Lockin to Weak Ferromagnetism in TbNi$_2$B$_2$C and ErNi$_2$B$_2$C

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This article describes a model in which ferromagnetism necessarily accompanies a spin-density-wave lockin transition in the boron carbide structure provided the commensurate phase wave vector satisfies $Q = (m/n)a^*$ with $m$ even and $n$ odd. The results account for the magnetic properties of TbNi$_2$B$_2$C, and are also possibly relevant also for those of ErNi$_2$B$_2$C.

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INTRODUCTION

The material TbNi$_2$B$_2$C is one of a number of rare earth boron carbide materials that display a fascinating variety of magnetic and superconducting phases. This particular material has a phase transition to a spin-density-wave phase at $T_N \approx 15$ K followed by the appearance of weak ferromagnetism at $T_{WFM} \approx 8$ K. A neutron diffraction study of the magnetic structure showed that the wave vector of the spin-density wave decreased with decreasing temperature in the spin-density wave phase until the transition temperature to the weak ferromagnetic phase was reached, at which point the spin-density-wave wave vector became approximately constant at $Q = 0.545a^*$. This behavior is characteristic of a continuous lockin transition to a commensurate phase occurring at $T_{WFM}$, the commensurate wave vector being $Q = 0.545a^* = (6/11)a^*$. This value for the locked in wave vector has been confirmed by recent high resolution magnetic X-ray scattering studies. There is some hysteresis observed at $T_{WFM}$, which is consistent with the idea that hysteresis can occur at a continuous commensurate-incommensurate transition.

Similarly, in ErNi$_2$B$_2$C, there is a transition to a spin density wave phase, followed apparently by a transition to a weak ferromagnetic phase as the temperature is lowered. The study of the weak ferromagnetism is made more complicated in this case because the magnetic transitions occur within a superconducting phase. Here also a lockin phase transition appears to accompany the transition to weak ferromagnetism. (The evidence for the lockin is that the spin-density-wave wave vector appears to become more or less independent of temperature below $T_{WFM}$). The lockin wave vector is close to $Q = 0.548a^* = (17/31)a^*$ according to and close to $Q = 0.550a^* = (11/20)a^*$ according to .

A curious aspect of the behavior of both TbNi$_2$B$_2$C and ErNi$_2$B$_2$C is that the apparent incommensurate-commensurate transition and the transition to weak ferromagnetism seem to occur at the same temperature. It is possible for two independent second-order phase transitions to occur at nearly the same temperature if the interactions in the system happen to have just the right values so that this happens. However, this is very unusual, and to have it happen in two different materials is even more unusual. This suggests that the lockin transition and the transition to weak ferromagnetism are not independent, but have a common origin. The purpose of this article is to describe a model in which weak ferromagnetism necessarily occurs simultaneously with a lockin transition in a spin-density-wave state, and that could therefore be relevant to the magnetic behavior of TbNi$_2$B$_2$C and ErNi$_2$B$_2$C. The model described here is related to that proposed in .

One of the results derived below is that weak ferromagnetism necessarily accompanies the lockin transition in the spin-density wave states of TbNi$_2$B$_2$C and ErNi$_2$B$_2$C only if the commensurate wave vector is of the form $Q = (m/n)a^*$, with $m$ an even integer and $n$ an odd integer. An independent microscopic calculation of the nature of commensurate phases of ErNi$_2$B$_2$C, including the possible occurrence of ferromagnetism, has been carried out in for a number of different commensurate wave vectors. These results for specific wave vectors are consistent with the general rule concerning the occurrence of ferromagnetism developed in this article.

The work described below makes use of a Ginzburg-Landau type of analysis. This is a useful way of obtaining results of general validity which depend on the symmetry of the problem. Thus, the approach is complementary to the microscopic type of calculation described in .

THE SPIN-DENSITY-WAVE PHASE

A model specifically related to TbNi$_2$B$_2$C will now be described. Above the spin-density-wave transition temperature the material TbNi$_2$B$_2$C has a body-centered tetragonal structure with space-group symmetry I4/mmm. The spin-density wave in TbNi$_2$B$_2$C is as-
sumed to be represented by the equation

$$S(\mathbf{r}) = \sum_{\mathbf{R}, \mathbf{\hat{B}}, n, i} [S_{n,i} e^{in\mathbf{Q}\cdot\mathbf{r}} + S_{n,i}^* e^{-in\mathbf{Q}\cdot\mathbf{r}}] \delta(\mathbf{r} - \mathbf{R} - \mathbf{\hat{B}}).$$

(1)

