$U(1)$ symmetry breaking and violated axial symmetry in $TlCuCl_3$ and other insulating spin systems

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We describe the Bose-Einstein condensate of magnetic bosonic quasiparticles in insulating spin systems using a phenomenological standard functional method for $T=0$. We show that results that are already known from advanced computational techniques immediately follow. The inclusion of a perturbative anisotropy term that violates the axial symmetry allows us to remarkably well explain a number of experimental features of the dimerized spin-1/2 system $TlCuCl_3$. Based on an energetic argument we predict a general intrinsic instability of an axially symmetric magnetic condensate toward a violation of this symmetry, which leads to the spontaneous formation of an anisotropy gap in the energy spectrum above the critical field. We, therefore, expect that a true Goldstone mode in insulating spin systems, i.e., a strictly linear energy-dispersion relation down to arbitrarily small excitations energies, cannot be observed in any real material.

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I. INTRODUCTION

The concept of the Bose-Einstein condensation (BEC), i.e., the occupation of a single-quantum state by a macroscopic number of bosons, has been extended from real bosonic particles to various types of quasiparticles with integer total spin.[1–11] Such discrete magnetic, electronic, or lattice excitations are then treated as a set of bosons forming a Bose gas.[9] These quasiparticles usually possess a small effective mass, which permits one to study BEC even at room temperature.[11] Both experiment[10,12] and theory[13,14] suggest that the occurrence of a BEC in a three-dimensional (3D) interacting Bose system has its origin in the spontaneous breaking of the $U(1)$ symmetry, thereby leading to a gapless and linear excitation spectrum in the long-wavelength limit, i.e., to a Goldstone mode.[15]

Experimental observations in a number of quantum spin systems can be explained within the theory of BEC, e.g., by a condensation of triplet states in dimerized spin-1/2 systems (hereafter called triplons) such as $TlCuCl_3$.[3,4,16] The bosonic character of these magnetic quasiparticles allows one to describe this spin-dimer system as a weakly interacting Bose gas. Inelastic neutron-scattering measurements in the condensate phase of $TlCuCl_3$ revealed, in accordance with theoretical investigations,[17] the presence of a seemingly gapless and linear excitation spectrum down to very low excitation energies of the order of 0.75 meV in $\mu_0 H = 14$ T.[4] This observation has been interpreted as a manifestation of the Goldstone mode.

In the last years various scenarios for the consequences of anisotropy on the properties of a magnetic BEC have been discussed in detail.[18–22] The presence of any kind of anisotropy will, in principle, explicitly break the rotational (i.e., axial) symmetry of the bosonic system.[21] The degree of spontaneous $U(1)$ symmetry breaking then depends on the order of magnitude of the anisotropic terms compared to the energy scale associated with the isotropic interactions of the Bose gas.[22] The relatively large triplon bandwidth in $TlCuCl_3$ (Ref. 23) exceeds the spin-nonconserving terms, such as an intradimer exchange (IE) anisotropy and a Dzyaloshinsky-Moriya (DM) anisotropy, by more than 2 orders of magnitude.[18,19,21] Nonetheless, the question to what extent the existing anisotropies in $TlCuCl_3$ do affect the magnetic phase diagram, the Goldstone mode, and other measurable quantities of $TlCuCl_3$ is still an issue under investigation.[18,19,21,24]

By taking a perturbative anisotropy term into account we will consider in the following the influence of an IE-like anisotropy that explicitly violates the axial symmetry, and we will study the consequences on the condensate phase of $TlCuCl_3$. The influence of a possible DM-type anisotropy[20,21] is not considered here.[25] Based on an energy consideration we will then argue that, as a consequence of an unavoidable magnetoelastic coupling, even an axially symmetric magnetic system is unstable toward a spontaneous violation of this symmetry as soon as the BEC state is formed.

