Consequences of spin-orbit coupling for the Bose-Einstein condensation of magnons

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Abstract. – In the first part we discuss how the BEC picture for magnons is modified by anisotropies induced by spin-orbit coupling. In particular we focus on the effects of antisymmetric spin interactions and/or a staggered component of the $g$ (gyromagnetic) tensor. Such terms lead to a gapped quasiparticle spectrum and a nonzero condensate density for all temperatures so that no phase transition occurs. We contrast this to the effect of crystal field anisotropies which are also induced by spin-orbit coupling. In the second part we study the field-induced magnetic ordering in TlCuCl$_3$ on a quantitative level. We show that the usual BEC picture does not allow for a good description of the experimental magnetisation data and argue that antisymmetric spin interactions and/or a staggered $g$ tensor component are still crucial, although both are expected to be tiny in this compound due to crystal symmetries. Including this type of interaction we obtain excellent agreement with experimental data.

Introduction. – It has been argued that the phase transition at a critical applied magnetic field in certain spin systems with an excitation gap $\Delta$ as for example integer-spin antiferromagnetic chains [1], weakly coupled two-leg ladders [2] or three-dimensional dimer systems [3,4] may be regarded as a Bose condensation. In these systems the lowest excited state is a triplet of massive bosons. A magnetic field $H$ causes a Zeeman splitting of the triplet with the lowest mode crossing the ground state at a critical field $H_c = \Delta/g\mu_B$. The ground state for $H > H_c$ then becomes a BEC of this low-energy boson. The density $n$ of the boson mode is directly related to the magnetisation per site $m = g\mu_B n$. In principle, spin-gap compounds offer therefore the exciting possibility to study BEC in a system where the density $n$ is tunable by the external magnetic field which acts as a chemical potential for the bosons. However, as the triplet excitations (magnons) interact via a hard-core repulsion a description as a weakly interacting Bose gas is only meaningful if the average distance between the magnons $l \sim a^{-1/3}$ is much larger than the s-wave scattering length $a$ which is the characteristic length scale representing the influence of the repulsive potential. This implies that $a/l \sim n^{1/3}a \ll 1$ so that the magnons have to be dilute. In this case the well-established gas approximation [5] which involves a systematic expansion in terms of the small parameter $n^{1/3}a$ is applicable and even the finite temperature properties of the interacting Bose gas can be studied analytically [6].
A dilute magnon gas is realized in the spin dimer system TlCuCl$_3$ in magnetic fields $H \sim 6 - 7$ T and considerable interest has focused onto this compound within recent years [3,4,7–10]. TlCuCl$_3$ has an excitation gap $\Delta \approx 0.7$ meV in zero magnetic field and a bandwidth $W \sim 6.3$ meV [8]. The dimers in this compound are formed by the $S = 1/2$ spins of the Cu$^{2+}$ ions and superexchange interactions are mediated by the Cl$^-$ ions. The crystal structure can be considered as two-leg ladders formed by these dimers. However, inelastic neutron scattering (INS) [8] has revealed that the magnons show a considerable dispersion in all spatial directions indicating that the interladder interactions are strong. TlCuCl$_3$ therefore has to be considered as a three-dimensional (3D) interacting dimer system. For fields $H \gtrsim H_c \sim 5.6$ T a long-range magnetic ordering below some critical temperature $T_c$ has been detected. It has been shown that this transition and the overall shape of the magnetisation curves as a function of temperature can be qualitatively described as the BEC of magnons [3]. Recently, the excitations for $H > H_c$ have been measured by INS and the lowest mode has been interpreted as the gapless Goldstone mode characteristic for a Bose condensed phase [11].

In the first part of this letter we will show how to include antisymmetric spin interactions or a staggered component of the $g$ tensor into a Hartree-Fock-Popov (HFP) treatment [3,6] of an interacting dilute Bose gas. Such terms will in general break the axial $U(1)$ symmetry of the system so that a Goldstone mode no longer exists. Furthermore the condensate density becomes nonzero for all temperatures so that no phase transition occurs. These findings are in agreement with [12] where the same type of anisotropy in a Haldane spin chain has been considered. We will discuss the differences to the effect of crystal field anisotropies which can also break $U(1)$ symmetry and affect Bose condensation [1]. Our analysis will allow us to investigate the effects of such anisotropies on the magnon density in 3D dimer systems at finite temperatures quantitatively and we present a detailed study of the field-induced magnetic ordering in TlCuCl$_3$ along these lines in the second part of this letter.

