Antiferromagnetic Magnons as Highly Squeezed Fock States underlying Quantum Correlations

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Employing the concept of two-mode squeezed states from quantum optics, we demonstrate a revealing physical picture for the antiferromagnetic ground state and excitations. Superimposed on a Néel ordered configuration, a spin-flip restricted to one of the sublattices is called a sublattice-magnon. We show that an antiferromagnetic spin-up magnon is comprised by a quantum superposition of states with $n + 1$ spin-up and n spin-down sublattice-magnons, and is thus an enormous excitation despite its unit net spin. Consequently, its large sublattice-spin can amplify its coupling to other excitations. Employing von Neumann entropy as a measure, we show that the antiferromagnetic eigenmodes manifest a high degree of entanglement between the two sublattices, thereby establishing antiferromagnets as reservoirs for strong quantum correlations. Based on these novel insights, we outline strategies for exploiting the strong quantum character of antiferromagetic (squeezed-)magnons.

Introduction.—As per the Heisenberg uncertainty principle, the quantum fluctuations of two non-commuting observables cannot simultaneously be reduced to zero. However, it is possible to generate a state with the quantum noise in one observable reduced below its ground state limit at the expense of enhanced fluctuations in the other observable [1, 2]. Considering a single mode or frequency of light, such states, generally called squeezed vacuum [1, 2], have proven instrumental in the detection of gravitational waves [3] with a sensitivity beyond the quantum ground state limit [4–6]. Furthermore, squeezed vacuum states have applications in quantum information [7–11] since they exhibit quantum correlations and entanglement. These are best represented and exploited via the two-mode squeezed vacuum states, where the two participating modes are entangled and correlated [1]. The widely studied [1, 2] single- and two-mode squeezed vacuums may be considered a special case, corresponding to zero photon number(s), of a wider class - squeezed Fock states [12, 13]. While investigated theoretically, the latter have been largely forgotten, probably owing to the experimental challenge of generating them. The squeezing concept applies to bosonic modes in general, and squeezed states of magnons [14–16] and phonons [17] have also been achieved experimentally.

The concept of squeezed Fock states [12, 13] has proven valuable in understanding the spin excitations of ordered magnets [18, 19]. Squeezed-magnons have been shown to be the eigen-excitations of a ferromagnet [18, 20]. A squeezed-magnon is comprised by a coherent superposition of the different odd number states of the spin-1 magnon [18, 19] [21]. This bestows it a noninteger average spin larger than 1. The relatively weak spin-nonconserving interactions, such as dipolar fields or crystalline anisotropy, underlie the magnon-squeezing in ferromagnets. These spin-nonconserving interactions were further found to result in two-sublattice magnets hosting excitations with spin varying continuously between positive and negative values [19]. In contrast, exchange interaction in a two-sublattice magnet leads to a strong squeezing effect, which does not affect the excitation spin and forms a main subject of the present Letter. Being eigen-excitations, squeezed-magnons are qualitatively distinct from the squeezed states of light discussed above, which are non-equilibrium states generated via an external drive. To emphasize this difference, we employ the terminology that "squeezed state of a boson" refers to a non-equilibrium state, while a "squeezed-boson" is an eigenmode [22].

Instigated by recent experimental breakthroughs [23–28], interest in antiferromagnets (AFMs) for practical applications has been invigorated [29–33]. Due to the well-known strong quantum fluctuations in AFMs, they have also been the primary workhorse of the quantum magnetism community [34]. The Néel ordered configuration, which is consistent with most of the experiments, is not the true quantum ground state of an AFM. Furthermore, quantum fluctuations destroy any order in a one-dimensional isotropic AFM. These and related general ideas applied to AFMs bearing geometrically frustrated interactions underlie quantum spin liquids [35–37], which are devoid of order in the ground state and host exotic, topologically non-trivial excitations embodying massive entanglement.