II. FUNCTIONAL METHOD

We describe the condensate at $T=0$ with a macroscopic wave function, a complex scalar field $\psi(\mathbf{r},\mathbf{t})$. Standard functional methods used to describe a dilute Bose gas in the classical limit at $T=0$ yield an extremal condition for the potential energy per dimer,[14] namely,

$$u(\psi) = -\mu\psi^{\dagger}\psi + \frac{v_0}{2}(\psi^{\dagger}\psi)\Big|^2_{\psi=\psi_0} = \min, \qquad (1)$$

where μ is the chemical potential, v_0 is a constant related to a repulsive short-range interaction,[13] and $\psi^{\dagger}$ is the complex conjugate of ψ. The minimum value ψ_0 then determines the condensate fraction $n_c(0)=\psi_0^{\dagger}\psi_0$, here defined as $n_c(0)=N_c/N_d$, with N_c the number of condensed triplons and N_d the number of dimers.

In a dimerized antiferromagnet, the chemical potential is $\mu = g\mu_B\mu_0(H - H_c)$, where μ_B is the Bohr magneton, g is the Landé g factor, and H_c is the critical magnetic field beyond which a triplet $S=1$ state is energetically equally favorable as the singlet $S=0$ state. This can be expressed in terms of the energy gap $\Delta = g\mu_B\mu_0 H_c$ separating at zero field the $S=1$ and the $S=0$ states, respectively.

In the case of an explicitly violated axial symmetry we may include a perturbation term $|\tilde{\gamma}|(\psi\psi + \psi^\dagger\psi^\dagger)$ (Refs. 1 and 21) to the potential energy that can arise in a real magnetic system from various sources such as anisotropic intradimer and interdimer interaction constants, J and $\tilde{J}$, respectively. For $TlCuCl_3$ we have, for example, $\mu_0 H_c \approx 5.6$ T,[26] $v_0/k_B = 315$ K,[27] and a $|\tilde{\gamma}|$ of the order of 0.01 meV,[18,21] depending on the orientation of the magnetic field $\mathbf{H}$ with respect to the crystal lattice.[18] Such an anisotropy term may arise from a pre-existing violated axial symmetry of the system or from a spontaneous distortion at the magnetic phase transition that we will discuss below. We, therefore, have to minimize

$$u(\psi) = -\mu\psi^\dagger\psi + |\tilde{\gamma}|(\psi\psi + \psi^\dagger\psi^\dagger) + \frac{v_0}{2}(\psi^\dagger\psi)^2, \qquad (2)$$

where we assume that H_c itself is at first unchanged by the presence of the small perturbative anisotropy $|\tilde{\gamma}| \ll \Delta$.

We first want to compare the results of this simple formalism with corresponding predictions from advanced Hartree-Fock (HF) computations and with experimental data on $TlCuCl_3$. Despite the simple formalism used here, we can later make specific predictions that would otherwise be more difficult to obtain.

III. RESULTS

A. Comparison with results from the Hartree-Fock calculations

Without any explicit anisotropy (i.e., $|\tilde{\gamma}|=0$) we obtain the well-known minimum value $\psi_0^\dagger\psi_0 = n_c(0) = \mu/v_0$.[3] The phase ϕ of $\psi_0 = |\psi_0|e^{i\phi}$ is not fixed in this case, leading to the $u(\psi)$ landscape sketched in Fig. 1 (left panel, "Mexican-hat potential"). However, any nonzero value for $|\tilde{\gamma}|$ locks the phase of ψ_0 to the imaginary axis (i.e., $\phi = \pm\pi/2$ and therefore ψ_0 =const) and leads to an optimum value $n_c(0) = (\mu + 2|\tilde{\gamma}|)/v_0$ in "Napoleon's hat potential" (see Fig. 1, right panel). The minimum potential energy becomes $u_{\min} = -(\mu + 2|\tilde{\gamma}|)^2/2v_0$ which is smaller than in the axially symmetric case. The saddle-point value for u on the real axis for $\mu > 2|\tilde{\gamma}|$ is $-(\mu - 2|\tilde{\gamma}|)^2/2v_0$. Note that these energy densities are all expressed per dimer. The corresponding energies per triplon are $u/n_c(0)$.