**General scenario.** – We want to restrict ourselves here to 3D spin-1/2 systems where the spin gap is due to some kind of explicit dimerisation. A useful approach to describe the excitations in such systems is the bond operator representation for spins introduced in [13]. The starting point is the strong coupling ground state $|s\rangle$ where each dimer at site $i$ is in singlet configuration $|i, s\rangle$. It is then natural to introduce operators $t_{i\alpha}^\dagger$ which create local triplet excitations $|i, \alpha\rangle = t_{i\alpha}^\dagger |i, s\rangle$ with $|i, +\rangle = -| \uparrow\downarrow\rangle$, $|i, -\rangle = | \downarrow\uparrow\rangle$ and $|i, 0\rangle = (| \uparrow\downarrow\rangle + | \downarrow\uparrow\rangle)/\sqrt{2}$. Due to hopping between the dimers the three triplet components acquire a dispersion $\Omega_{k_0} = \Omega_{k_0} - og\mu H$ with a band minimum $\Omega_{q_0, \alpha} = \Delta - og\mu H$ at momentum $q_0$ depending on microscopic details of the exchange interactions. The triplets are subject to a hard-core constraint which can be taken into account by introducing an infinite on-site repulsion

$$H_U = U \sum_{i,\alpha,\beta} t_{i\alpha}^\dagger t_{i\beta}^\dagger t_{i\alpha} t_{i\beta}, \quad U \to \infty$$

with $\alpha, \beta = -, 0, +$. Note that the renormalisation of the triplet dispersion $\Omega_{k_0}$ due to the two-particle scattering vertex $v(\mathbf{k}, \omega)$ corresponding to the interaction (1) can be calculated exactly in the dilute limit by a summation of ladder diagrams [14]. For magnetic fields $H \gtrsim H_c$ and temperatures $T < \Delta$ it is sufficient to take only the lowest triplet mode ($\alpha = +$) into account. In this case only particles near the band minimum at $q_0$ are excited so that the generally energy and momentum dependent $v(\mathbf{k}, \omega)$ can be replaced by a constant $v_0 = v(q_0, 0)$. With the definitions $\epsilon_k \equiv \Omega_{k_0} - \Delta$ and $t_k \equiv t_{k,+}$ the Hamiltonian for the lowest mode is given by

$$H = \sum_k (\epsilon_k - \mu_0) t_k^\dagger t_k + \frac{v_0}{2} \sum_{k,k',q} t_{k+q}^\dagger t_{k'}^\dagger t_q t_{k+k'}$$

(2)
where $\mu_0 = g\mu_B(H - H_c)$. Here we want to consider a perturbation linear in $t, t^\dagger$

$$\mathcal{H}' = i\gamma(t_{q_0} - t^\dagger_{q_0})$$

(3)

where $\gamma$ is a small parameter. This term respects parity and time reversal symmetry and will therefore be non-zero in general if not forbidden by additional crystal symmetries. Clearly its effect will be 

\textit{non-perturbative} at fields $H \sim H_c$ because it mixes the singlet ground state and the triplet soft mode. Physically such a term can originate from Dzyaloshinsky-Moriya (DM) interactions $\sim \mathbf{D} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$ between sites $i, j$ where $\mathbf{D}$ is the DM vector. For illustration we consider the two-leg spin-ladder shown in fig. 1. In the limit $J_\perp \gg J$ the triplet dispersion for this model is given by $\Omega_{k\alpha} = J_\perp + J\cos k - \alpha g\mu_B H$ and has a minimum at $q_0 = \pi$. Consider now the additional antisymmetric exchange

$$\mathcal{H}_{DM} = D_x \sum_i (-1)^i (S^l_{iy} S^r_{iz} - S^l_{iz} S^r_{iy})$$

(4)

where indices $l, r$ label the two spins within the dimer as shown in fig. 1. Using the identity [13] $S^l_{x} = (\pm t_{x} \pm t_{y}^l - i\epsilon_{\alpha\beta\gamma} t_{y}^l t_{z})/2$ with $\alpha = x, y, z$ this is easily transformed into $\mathcal{H}_{DM} = iD_x \sum_i (-1)^i(t_{tx} - t_{ty})/2$. Using spiral instead of cartesian indices we find in momentum space $\mathcal{H}_{DM} \sim iD_x(t_\pi - t_\beta)/2$ where $t$ denotes now the lowest triplet mode ($\alpha = +$) as in (2).

