Spin order and entropy in antiferromagnetic films subjected to magnetic fields

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Abstract. Using systematic effective field theory, we explore the properties of antiferromagnetic films subjected to magnetic and staggered fields that are either mutually aligned or mutually orthogonal. We provide low-temperature series for the entropy density in either case up to two-loop order. Invoking staggered, uniform and sublattice magnetizations of the bipartite antiferromagnet, we investigate the subtle order–disorder phenomena in the spin arrangement, induced by temperature, magnetic and staggered fields—some of which are quite counterintuitive. In the figures we focus on the spin-1/2 square-lattice antiferromagnet, but our results are valid for any other bipartite two-dimensional lattice.

Keywords: rigorous results in statistical mechanics

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1. Introduction

The investigation of order–disorder phenomena in low-dimensional quantum antiferromagnets represents an important ongoing topic in theoretical and experimental condensed matter research. Out of the various counterintuitive effects discovered over the years, one phenomenon concerns the increase of the uniform magnetization with temperature—taking place not only in two-dimensional systems but also in antiferromagnetic spin chains and ladders. For example, according to reference [1], the magnetization of the critical spin-$\frac{1}{2}$ antiferromagnetic chain initially increases as temperature rises. The effect has been confirmed experimentally (see, e.g., reference [2]). Then the behavior of the spin-$1$ antiferromagnetic chain is even more intriguing: the magnetization first drops as temperature rises, goes through a minimum, and then grows. Similar behavior of the uniform magnetization has been reported for the spin-$\frac{1}{2}$ two-leg spin ladder in references [3, 4].

The initial increase of the magnetization with temperature has furthermore been documented for the spin-$\frac{1}{2}$ square-lattice antiferromagnet that presents this counterintuitive phenomenon in very weak magnetic fields, according to references [5, 6] based on Monte Carlo simulations, and reference [7] relying on exact diagonalization. Apparently, the literature on this counterintuitive effect occurring in two-dimensional systems
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is scarce. Moreover, the aforementioned references only include an external magnetic field, but do not take into account a staggered field.

In the present study, using systematic magnon effective field theory, we address two-dimensional quantum Heisenberg antiferromagnets, with the intention to explore the non-monotonous behavior of the magnetization with temperature in a more general setting. To this end we consider bipartite two-dimensional antiferromagnets subjected to magnetic and staggered fields that are either mutually parallel or mutually orthogonal. We are not aware of any theoretical or Monte Carlo based references that have explored counterintuitive effects for this more general setting that allows for two different types of external fields.

Our motivation for including the staggered field—and for choosing these two specific configurations of fields—is that they represent the two most studied examples of collinear antiferromagnetic spin arrangement. It should be emphasized that the relevance of the staggered field lies in the fact that it takes the role of an ‘effective’ anisotropy field that mimics the magnetic anisotropy. It hence determines the direction of the ‘easy axis’ along which the spontaneous staggered magnetization at $T = 0$ aligns (see, e.g., reference [8]). The direction of the external magnetic field is then chosen with respect to this ground state configuration—here we consider magnetic fields that are either aligned with, or, orthogonal to, the ‘easy axis’. While we address two-dimensional systems, it should be noted that experimental (three-dimensional) samples often behave as quasi two-dimensional: the essential physics takes place in a plane and the interactions in the direction transverse to it are weak. Still, these interactions may give rise to a preferred axis of spin antialignment—as a consequence, an easy axis also emerges in (quasi) two-dimensional systems.

From a more general perspective, the present work is part of an ongoing program the aim of which it is to systematically analyze the thermodynamic properties of antiferromagnetic systems using magnon effective field theory. While three-dimensional antiferromagnets have been discussed within this framework in references [9–12]—and more recently in references [13–16]—here we continue exploring antiferromagnetic films.

Earlier effective field theory studies on antiferromagnetic films include references [9, 17–20]. More recent articles are references [21–23]. In the present investigation we consider antiferromagnetic films subjected to magnetic and staggered fields that are either mutually aligned or mutually orthogonal. A thorough and rigorous analysis of this situation—in particular at the two-loop level that is our objective—still appears to be lacking which motivates our work. New analytic results comprise low-temperature series for the entropy density and the sublattice magnetizations of the bipartite two-dimensional antiferromagnet up to two-loop order. As it turns out, these low-temperature representations are parameter free in the sense that they do not involve next-to-leading order (NLO) effective constants, but solely depend on the spin stiffness and the zero-temperature staggered magnetization, i.e., the order parameter.

1Canted antiferromagnetic phases where ‘up-spins’ and ‘down-spins’ are not oriented antiparallel do not fall into this category and are hence outside the scope of the present analysis.
It should be pointed out that the path we pursue here—based on the systematic effective Lagrangian method—is not the conventional choice to address antiferromagnetic systems. Rather, more standard techniques to study antiferromagnetic films are modified spin-wave theory [24–32], numerical simulations [5, 6, 33], exact diagonalization and other approaches [7, 34–42].

Our main theme is the exploration of order–disorder phenomena in the spin arrangement that are induced by temperature and by the magnetic and staggered fields. To this end, referring specifically to the spin-$\frac{1}{2}$ square-lattice antiferromagnet, we provide plots for the entropy density, as well as the uniform, staggered and sublattice magnetizations.

Before delving into the effects at finite temperature, we first survey the situation at $T = 0$. In both cases—mutually parallel and orthogonal fields—staggered, uniform and sublattice magnetizations grow when the magnetic or the staggered field become stronger, which can be understood by suppression of quantum fluctuations.

We then analyze the behavior of the same observables at finite—but first fixed—temperature by varying magnetic and staggered field strength. Regarding the entropy density we identify two opposite tendencies: (1) if the staggered field becomes stronger, the entropy decreases, and (2) if the magnetic field becomes stronger, the entropy increases. Naively, the staggered field enhances spin order by enforcing antialignment of the spins, whereas the magnetic field perturbs the antialigned array of spins. These observations that apply to mutually parallel and mutually orthogonal fields alike, are confirmed by the finite-temperature behavior of the staggered, uniform and sublattice magnetizations.

In both configurations of external fields a finite-temperature uniform magnetization is induced that initially even grows as temperature rises—this outcome is rather counterintuitive. While all effects have been illustrated by the spin-$\frac{1}{2}$ square-lattice antiferromagnet, any other bipartite two-dimensional antiferromagnet behaves in a qualitatively similar way. Our results and the observed order–disorder phenomena are universal in this sense—microscopic details are merely encoded in the concrete values the spin stiffness and zero-temperature staggered magnetization.

The paper is organized as follows. In section 2, after a few general remarks on antiferromagnetic films in magnetic fields aligned with the order parameter, we derive the two-loop representation for the entropy density and examine its behavior in external fields. In the same section we then investigate in detail how temperature, magnetic and staggered fields influence spin order by providing various figures that include uniform, staggered and sublattice magnetizations for the spin-$\frac{1}{2}$ square-lattice antiferromagnet. Some effects are quite counterintuitive. Along the same lines—and by providing analogous figures—in section 3, we analyze antiferromagnetic films subjected to magnetic and staggered fields that are mutually orthogonal, and point out similarities and differences with respect to the configuration of mutually parallel external fields. In section 4 we conclude. Finally, in appendix A—for self-consistency—we list the relevant kinematical functions for antiferromagnets in mutually orthogonal external fields.