We here develop the squeezing picture for the ground state and excitations of a simple, two-sublattice AFM. It continuously connects and allows a unified understanding of classical and quantum as well as ordered and disordered antiferromagnetic states. We show that the AFM eigenmodes are obtained by pairwise, two-mode squeezing of sublattice-magnons, the spin-1 excitations delocalized over one of the two sublattices. Focusing on spatially uniform modes, the antiferromagnetic ground state is a superposition of states with equal number of spin-up and -down sublattice-magnons [Fig. 1(a) and (c)]. The result is a diminished net spin on each sublattice by an amount dictated by the degree of squeezing, parametrized by the non-negative squeeze parameter r. Similarly, a spin-up AFM (squeezed-)magnon is comprised by a superposition of states with $n+1$ spin-up and n spin-down sublattice-magnons [Fig. 1(b) and (c)]. Thus, despite its unit net spin, it carries enormous spins on each sublattice which allows it to couple strongly with other excitations via a sublattice-spin mediated interaction (Fig. 2). Owing to a perfect correlation between the two sublattice-magnon numbers, AFM squeezed-magnons are shown to embody entanglement quantified by von Neumann entropy [1, 38] increasing monotonically with r (Fig. 3). The degree of squeezing and entanglement embodied by these eigenmodes is significantly larger than that in hitherto achieved non-equilibrium states. We also comment on existing experiments [39, 40], where this squeezing-mediated coupling enhancement (Fig. 2) has been observed, and strategies for exploiting the entanglement contained in antiferromagnetic magnons.

AFM eigenmodes as squeezed Fock states.—We consider a Néel ordered ansatz with sublattice A and B spins pointing along $\hat{\boldsymbol{z}}$ and $-\hat{\boldsymbol{z}}$, respectively. The antiferromagnetic Hamiltonian may then be expressed in terms of the corresponding sublattice-magnon ladder operators $\tilde{a}_{\boldsymbol{k}}, \tilde{b}_{\boldsymbol{k}}$ as [19, 41]:

$$\tilde{H} = \sum_{\boldsymbol{k}} A_{\boldsymbol{k}} \left(\tilde{a}_{\boldsymbol{k}}^\dagger \tilde{a}_{\boldsymbol{k}} + \tilde{b}_{\boldsymbol{k}}^\dagger \tilde{b}_{\boldsymbol{k}} \right) + C_{\boldsymbol{k}} \left(\tilde{a}_{\boldsymbol{k}} \tilde{b}_{-\boldsymbol{k}} + \tilde{a}_{\boldsymbol{k}}^\dagger \tilde{b}_{-\boldsymbol{k}}^\dagger \right), \quad (1)$$

where we assume inversion symmetry and disregard applied magnetic fields, for simplicity. Consistent with the assumed Néel order, sublattice B (A) magnons represented by $\tilde{b}_{\boldsymbol{k}}$ ($\tilde{a}_{\boldsymbol{k}}$) are spin-up (-down). In addition to the general considerations captured by Eq. (1), we will obtain specific results for a uniaxial, easy-axis AFM described by:

$$\tilde{H}_{\rm uni} = \frac{J}{\hbar^2} \sum_{i,\boldsymbol{\delta}} \tilde{\boldsymbol{S}}_{\rm A}(\boldsymbol{r}_i) \cdot \tilde{\boldsymbol{S}}_{\rm B}(\boldsymbol{r}_i + \boldsymbol{\delta}) - \frac{K}{\hbar^2} \sum_i \left[\tilde{S}_{{\rm A}z}(\boldsymbol{r}_i) \right]^2 - \frac{K}{\hbar^2} \sum_j \left[\tilde{S}_{{\rm B}z}(\boldsymbol{r}_j) \right]^2. \quad (2)$$

Here, the positive parameters J and K account for intersublattice antiferromagnetic exchange and easy-axis anisotropy, respectively. $\tilde{\boldsymbol{S}}_{\rm A,B}$ represent the respective spin operators, $\boldsymbol{r}_i$ ($\boldsymbol{r}_j$) runs over the sublattice A (B),

