments[1] are unable to distinguish between the magnetic structures that correspond to the six- and twelve-dimensional representations. We take the simplest approach and construct a theory based on the six-dimensional representation which corresponds to a noncollinear structure, termed Keffer type-III ordering.[6] A theory for the twelve-dimensional representation will be presented in a future publication.

The order parameter components are denoted $\phi_i, \bar{\phi}_i$ for $i = 1,2,3$ (x,y,z) corresponding to the two possible orderings along each of the three cubic axes. A schematic of one of these structures is shown in Fig 1; the remaining five are obtained from it by applying the symmetry operations of $F\bar{4}3m$. Standard group theoretical techniques[4,7,8] then yield the part of the free energy depending only on the magnetic order and given by

$$F_m = \int d\mathbf{x} \left(\sum_{i=1}^{3} [r(\phi_i^2 + \bar{\phi}_i^2) + \beta(\phi_{i,i}^2 + \bar{\phi}_{i,i}^2) + \gamma(\phi_{i,ii}^2 + \bar{\phi}_{i,ii}^2)] + u \left(\sum_{i=1}^{3} \phi_i^2 + \bar{\phi}_i^2 \right)^2 + v \sum_{i=1}^{3} (\phi_i^2 + \bar{\phi}_i^2)^2 + w \sum_{i=1}^{3} \phi_i^2 \bar{\phi}_i^2 \right), \tag{1}$$

where we use the notation $\phi_{i,i} = \partial\phi_i/\partial x_i$ and $\phi_{i,ii} = \partial^2\phi_i/\partial x_i^2$ ($x_1 = x, x_2 = y, x_3 = z$). Consistent with the nature of incommensurate phases in many other magnetic systems,[9] we have only considered spatial derivatives in the direction of the unit cell doubling. Thus we have omitted from Eq. (1) symmetry allowed terms of the form $\phi_{i,k}^2$ and $\phi_{i,kk}^2$ for $i \neq k$. We further note that the presence of the commensurate (type-III) phase in the absence of strain requires $\beta, \gamma > 0$.

The effect of the lattice mismatch between magnetic and nonmagnetic layers is included by treating the magnetic layers as a bulk zinc-blende crystal subjected to a uniform stress arising from the mismatch. Microscopically, this stress derives from the requirement, for epitaxial growth, of a nonvarying growth plane lattice constant and thus the MnSe or MnTe bonds will stretch and bend accordingly. Specifically, we assume a uniform stress in the growth plane which we take to be the x-y plane. The stress, denoted τ, couples to the strain field giving rise to the free-energy term

$$F_\tau = -(A_{xx} + A_{yy})\,\tau, \tag{2}$$

where $A_{ij} = \frac{1}{2}(\partial u_i/\partial x_j + \partial u_j/\partial x_i)$ with u_i the displacement in the ith direction. The external stress then couples to the order parameter indirectly through the coupling of the strain field to the order parameter components. To lowest order in the strain field these terms are generically of the form $\phi_i\phi_j A_{kk}$, and $\phi_{i,i}\phi_{j,j}A_{kk}$. One then finds the magnetoelastic part of the free energy,

$$F_{m\text{-}e} = \int d\mathbf{x} \sum_{i=1}^{3} \left[\left(B_1 A_{ii} + \sum_{j\neq i} B_2 A_{jj} \right) (\phi_i^2 + \bar{\phi}_i^2) + \left(D_1 A_{ii} + \sum_{j\neq i} D_2 A_{jj} \right) (\phi_{i,i}^2 + \bar{\phi}_{i,i}^2) \right], \tag{3}$$

where B_1, B_2, D_1, D_2 are magnetoelastic constants. We also include the elastic part of the free energy in the harmonic approximation given by

$$F_e = \tfrac{1}{2}C_{11}(A_{xx}^2 + A_{yy}^2 + A_{zz}^2) + C_{12}(A_{xx}A_{yy} + A_{xx}A_{zz} + A_{yy}A_{zz}) + 2C_{44}(A_{xy}^2 + A_{xz}^2 + A_{yz}^2), \tag{4}$$

where C_{11}, C_{12}, C_{44} are the elastic constants.[10] The total Landau free energy, denoted F_0, is then given by

$$F_0 = F_m + F_{m-e} + F_e + F_\tau. \tag{5}$$

By first minimizing the free energy with respect to the strain components one obtains the effective free energy given by

$$F = \int d\mathbf{x} \Bigg[r_1(\tau)\,[\phi_1^2 + \bar{\phi}_1^2 + \phi_2^2 + \bar{\phi}_2^2] + r_2(\tau)\,[\phi_3^2 + \bar{\phi}_3^2] + \beta_1(\tau)\,[\phi_{1,x}^2 + \bar{\phi}_{1,x}^2 + \phi_{2,y}^2 + \bar{\phi}_{2,y}^2] + \beta_2(\tau)\,[\phi_{3,z}^2 + \bar{\phi}_{3,z}^2]$$
$$+u \left(\sum_{i=1}^{3} \phi_i^2 + \bar{\phi}_i^2 \right)^2 + v \sum_{i=1}^{3} (\phi_i^2 + \bar{\phi}_i^2)^2 + w \sum_{i=1}^{3} \phi_i^2 \bar{\phi}_i^2 + \gamma \sum_{i=1}^{3} (\phi_{i,ii}^2 + \bar{\phi}_{i,ii}^2) \Bigg], \tag{6}$$

where $r_1(\tau), r_2(\tau), \beta_1(\tau), \beta_2(\tau)$ are linear in τ and are functions of the elastic and magnetoelastic constants. In deriving Eq. (6) we have dropped terms of the form $\phi_i^2\phi_{j,j}^2$ and $\phi_{i,i}^2\phi_{j,j}^2$ which substantially complicate the analysis and can be shown not to alter the qualitative nature of the phase diagram.