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2. Antiferromagnetic films in mutually parallel staggered and magnetic fields

2.1. Preliminaries

Within the microscopic point of view, antiferromagnetic films are captured by the quantum Heisenberg model that incorporates an external magnetic ($\vec{H}$) and a staggered ($\vec{H}_s$) field as

$$\mathcal{H} = -J \sum_{n.n.} \vec{S}_m \cdot \vec{S}_n - \sum_n \vec{S}_n \cdot \vec{H} - \sum_n (-1)^n \vec{S}_n \cdot \vec{H}_s, \quad J < 0, \quad J = \text{const.} \quad (2.1)$$

The notation ‘$n.n.$’ indicates that the sum refers to nearest neighbor spins only. While our focus in the numerical analysis is square-lattice geometry, we allow for a general bipartite lattice.

In the absence of external fields, two degenerate spin-wave branches (magnon modes) emerge, obeying the dispersion law

$$\omega(\vec{k}) = v|\vec{k}| + O(\vec{k}^3), \quad \vec{k} = (k_1, k_2). \quad (2.2)$$

The quantity $v$ is the spin-wave velocity. The specific configuration of external fields in the present section is

$$\vec{H} = (H, 0, 0), \quad \vec{H}_s = (H_s, 0, 0), \quad H, H_s > 0, \quad (2.3)$$

i.e., magnetic and staggered fields are aligned, and they both point into the direction of the order parameter (staggered magnetization at $T = 0$). In presence of these fields, the magnon dispersion relations exhibit an energy gap and their degeneracy is lifted$^2$,

$$\omega_+ = \sqrt{\vec{k}^2 + \frac{M_s H_s^2}{\rho_s}} + H, \quad \omega_- = \sqrt{\vec{k}^2 + \frac{M_s H_s^2}{\rho_s}} - H. \quad (2.4)$$

The leading-order effective constants that appear in the dispersion relations are $M_s$ (staggered magnetization at $T = 0$ and infinite volume) as well as $\rho_s$ (spin stiffness).

An important point to emphasize is that the lower spin-wave excitation $\omega_-$ acquires negative values, unless the stability criterion

$$H_s > \frac{\rho_s}{M_s} H^2 \quad (2.5)$$

is met. The reason why the system becomes unstable is because in the situation where magnetic and staggered fields are parallel, we have a conflicting situation. In general, the staggered field wants to align the order parameter in its own direction, whereas the magnetic field wants to orient the order parameter in a plane transverse to its own direction. In the present configuration of parallel staggered and magnetic fields, the two effects therefore compete. In particular, if the condition (2.5) is not satisfied

$^2$Note that in equation (2.4)—much like in subsequent effective field theory expressions—the spin-wave velocity $v$ is set to one.
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and the magnetic field becomes too strong, a spin–flop transition occurs: the staggered magnetization vector changes its orientation and a new configuration orthogonal to the external magnetic field is realized. Throughout this section referring to mutually parallel magnetic and staggered fields, we assume the stability criterion is satisfied, such that a spin–flop transition does not occur.

The following discussion of entropy and spin order relies on the representation of the free energy density derived in reference [22]. The interested reader is invited to consult this reference for technical details regarding the evaluation of the partition function. Here we merely quote the final renormalized two-loop free energy density of antiferromagnetic films subjected to mutually parallel staggered and magnetic fields (as defined by equation (2.3)),

\[
\hat{g}_0 = T^3 \int_0^\infty d\lambda \lambda^{-5/2} e^{-\lambda m^2/4t^2} \left\{ \sqrt{\lambda} \theta_3 \left( \frac{m_H \lambda}{2t}, e^{-\pi \lambda} \right) e^{m_H \lambda / 4t^2} - 1 \right\} = T^3 \hat{h}_0, \\
\hat{g}_1 = \frac{T}{4\pi} \int_0^\infty d\lambda \lambda^{-3/2} e^{-\lambda m^2/4t^2} \left\{ \sqrt{\lambda} \theta_3 \left( \frac{m_H \lambda}{2t}, e^{-\pi \lambda} \right) e^{m_H \lambda / 4t^2} - 1 \right\} = T \hat{h}_1, \\
\hat{g}_2 = \frac{1}{16\pi^2 T} \int_0^\infty d\lambda \lambda^{-1/2} e^{-\lambda m^2/4t^2} \left\{ \sqrt{\lambda} \theta_3 \left( \frac{m_H \lambda}{2t}, e^{-\pi \lambda} \right) e^{m_H \lambda / 4t^2} - 1 \right\} \equiv \frac{\hat{h}_2}{T}. 
\]  

These quantities involve the Jacobi theta function

\[
\theta_3(u, q) = 1 + 2 \sum_{n=1}^\infty q^n \cos(2nu),
\]  

and depend on three dimensionless parameters,

\[
m \equiv \frac{\sqrt{M_s H_s}}{2\pi \rho_s^{3/2}}, \quad m_H \equiv \frac{H}{2\pi \rho_s}, \quad t \equiv \frac{T}{2\pi \rho_s}. 
\]  

Since the common denominator,

\[
2\pi \rho_s \approx J, 
\]  

\[3\] The function \(\hat{g}_2\) does not show up in the free energy density, but it is relevant in the staggered and sublattice magnetizations (see below).
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approximately corresponds to the exchange coupling $J$ that sets the microscopic scale, the parameters $m, m_H, t$ ought to be small as the effective field theory describes the physics in the low-energy sector.

Apart from the leading-order effective constants $M_s$ and $\rho_s$ in the free energy density, equation (2.6), in the zero-temperature piece $z_0$, two additional effective constants—$k_2$ and $k_3$—show up: so-called NLO effective constants. All these constants depend on the lattice geometry. Concrete numerical values for $\rho_s$ and $M_s$, as well as for the spin-wave velocity $v$, are available for the spin-$\frac{1}{2}$ square-lattice antiferromagnet (see reference [43]),

\[ \rho_s = 0.1808(4)J, \quad M_s = 0.30743(1)/a^2, \quad v = 1.6585(10)J a, \quad (2.12) \]

and for the spin-$\frac{1}{2}$ honeycomb-lattice antiferromagnet (see reference [44]),

\[ \rho_s = 0.102(2)J, \quad \tilde{M}_s = 0.2688(3), \quad v = 1.297(16)J a, \quad (2.13) \]

with

\[ \tilde{M}_s = \frac{3\sqrt{3}}{4} M_s a^2. \quad (2.14) \]

Furthermore, for the spin-$\frac{1}{2}$ square-lattice antiferromagnet, the relevant combination $k_2 + k_3$ of NLO effective constants that matters at zero temperature, has been determined in reference [43] as

\[ \frac{k_2 + k_3}{v^2} = \frac{-0.0037}{2\rho_s} = \frac{-0.0102}{J}. \quad (2.15) \]

It should be emphasized that NLO effective constants ($k_2, k_3$) only show up in the (zero-temperature) vacuum energy density $z_0$. The finite-temperature physics of the system, up to two-loop order, is thus fully described in terms of the leading-order effective constants $\rho_s$ and $M_s$. The only difference between, e.g., square- and honeycomb-lattice antiferromagnets consists in the concrete numerical values of $\rho_s$ and $M_s$.

The effective field theory is tied to the low-energy sector where the parameters $m, m_H, t$—defined in equation (2.10)—are small. In the present investigation we consider values up to

\[ m, m_H, t \lesssim 0.4. \quad (2.16) \]

A further constraint is imposed by the stability criterion, equation (2.5), which we implement by choosing the parameter region of external fields as

\[ m > m_H + \delta, \quad \delta = 0.1. \quad (2.17) \]

From a physical point of view, the staggered field is always present in a real magnetic sample because anisotropies or other effects give rise to an ‘easy axis’ that selects the direction of the staggered magnetization order parameter. In contrast to the staggered field whose numerical value is fixed in a concrete antiferromagnetic sample, the external magnetic field can be tuned in an experiment. The stability criterion then just means that the strength of the magnetic field should not exceed the value determined by equation (2.17). It guarantees that we are away from the spin–flop transition that

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would lead to another ground state configuration not covered by the present analysis. Therefore, the stability criterion is not a severe limitation of our approach—it just defines in which parameter domain the configuration of mutually parallel staggered and magnetic fields is stable. Experimentally, as witnessed by reference [45], this situation is for example realized in K₂MnF₄ that has been probed with mutually parallel magnetic and staggered fields.