A vanishing $n_c(0)$ is realized when $(\mu + 2|\tilde{\gamma}|) = 0$. As a consequence, the gap field H_c that would be observed in an ideal system with axial symmetry is renormalized to a value $H_c^{\text{expt}} = H_c - \Delta H_c$ (with $\Delta H_c = 2|\tilde{\gamma}|/g\mu_0\mu_B$) above which condensation occurs. Taking a reasonable value for $|\tilde{\gamma}| \approx 0.01$ meV for $TlCuCl_3$ (Refs. 18 and 21) and $\mathbf{H} \| b$ with $g = 2.06$,[18] we obtain a renormalization of the critical field due to $|\tilde{\gamma}|$ alone by $\mu_0\Delta H_c \approx 0.2$ T for this particular magnetic-field direction.

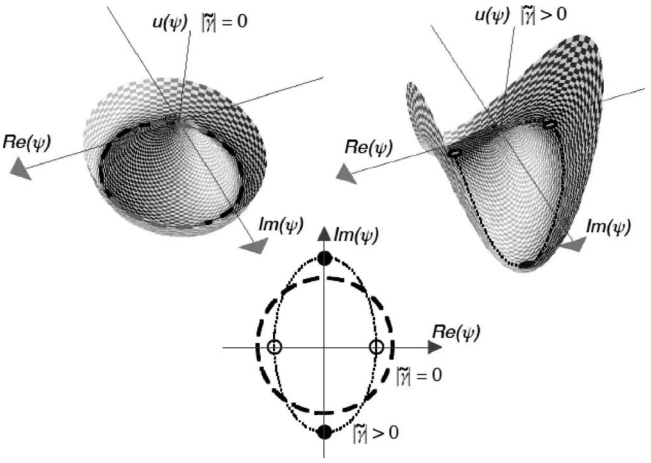

FIG. 1. Potential energy u as a function of ψ for axial symmetry (left panel, Mexican-hat potential) and violated axial symmetry (right panel, Napoleon's hat potential), respectively. In the symmetric case the minimum value $u_{\min} = -\mu^2/2v_0$ is realized along a circle (dashed line), while in the anisotropic case isolated minima $u_{\min} = -(\mu + 2|\tilde{\gamma}|)^2/2v_0$ are on the imaginary axis (filled circles).

The resulting condensate fraction, $n_c(0) = \mu^{\text{expt}}/v_0$, where $\mu^{\text{expt}} = g\mu_B\mu_0(H - H_c^{\text{expt}})$, is in full agreement with Hartree-Fock calculations for spin dimer systems.[3] In Fig. 2(a) we show the triplon condensate fraction $n_c(0)$ as deduced from our magnetization $M(T,H)$ data of $TlCuCl_3$,[28] see Fig. 2(b), that we have already corrected for a small fraction $\tilde{n}$ of noncondensed triplons.[3] These data have been obtained from the simple relation $M(0) = g\mu_B n(0) N_d$ [with N_d as the number of dimers and $n(0)$ as the total triplon fraction at $T=0$],[3] without assuming any specific value for v_0. A linear fit to the data in the dilute limit $[\mu_0 H_c < \mu_0 H \lesssim 7.5$ T (Ref. 21)] yields $\mu_0 H_c^{\text{expt}} = 5.501 \pm 0.003$ T and $v_0/k_B = 311.4 \pm 0.5$ Km^3 which is in very good agreement with available literature values.[21,27,29] The deviation from the linear behavior at larger magnetic fields can be attributed to the contribution of higher triplet states.[5] Our simple formalism does not include the influence of such triplet states nor does it allow for a determination of $\tilde{n}$ itself; but this latter correction is of the order of a few percent at most in our data,[3,28] as it is typical for a weakly interacting Bose gas, see inset of Fig. 2(a).

Focusing further on the effects of an explicit violation of axial symmetry, the condensate fraction at $T=0$ is changed by $2|\tilde{\gamma}|/v_0$ in our calculation, see Figs. 2(c) and 2(d). If we use the fact that the total triplon fraction $n(0) \approx n_c(0)$ and take the result $n(T_c^{\text{BE}}) = n(0)/2$ from Ref. 3 assuming a quadratic triplon dispersion relation, we obtain a shift $|\tilde{\gamma}|/v_0$ in $n(T_c^{\text{BE}})$ that is again in full agreement with the corresponding HF calculations,[21] see again Figs. 2(c) and 2(d).