In principle, one can now obtain the phase diagram from the mean-field ground state of Eq. (6) as a function of τ. Given our incomplete knowledge of the Landau-Ginzburg parameters, however, such an approach is not possible. In particular, one finds that there are seventeen distinct commensurate states, i.e., local minima, that arise from the possibility of unit cell doubling along one, two, or three simultaneous spatial directions. In the experiments on both compressively strained and unstrained samples, however, one sees only commensurate phases with unit cell doubling along a single direction. Such states are described by nonzero values of ϕ_i and $\bar{\phi}_i$ for a given value of i, with the remaining four components set to zero. We thus restrict our mean-field solutions to be of the above form, which we denote by $[\phi_i(x_i), \bar{\phi}_i(x_i)]$.

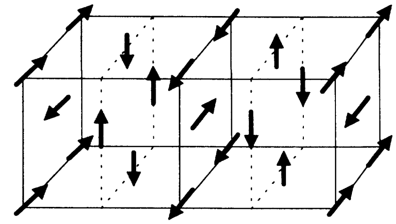

FIG. 1. Magnetic unit cell for Keffer type–III ordering.

[Type-III phases have no spatial dependence and are denoted $(\phi_i, \bar{\phi}_i)$.]

Further progress requires some knowledge about the parameters $r_1(\tau), r_2(\tau)$, which determine the ordering direction, and $\beta_1(\tau), \beta_2(\tau)$, which control whether the phase is commensurate or incommensurate. We find $\beta_1 = \beta + \Delta_1\tau$ and $\beta_2 = \beta + \Delta_2\tau$ where Δ_1 and Δ_2 are functions of the elastic and magnetoelastic constants and $\beta > 0$. In order to account for the commensurate state seen for the compressively strained systems ZnSe/MnSe and ZnTe/MnTe, one requires $\Delta_1, \Delta_2 < 0$ ($\tau < 0$ for compressive strain). One then concludes that for sufficiently large tensile strain ($\tau > 0$), an incommensurate phase may exist. Thus the Landau-Ginzburg theory naturally predicts a strain-driven commensurate-incommensurate transition, as observed experimentally.

The coefficients $r_1(\tau)$ and $r_2(\tau)$ are given by

$$r_1(\tau) = r + \alpha\tau[\kappa_1(B_1 + B_2) - \kappa_2 B_2],$$
$$r_2(\tau) = r + \alpha\tau[2\kappa_1 B_2 - \kappa_2 B_1], \tag{7}$$

where

$$\kappa_1 \equiv \frac{(C_{11}^2 - C_{12}^2)}{C_{11}}, \quad \kappa_2 \equiv \frac{C_{12}\,(C_{11} - C_{12})}{C_{11}},$$
$$\alpha \equiv \frac{C_{11}}{(2C_{12}^3 + C_{11}^3 - 3C_{11}C_{12}^2)}.$$

One can show that the positive definiteness of the elastic constants implies $\alpha > 0$ and thus the relationship between $r_1(\tau)$ and $r_2(\tau)$ is specified given the relationship between the quantities in the brackets in Eq. (7). Such a relationship can be found from our knowledge of magnetostriction in MnTe. Experiments[2] show the presence of magnetostriction such that for a type-III domain with doubled unit cell along x, one has the spontaneous strain field: $A_{xx} > 0, A_{yy} = A_{zz} < 0$. For a commensurate phase oriented along x, and in the absence of external strain, the Landau-Ginzburg theory yields

$$A_{xx} = \frac{\phi_1^2[\kappa_1 B_1 - 2\kappa_2 B_2]}{(\kappa_2^2 - \kappa_1^2)},$$
$$A_{yy} = A_{zz} = \frac{\phi_1^2[(\kappa_1 - \kappa_2)B_2 - \kappa_2 B_1]}{(\kappa_2^2 - \kappa_1^2)}. \tag{8}$$

From the extrapolated elastic constants of MnTe obtained from the known elastic constants of the diluted magnetic semiconductor alloy $Zn_{1-x}Mn_xTe$,[11,12] one finds $\kappa_1 > \kappa_2$ (the same relation applies for MnSe). Requiring that A_{xx}, A_{yy}, A_{zz} in Eq. (8) agree with the experiments, one finds that $r_2(\tau) < r_1(\tau)$ for compressive strain and $r_1(\tau) < r_2(\tau)$ for tensile strain.