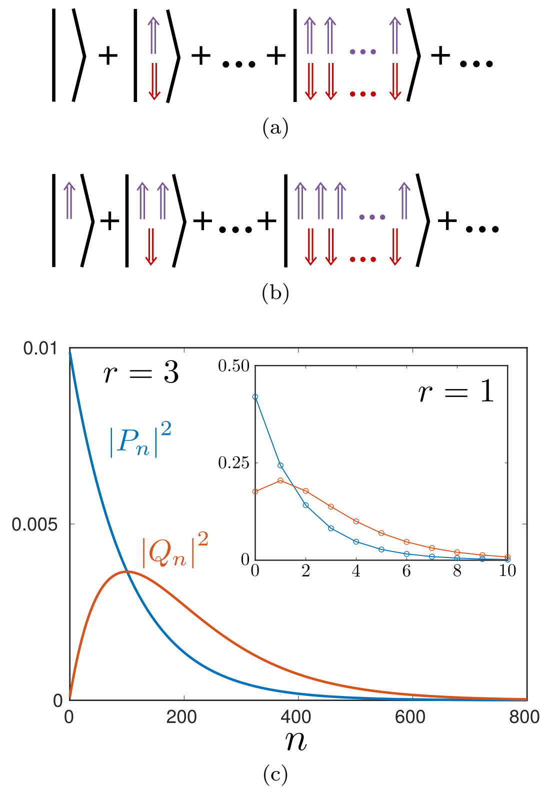

FIG. 1. Schematic depiction of spatially uniform antiferromagnetic (a) vacuum and (b) spin-up eigenmodes. (a) The vacuum mode, represented as $|0\rangle_{\rm sq} = \sum_n P_n \, |n,n\rangle_{\rm sub}$, is a superposition over states with equal number of spin-up and -down sublattice-magnons. (b) The spin-up squeezed-magnon, represented as $|\uparrow\rangle_{\rm sq} = \sum_n Q_n \, |n+1,n\rangle_{\rm sub}$, is comprised by states with one extra spin-up sublattice-magnon. (c) Squared amplitudes corresponding to the sublattice-magnon states constituting the uniform squeezed vacuum and spin-up eigenmodes for squeeze parameters of 3 (main) and 1 (inset).

and $\boldsymbol{\delta}$ are vectors to the nearest neighbors. Executing Holstein-Primakoff transformations [42] and switching to Fourier space, Eq. (2) reduces to Eq. (1) apart from a constant energy offset [19, 43], with $A_{\boldsymbol{k}} = JSz + 2KS$ and $C_{\boldsymbol{k}} = JSz\gamma_{\boldsymbol{k}}$. Here, S is the spin on each site, z is the coordination number, and $\gamma_{\boldsymbol{k}} \equiv (1/z) \sum_{\boldsymbol{\delta}} \exp{(i\boldsymbol{k}\cdot\boldsymbol{\delta})}$.

The Hamiltonian [Eq. (1)] is diagonalized to $\tilde{H} = \sum_{\boldsymbol{k}} \epsilon_{\boldsymbol{k}} \left(\tilde{\alpha}_{\boldsymbol{k}}^\dagger \tilde{\alpha}_{\boldsymbol{k}} + \tilde{\beta}_{\boldsymbol{k}}^\dagger \tilde{\beta}_{\boldsymbol{k}} \right)$ via a Bogoliubov transformation [42] described by [44]:

$$\tilde{\alpha}_{\boldsymbol{k}} = u_{\boldsymbol{k}} \tilde{a}_{\boldsymbol{k}} + v_{\boldsymbol{k}} \tilde{b}_{-\boldsymbol{k}}^\dagger, \quad \tilde{\beta}_{\boldsymbol{k}} = u_{\boldsymbol{k}} \tilde{b}_{\boldsymbol{k}} + v_{\boldsymbol{k}} \tilde{a}_{-\boldsymbol{k}}^\dagger, \quad (3)$$

$$u_{\boldsymbol{k}} = \sqrt{\frac{A_{\boldsymbol{k}} + \epsilon_{\boldsymbol{k}}}{2\epsilon_{\boldsymbol{k}}}}, \qquad v_{\boldsymbol{k}} = \sqrt{\frac{A_{\boldsymbol{k}} - \epsilon_{\boldsymbol{k}}}{2\epsilon_{\boldsymbol{k}}}}, \quad (4)$$