We may relate the minimum value $u_{\min}$ at $T=0$ to the transition temperature T_c^{BE} if we assume that $|u_{\min}|/n_c(0)$, the energy gain per triplon upon condensation, is proportional to $k_B T_c^{\text{BE}}$ with a field-dependent proportionality factor of the order of unity. Any nonzero $|\tilde{\gamma}|$ leads to an increase in T_c^{BE} as compared to the axially symmetric case, see Fig. 2(d). This trend can be clearly seen in the calculated $M(T,H)$ curves from Ref. 21, where the minimum in M that is usually taken as a criterion to define T_c^{BE} is shifted toward higher tempera-

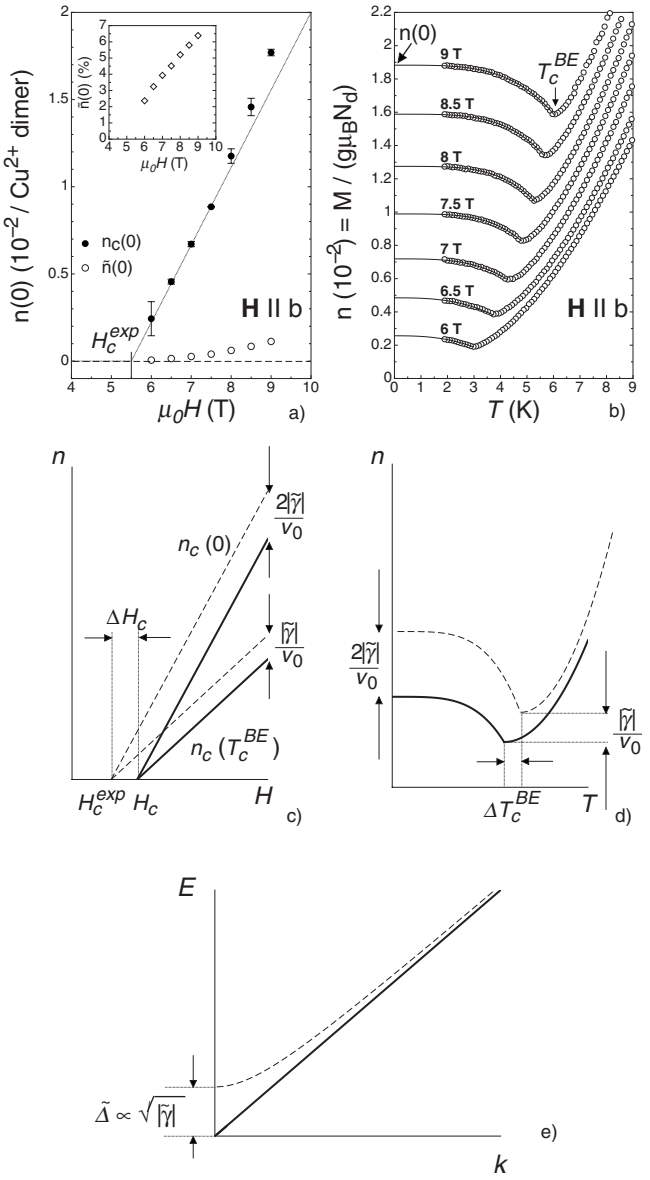

FIG. 2. (a) Condensate fraction $n_c(0)$, noncondensed triplon fraction $\tilde{n}(0)$, and the percentage of noncondensed triplons (inset) (Ref. 28). (b) Triplon-fraction $n(0)$ data obtained from magnetization $M(T)$ data for $\mathbf{H}\|b$ (see arrow for the $\mu_0 H=9$ T data and Ref. 28). (c) Schematic representations of the effects of violated axial symmetry on the triplon fraction at $T=0$ and at $T=T_c^{\mathrm{BE}}$, (d) on the triplon fraction at fixed magnetic field $H>H_c$ (Refs. 3, 20, and 21), and (e) on the excitation spectrum $E(k)$ (axes are not to scale). Solid and dashed lines represent an axially symmetric system and a system with violated axial symmetry, respectively.

tures as soon as $|\tilde{\gamma}|>0$. If we again take $|\tilde{\gamma}|=0.01$ meV we obtain a shift in T_c^{BE} of the order of $\Delta T_c^{\mathrm{BE}} \approx |\tilde{\gamma}|/k_B \approx +0.1$ K, which has to be compared to the result of the more precise HF calculations with $\Delta T_c^{\mathrm{BE}} \approx +0.5$ K.[21]