Here $\mathbf{R}$ labels the unit cell, $\mathbf{\hat{B}} = 0, \frac{1}{2}(a + b + c)$ labels the magnetic ions in the crystallographic unit cell (at the corner and body center positions), $n$ labels the harmonic, and $i = a, b, c$ labels the symmetry-equivalent wave vectors $\mathbf{Q}_a = (Q, 0, 0)$ and $\mathbf{Q}_b = (0, Q, 0)$, where $Q \approx 0.55a^\ast$. Since the spin-density wave in TbNiz2B2C is thought to be longitudinally polarized[12], i.e. $S_1$ is parallel to $\mathbf{Q}_i$ (see however [13]), the nonzero components of the primary order parameter $S_{1,i}$ (describing the first harmonic) are $S_{1,a,x}$ and $S_{1,b,y}$, which will be denoted below simply by $S_x$ and $S_y$, respectively. It is important to remember, however, that $S_x$ and $S_y$ are complex numbers giving the amplitude and phase of the spin-density waves with wave vectors $\mathbf{Q}_a$ and $\mathbf{Q}_b$, respectively. (A similar model relevant to ErNiz2B2C would involve $S_{1,a,y}$ and $S_{1,b,x}$ since the spin-density wave in this material is transversely polarized[1].)

Given the above description of the spin-density wave, the Landau free energy describing a second-order phase transition to the spin-density-wave phase has the form

$$F = A(|S_x|^2 + |S_y|^2) + B(|S_x|^2 + |S_y|^2)^2 + C|S_x|^2|S_y|^2,$$

(2)

where $A \propto (T - T_N)$, with $T_N$ being the transition temperature. The constants $B$ and $C$ must satisfy $B > 0$ and $4B + C > 0$ for stability. If $C < 0$ a tetragonal double-$Q$ spin-density wave phase with $|S_x| = |S_y| \neq 0$ occurs, while if $C > 0$ an orthorhombic single-$Q$ phase with either $|S_x| \neq 0$ or $|S_y| \neq 0$, but not both, occurs. Below $T_N$ in TbNiz2B2C, the structure becomes orthorhombic[1, 13] so it is clearly the single-$Q$ state that occurs in this material. A given material normally contains both single-$Q$ domains (in spatially different regions of the crystal). In what follows we study explicitly only the properties of the single-$Q$ domain characterized by a non-zero $S_x$; the properties of the domain characterized by $S_y$ follow immediately by rotating by $\pi/2$ about the $c$-axis.

Spin-density-wave harmonics of order $2n + 1$ with $n$ an integer are induced by contributions to the free energy proportional to $S_{2n+1,x}(S_x)^{2n+1}$ and $S_{2n+1,z}(S_z)^{2n+1}$. Since this term is linear in $S_{2n+1,x}$, the value of $S_{2n+1,x}$ which minimizes the free energy must be non-zero. There are other terms in the free energy that are also linear in $S_{2n+1,x}$ and which also contribute to the induction of the $(2n + 1)^{th}$ harmonic. However, since we are only interested here in demonstrating that the symmetry of the problem requires the existence of the $(2n + 1)^{th}$ harmonic (and do not attempt to evaluate its magnitude) it is sufficient to consider only one example of the terms linear in $S_{2n+1,x}$. Similarly, throughout this article, only one example of the type of term necessary for our purpose will be given. The wave vector associated with $S_{2n+1,x}$ is $(2n + 1)Q_x$. Similarly, charge-density-wave (or, equivalently, longitudinal lattice-displacement-wave) harmonics of order $2n$ and complex charge-density-wave amplitude $\rho_2$ are induced by terms in the free energy proportional to $\rho_2(S_x S_y^{2n-1})^r + \rho_2^*(S_z S_y^{2n-1})$. The conclusions of this paragraph are well known from studies of the spin-density-wave state of chromium.[15]

**LOCKIN TO WEAK FERROMAGNETISM**

Consider a single-$Q$ spin-density-wave phase characterized by a nonzero $S_x$, as described above. A lock-in transition is obtained by adding so-called lock-in terms proportional to $S_x^r$ and $(S_x^r)^p$ ($p$ is an integer) to this free energy. Since the free energy must be invariant with respect to a translation of the spin-density-wave by the displacement $\frac{1}{2}(a + b + c)$, these terms are allowed only for $Q = 2(m/p)a^\ast$, where $m$ (as well as $p$) is an integer. (The factor 2 in the expression $Q = 2(m/p)a^\ast$ appears because the unit cell is body centered.) Thus if $Q$ is close to satisfying this commensurability condition for some $m$, it sometimes pays the system to adjust its $Q$ to exactly satisfy it, so as to be able gain energy from the presence of the lock-in terms in the free energy. If $p$ is odd, however, terms proportional to $S_x^p$ can not exist by themselves in the free energy since they are not invariant with respect to time reversal. The solution to this problem for odd $p$ is to consider terms such as $S_x^p S_{0x}$,[10] where $S_{0x}$ is the ferromagnetic component of the spin density, which are invariant under time reversal, and since $Q = 2(m/p)a^\ast$, are also invariant under body-centered Bravais lattice translations. Note that symmetry requires the ferromagnetic moment to be in the same direction as the spin-density-wave polarization vector [in this case both are directed along the x (or a) axis]. Thus, for $p$ odd, the terms stabilizing a lock-in transition are