This is exactly of the proposed form (3) with the triplet operators $t_\pi^{(1)}$ acting at the band minimum. It can be shown by an exact transformation that the considered DM interaction produces an effective staggered field if a uniform magnetic field is applied [15]. A staggered magnetic field can also originate directly from a staggered $g$ tensor [12]. Both effects are induced by spin-orbit coupling and can contribute to (3). Note, however, that the interaction itself is forbidden by symmetry if there is an inversion center in the middle of each dimer irrespective of the origin of this term.

Next we diagonalize (2) with the perturbation (3) included, treating the interaction between non-condensed magnons in the one-loop (HFP) approximation. We want to emphasize again that taking only these diagrams into account is not an uncontrolled approximation but instead the first order in a systematic expansion in the gas parameter $n$. First, we introduce new operators $c_k$ by $t_k = c_k + i\delta_k q_0 \eta$ where $\eta$ is a real number. The density of condensed magnons per dimer $n_0$ is then given by $n_0 = \eta^2$. Ignoring a momentum independent term, the Hamiltonian splits into 2 parts $\mathcal{H} = \mathcal{H}_{\text{lin}} + \mathcal{H}_{\text{bilin}}$ with $\mathcal{H}_{\text{lin}} = i(2\tilde{n} \eta \eta + v_0 \eta^3 - \mu_0 \eta - \gamma)(c_{q_0}^\dagger - c_{q_0})$ where $\tilde{n}$ denotes the density of non-condensed magnons per dimer and

$$\mathcal{H}_{\text{bilin}} = \sum_k \{A_k c_k^\dagger c_k - \Sigma_{12}(c_k^\dagger c_k + h.c.)/2\}.$$  

(5)

Here $A_k = \epsilon_k - \mu_0 + \Sigma_{11}$ with the normal self-energy $\Sigma_{11} = 2v_0 \tilde{n} + 2v_0 \eta^2$ whereas the anomalous self-energy is given by $\Sigma_{12} = v_0 \eta^2$. By a Bogoliubov transformation we find the quasiparticle...
spectrum $E_k = (A_k^2 - \Sigma_{12}^2)^{1/2}$. Additionally we have to demand that $\mathcal{H}_{\text{lin}}$ vanishes

$$-\mu_0 \eta - \gamma + \eta (\Sigma_{11} - \Sigma_{12}) = 0. \quad (6)$$

To stress the differences between BEC with and without $\mathcal{H}'$ we first briefly summarise the case $\gamma = 0$ which is the usual BEC scenario for a weakly interacting Bose gas in the HFP approximation. In this case we have to distinguish further between the case $\eta = 0$, i.e., no magnons are condensed and the case with nonzero condensate density $\eta \neq 0$. For $\eta = 0$ the subcondition $\mathcal{H} = 0$ is identically fulfilled, the anomalous self-energy $\Sigma_{12}$ vanishes and the quasiparticle spectrum is identical to the bare triplet spectrum with an effective chemical potential $\mu = \mu_0 - 2v_0 \tilde{n}$. The density of magnons in this phase is therefore given by the usual Bose distribution $\tilde{n} = (1/N) \sum_k [\exp(\beta(\epsilon_k - \mu)) - 1]^{-1}$. BEC occurs when the chemical potential $\mu$ vanishes so that the density $n_c$ at the critical point is given by $n_c = \mu_0/2v_0$. At temperatures below the critical point $(T < T_c)$ $\eta$ will be nonzero and the condition $\mathcal{H} = 0$ for $\gamma = 0$ becomes equivalent to the Hugenholtz-Pines theorem [6] $\mu_0 = \Sigma_{11} - \Sigma_{12}$ and guarantees the existence of a Goldstone mode, i.e., the quasiparticle spectrum $E_k = \sqrt{\epsilon_k^2 + 2\epsilon_k n_0 v_0}$ is gapless. The number of non-condensed magnons $\tilde{n}$ in this phase is given by