As a consequence of the anisotropy term $|\tilde{\gamma}|$ the original invariance of $u(\psi)$ with respect to a transformation $\psi \rightarrow \psi e^{i\phi}$ is lifted. The maximum variation in the potential energy per dimer u along the ellipsoid contour with local minima in the radial $|\psi|$ direction, see Fig. 1, is $4\mu|\tilde{\gamma}|/v_0$ or $\tilde{\Delta} \approx 4|\tilde{\gamma}|$ per ground-state triplon. This determines the order of magnitude of an anisotropy gap that can be calculated to $\tilde{\Delta}=\sqrt{8|\tilde{\gamma}| g\mu_B \mu_0 (H-H_c)}$.[21,30]

Excitations above the ground state ψ_0 with energies below $\tilde{\Delta}$ will clearly not show the typical gapless Goldstone-type behavior as expected for an axially symmetric system. For $TlCuCl_3$ we calculate, with $|\tilde{\gamma}|=0.01$ meV,[18,21] a gap $\tilde{\Delta} \approx 0.3$ meV for $\mu_0 H=14$ T along b, which is somewhat below what has been resolved in inelastic neutron-scattering measurements.[4] For excitation energies larger than $\tilde{\Delta}$ the presence of an anisotropy gap may remain unnoticed, see Fig. 2(e).

In $TlCuCl_3$ such a gap may arise from a pre-existing anisotropy that is already present in $H=0$.[19] In the following we argue, however, that even a perfectly axially symmetric magnetic system is unstable toward a spontaneous violation of this symmetry at H_c, which inevitably leads to the formation of a small anisotropy gap $\tilde{\Delta}$ above H_c of real materials.

B. Instability of the condensate toward violation of axial symmetry

A striking fact in our analysis is that the minimum potential energy per dimer is *smaller* with a nonzero $|\tilde{\gamma}|$ than in an analogous axially symmetric system with $|\tilde{\gamma}|=0$. This means that a distortion of the original crystal symmetry may spontaneously occur at H_c together with an increase in $|\tilde{\gamma}|$, provided that the total energy, including both magnetic and crystal-lattice contributions, is lowered along with this distortion. This argument is so general that it should be applicable to all insulating spin systems that are supposed to show a Bose-Einstein condensation of magnetic bosonic quasiparticles. As we do not make any specific assumptions on the microscopic arrangement of the spin-carrying atoms, one cannot make any more precise universal statement about the details of the resulting lattice distortion.

The gain in potential energy per dimer upon condensation in combination with this simultaneous distortion is $2|\tilde{\gamma}|^2/v_0$, see Fig. 3(a). If the critical field is approached from below with H increasing, the parameter $|\tilde{\gamma}|$ may therefore jump discontinuously from zero to its optimum value either at H_c or at a transition field $H_c^{\star}$ with $H_c^{\mathrm{expt}} \leq H_c^{\star} \leq H_c$, while with H decreasing the transition can take place at a different field in the same magnetic-field interval, see gray shaded area in Figs. 3(a) and 3(b). In an ideal situation with a perfect axial symmetry in $H=0$, the critical field H_c corresponds to a "normal-state" value with $|\tilde{\gamma}|=0$ that is determined only by the gap energy, while in the condensate phase the effective critical field is $H_c^{\mathrm{expt}}=H_c-\Delta H_c<H_c$, lowered by $\Delta H_c=2|\tilde{\gamma}|/g\mu_B\mu_0$ with respect to H_c due to the increase in $|\tilde{\gamma}|$. If the transition occurs at a transition field $H_c^{\star}$ that is strictly larger than H_c^{expt}, one will observe at $T=0$ corresponding small discontinuities in u [$\Delta u \leq 2|\tilde{\gamma}|^2/v_0$, see Fig. 3(a)], n_c [$\Delta n_c \leq 2|\tilde{\gamma}|/v_0$, see Fig. 2(c)], and M ($\Delta M=\Delta n_c g\mu_B N_d$), which would qualify the transition as of weakly first order with a maximum observable hysteresis width ΔH_c. The occurrence of hysteretic effects in a real material may depend, however, on further conditions that are not considered here, such as material-quality issues or the relevance of possible

