Comment on "Magnetic structure and magnetization of z-axis helical Heisenberg antiferromagnets with XY anisotropy in high magnetic fields transverse to the helix axis at zero temperature"

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In two recent papers [Phys. Rev. B **99**, 214438 (2019); Phys. Rev. B **96**, 104405 (2017)], Johnston employs his *unified molecular field theory* to study the phase diagram of a classical one-dimensional magnet with exchange frustration in dependence of the anisotropy and magnetic field. Here, it is argued that the assumptions made about the stable configurations are too restrictive and not well justified.

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A recent article by Johnston [1] is devoted to the study of the minimum-energy configurations of a classical magnetic chain with first- and second-neighbor exchange, subjected to easy-plane anisotropy and in-plane magnetic field. It follows a previous paper [2] where the case of infinite anisotropy, the planar model, was considered. The physical system is in itself interesting, especially in the frustrated case with antiferromagnetic (AFM) second-neighbor exchange, which shows a variety of phases, from helical ordering to fan configurations, and so on.

Equations (A18)–(A20) of [1] take the second-neighbor exchange coupling as the energy unit and give the dimensionless Hamiltonian of the system, which, disregarding irrelevant constants, can be written with simpler notations as

$$\mathcal{H} = \sum_n \big[\boldsymbol{s}_n\cdot(-J\boldsymbol{s}_{n+1} + \boldsymbol{s}_{n+2}) + A s_n^{z\,2} - h s_n^x\big], \tag{1}$$

where $\boldsymbol{s}_n$ are unit vectors representing the magnetic moments or spins, J (same as $-J_{12}$ in [1]) is the nearest-neighbor exchange, $A > 0$ ($h_{\rm A}^{**}$ in [1]) is the easy-plane anisotropy that favors alignment in the xy plane, and the field $h > 0$ (h_x^{**} in [1]) favors alignment along the positive x axis. For $h = 0$ the minimum-energy configuration is a helix with axis along z and spins in the xy plane, $\boldsymbol{s}_n = (\cos kn, \sin kn, 0)$, with the angle k (reciprocal of the helix pitch) such that $4\cos k = J$; if $|J| \geqslant 4$ the configuration is ferromagnetic or AFM, according to the sign of J. Of course, for zero anisotropy helices with axes in any direction are degenerate minima.

For $A = \infty$ the z components of the spins are vanishing and one indeed has the strictly *planar model*: the two-component spins can be described by angles, $\boldsymbol{s}_n = (\cos\varphi_n, \sin\varphi_n, 0)$, and the simplified Hamiltonian reads

$$\mathcal{H} = \sum_n [-J\cos(\varphi_{n+1}-\varphi_n) + \cos(\varphi_{n+2}-\varphi_n) - h\cos\varphi_n]. \tag{2}$$

In the presence of nonzero field the only exact result known for the planar model is that if h is larger than a critical value $h_{\rm c} = 4\,(1 - J/4)^2$ the spins are aligned along it, which is the paramagnetic phase (PM); indeed the spin-wave dispersion $\omega_q = h + 4\sin^2\frac{q}{2}(J - 4\cos^2\frac{q}{2})$ is minimal at $q = k$ with a gap $\omega_k = h - h_{\rm c}$ that closes to zero at the critical field. The latter is correctly given as $h_{\rm c} = 16\sin^4\frac{k}{2}$ in Eq. (3) of [1], earlier obtained in [2] as Eqs. (21); however, the author attributes this result to himself, as "inferred" from a series of data he got for a discrete set of values of k [2]. Actually, he does not recognize that $h_{\rm c}$ has been known since 1962, when it appeared in a slightly different form, namely, $h_{\rm c} = 4(1 - \cos k)^2$, in Eq. (3.11) of the pioneering paper (cited in both [1] and [2]) by Nagamiya, Nagata, and Kitano [3] (NNK).

When both A and h are turned on, the minimization problem strongly increases in complexity. Johnston faces it by the *unified molecular field theory* he recently developed: it is manifest that the approach consists in numerically looking for classical minimum-energy configurations subjected to imposed constraints, namely, the number of variables in the minimization problem is reduced by means of *assumptions* regarding the possible structures. This is obviously legitimate, provided the assumptions are physically well-founded. Instead, in my opinion, some of them are unjustified, since "fans," "deformed helices," and "spherical ellipses" with rigidly periodic orderings, are taken as best candidates for minimum-energy configurations in absence of convincing theoretical or numerical arguments.

Assumption about the commensurability of k. In order to minimize the number of parameters, Johnston assumes that $k = 2\pi/N$ (or an integer multiple of this) with integer N such that the helix structure has exact periodicity N when A or/and h vanish. This reduces to N the number of spins to be considered, but it is to be noted that in reality k is determined by the ratio J of the exchange constants, which is a material property and hardly would give commensurate values. While this is a minor sin, as numerically treating incommensurate k is much harder, postulating the same periodicity to persist when both A and h are nonzero is a strong assumption, in favor of which no convincing reasons are provided in [1] and [2].

Assumption for the fan structure for $A = \infty$. In Eq. (3.6) of [3] the fan structure is described as a sinusoidal distortion,

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$\phi_n = \phi_{\max}\sin(nkd)$; however, the NNK derivation is valid only for fields very close to the critical value, $h_c - h \ll 1$, such that only Fourier modes with a wave vector closest to k are unstable. Postulating the same shape for smaller fields, as done in [2], where only $\phi_{\max}$ is taken as the minimization parameter, lacks any justification. As a matter of fact, early numerical transfer-matrix studies [4] even showed that there is no fan phase if the nearest-neighbor exchange is AFM; namely, if $J < 0$ and $k > \pi/2$.