2.2. Entropy density

In this subsection we derive the low-temperature representation for the two-loop entropy density and study how it is affected by the external fields. The entropy density \( s \) of antiferromagnetic films subjected to mutually parallel staggered and magnetic fields is readily obtained from the finite-temperature part of the free energy density, \( z - z_0 \), via

\[
s = \frac{d}{dT} (z_0 - z), \tag{2.18}
\]

with the result

\[
s(t, m, m_H) = s_1 T^2 + s_2 T^3 + O(T^4). \tag{2.19}
\]

The respective coefficients are

\[
s_1 = \frac{t^2}{2} \frac{d^2\hat{h}_{-1}}{dm_H^2} + \frac{mm_Ht^2}{4} \frac{d^3\hat{h}_{-1}}{dm_H^3} - m_H \left( 1 + \frac{m}{2} \right) \frac{d\hat{h}_0}{dm_H}
- mm_H^2 \frac{d^2\hat{h}_0}{dm_H^2} + \frac{mm_H^3}{t^2} \frac{d\hat{h}_1}{dm_H}, \tag{2.20}
\]

\[
s_2 = -\frac{m_H t^2}{2\rho_s} \frac{d^2\hat{h}_0}{dm_H^2} \frac{d\hat{h}_0}{dm_H} + \frac{m_H^2 t^2}{\rho_s} \frac{d\hat{h}_1}{dm_H} \frac{d\hat{h}_0}{dm_H} - \frac{m_H t^2}{2\rho_s} \frac{d\hat{h}_1}{dm_H} \frac{d^3\hat{h}_{-1}}{dm_H^3}
+ \frac{2m_H^2 \hat{h}_1}{\rho_s} \frac{d^2\hat{h}_0}{dm_H^2} - \frac{2m_H^3 \hat{h}_1}{\rho_s} \frac{d\hat{h}_1}{dm_H} + \frac{m_H \hat{h}_1}{\rho_s} \frac{d\hat{h}_0}{dm_H}, \tag{2.21}
\]

The kinematical function \( \hat{h}_{-1} \) reads

\[
\hat{h}_{-1} = \frac{\hat{g}_{-1}}{T^5} = 4\pi \int_0^\infty d\lambda \lambda^{-7/2} e^{-\lambda m^2/4t^2} \left\{ \sqrt{\lambda} \theta_3 \left( \frac{m_H \lambda}{2t}, e^{-\pi\lambda} \right) e^{m_H \lambda/4t^2} - 1 \right\}. \tag{2.22}
\]

In the derivation of the above representation for the entropy density we have used the relation

\[
\frac{d\hat{g}_r}{dT} - \frac{1}{2T} \frac{d^2\hat{g}_{r-1}}{dH^2} - \frac{H}{T} \frac{d\hat{g}_r}{dH}, \tag{2.22}
\]
which is based on the identities

\begin{align}
    e^{-\frac{2}{\kappa^2}n e^{\mp n \beta H}} &= \frac{1}{\beta} \frac{\partial}{\partial H} \left\{ e^{-\frac{2}{\kappa^2}e^{\mp n \beta H}} \right\}, \\
    e^{-\frac{2}{\kappa^2} n^2 e^{\mp n \beta H}} &= \frac{1}{\beta^2} \frac{\partial^2}{\partial H^2} \left\{ e^{-\frac{2}{\kappa^2}e^{\mp n \beta H}} \right\}.
\end{align}

(2.23)

Recall that the kinematical functions $\tilde{g}_0, \tilde{g}_1, \tilde{g}_2$ are defined in equation (2.8).

In figure 1, for the temperatures $t = \{0.2, 0.4\}$, we depict the entropy density, i.e., the quantity

\[ s_1 T^2 + s_2 T^3, \]

as a function of magnetic ($m_H$) and staggered ($m$) field strength. Roughly, two opposite tendencies can be identified: (1) if the staggered field becomes stronger, the entropy decreases, and (2) if the magnetic field becomes stronger, the entropy increases. Naively, one would conclude that the staggered field establishes spin order by enforcing antialignment of the spins, while the magnetic field perturbs the antialigned array of spins. However, as the figure referring to the more elevated temperature $t = 0.4$ indicates, in stronger fields subtle effects emerge. To interpret these observations, let us also consider how uniform and staggered magnetization—as well as the individual sublattice magnetizations—behave when magnetic field, staggered field and temperature are varied.

2.3. Order–disorder effects

The low-temperature series for the staggered magnetization $M_s$ and the uniform magnetization $M$ have been derived in reference [22]. The staggered magnetization takes
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\[ M_s(t, m, m_H) = M_s(0, m, m_H) + \tilde{\sigma}_1 T + \tilde{\sigma}_2 T^2 + \mathcal{O}(T^3), \tag{2.25} \]

with coefficients

\[ \tilde{\sigma}_1(t, m, m_H) = \frac{M_s}{\rho_s} \hat{h}_1, \]
\[ \tilde{\sigma}_2(t, m, m_H) = M_s \left\{ \frac{m_H}{\rho_s} \frac{\partial \hat{h}_0}{\partial m_H} \frac{\partial \hat{h}_1}{\partial m_H} + \frac{m_H t}{8\pi \rho_s m} \frac{\partial \hat{h}_0}{\partial m_H} + \frac{mm_H}{4\pi \rho_s t} \hat{h}_0 \frac{\partial \hat{h}_1}{\partial m_H} \right. \]
\[ \left. - \frac{2m_H^2}{\rho_s t} \hat{h}_1 \hat{h}_2 - \frac{m_H^2}{2\pi \rho_s t^2} \hat{h}_1 - \frac{m_H^2}{2\pi \rho_s t^2} \hat{h}_1 \right\}, \tag{2.26} \]

and the uniform magnetization amounts to

\[ M(t, m, m_H) = M(0, m, m_H) + \hat{\sigma}_1 T + \hat{\sigma}_2 T^2 + \mathcal{O}(T^3), \tag{2.27} \]

with coefficients

\[ \hat{\sigma}_1(t, m, m_H) = 2\pi \rho_s t^2 \frac{\partial \hat{h}_0}{\partial m_H}, \]
\[ \hat{\sigma}_2(t, m, m_H) = -2\pi t^2 \hat{h}_1 - 2m_H^2 \frac{\partial \hat{h}_1}{\partial m_H} \frac{\partial \hat{h}_0}{\partial m_H} - 2m_H t^2 \hat{h}_1 \frac{\partial^2 \hat{h}_0}{\partial m_H^2} \]
\[ + \frac{mt}{2} \frac{\partial \hat{h}_0}{\partial m_H} + \frac{mm_H t}{2} \frac{\partial^2 \hat{h}_0}{\partial m_H^2} + 4\pi m_H \frac{\hat{h}_1}{2} + 4\pi m_H^2 \frac{\hat{h}_1}{2} + 4\pi m_H^2 \frac{\hat{h}_1}{2} \frac{\partial \hat{h}_1}{\partial m_H} \]
\[ - \frac{2mm_H^2}{t} \hat{h}_1 - \frac{mm_H^2}{t} \frac{\partial \hat{h}_1}{\partial m_H}. \tag{2.28} \]

Although the discussion below mainly concerns effects caused by finite temperature, for completeness and clarity we also quote the results for the staggered and uniform magnetization at zero temperature. These are

\[ \frac{M_s(0, m, m_H)}{M_s} = 1 + \frac{m}{2} + \frac{m_H^2}{4} + 8\pi^2 \rho_s (k_2 + k_3) m^2, \tag{2.29} \]

and

\[ M(0, m, m_H) = \pi \rho_s^2 m^2 m_H, \tag{2.30} \]

respectively. The numerical value of the combination \( k_2 + k_3 \) of NLO effective constants for the spin-\( \frac{1}{2} \) square-lattice antiferromagnet is given in equation (2.15). While our effective results apply to any bipartite lattice, in the plots that follow, we specifically refer to square-lattice geometry and spin-\( \frac{1}{2} \), since only for this system all relevant effective constants are known.