where $\epsilon_{\boldsymbol{k}} = \sqrt{A_{\boldsymbol{k}}^2 - C_{\boldsymbol{k}}^2}$. $\tilde{\alpha}_{\boldsymbol{k}}$ and $\tilde{\beta}_{\boldsymbol{k}}$ represent the spin-down and -up eigenmodes of the AFM, which are subsequently called squeezed-magnons. Denoting the resulting antiferromagnetic vacuum or ground state wavefunction by $|\mathrm{G}\rangle_{\mathrm{sq}}$, we have $\tilde{\alpha}_{\boldsymbol{k}} |\mathrm{G}\rangle_{\mathrm{sq}} = \tilde{\beta}_{\boldsymbol{k}} |\mathrm{G}\rangle_{\mathrm{sq}} = 0$ for all $\boldsymbol{k}$.

Let us first consider the spatially uniform modes, i.e. $\boldsymbol{k} = \boldsymbol{0}$. We denote states in the corresponding reduced subspaces via $|N_{b_{\boldsymbol{0}}}, N_{a_{\boldsymbol{0}}}\rangle_{\mathrm{sub}}$ and $|N_{\beta_{\boldsymbol{0}}}, N_{\alpha_{\boldsymbol{0}}}\rangle_{\mathrm{sq}}$, where $N_{b_{\boldsymbol{0}}}$ denotes the number of spin-up sublattice-magnons and so on. Within the reduced subspaces, the Néel ordered state is thus denoted by $|0,0\rangle_{\mathrm{sub}}$, while the antiferromagnetic ground state obtained above is represented by $|0,0\rangle_{\mathrm{sq}}$. We define the relevant two-mode squeeze operator [1]: $\tilde{S}_2(r_{\boldsymbol{0}}) \equiv \exp\left(r_{\boldsymbol{0}}\tilde{a}_{\boldsymbol{0}}\tilde{b}_{\boldsymbol{0}} - r_{\boldsymbol{0}}\tilde{a}_{\boldsymbol{0}}^{\dagger}\tilde{b}_{\boldsymbol{0}}^{\dagger}\right)$, with the non-negative squeeze parameter $r_{\boldsymbol{0}}$ given via $u_{\boldsymbol{0}} \equiv \cosh r_{\boldsymbol{0}}$ and $v_{\boldsymbol{0}} \equiv \sinh r_{\boldsymbol{0}}$ [Eq. (4)] [45]. Employing the identities [1, 18]:

$$\tilde{\alpha}_{\boldsymbol{0}} = \tilde{S}_2(r_{\boldsymbol{0}})\tilde{a}_{\boldsymbol{0}}\tilde{S}_2^{-1}(r_{\boldsymbol{0}}), \quad \tilde{\beta}_{\boldsymbol{0}} = \tilde{S}_2(r_{\boldsymbol{0}})\tilde{b}_{\boldsymbol{0}}\tilde{S}_2^{-1}(r_{\boldsymbol{0}}), \tag{5}$$

where $\tilde{\alpha}_{\boldsymbol{0}}$ and $\tilde{\beta}_{\boldsymbol{0}}$ are given by Eq. (3), into the condition $\tilde{\alpha}_{\boldsymbol{0}} |0,0\rangle_{\mathrm{sq}} = \tilde{\beta}_{\boldsymbol{0}} |0,0\rangle_{\mathrm{sq}} = 0$, we obtain:

$$|0,0\rangle_{\mathrm{sq}} = \tilde{S}_2(r_{\boldsymbol{0}}) |0,0\rangle_{\mathrm{sub}}. \tag{6}$$

Thus, the uniform modes antiferromagnetic ground state is a two-mode squeezed vacuum of sublattice-magnons. Working along the same lines as above, it is straightforward to show that $|m,n\rangle_{\mathrm{sq}} = \tilde{S}_2(r_{\boldsymbol{0}}) |m,n\rangle_{\mathrm{sub}}$, thereby demonstrating the antiferromagnetic eigenmodes to be two-mode squeezed sublattice-magnon Fock states. Therefore, the eigenmodes are henceforth called "squeezed-magnons".