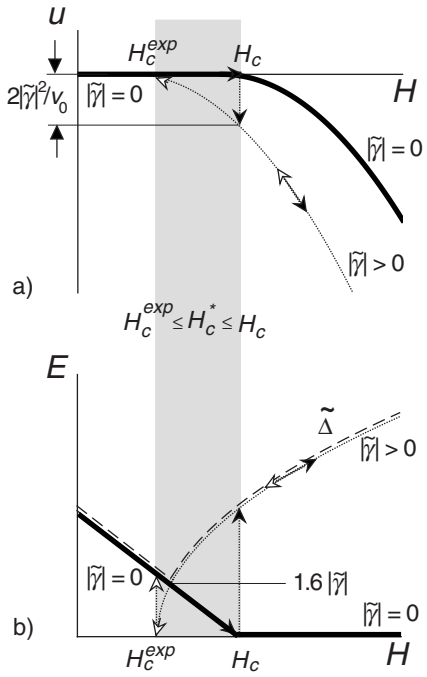

FIG. 3. (a) Potential energy per dimer and (b) energy-level scheme of the lowest triplon branch of an axially symmetric system ($|\tilde{\gamma}|=0$, thick solid lines) and of a system that shows a spontaneous axial distortion ($|\tilde{\gamma}|>0$) above $H_c^{\star}$ (thin lines and arrows, axes are not to scale), respectively. The gray shaded area indicates the maximum hysteresis width of a possible weakly first-order transition at $H_c^{\star}$ with $H_c^{\mathrm{expt}} \le H_c^{\star} \le H_c$ (filled arrows: H increasing; open arrows: H decreasing). The dashed line in (b) reproduces the trend in the ESR data from Refs. 18 and 19 for $TlCuCl_3$ and $\mathbf{H} \| b$ (see text).

quantum fluctuations at $T=0$ to the order of the phase transition.

Even if a nonzero $|\tilde{\gamma}|$ evolves continuously above the critical field as a function of H along with a continuous structural distortion with no detectable hysteresis, one should still be able to distinguish between the normal-state H_c extracted from the experimental data taken below H_c^{expt} and a $H_c^{\mathrm{expt}} < H_c$ from corresponding data taken well above H_c, respectively, to obtain an estimate for $|\tilde{\gamma}|$ from the resulting difference ΔH_c. In any case, if $H_c^{\mathrm{expt}} < H_c^{\star} < H_c$, one expects a finite-energy difference $\le 1.6|\tilde{\gamma}|$ at the transition field $H_c^{\star}$ between the lowest-triplon state and the singlet states, respectively, see Fig. 3(b).

A possible pre-existing anisotropy that may already be present at $H=0$ (which is likely to be the case in $TlCuCl_3$) can be easily included in this formalism by identifying H_c with a renormalized normal-state value that already contains this pre-existing anisotropy. Any additional $|\tilde{\gamma}|$ that may develop together with the lattice distortion around this critical field will somewhat change the value of the anisotropy gap $\tilde{\Delta}$. The difference ΔH_c, however, and possible discontinuities in u, n_c, and M are determined by the additional $|\tilde{\gamma}|$ alone.

It is remarkable that electron-spin resonance (ESR) data taken on $TlCuCl_3$ do indeed show a clearly gapped behavior at and above $\mathbf{H}=H_c^{\mathrm{expt}} \| b$,[18,19] as we sketched in Fig. 3(b). Moreover, the ESR frequencies due to the lowest triplon gap in the normal phase for $\mathbf{H} \| b$ extrapolate to zero at a somewhat larger H_c (by $\mu_0 H_c \approx 0.2$ T) than the square-root-like gap that we attribute to $\tilde{\Delta}(H)$ in the condensate phase and that can be fitted nicely with a $|\tilde{\gamma}| \approx 0.016$ meV, see energy-level scheme in Fig. 3(b). Our scenario may also be an explanation for the observed abrupt changes in the ^{35}Cl quadrupole shift[24] that has been interpreted as an indication of a weakly first-order lattice deformation, as well as for the pronounced hysteretic behavior (with a $\mu_0 \Delta H_c \approx 0.2-0.3$ T) of the observed peaks in the sound-attenuation data of $TlCuCl_3$ at $H_c(T)$.[31] These observations may indicate that $|\tilde{\gamma}|$ of $TlCuCl_3$ is indeed larger for $H > H_c$ than well below this value.