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Apart from $M_s$ and $M$, it is illuminating to also consider the individual sublattice magnetizations $M_A$ and $M_B$. On a bipartite lattice they can be extracted from the uniform and staggered magnetization as

$$M_A(t, m, m_H) = \frac{M(t, m, m_H) + M_s(t, m, m_H)}{2},$$

$$M_B(t, m, m_H) = \frac{M(t, m, m_H) - M_s(t, m, m_H)}{2}. \tag{2.31}$$

By definition, $M_A$ is positive (‘A-spins point up’) and $M_B$ is negative (‘B-spins point down’). While figures for the zero-temperature uniform and staggered magnetizations in external fields have been presented in reference [22] (figures 3 and 4, respectively), here, in figure 2, we depict the zero-temperature sublattice magnetizations $M_A(0, m, m_H)$ and $|M_B(0, m, m_H)|$. One observes that both $M_A$ and $|M_B|$ increase when the magnetic or the staggered field become stronger. The explanation is via suppression of quantum fluctuations caused by the external fields. Note that the magnitude of the net external field on sublattice $A$ is not the same as on sublattice $B$. The net external field on sublattice $A$ is given by the sum $H_s + H$, whereas on sublattice $B$ it is given by the difference $H_s - H$. Due to the stronger net external field on sublattice $A$, the suppression of quantum fluctuations on sublattice $A$ is more pronounced. Accordingly, the zero-temperature sublattice magnetization $M_A$ is larger than the zero-temperature sublattice magnetization $|M_B|$ for any values of fixed external fields.

We now turn to finite temperature where rather subtle effects emerge. In figure 3 we first show staggered and uniform magnetization as a function of $m_H$ and $m$ for the temperatures $t = 0.2$ and $t = 0.4$, as in the previous plots for the entropy density. More precisely, we depict the sum of 1-loop and 2-loop contributions given in equations (2.25).
Figure 3. Staggered and uniform magnetization for the spin-\(1/2\) square-lattice antiferromagnet at the temperatures \(t = 0.2\) (upper panel) and \(t = 0.4\) (lower panel): dependence on mutually parallel magnetic \((m_H)\) and staggered \((m)\) fields.

and (2.27),

\[
M_s : \tilde{\sigma}_1 T + \tilde{\sigma}_2 T^2,
\]

\[
M : \hat{\sigma}_1 T + \hat{\sigma}_2 T^2,
\]  

(2.32)

without superimposing the dominant \(T = 0\) contributions. The plots in figure 3 hence capture the change of staggered and uniform magnetizations when temperature is raised from \(t = 0\) to \(t = \{0.2, 0.4\}\). These are the relevant quantities to be compared with entropy density where the zero-temperature piece is excluded as well\(^4\). As expected, the change in the staggered magnetization is negative, i.e., the staggered magnetization drops when temperature is raised—while magnetic and staggered field strengths held fixed—due to the thermal fluctuations. The decrease is most pronounced in weak fields where the thermal disruption of spin order is strongest.

Interestingly, in the uniform magnetization we observe a qualitatively different pattern: if temperature is raised from \(t = 0\) to \(t = \{0.2, 0.4\}\)—while magnetic and staggered

\(^4\)Notice that uniform and sublattice magnetizations in the plots are given in the same units as the staggered magnetization: \(1/a^2\).
fields held fixed—the change in the uniform magnetization is positive. Here the combined effect of thermal fluctuations and suppression of quantum fluctuations by the external fields is rather counterintuitive: the uniform magnetization is enhanced—and not weakened. The enhancement becomes larger as the magnetic field strength increases. However, the enhancement does not grow monotonously: at more elevated temperatures ($t = 0.4$)—and in stronger external fields—the enhancement becomes less distinctive.

These findings regarding the staggered and the uniform magnetization are consistent with the previous observation (see figure 1) that the entropy density drops when the staggered field becomes stronger: the antialigned spin pattern is enforced. Likewise, they are consistent with the observation that the entropy density grows when the magnetic field becomes stronger: the antialigned spin pattern is perturbed. Note that the correlation is between entropy density and the staggered (and not the uniform) magnetization—the effects in the uniform magnetization are much less pronounced: there is about a one order of magnitude difference between the changes in $M_s$ and $M$ in the respective plots of figure 3. Also, at the more elevated temperature $t = 0.4$ and in stronger staggered fields, the entropy density only initially grows when $H$ becomes stronger, but then starts to drop (rhs of figure 1)—much like the decrease in the staggered magnetization becomes less distinctive when $H$ gets stronger (rhs and lower panel of figure 3).

This subtle interplay between entropy and spin order can also be appreciated in the behavior of the individual sublattice magnetizations. As figure 4 indicates, the sublattice magnetizations $M_A$ and $|M_B|$ both behave in a qualitatively similar way as the staggered magnetization: the thermal perturbation of the antialigned spins is most drastic in weak fields. Analogously, at more elevated temperatures ($t = 0.4$)—and in stronger external fields—the decrease in both $M_A$ and $|M_B|$ gets less distinctive as the magnetic field becomes stronger.

So far we have been focusing on just two fixed temperatures. Let us now investigate in more detail how entropy and magnetizations vary with temperature. To explore this situation, we consider two representative points in parameter space $\{m, m_H\}$ that we choose as $\{0.3, 0.05\}$ and $\{0.3, 0.2\}$, respectively. For each point we evaluate entropy density, uniform magnetization, staggered magnetization and the sublattice magnetizations as a function of temperature. Note that all magnetizations are the magnetizations induced by finite temperature, i.e., we consider the changes of $M, M_s, M_A, M_B$ when temperature is raised from $t = 0$ to $t \neq 0$.

First of all, if no magnetic field is present, the uniform magnetization is zero and the sublattice magnetizations $M_A$ and $|M_B|$ are identical for arbitrary temperatures and arbitrary staggered field strength. However, in presence of a magnetic field aligned with the order parameter, interesting effects take place, as we illustrate in figure 5. The upper panel refers to the first point $\{m, m_H\} = \{0.3, 0.05\}$ that corresponds to weak magnetic field. Remarkably, a net uniform magnetization is induced that even gets larger at higher temperatures. This is quite counterintuitive: one would rather expect thermal fluctuations to prevent emergence of a uniform magnetization, in particular, at more elevated temperatures. Still, as temperature continues to rise, the uniform magnetization increases less rapidly and then eventually starts to drop (not shown in the figure).
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Figure 4. Sublattice magnetizations $M_A$ and $|M_B|$ for the spin-$\frac{1}{2}$ square-lattice antiferromagnet at the temperatures $t = 0.2$ (upper panel) and $t = 0.4$ (lower panel): dependence on mutually parallel magnetic ($m_H$) and staggered ($m$) fields.