Isotropic system with field. In the absence of anisotropy, $A = 0$, symmetry suggests an unambiguous stable configuration for $h > 0$: the helix axis is aligned with the field and the spins are uniformly canted along its direction, as in Fig. 2 of [1] ("canted helix"). As pointed out above, for zero field the helix can have any direction: it is improper to assert (Sec. II of [1]) that the moments lie in the xy plane and that an infinitesimal field causes the helix axis to "flop by 90° into the yz plane." From the Hamiltonian (1) one finds the average energy per spin $(-J\cos k + \cos 2k)(1 - s^{x2}) + (1-J)s^{x2} - hs^x = \text{const} + h_c s^{x2}/2 - hs^x$, so the magnetization is $s^x = h/h_c$ and the saturation field, not mentioned in [1], equals the critical field of the planar magnet.

Assumption of the "spherical ellipse" configuration. In Sec. II of [1] it is proposed, as a *Gedankenexperiment*, to include a small easy-plane anisotropy $A > 0$ from the above canted helix and it is hypothesized that the effect is a distortion yielding the "spherical ellipse" (SE) configuration, Eq. (1) of [1]. In the SE the components s_n^x are allowed to be alternatively positive or negative [Eq. (1h)], a hardly convincing assumption, since the spins follow the field and there is no reason to allow for any s_n^x to be negative, even in the AFM case ($J < 0$) where the antiferromagnetic exchange is already satisfied by the helix pitch k being larger than $\pi/2$. Moreover, the similar assumption of the SE made by NNK holds in the limit of infinitesimal anisotropy only.

On the other hand, a similar *Gedankenexperiment* could be made by imagining to start from the anisotropic system with the helix in the xy plane and then switch the field on: in this case one would expect a deformation of the helix that changes the angles between pairs of subsequent spins in a nonuniform way, such as in Eq. (16) and Fig. 7 of [2]: this configuration is quite different from the double-sided SE picture, even with the SE minor axis shrunk to zero (fan).

In my opinion, the assumption of the SE/fan configurations assumed in [1] is too strict to be able to describe the actual behavior of the magnetic chain for finite anisotropy and field. Even in the simplified planar case ($A = \infty$) this is a challenging task, as the minimum-energy configurations are likely to be far more complex. For instance, it is known that the closely related, though in principle much simpler, problem of the spin-flop transition in a chain with just nearest-neighbor AFM exchange, deals with bewilderingly complex minimum-energy configurations, which can even involve spatial chaos [5–9]: such configurations are incompatible with assumptions of regularity or periodicity. Hence, *unified molecular field theory* has no chance to face such simpler problem.

Coming back to the "simple" planar magnet (2), it is shown in Ref. [10] that just for zero field there are plenty of metastable configurations (relative energy minima), where a numerical minimization algorithm can get stuck. This risk is likely to worsen when a finite field is included, which is the purpose of [2]. It may be worth trying a generalization for finite h of the theoretical approach proposed in Ref. [10]. From the Hamiltonian (2) one finds the stationarity equations,

$$\partial_{\varphi_n}\mathcal{H} = -J[\sin(\varphi_{n+1}-\varphi_n) - \sin(\varphi_n-\varphi_{n-1})] + \sin(\varphi_{n+2}-\varphi_n) - \sin(\varphi_n-\varphi_{n-2}) + h\sin\varphi_n = 0. \tag{3}$$

In principle, these have to be solved and the solutions should be classified in order to identify relative and absolute minima; not an easy task. Note that these equations determine a stationary solution $\{\varphi_n\}$ from any four arbitrary consecutive angles, so that infinite stationary configurations can be devised. To obtain a recursive map, as in the cited papers, one can set $\alpha_n \equiv \varphi_n - \varphi_{n-1}$, $\beta_n \equiv \alpha_{n+1} + \alpha_n$, and $s_n \equiv \sin\beta_n - \sin\beta_{n-1}$, so that Eqs. (3) together with these definitions take the form of a $\mathbb{R}^4 \to \mathbb{R}^4$ map

$$\begin{aligned} s_{n+1} &= -s_n + J\sin(\beta_n-\alpha_n) - J\sin\alpha_n - h\sin\varphi_n,\\ \beta_{n+1} &= \sin^{-1}(\sin\beta_n + s_{n+1}),\\ \alpha_{n+1} &= \beta_n - \alpha_n,\\ \varphi_{n+1} &= \beta_n - \alpha_n + \varphi_n. \end{aligned} \tag{4}$$

The map is invariant under reflection $(s, \beta, \alpha, \varphi) \to (-s, -\beta, -\alpha, -\varphi)$ and has a fixed point (0,0,0,0) that identifies the uniform PM phase, stable for $h \geqslant h_c$; for $h = 0$ one has also the helix fixed point, $(0, 2k, k, nk)$. It is not the aim of this Comment to push forward with this analysis in the hard terrain for $0 < h < h_c$, enclosed between the regular k-pitch helix ($h = 0$) and the PM phase ($h = h_c$).

On the other hand, since any minimum configuration must be a solution of the above map, an easier task would be checking to what extent the "constrained" minimum configurations obtained in [2] are compatible with Eq. (3) or (4). In the same way, as one can easily obtain stationarity equations for the three-dimensional spin model (1), an analogous test for the minimum-energy configurations guessed in [1] can be devised.

In conclusion, the purpose of this Comment is to draw attention to the fact that papers [1] and [2] deal with the problem of the phase diagram of the one-dimensional frustrated magnet in a relatively naïve way and their results have to be handled with caution. Anyhow, the frustrated magnet described by (1) is an intriguing and challenging subject that deserves further research.

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