Based on the analysis above, it becomes evident that the antiferromagnetic ground state is obtained by pairwise, two-mode squeezing of the Néel ordered state:

$$|\mathrm{G}\rangle_{\mathrm{sq}} = \left[\prod_{\boldsymbol{k}} \tilde{S}_2\left(r_{\boldsymbol{k}}\right)\right] |\text{Néel}\rangle_{\mathrm{sub}}, \tag{7}$$

where $\tilde{S}_2\left(r_{\boldsymbol{k}}\right) \equiv \exp\left(r_{\boldsymbol{k}}\tilde{a}_{\boldsymbol{k}}\tilde{b}_{-\boldsymbol{k}} - r_{\boldsymbol{k}}\tilde{a}_{\boldsymbol{k}}^{\dagger}\tilde{b}_{-\boldsymbol{k}}^{\dagger}\right)$, with the squeeze parameters $r_{\boldsymbol{k}}$ given via $u_{\boldsymbol{k}} = u_{-\boldsymbol{k}} \equiv \cosh r_{\boldsymbol{k}}$. The $\tilde{\alpha}_{\boldsymbol{k}}$ eigenmode is thus a two-mode ($\tilde{a}_{\boldsymbol{k}}$ and $\tilde{b}_{-\boldsymbol{k}}$) squeezed-magnon, and similar for $\tilde{\beta}_{\boldsymbol{k}}$ eigenmode. Due to this mathematical equivalence, it suffices to analyze the spatially uniform eigenmodes, which is what we focus on in the following.

Spatially uniform eigenmodes.—For ease of notation, we denote the wavefunctions for spatially uniform squeezed vacuum by $|0\rangle_{\mathrm{sq}}$ and spin-up squeezed-magnon by $|\uparrow\rangle_{\mathrm{sq}}$, while the corresponding squeeze parameter is denoted by r. Considering a uniaxial AFM [Eq. (2)], we obtain $\cosh r \approx (1/2)(Jz/K)^{1/4}$ [Eq. (4)], which translates to $r \approx 3$ for a typical ratio of $J/K \sim 10^4$. To get a feel for numbers, the most squeezed vacuum state of light generated so far corresponds to a squeeze parameter of about 1.7 [2, 46]. Furthermore, in the limit $K \to 0$, the squeeze parameter is found to diverge. This feature is general and a direct consequence [Eq. (4)] of the Goldstone theorem, according to which $\epsilon_{\boldsymbol{0}} \to 0$ in the limit of isotropy.

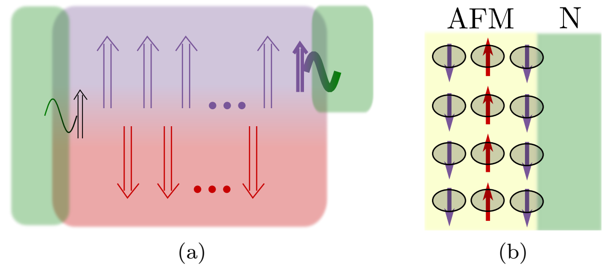

FIG. 2. (a) An external excitation bath (shaded green) interacts weakly with the AFM squeezed-magnon if coupled via its unit net spin (left), but strongly if exposed to only one of the sublattices (right). (b) Schematic depiction of a metal (N) coupled to an AFM via a fully uncompensated interface.

Employing the relation $\tilde{\alpha}_{\boldsymbol{0}} |0\rangle_{\mathrm{sq}} = (\cosh r \ \tilde{a}_{\boldsymbol{0}} + \sinh r \ \tilde{b}_{\boldsymbol{0}}^{\dagger}) |0\rangle_{\mathrm{sq}} = 0$, the squeezed vacuum is obtained in terms of the uniform sublattice-magnons subspace [1]:

$$|0\rangle_{\mathrm{sq}} = \sum_{n=0}^{\infty} \frac{(-\tanh r)^n}{\cosh r} |n,n\rangle_{\mathrm{sub}} \equiv \sum_n P_n |n,n\rangle_{\mathrm{sub}}. \tag{8}$$

The ensuing wavefunction is schematically depicted in Fig. 1(a) and the distribution over constituent states is plotted in Fig. 1(c). With an increasing r, the number of states that contribute substantially to the superposition increases monotonically. This presence of sublattice-magnons in the ground state constitutes quantum fluctuations.