The present picture may also account for the first-order-like features that have been seen in the x-ray data of the spin-ladder compound $Cu_2(C_5H_{12}N_2)_2Cl_4$ (Ref. 32) and in the magnetocaloric effect of the axially symmetric $S=1$ system $NiCl_2$-4SC$(NH_2)_2$ (Ref. 33) at the respective magnetic phase transitions. It is also not unreasonable to assume that the observed gap feature in the ESR data of this latter compound at $\mu_0 H=8$ T (Ref. 34) is also related to a possible lattice distortion.

Such a spontaneous distortion arising from the interplay between the emerging magnetic Bose-Einstein condensate and its host crystal that lowers the total energy, with a tendency to increase or *even create* an anisotropy perpendicular to the external magnetic field above H_c even in a perfectly axially symmetric system, is rather unique and is reminiscent of the spin-Peierls instability in one-dimensional magnetic chains. A spin-Peierls-type scenario has indeed been suggested to explain the NMR data on the spin-ladder compound $Cu_2(C_2H_{12}N_2)_2Cl_4$ around its critical magnetic field.[35,36] Such an instability should be a universal feature of magnetic BEC systems at their magnetic phase transition, and it is not expected to occur in axially symmetric Bose gases composed of real particles such as superfluid ^{4}He or atomic condensates, where the condensate cannot create an axial-symmetry-breaking term by itself.

In a microscopic picture, the tendency of a magnetic condensate to spontaneously violate the axial symmetry can be interpreted as a natural consequence of the transverse magnetic ordering that develops in the condensate phase and that locks to the crystal lattice due to unavoidable magnetoelastic coupling.[3,37,38] As soon as the transverse magnetic moments point to a specific energetically preferred crystal direction, the phase ϕ that is associated with the angle between these moments and the crystal axes[16] is indeed fixed [in $TlCuCl_3$ with an angle $\alpha \approx 39°$ to the a axis (Ref. 37)], and the magnetic analog to a supercurrent velocity $\mathbf{v}_s = \hbar / m^{\star} \nabla \phi$ (where $m^{\star}$ is the effective mass of a triplon) is naturally zero for excitation energies below the anisotropy gap $\tilde{\Delta}$. This gap covers an excitation-frequency range that is crucial for experiments that rely on the existence of a long-lived phase-coherent condensate,[9] such as the detection of a long-lived spin supercurrent as in ^{3}He–B,[39,40] of macroscopic second-sound-like oscillations as observed in superfluid ^{4}He,[41] or of stable vortex-like structures as they have been observed in superfluid ^{4}He and in atomic condensates.[42] To achieve a lifetime of the order of seconds for a phase-coherent condensate, a corresponding anisotropy gap may not exceed a few femtoelectron volts.

In the absence of a Goldstone mode (for which all values of the phase ϕ have to be energetically equivalent), the quantity $\psi_0^\dagger \psi_0$ in the zero-frequency limit does no longer represent a condensate fraction, but it is rather related to the order parameter characterizing the antiferromagnetic state.[1]

IV. CONCLUSIONS

We have analyzed the spontaneous symmetry breaking in a Bose gas of magnetic bosonic quasiparticles in insulating spin systems based on simple functional methods in the classical limit. Our results reproduce several results from earlier HF approximated computations,[3,20,21] and various experimental findings in $TlCuCl_3$ such as the occurrence and the magnitude of an anisotropy gap[18,19] and a weakly first-order-like behavior at the magnetic phase transition[24,31] can be explained. On the basis of an energetic argument we expect that all magnetic BEC systems in insulating spin systems are intrinsically unstable toward a spontaneous anisotropic distortion perpendicular to the external magnetic field, which leads to the formation of an anisotropy gap that is seriously limiting the lifetime of a phase-coherent condensate.

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