On the other hand, the staggered magnetization diminishes when temperature rises, according to intuition. In figure 5 we furthermore depict the sublattice magnetizations $M_A$ and $|M_B|$ that also drop when temperature increases. The essential point is that $M_A$ and $|M_B|$ do not drop alike, such that a net uniform magnetization is created in presence of a magnetic field.

Trying to correlate these findings with the behavior of the entropy density, we note that information on microscopic order is provided by the staggered magnetization, because it measures the total of aligned spins along the order parameter axis. Following this simple picture, the decrease of the staggered magnetization with temperature is related to the increase of entropy with temperature:

$$-\frac{dM_s}{dT} \propto \frac{ds}{dT}. \quad (2.33)$$

According to figure 5 this is indeed the case. For the specific point $\{m, m_H\} = \{0.3, 0.05\}$, the entropy density first remains constant, up to about $t \approx 0.07$, but then starts to increase. Likewise, staggered and sublattice magnetization first remain constant, up to
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Figure 5. Spin-$\frac{1}{2}$ square-lattice antiferromagnet in mutually parallel magnetic and staggered fields. Lhs: temperature dependence of uniform (magenta), staggered (brown), as well as sublattice magnetizations $M_A$ (blue) and $|M_B|$ (green). Rhs: temperature dependence of entropy density. Upper panel: $\{m, m_H\} = \{0.3, 0.05\}$. Lower panel $\{m, m_H\} = \{0.3, 0.2\}$.

about $t \approx 0.07$, but then start to decrease. Although the counterintuitive emergence of uniform magnetization hints at creation of spin order, the entropy increases because the destruction of spin antialignment—reflected by the staggered magnetization—is the dominant effect.

Let us consider the second point $\{m, m_H\} = \{0.3, 0.2\}$ that corresponds to stronger magnetic field, but still fulfills the stability criterion, equation (2.5). According to the lower panel of figure 5, the splitting between the curves for $M_A$ and $|M_B|$, induced by the magnetic field, is now more pronounced. Accordingly, this larger asymmetry creates a larger uniform magnetization, as compared to the previous point $\{m, m_H\} = \{0.3, 0.05\}$. The staggered magnetization and the sublattice magnetizations $M_A$ and $|M_B|$ again decrease with temperature while the entropy density grows. The overall pattern is thus the same: the destruction of the arrangement of antialigned spins is predominant. It should be noted that the perturbation of spin order in the stronger magnetic field is more pronounced: compared to the previous point, the entropy here starts to increase—and the staggered magnetization starts to decrease—already around the temperature $t \approx 0.03$, whereas for the previous point we had $t \approx 0.07$. 

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Our findings are consistent with the experimental data presented in reference [45] where the thermomagnetic properties of the quasi two-dimensional antiferromagnet K₂MnF₄ have been studied. The sublattice magnetizations decrease with temperature, but not alike: the down-sublattice magnetization drops more rapidly such that a net uniform magnetization is induced that increases with temperature. In accordance with our findings, this counterintuitive effect is more pronounced in stronger magnetic fields. Regarding the theoretical side, we are not aware of any references—except for reference [22]—that have reported such order–disorder phenomena in antiferromagnetic films subjected to magnetic fields aligned with nonzero staggered fields.

3. Antiferromagnetic films in mutually orthogonal staggered and magnetic fields

3.1. Preliminaries

It is instructive to discuss the topic of entropy and spin order also for antiferromagnetic films that are subjected to a magnetic field orthogonal to the staggered field. While this general setting has been analyzed in references [21, 23] within effective field theory, entropy density as well as order–disorder phenomena in the staggered and uniform magnetizations have not been addressed in that reference—this is the objective of the present section.

Note that in the configuration of mutually orthogonal staggered and magnetic fields, no stability criterion (as in the case of mutually parallel staggered and magnetic fields) must be met. The magnetic field orients the order parameter in a plane transverse to its own direction—and out of all directions in this plane, the staggered field aligns the order parameter into its proper direction. In this situation the ground state will be hence be aligned with the staggered field regardless of the strength of the magnetic field. No conflicting tendencies arise.

The specific configuration of external fields now is

\[ \vec{H} = (0, H, 0), \quad \vec{H}_s = (H_s, 0, 0), \quad H, H_s > 0, \]  

i.e., magnetic and staggered fields are mutually orthogonal. The staggered field points again into the direction of the order parameter.

An essential difference with respect to the configuration of mutually parallel fields is that in the case of mutually orthogonal fields, the two magnons satisfy the dispersion laws

\[ \omega_1 = \sqrt{\vec{k}^2 + \frac{M_s H_s}{\rho_s} + H^2}, \]

\[ \omega_II = \sqrt{\vec{k}^2 + \frac{M_s H_s}{\rho_s}}, \]  

i.e., one of the magnons does not take notice of the magnetic field. As a consequence the low-energy physics is different. Readers interested in the derivation of the corresponding
free energy density are referred to reference [21]. Here we merely quote the two-loop result,

\[
z = z_0 - \frac{1}{2} \left\{ g_0^I + g_0^{II} \right\} + \frac{M_s H_s}{16\pi\rho_s^2} \left\{ \sqrt{\frac{M_s H_s}{\rho_s}} + H^2 - \sqrt{\frac{M_s H_s}{\rho_s}} \right\} g_1^I \\
+ \frac{H^2}{4\pi\rho_s} \sqrt{\frac{M_s H_s}{\rho_s}} + H^2 g_1^I - \frac{M_s H_s}{16\pi\rho_s^2} \left\{ \sqrt{\frac{M_s H_s}{\rho_s}} + H^2 - \sqrt{\frac{M_s H_s}{\rho_s}} \right\} g_1^{II} \\
- \frac{M_s H_s}{8\rho_s^2} \left\{ (g_1^I)^2 - 2g_1^I g_1^{II} + (g_1^{II})^2 \right\} - \frac{H^2}{2\rho_s} (g_1^I)^2 + \frac{2}{\rho_s} s(\sigma, \sigma_H)T^4,
\]

(3.3)

where the vacuum energy density \(z_0\) is

\[
z_0 = -M_s H_s - \frac{1}{2} \rho_s H^2 - (k_2 + k_3) \frac{M_s^2 H^2}{\rho_s^3} - k_1 \frac{M_s H_s}{\rho_s} H^2 - (e_1 + e_2) H^4 \\
- \frac{1}{12\pi} \left\{ \left( \frac{M_s H_s}{\rho_s} + H^2 \right)^{3/2} + \left( \frac{M_s H_s}{\rho_s} \right)^{3/2} \right\} - \frac{M_s^2 H^2}{64\pi^2 \rho_s^3} \\
- \frac{5M_s H_s^2}{128\pi^2 \rho_s^5} - \frac{H^4}{32\pi^2 \rho_s^3} + \frac{M_s^2 H^2}{64\pi^2 \rho_s^5} \mathcal{I}_s \mathcal{H} + H^2,
\]

(3.4)

and the kinematical Bose functions \(g_1^{II}\) and the sunset function \(s(\sigma, \sigma_H)\) are provided in appendix A. The vacuum energy density—in addition to \(k_2\) and \(k_3\) that already showed up in the previous case of mutually parallel fields—contains the NLO effective constants \(e_1, e_2, k_1, k_3\).