$$\tilde{n} = -\frac{1}{2} + \frac{1}{N} \sum_k \frac{\epsilon_k + n_0 v_0}{2E_k} \coth \left( \frac{\beta E_k}{2} \right) \quad (7)$$

whereas the number of condensed magnons can be determined from eq. (6) which has to be solved self-consistently together with (7).

Now we want to compare this with the case $\gamma \neq 0$. From eq. (6) it follows immediately that this changes the situation qualitatively irrespective of the magnitude of $\gamma$ because $\eta$ must be nonzero for all temperatures so that there will be always condensed magnons and no phase transition will occur. Furthermore there is no longer a Hugenholtz-Pines theorem and consequently the quasiparticle spectrum

$$E_k = \sqrt{(\epsilon_k + \gamma)/\sqrt{\eta}^2 + 2(\epsilon_k + \gamma)/\sqrt{\eta} v_0 n_0} \quad (8)$$

is gapped. This is expected because the component of the DM interaction perpendicular to the applied field $H$ breaks the $U(1)$ symmetry so that there is no longer a Goldstone mode. Using the spectrum (8), the non-condensed magnon density $\tilde{n}$ can be again calculated by eq. (7) with $\epsilon_k$ being replaced by $\epsilon_k + \gamma/\sqrt{\eta}$. The condensed density is determined by eq. (6) so that there are again two equations which have to be solved self-consistently.

Finally we want to discuss the difference between the perturbation $\mathcal{H}$ originating from DM interactions or a staggered $g$ tensor and single-ion anisotropies $\sim D(S_i^z)^2 + E[(S_i^x)^2 - (S_i^y)^2]$. (Exchange anisotropies have the same effect as single-ion anisotropies) In terms of triplet operators a single-ion anisotropy produces a perturbation bilinear in the triplet operators $\mathcal{H}_{\text{pert}} = \tilde{\gamma}(t_k t_{-k} + h.c.)$ which results from the $E$-term provided that the magnetic field is along the $z$-axis. Here $\tilde{\gamma}$ is again a small parameter and we assume $\tilde{\gamma} > 0$ without loss of generality. Consequently the vanishing for the linear terms now becomes $-\mu_0 \eta - 2\tilde{\gamma} \eta + \eta (\Sigma_{11} - \Sigma_{12}) = 0$ and $\tilde{\gamma} \neq 0$ does not imply $\eta \neq 0$ as before. Therefore a phase transition will still occur and $n_0 = 0$ for $T > T_c$. The quasiparticle gap will vanish exactly at the critical point, however, it will reopen again below $T_c$. Additionally, the critical point itself will be slightly shifted. Although in both cases the quasiparticle spectrum is gapped for $n_0 \neq 0$, the gap only weakly depends on the condensate density for the single-ion anisotropy case contrary to what was found before (see eq. (8)).
Fig. 2 – Experimental magnetisation curves (symbols) for different magnetic fields $H = 5.9, 6.0, 6.1, \cdots, 7$ T taken from ref. [3] compared to the theoretically calculated (solid lines) with $\Delta = 0.67$ meV where (a) $v_0 = 9.8$ meV and (b) $v_0 = 25$ meV. In (c) the result with (3) included is shown where $\gamma = 10^{-3}$ meV, $\Delta = 0.72$ meV and $v_0 = 27$ meV. (d) Quasiparticle spectrum (8) (solid line) for $H = 14$ T ($n_0 \approx 2.3 \cdot 10^{-2}$ at $T \lesssim 1.5$ K) with all other parameters as in (c). INS data for $T = 50$ mK (circles) and $T = 1.5$ K (squares) taken from ref. [11] are shown for comparison.