$$F_{lockin} = A_0 |S_x|^2 + B_0 |S_x|^p \cos(p\phi) S_{0x},$$

(3)

where $A_0 > 0$ is expected, and $S_x = |S_x| \exp(i\phi)$. These terms must be added to the original free energy given above. Because the second term in $F_{lockin}$ is linear in $S_{0x}$, the free energy has its minimum at a non zero value of $S_{0x}$ in the locked in phase, and weak ferromagnetism is thereby necessarily induced at an odd-$p$ lock-in phase transition. For a continuous lock-in transition corresponding to even $p$, ferromagnetism will not automatically be induced, and thus would not be expected to occur simultaneously with the lock-in. It should be noted that the lock-in terms of Eq. 3 exist in the commensurate phase free energy independently of whether the transition to the commensurate phase is first or second order. Thus, ferromagnetism necessarily occurs in any commensurate phase with $Q = 2(m/p)a^\ast$ where $p$ is odd.
If $B_{0} > 0$ is assumed, the minimum of $F_{\text{lockin}}$ occurs for $p\phi = 2r\pi$ and $S_{0x} < 0$, or for $p\phi = (2r+1)\pi$ and $S_{0x} > 0$, where $r$ is an integer. Assuming $p = 11$, as is appropriate for TbNi$_2$B$_2$C (since $Q = 2(m/p)a^{*} = (6/11)a^{*}$ in the locked in phase), and taking $r = 1, 2, ...11$ in $p\phi = 2r\pi$, one finds eleven distinct commensurate domains, each with $S_{0x} < 0$. The new commensurate unit cell has $a$-axis length $11a$, and the eleven distinct domains are obtained by starting with one of them, and then producing the others by translating the spin-density-wave structure by the original lattice constant $a$ ten times. Translating each of these different commensurate domains by the displacement $\frac{1}{2}(a + b + c)$ gives a corresponding domain with all the spin directions (including the ferromagnetic component $S_{0x}$) reversed in sign. An incommensurate phase can be viewed as a sequence of commensurate domains separated by domain walls, which are often called discommensurations. In a continuous incommensurate to commensurate transition, the domain walls are swept out to the sample boundary as the temperature approaches the transition temperature. In a commensurate to incommensurate transition, domain walls must be nucleated in the commensurate phase to form the incommensurate phase. Pinning of the domain walls, as well as the energy barrier required to nucleate a domain wall, contribute to hysteresis effects for both first order and second order transitions. The lockin transition will be first order if the interaction between domain walls is repulsive (e.g. as in \cite{13}). In a continuous incommensurate to commensurate transition, the domain walls are swept out to the sample boundary as the temperature approaches the transition temperature.

**MAGNETIC EVEN-ORDER HARMONICS**

Now consider a magnetic state in which there is a ferromagnetic moment $S_{0x}$ and a spin-density-wave (which may be either commensurate or incommensurate for the purposes of this section) characterized by the complex amplitude $S_{x}$. Spin-density-wave harmonics of (even) order $2n$ and complex amplitude $S_{2n,x}$ are induced in the combined ferromagnetic and spin-density-wave states by terms in the free energy given by

$$F_{\text{even}} = S_{0x}[S_{2n,x}(S_{x}^{*})^{2n} + S_{2n,x}^{*}S_{x}^{2n}].$$

Since this contribution to the free energy is linear in $S_{2n,x}$, magnetic even-order harmonics are necessarily present in a state containing both a spin-density wave and ferromagnetism. Conversely, a state containing both a spin-density wave and one of its even harmonics, necessarily contains a ferromagnetic moment.

It has been found that, in ErNi$_2$B$_2$C, even-order magnetic harmonics are present below $T_{WF\text{FM}}$. This very unusual result is explained by the discussion of the preceding paragraph.