The Landau-Ginzburg theory can now be solved for the case of commensurate phases ($\beta_1, \beta_2 > 0$). For compressive strain the ground state is of the form $(\phi_3, \bar{\phi}_3)$ as seen in the experiments on ZnSe/MnSe and ZnTe/MnTe. For tensile strain we find a commensurate phase of the form $(\phi_1, \bar{\phi}_1)$ or $(\phi_2, \bar{\phi}_2)$ not yet seen in experiment and characterized as a type-III phase with doubled unit cell along a cubic axis in the growth plane. One finds that the paramagnetic-antiferromagnetic phase line for compressive strain is determined from $r_2(\tau) = 0$ and for tensile strain by $r_1(\tau) = 0$. In addition the phase boundary between $(\phi_3, \bar{\phi}_3)$ and $(\phi_1, \bar{\phi}_1)$, $(\phi_2, \bar{\phi}_2)$ is found to be first order and occurs at zero built-in strain (see Fig. 2). At exactly zero strain we find, as in the experiments on MnTe, three degenerate commensurate phases corresponding to type-III domains oriented along each of the three cubic axes.

In the case of tensile strain with either or both of $\beta_1, \beta_2 < 0$ the possibility of an incommensurate phase exists, wherein the order parameter acquires a spatial dependence. Given the experiments on ZnTe/MnSe find such a phase (helical) with a wave vector in either the x or y direction, we assume $\beta_1 < \beta_2$ and $\beta_1 < 0$, so that the mean-field ground state is of the form $(\phi_1(x), \bar{\phi}_1(x))$ or $(\phi_2(y), \bar{\phi}_2(y))$. The effective free energy is then

$$\mathrm{F} = \int dx \Big(r_1(\tau)\,[\phi_1^2 + \bar{\phi}_1^2] + \beta_1(\tau)\,[\phi_{1,x}^2 + \bar{\phi}_{1,x}^2] + \gamma(\phi_{1,xx}^2 + \bar{\phi}_{1,xx}^2) + (u+v)\,(\phi_1^2 + \bar{\phi}_1^2)^2 + w\phi_1^2\bar{\phi}_1^2 \Big), \tag{9}$$

which is equivalent to the free energy of a tetragonal crystal with an easy plane of magnetization.

The thermodynamic properties of such a system were studied by Michelson.[13] Although the mean-field ground state cannot be found exactly, one can obtain an asymptotic solution[13] valid near the paramagnetic-antiferromagnetic phase line. For $w < 4(u+v)$ one finds the helical state seen in ZnTe/MnSe described by $\phi_1 = A\cos(k_0 x)$ and $\bar{\phi}_1 = \pm A\sin(k_0 x)$ with $k_0 = (-\beta_1/2\gamma)^{1/2}$ [for $w > 4(u+v)$ the solution is a transverse sinusoid[13]]. We find that the wavelength λ of the physical helix is related to the wave vector of the order parameter modulation, k_0, by $\lambda = 2a/[1 - (k_0 a/\pi)]$ where a is the lattice constant in the growth plane. In addition one finds that the phase line between the helical state and the type-III state $(\phi_1, \bar{\phi}_1)$, $(\phi_2, \bar{\phi}_2)$ is first order for $w \neq 0$ and

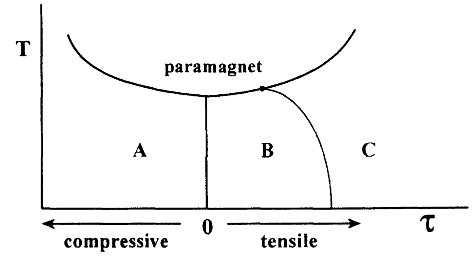

FIG. 2. Phase diagram in the temperature-strain plane. Phase A is a type-III antiferromagnet with doubled unit cell along the growth (001) direction; phase B is a type-III antiferromagnet with doubled unit cell along a cubic axis in the growth plane; phase C is a helical phase with wave vector along a cubic axis in the growth plane. The triple point joining the paramagnetic, helical, and growth plane type-III phases is a Lifshitz point.

second order for $w = 0$. Thus as one crosses this phase boundary, the wavelength of the helix, λ, will change discontinuously for $w \neq 0$ and continuously for $w = 0$. In particular, for $w \neq 0$, one finds that $\Delta k_0 \propto r_1^{1/4}$ where Δk_0 is the magnitude of the discontinuity. It is then clear that the triple point connecting the paramagnetic-(type-III) phase line, the paramagnetic-helical phase line, and the helical-(type-III) phase line (see Fig. 2) is a Lifshitz point[14] characterized by the fact that $k_0 \to 0$ continuously as this point is approached.

The above predictions of the Landau-Ginzburg theory are compactly summarized in Fig. 2 which is the phase diagram for this new class of semiconductor antiferromagnets as a function of the built-in strain field. As mentioned in the introductory remarks, experiments have seen the type-III phase oriented along the growth (001) direction as well as the helical phase. Systems with weaker tensile strain than ZnTe/MnSe should allow access to our predicted type-III phase with doubled unit cell along one of the cubic axes in the growth plane and should also permit a study of the nature of the commensurate-incommensurate phase boundary.

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