$\text{TlCuCl}_3$. For $\text{TlCuCl}_3$ detailed magnetisation measurements have been performed for magnetic fields $H \gtrsim H_c$ with typical magnon densities $n \sim 10^{-3}$ [3] satisfying the condition of diluteness. This compound is therefore particularly suited to test the predictions of the BEC theory within the HFP approximation even quantitatively. This is the purpose of the remainder of this letter. $\text{TlCuCl}_3$ crystallises in the space group $P2_1/c$ with two dimers per unit cell formed by the $S = 1/2$ spins of the Cu$^{2+}$ ions [8]. Because the centre of each dimer is an inversion centre, the interaction (3) is forbidden by symmetry making, apparently, a description as a usual BEC valid. To do so it is not sufficient to use a magnon dispersion of the form $\Delta + k^2/2m$ [3]. Although a quadratic dispersion is indeed expected close to the band minimum its applicability is restricted to small excitation energies and we find that this simplification here is only justified for temperatures $T < 1$ K which is well below the experimental temperature range (1). Matsumoto et al. [7] have successfully applied the afore mentioned bond-operator technique to describe the real triplet dispersion in $\text{TlCuCl}_3$. However, we have found that the mean-field treatment of the hard-core constraint (4) used in [7] is not sufficient for a quantitative comparison with the magnetisation measurements in [3]. We therefore have calculated the renormalisation of the dispersion due to the constraint in a systematic way by a summation of ladder diagrams [14]. Doing so we find a considerable renormalisation of the superexchange parameters compared to ref. [7]. In particular this treatment allows us to calculate the scattering amplitude $v_0$ directly and we find $v_0 = 1.6 W = 9.8$ meV [16]. Additionally we note that the gap cannot be determined very accurately from the INS data so that we have used $H_c = 5.6$ T and $g = 2.06$ [17] which yield $\Delta = g\mu_B H_c = 0.67$ meV. This value was used as a constraint when calculating the renormalised dispersion. Having the renormalised spectrum and $v_0$ at hand we can calculate the magnon density and therefore the magnetisation as a function of temperature within the standard BEC picture. As we are interested in temperatures $T \ll v_0$ we can treat $v_0$ as temperature independent. The result is shown in fig. 2a. Quite obviously the calculated magnetisation curves do not agree with the experimental data. We want to point out that although the jump in the magnetisation at

\footnote{The experimentally observed deviation from the universal BEC power law $n_c \sim T_c^{-\alpha}$ with $\alpha = 3/2$ is therefore not astonishing because this exponent is a direct consequence of a quadratic dispersion.}
the transition point is known to be an artefact of the HFP treatment, this approximation is reliable for a dilute Bose gas apart from the small temperature interval \(|T - T_c| \lesssim n^{1/3}aT_c|\) [6] where we find \(n^{1/3}a \lesssim 0.1\). Especially the huge overestimation of the magnetisation at \(T = 0\) is definitely not due to an invalid approximation.

Taking \(v_0\) as a fitting parameter we can obtain good agreement with experiment up to the minima of the magnetisation curves with \(v_0 = 25 \text{ meV}\) yielding the best result shown in fig. 2. A renormalisation of \(v_0\) up to this value is by no means implausible as our calculation of \(v_0\) takes only magnon-magnon interactions into account. Raman spectroscopy, however, has revealed that low-energy optical phonons exist in TlCuCl\(_3\) which interact with the spin system [9]. As a consequence a reduction of the bare magnon bandwidth \(W_0\) due to polaronic effects is expected so that \(v_0\), which is basically given by \(W_0\), might be in fact much larger. A reduction of the bandwidth by a factor 2-3 is not exceptional and known in polaron physics for a long time [18]. However, even when using \(v_0 = 1.6 W_0 = 25 \text{ meV}\) the theoretically calculated magnetisations are still about 50\% too large at \(T = 0\). More important, the standard BEC scenario always predicts a sharp increase of the magnetisation below \(T_c\) which qualitatively disagrees with experiment where only a slight and smooth upturn is visible.