**DISCUSSION**

The model described above appears to account well for the simultaneous continuous lockin and weak ferromagnetic phase transitions which occur at approximately 8K in TbNi$_2$B$_2$C. In particular, the commensurate wave vector $Q = (6/11)a^{*}$ is of the form $Q = (m/n)a^{*}$ with $m$ even and $n$ odd. Thus, the continuous lockin transition is necessarily accompanied by the presence of weak ferromagnetism. The direction of the induced weak ferromagnetic moment is required by symmetry to be in the same (or opposite) direction as the polarization vector of the spin-density wave, which is consistent with the observation that the induced ferromagnetic moment is in the basal plane. If there are ferromagnetic domains present, the lowest free energy type of domain structure is expected to have domains of different ferromagnetic spin orientations associated with commensurate regions shifted relative to one another by the displacement $\frac{1}{2}(a + b + c)$. Even harmonics have been observed very recently \cite{13} both above and below the transition temperature to weak ferromagnetism, and it would be of interest to determine the magnetic character of these satellites as a function of temperature. (In the model described above, even harmonics of magnetic character should occur only in the weakly ferromagnetic phase). Also, \cite{13} has presented evidence that the spin-density-wave phase of TbNi$_2$B$_2$C may not be perfectly longitudinal, at least at some temperatures. A reorientation of the spin polarization should not affect the general arguments of this article concerning the lockin to weak ferromagnetism.

Some features of the weakly ferromagnetic phase observed in ErNi$_2$B$_2$C are accounted for by the above work. For example, the even harmonics which occur in the ferromagnetic phase \cite{8, 9}, and which have the unusual characteristic of being magnetic, are shown to necessarily ac-
company the presence of ferromagnetism combined with a spin-density wave.

However, the observed magnitude $Q$ of the locked in wave vector in ErNi$_2$B$_2$C does not obviously satisfy the requirement $Q = (m/p)a^*$, with $m$ even and $p$ odd, derived above for continuous phase transition to a locked phase that is also ferromagnetic. The commensurate wave vector found in $\text{[5]}$ is $Q = 0.548a^* = (17/31)a^*$, whereas the commensurate wave vector found in $\text{[7]}$ is $Q = 0.550a^* = (11/20)a^*$. It should be noted, however, that the value $Q = (28/51)a^* = 0.549a^*$ $\text{[1]}$, and the value $Q = (16/29)a^* = 0.5517a^*$ would both produce a locked in weakly ferromagnetic phase as described above.

The question now arises as to what is the significance of variations of wave vector of the order of $\delta Q \approx 0.001$. The transition from an incommensurate to a commensurate phase is brought about by sweeping the domain walls between different commensurate regions out of the crystal. If it turns out that the pinning of the domain walls does not allow this process to be completed, so that there remain domain walls separated on average by a distance 1000$a$, then this will cause a change in the observed $Q$ of order $\delta Q \approx 0.001a^*$. Clearly, the interpretation of the locked in phase of TbNi$_2$B$_2$C, where the commensurate wave vector involves smaller integers and the absolute precision of its measurement is less significant, is more straightforward than the interpretation of the locked in phase of ErNi$_2$B$_2$C.

It is also of interest to compare the results of this article with those of the microscopic mean field calculation of $\text{[1]}$. Ref. $\text{[1]}$ studied a number of commensurate spin-density-wave structures having $Q$ close to $Q = 0.55a^*$ for a detailed model of ErNi$_2$B$_2$C. The commensurate structures of that article having $Q = (6/11)a^*$ and $Q = (4/7)a^*$ both had ferromagnetic moments, whereas the structure with $Q = (5/9)a^*$ had no ferromagnetic moment. These results are consistent with the general rule stated above that weak ferromagnetism is necessarily present in a commensurate phase with $Q$ of the form $Q = (m/n)a^*$ where $m$ is even and $n$ is odd. On the other hand, Ref. $\text{[1]}$ finds a first order phase transition within the $Q = (11/20)a^*$ commensurate phase from a non-ferromagnetic to a weakly ferromagnetic state. This does not contradict the general rule given above, since the rule does not require a commensurate phase of this wave vector to be ferromagnetic, nor does it prevent a particular choice of the model interactions from yielding ferromagnetism at this wave vector. In this case, however, there is no reason for the ferromagnetism to occur simultaneously with the lockin (and $\text{[1]}$ does not argue that it does). Finally, it is interesting to note that certain aspects of the neutron scattering data of $\text{[1]}$ were explained in $\text{[1]}$ by invoking a commensurate phase with $Q = (28/51)a^*$. Interestingly, the symmetry arguments of this article require that such a phase is weakly ferromagnetic. (Ref. $\text{[1]}$ is silent on the question of the ferromagnetism of their $Q = (28/51)a^*$ phase.)

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