### 3.2. Entropy density

Let us first look at the entropy density that one readily derives from the two-loop free energy density, equation (3.3), as

\[
s(t, m, m_H) = \frac{1}{2} \left( \frac{dg_0^I}{dT} + \frac{dg_0^{II}}{dT} \right) - \pi^2 \rho_s^2 m^2 \left( \sqrt{m^2 + m_H^2} - m \right) \frac{dg_1^I}{dT} \\
- 2\pi \rho_s^2 m^2 \sqrt{m^2 + m_H^2} \frac{dg_1^I}{dT} + \pi^2 \rho_s^2 m^2 \left( \sqrt{m^2 + m_H^2} - m \right) \frac{dg_1^{II}}{dT} \\
+ 2\pi \rho_s m^2 \left( g_1^I \frac{dg_1^I}{dT} + g_1^{II} \frac{dg_1^{II}}{dT} - g_1^{II} \frac{dg_1^I}{dT} - g_1^I \frac{dg_1^{II}}{dT} \right) \\
+ 4\pi^2 \rho_s m_H^2 g_1^I \frac{dg_1^I}{dT} - \frac{2}{\rho_s} \frac{ds(\sigma, \sigma_H)}{dT} T^4 - \frac{8}{\rho_s} s(\sigma, \sigma_H)T^3.
\]

(3.5)

It should be stressed that NLO effective constants only show up in the (zero-temperature) vacuum energy density \(z_0\). The finite-temperature behavior of the system,
3. Order–disorder effects

We first comment on a qualitative difference between antiferromagnetic films in mutually orthogonal and mutually parallel fields. When the fields are parallel, the relative strengths of magnetic and staggered field cannot take arbitrary values—rather a stability criterion must be obeyed. Moreover, uniform, staggered and sublattice magnetizations are all measured with respect to the same axis that is defined by the staggered magnetization vector at zero temperature, i.e., the order parameter.

On the other hand, if magnetic and staggered fields are orthogonal, the magnetic field can take larger values than the staggered field—no stability criterion must be met. Also, while staggered and sublattice magnetizations are measured with respect to the order parameter axis, the uniform magnetization that is induced by the external magnetic field, points orthogonal to the order parameter. In such a configuration of external fields, no uniform magnetization in the direction of the order parameter can be created by the magnetic field whose effect rather is to tilt the spins in its proper
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To begin with, we analyze the situation at zero temperature, where staggered and uniform magnetizations are given by (see reference [21])

$$\frac{M_s(0, m, m_H)}{M_s} = 1 + \frac{m}{4} + \frac{\sqrt{m^2 + m_H^2}}{4} + \frac{m^2}{8} + \frac{5m_H^2}{32} - \frac{m^3}{8\sqrt{m^2 + m_H^2}}$$

$$- \frac{3mm_H^2}{32\sqrt{m^2 + m_H^2}} + 8\pi^2 \rho_s(k_2 + k_3)m^2 + 4\pi^2 \rho_s k_1 m_H^2,$$

$$m = \frac{\sqrt{M_s H}}{2\pi \rho_s^{1/2}}, \quad m_H = \frac{H}{2\pi \rho_s}, \quad M_s = M_s(0, 0, 0),$$

(3.6)

and

$$\frac{M(0, m, m_H)}{\rho_s^{3/2}} = 2\pi m_H + \pi m_H \sqrt{m^2 + m_H^2} + \pi m_H^3 + \frac{5\pi}{8} m^2 m_H$$

$$- \frac{\pi m^3 m_H}{8\sqrt{m^2 + m_H^2}} + 32\pi^3 \rho_s (e_1 + e_2)m_H^3 + 16\pi^3 \rho_s k_1 m^2 m_H,$$

(3.7)

respectively. The staggered magnetization, in addition to the combination $k_2 + k_3$ of NLO effective constants that showed up previously in equation (2.29), here also depends on $k_1$. Moreover, the uniform magnetization involves the NLO effective constants $e_1$ and $e_2$ that were absent in the case of mutually parallel fields, equation (2.30). Unlike for the combination $k_2 + k_3$, the numerical values of $k_1, e_1, e_2$ are not available from Monte Carlo simulations. Estimates based on scaling arguments given in reference [46, 47] however show that their absolute values are small—of the order of $0.001 \rho_s^{-1}$—while their respective signs remain open. Since we are dealing with only minor corrections, in the following figure referring to $T = 0$, we neglect these contributions. As we will see, at $T \neq 0$, all these NLO effective constants are absent, such that the finite-temperature results for $M_s$ and $M$—much like the entropy density, equation (3.5)—are parameter free.

In figure 7, we depict the dependence of the staggered and uniform magnetizations on $m$ and $m_H$ at $T = 0$. The staggered magnetization $M_s(0, m, m_H)$ grows when the magnetic or the staggered field become stronger—much like in the previous case of mutually parallel external fields. The enhancement of the order parameter can be explained again via suppression of quantum fluctuations caused by the external fields. Although here the magnetic field points orthogonal to the staggered magnetization vector, it also supresses quantum fluctuations. The overall suppression of quantum fluctuations is of the same magnitude as in the case of mutually parallel fields: following figure 2, the increase in the sublattice magnetizations (strong fields compared to weak fields) is about $0.035a^{-2}$ which adds up to $0.07a^{-2}$ for the staggered magnetization—this is about the
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Figure 7. Staggered magnetization $M_s$ and uniform magnetization $M$ for the spin-1/2 square-lattice antiferromagnet in mutually orthogonal magnetic ($m_H$) and staggered ($m$) fields at $T = 0$.

same change one observes in the staggered magnetization in figure 7. Finally, the uniform magnetization $M(0, m, m_H)$—as expected—is zero when $H = 0$, but continuously grows when the magnetic field strength increases. The dependence on the staggered field is tiny, but it also slightly suppresses quantum fluctuations.

Let us now turn to finite temperature where the expansions for the staggered and uniform magnetization take the form

$$M_s(t, m, m_H) = M_s(0, m, m_H) + \tilde{\sigma}_1 T + \tilde{\sigma}_2 T^2 + O(T^3),$$

$$M(t, m, m_H) = M(0, m, m_H) + \hat{\sigma}_1 T + \hat{\sigma}_2 T^2 + O(T^3),$$

(3.8)

with

$$\tilde{\sigma}_1(t, m, m_H) = -\frac{M_s}{2\rho_s} (h_1^{\parallel} + h_1^{\perp}),$$

$$\hat{\sigma}_1(t, m, m_H) = -2\pi\rho_s m_H h_1^{\parallel}.$$  

(3.9)

We refrain from listing the rather lengthy expressions for the coefficients $\tilde{\sigma}_2$ and $\hat{\sigma}_2$ that can be obtained trivially from the free energy density, equation (3.3), via

$$M_s(T, H_s, H) = -\frac{\partial z(T, H_s, H)}{\partial H_s},$$

$$M(T, H_s, H) = -\frac{\partial z(T, H_s, H)}{\partial H}.$$  

(3.10)

Plots for the staggered and the uniform magnetization as functions of $m_H$ and $m$ for the temperatures $t = 0.2$ and $t = 0.4$ are provided in figure 8. We depict again the sum of 1-loop and 2-loop contributions given in equation (3.8),

$$M_s : \tilde{\sigma}_1 T + \tilde{\sigma}_2 T^2,$$

$$M : \hat{\sigma}_1 T + \hat{\sigma}_2 T^2,$$

(3.11)
Figure 8. Staggered and uniform magnetization for the spin-$\frac{1}{2}$ square-lattice anti-ferromagnet at the temperatures $t = 0.2$ (upper panel) and $t = 0.4$ (lower panel): dependence on mutually orthogonal magnetic ($m_H$) and staggered ($m$) fields.

without superimposing the dominant $T = 0$ contributions. The plots thus capture the change of staggered and uniform magnetizations when temperature is raised from $t = 0$ to $t = \{0.2, 0.4\}$. The change in the staggered magnetization, as expected, is negative: it drops on account of the thermal fluctuations and the decrease is most drastic in weak fields where the thermal disruption of spin order is strongest. This is consistent with the previous observation (see figure 6) that the entropy density drops when the staggered field becomes stronger: the antialigned spin pattern is enforced. Unlike in mutually parallel external fields (compare with figure 3), at more elevated temperatures ($t = 0.4$) and in stronger external fields, the effect of the magnetic field is almost nil.