In recent ESR measurements [17] a direct singlet-triplet transition has been observed which would be usually forbidden by spin conservation. This yields some indication that a small perturbation of the form is still present. The reason might be small static distortions which violate the exact inversion symmetry within a dimer so that \(H'\), although certainly tiny, becomes nonzero. We therefore tried to fit the measured magnetisation curves using the outlined BEC theory with \(H'\) included. Because any magnetic field will then cause a finite magnetisation, \(H_c\) is no longer well defined. However, fixing the gap in a range consistent with the INS data we can still use \(H_c = \Delta/g\mu_B\) as a formal definition. With \(\Delta = 0.72 \text{ meV}, v_0 = 27 \text{ meV}\) and a DM interaction \(\gamma = 10^{-3} \text{ meV}\) we can obtain excellent agreement with experiment as shown in fig. 2. Most important, even this extremely small perturbation yields smooth minima and a slow increase of the magnetisation at temperatures below the minima consistent with experiment. If \(\gamma\) is indeed nonzero we expect a gapped quasiparticle spectrum. In fig. 2 we therefore compare the spectrum with the INS data taken from ref. [11]. At low excitation energies our result is in perfect agreement with these data. The deviations at higher energies are expected because our approximation \(v(k, \omega) = v(q_0, 0)\) in eq. (2) is then no longer justified. The predicted gap \(\Delta \sim 0.1 \text{ meV}\) is only a factor 2 smaller than the lowest measured excitation energies making it perhaps accessible in future studies. We also mention that in systems where DM interactions are allowed by symmetry they are typically of the order \(\sim J(g - 2)/g\) where \(J\) is the isotropic superexchange [19]. For a DM interaction within a TlCuCl\(_3\) dimer this estimate yields \(\gamma \sim 0.2 \text{ meV}\) which is two orders of magnitude larger than the term considered here supporting our statement that tiny violations of inversion symmetry are sufficient. However, a nonzero anisotropy term raises questions about the orientation of the DM vector \(D\). If \(D\) would be oriented along a specific axis throughout the crystal as in our example in fig. 1 it would be possible to restore \(U(1)\) symmetry by applying the magnetic field along the same axis. In this configuration a sharp phase transition would be visible. However, such dependencies on field direction have not been reported for TlCuCl\(_3\). Another possibility would be that small static distortions lead to domains with different orientations of \(D\). In this case a component perpendicular to \(H\) could exist for each field direction. Provided that indeed no anisotropy with respect to the field direction exists we believe this is the most probable scenario taking the tininess of the needed DM term into account. In principle it is also allowed by symmetry to construct a DM term from the magnetic field itself, i.e. it is possible that the magnetic field induces the anisotropy and therefore determines the orientation of \(D\). Finally we want to discuss a single-ion or exchange anisotropy term as an
alternative possible perturbation. As already mentioned before a sharp phase transition will still occur with such a term included. Additionally, the quasiparticle gap in the condensed phase will be almost independent of \( n_0 \) and small for a small perturbation. We therefore do not expect any qualitative change for the magnetisation curves. Explicit calculations indeed show that such a term basically yields a small shift of the critical temperature leaving the shape of the magnetisation curves otherwise unchanged (data not shown).

Conclusions. – In summary, we have studied how a term \( \mathcal{H}' \) caused by antisymmetric spin interactions and/or a staggered \( g \) tensor affects BEC in spin-gap systems. This interaction directly mixes the singlet with the triplet soft mode so that its effect is non-perturbative at magnetic fields \( H \sim H_c \). Treating the magnon-magnon interaction in the one-loop approximation we have found a gapped quasiparticle spectrum compared to the gapless spectrum without \( \mathcal{H}' \) and a nonzero condensate density for all temperatures consistent with findings in ref. [12] for Haldane spin chains. Differences to the case of crystal field anisotropies have also been discussed. We have then performed a detailed quantitative study of the field induced magnetisation process in \( \text{TlCuCl}_3 \) and found that the scattering amplitude \( v_0 \) seems to be much larger than expected from magnon-magnon interactions alone. We see this as a further confirmation that spin-phonon coupling is important in this compound. Finally we have pointed out that even in \( \text{TlCuCl}_3 \) where \( \mathcal{H}' \) is expected to be tiny due to symmetry it seems to remain crucial to obtain a quantitative correct description of the experimental data.

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