The change in the uniform magnetization, when temperature is raised from $t = 0$ to $t = 0.2$, is negative—as one would expect. Remarkably, when temperature is raised from $t = 0$ to the more elevated temperature $t = 0.4$, things are less intuitive: the uniform magnetization may be enhanced or damped. In particular, the observation that the enhancement takes place in weaker external fields is quite counterintuitive. Note however that the effects induced in the uniform magnetization are minor compared to the effects in the staggered magnetization: there is about a two order of magnitude
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difference between the changes in $M_s$ and $M$ in the respective plots of figure 8. Also, compared to the case of mutually parallel fields (see rhs of figure 3), the effects in the uniform magnetization are much less pronounced here: it is more difficult to induce a net magnetization when the magnetic field points orthogonal to the order parameter.

Let us finally explore in more detail how entropy and magnetizations vary with temperature. To this end we consider the same two representative points in parameter space $\{m, m_H\} = \{0.3, 0.05\}$ and $\{m, m_H\} = \{0.3, 0.2\}$, respectively, and evaluate entropy density, uniform magnetization and staggered magnetization as functions of temperature. The quantities $M_A$ and $M$ again refer to the magnetizations induced by finite temperature, i.e., correspond to the response of the system when temperature is raised from $t = 0$ to $t \neq 0$.

In the absence of a magnetic field, the uniform magnetization is zero for arbitrary temperatures and arbitrary staggered field strength. In presence of a magnetic field—oriented perpendicular to the order parameter—a uniform magnetization is induced that slightly increases at more elevated temperatures according to figure 9. But note that even in stronger magnetic fields corresponding to the point $\{m, m_H\} = \{0.3, 0.2\}$, the effect is very weak as compared to the uniform magnetization induced in the case of mutually parallel fields (see lhs of figure 5). Still, we are dealing with the analogous counterintuitive phenomenon because one would rather expect thermal fluctuations to preclude creation of a uniform magnetization.

The staggered magnetization, on the other hand, decreases when temperature rises, as expected. The decrease is of the same magnitude as for mutually parallel fields (see lhs of figure 5)\(^6\). The sublattice magnetizations $M_A$ and $|M_B|$ are the same for arbitrary temperature and for any point $\{m, m_H\}$ in parameter space, and just are half of the staggered magnetization. As a consequence, due to this symmetric situation, no uniform magnetization can be created in the direction of the order parameter: the uniform magnetization induced by the magnetic field points orthogonal to the staggered magnetization.

In the case of mutually parallel fields, the staggered magnetization served as an indicator for the total of aligned spins along the order parameter axis. If the external fields are mutually orthogonal, in addition, we have to take into account the order created through the uniform magnetization pointing into the direction of the magnetic field. Trying to correlate spin order with entropy, naively, the change of entropy density with temperature is related to the decrease of the staggered magnetization and to the emergence of the uniform magnetization as

$$\frac{dM_s}{dT} \propto \frac{ds}{dT}, \quad -\frac{dM}{dT} \propto \frac{ds}{dT}.$$  \hspace{1cm} (3.12)

Recall that $M_s$ and $M$ are the staggered and uniform magnetizations induced by finite temperature, i.e., reflecting the changes in these quantities when temperature is raised from $t = 0$ to $t \neq 0$.

Figure 9 just evidences this scenario: the staggered magnetization decreases with temperature while the entropy density increases—the destruction of the arrangement of

\(^6\)In order for the tiny effects in the uniform magnetization to become visible, in figure 9 we have drawn up to $t = 0.4$, whereas in figure 5 we plotted up to $t = 0.3$. 

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antialigned spins is the dominant effect. As the figure shows, the second contribution in equation (3.12) is negligible, such that the correlation is again between entropy density and staggered magnetization. Note that the behavior of the entropy density—as well as the staggered magnetization—for both temperatures \( t = \{0.2, 0.4\} \) is qualitatively the same: there is no ‘retardation effect’ in the increase (decrease) of entropy density (staggered magnetization) in weaker magnetic fields—in contrast to the previous case of mutually parallel fields (see figure 5), where the perturbation of spin order in the stronger magnetic field was more pronounced.

A final remark concerns the validity domain of our effective analysis. As we have mentioned before, it applies to the domain where temperature and external fields—quantified by the parameters \( t, m, m_H \)—are small. In the case of mutually aligned magnetic and staggered fields we furthermore have the stability criterion that we implemented by the condition \( m > m_H + 0.1 \). In general, as a consequence of the Mermin–Wagner theorem, the staggered field in our effective analysis cannot be arbitrarily small at finite temperature—in particular it cannot be zero\(^7\). While the stability criterion already guarantees

\(^7\)For a detailed discussion of why the effective approach fails in small staggered fields, the interested reader may want to consult figures 2 and 3 of reference [48] and the information provided therein.

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that we never leave the domain where our effective analysis is valid, in the case of mutually orthogonal fields we have been careful in the plots by not going below the value $m = 0.1$ either. For this parameter region we are not aware of any references—neither theoretical nor experimental—that have reported the counterintuitive effects we have described above.

4. Conclusions

In the present systematic effective field theory investigation of two-dimensional bipartite antiferromagnets subjected to magnetic and staggered fields, we focused on order–disorder phenomena by examining the behavior of the staggered, uniform and sublattice magnetizations as well as the entropy density.

The first part dealt with the specific configuration of mutually aligned magnetic and staggered fields. In the entropy density at fixed temperature we identified two opposite tendencies: (1) if the staggered field becomes stronger, the entropy decreases, and (2) if the magnetic field becomes stronger, the entropy increases. Naively, the staggered field establishes spin order by enforcing antialignment of the spins, whereas the magnetic field perturbs the antialigned array of spins. These observations are confirmed by the finite-temperature behavior of the staggered, uniform and sublattice magnetizations. Remarkably, since the two sublattices of the bipartite antiferromagnet are affected by the magnetic field in a nonsymmetric way, a finite-temperature uniform magnetization is induced that initially even grows as temperature increases—this outcome is rather counterintuitive.

In the second part we studied the configuration of mutually orthogonal magnetic and staggered fields. Overall, we find that the entropy density at fixed temperature again diminishes when the staggered field grows, and that it increases when the magnetic field grows: likewise the staggered field establishes spin order by enforcing antialignment of the spins, whereas the magnetic field perturbs this pattern. The finite-temperature uniform magnetization that is induced perpendicular to the order parameter axis, slightly grows as temperature increases, but here this counterintuitive phenomenon is tiny. The decrease of the staggered magnetization with temperature along the order parameter axis is the dominant effect: microscopic order is destroyed.

While concrete plots referred to the spin-$\frac{1}{2}$ square-lattice antiferromagnet, our results and observations apply to any other two-dimensional bipartite lattice. In this perspective the effective field theory analysis is universal—details rely on the concrete values of the spin stiffness and the zero-temperature staggered magnetization.

In conclusion, comparing the two different configurations of external fields antiferromagnets are subjected to, analogous characteristics concern the emergence of counterintuitive phenomena, but the impact of the magnetic field in the case of mutually parallel fields is more pronounced.

From an experimental perspective, the question is which antiferromagnetic materials are adequately described by the extended Heisenberg model, i.e., the Heisenberg Hamiltonian (2.1) containing magnetic and staggered fields which represents the starting point of our effective analysis. Various realizations are known in nature: these comprise

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three-dimensional antiferromagnets such as MnF$_2$ and MnO (see, e.g., references [8, 49, 50]), or quasi two-dimensional antiferromagnets K$_2$MnF$_4$ (see, e.g., reference [45]) and Rb$_2$MnF$_4$ (see, e.g., references [52, 53])$^8$. Furthermore, apart from the prominent (undoped) square-lattice antiferromagnet La$_2$CuO$_4$, novel quasi two-dimensional samples that contain organic compounds, have been synthesized and studied experimentally (see, e.g., references [51, 55]). As the authors of references [51, 55] point out, these samples represent excellent model systems for the two-dimensional spin-$\frac{1}{2}$ Heisenberg antiferromagnet. All aforementioned quasi two-dimensional materials can in principle serve as experimental testing grounds of the effective field theory results presented here.

Unfortunately, regarding the actual experimental side on antiferromagnetic materials—and mutually parallel magnetic and staggered fields—we are only aware of reference [45], where the counterintuitive behavior of the magnetization has been observed in the quasi two-dimensional sample K$_2$MnF$_4$ and been compared with the predictions of first-order Oguchi renormalized spin-wave theory. Our results are perfectly consistent with the experimental data presented there. In particular, the experimentally observed decrease of the sublattice magnetizations with temperature—more rapidly for the down-sublattice than for the up-lattice magnetization—and the fact that the resulting counterintuitive effect is more pronounced in stronger magnetic fields, is in perfect agreement with our findings.

It should be pointed out that our effective analysis goes beyond first-order Oguchi renormalized spin-wave theory as the latter neglects higher-order corrections. In the concluding section of reference [45] the authors point out that first-order Oguchi renormalized spin-wave theory describes the experimental data quite well, although further improvement may possibly be obtained by incorporating higher-order corrections. The present effective field theory investigation takes into account such higher-order corrections in a fully systematic way and hence paves the way for such an endeavor—not only for K$_2$MnF$_4$, but also for other (quasi) two-dimensional antiferromagnetic samples that may be probed in future experiments. Regarding the theoretical side—and still referring to mutually parallel magnetic and staggered fields—we are not aware of any references (beyond the calculations presented in reference [45]) that have reported the counterintuitive increase of the magnetization with temperature.

Turning to the case of mutually orthogonal magnetic and staggered fields, we are unaware of any references—theoretical or experimental—that have reported the counterintuitive effects we have described within effective field theory: apparently our findings are new. Still, the case of zero staggered field, not covered by our analysis$^9$, is reminiscent of the configuration of mutually orthogonal magnetic and staggered fields, because the staggered magnetization lies in a plane orthogonal to the external magnetic field. Indeed, for this situation where only a magnetic field is present, the initial increase of the magnetization with temperature has been reported for the spin-$\frac{1}{2}$ square-lattice antiferromagnet in a theoretical study relying on exact diagonalization (see reference [7]) as well as in Monte Carlo simulations (see references [5, 6]).

$^8$ A more extensive list of quasi two-dimensional antiferromagnets is provided by table I of reference [54].

$^9$ As we mentioned previously, the staggered field cannot be turned off in our analysis as we would leave the domain of validity of our effective field theory approach. Nonetheless, the case of zero staggered field is also accessible within the effective field theory framework. But one has to resort to an alternative type of perturbative expansion, called $\epsilon$-expansion [56–59].
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Aside from experimental verification, numerical simulations of the two-dimensional antiferromagnetic Heisenberg model in presence of staggered and magnetic fields, indeed provide an alternative route to test and complement our effective field theory predictions. Since the ‘clean’ Heisenberg Hamiltonian does not contain terms related to impurities, spin–orbit couplings, lattice anisotropies (to name just a few effects that may be present in a real antiferromagnetic sample), numerical simulations allow one to detect and characterize the non-monotous behavior of the uniform magnetization with temperature in an idealized model situation.

Finally, it would be interesting to extend the present effective field theory investigation into various directions. One question is whether the enhancement of the uniform magnetization with temperature—or emergence of spin order—also takes place in three-dimensional antiferromagnets. Furthermore, while our current study refers to the Heisenberg exchange model in presence of external magnetic and staggered fields, one may want to take into account further types of interactions like spin–orbit couplings, Dzyaloshinskii–Moriya interactions—or consider the XY-model. Respective work is in progress.

Appendix A. Kinematical functions and sunset function for antiferromagnetic films in mutually orthogonal magnetic and staggered fields

The dimensionless kinematical Bose functions

\[
\begin{align*}
  h_0^{I,II} &= \frac{g_0^{I,II}}{T^3}, & h_1^{I,II} &= \frac{g_1^{I,II}}{T}, & h_2^{I,II} &= g_1^{I,II}T
\end{align*}
\]  

(A.1)

for magnon I and magnon II are

\[
\begin{align*}
  h_0^I(H_s, H, T) &= \frac{4\pi^2(\sigma^2 + \sigma_H^2)^{3/2}}{3} - 2\sqrt{\sigma^2 + \sigma_H^2}Li_2(e^{2\pi\sqrt{\sigma^2 + \sigma_H^2}}) \\
  &\quad + \frac{1}{\pi}Li_3(e^{2\pi\sqrt{\sigma^2 + \sigma_H^2}}) + 2\pi(\sigma^2 + \sigma_H^2) \\
  &\quad \times \left\{ \log(1 - e^{-2\pi\sqrt{\sigma^2 + \sigma_H^2}}) - \log(1 - e^{2\pi\sqrt{\sigma^2 + \sigma_H^2}}) \right\}, \\
  h_1^I(H_s, H, T) &= -\frac{1}{2\pi} \log\left(1 - e^{-2\pi\sqrt{\sigma^2 + \sigma_H^2}}\right), \\
  h_2^I(H_s, H, T) &= \frac{1}{8\pi^2\sqrt{\sigma^2 + \sigma_H^2}}\left(e^{2\pi\sqrt{\sigma^2 + \sigma_H^2}} - 1\right).
\end{align*}
\]  

(A.2)
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\[
\begin{align*}
    h_0^\Pi(H_s, 0, T) &= \frac{4\pi^2\sigma^3}{3} + 2\pi\sigma^2 \left\{ \log(1 - e^{-2\pi\sigma}) - \log(1 - e^{2\pi\sigma}) \right\} \\
    &\quad - 2\sigma Li_2(e^{2\pi\sigma}) + \frac{1}{\pi} Li_3(e^{2\pi\sigma}), \\
    h_1^\Pi(H_s, 0, T) &= -\frac{1}{2\pi} \log \left(1 - e^{-2\pi\sigma}\right), \\
    h_2^\Pi(H_s, 0, T) &= \frac{1}{8\pi^3\sigma(e^{2\pi\sigma} - 1)},
\end{align*}
\]

respectively, where \( Li_2 \) and \( Li_3 \) are polylogarithms. All quantities are expressed in terms of the two dimensionless parameters \( \sigma_H \) and \( \sigma \),

\[
\sigma_H = \frac{H}{2\pi T}, \quad \sigma = \sqrt[2]{\frac{M_s H_s}{2\pi \rho_s T}},
\]

which in turn are related to \( m_H \) and \( m \) as

\[
\sigma_H = \frac{\rho_s}{T} m_H, \quad \sigma = \frac{\rho_s}{T} m.
\]

The dimensionless sunset function \( s(\sigma, \sigma_H) \) is quite involved and we refer the reader to reference [21], where its definition is given in (B14) and a plot is provided in figure 3.

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