A similar representation for the spin-up squeezed-magnon is obtained via $|\uparrow\rangle_{\mathrm{sq}} = \tilde{\beta}_{\boldsymbol{0}}^{\dagger} |0\rangle_{\mathrm{sq}} = (\cosh r \ \tilde{b}_{\boldsymbol{0}}^{\dagger} + \sinh r \ \tilde{a}_{\boldsymbol{0}}) |0\rangle_{\mathrm{sq}}$ and Eq. (8):

$$|\uparrow\rangle_{\mathrm{sq}} = \sum_{n=0}^{\infty} \frac{\sqrt{n+1}\,(-\tanh r)^n}{\cosh^2 r} |n+1,n\rangle_{\mathrm{sub}}, \\ \equiv \sum_n Q_n |n+1,n\rangle_{\mathrm{sub}}. \tag{9}$$

A schematic depiction and the distribution over constituent states are shown in Fig. 1(b) and (c). In stark contrast with the squeezed vacuum, where the contribution from states decreases monotonically with n, the highest contribution to the superposition here comes from $n \approx \sinh^2 r$. No such peak exists for weak squeezing when $\sinh r < 1$. The average number of spin-up magnons comprising a squeezed-magnon is evaluated as $\cosh^2 r + \sinh^2 r$. Thus, a typical AFM squeezed-magnon, corresponding to $r \approx 3$ estimated above, is comprised by around 200 spin-up magnons on one sublattice and nearly the same number of spin-down magnons on the other. It is thus an enormous excitation, despite its unit net spin.

Enhanced interaction.—This enormous nature of the AFM squeezed-magnon reveals an approach to exploit it. When it couples to excitations, such as itinerant

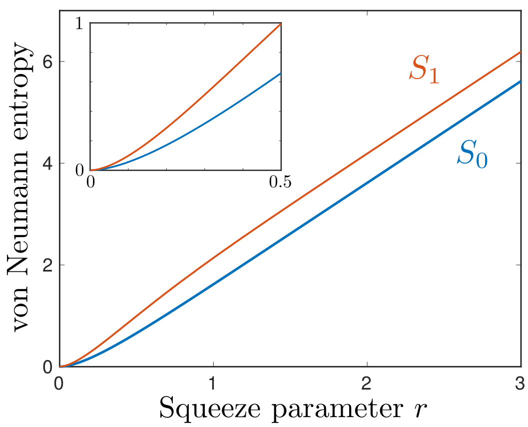

FIG. 3. Entanglement between the two constituent sublattice-magnons quantified via von Neumann entropy for the squeezed vacuum (S_0) and magnon (S_1) eigenmodes. The inset shows a zoom-in of the small r range.

electrons or phonons, via its net spin, the interaction strength is proportional to the relatively small unit spin. On the other hand, if an interaction is mediated via the sublattice-spin, it will be greatly enhanced (by a factor $\sim 2\cosh^2 r \approx 200$ for $r \approx 3$) on account of its large sublattice spin content [Fig. 2(a)]. Such a situation arises, for example, when an AFM is exposed to a metal via an uncompensated interface [Fig. 2(b)] [25, 47–49]. This effect provides a physical picture for the theoretically encountered enhancement in spin pumping current from AFM into an adjacent conductor coupled asymmetrically to the two sublattices [48]. The same mechanism has also been exploited in predicting an enhanced magnon-mediated superconductivity in a conductor bearing an uncompensated interface with an AFM [50].

Entanglement.—In a two-mode squeezed vacuum, the participating modes are entangled with the degree of entanglement quantified by the von Neumann entropy [1, 38] S_0:

$$\begin{aligned} S_0 = & -\sum_n |P_n|^2 \log\left(|P_n|^2\right), \\ = & \ 2\log(\cosh r) - 2(\sinh^2 r)\log(\tanh r). \end{aligned} \tag{10}$$

Such two-mode squeezed vacuum states of light have been exploited for obtaining useful entanglement [7]. It is not clear if the high entanglement content of our squeezed-magnon vacuum can be used. However, the squeezed-magnons themselves embody strong entanglement, quantified by an even larger von Neumann entropy $S_1 = -\sum_n |Q_n|^2 \log(|Q_n|^2)$ (Fig. 3), which may be transfered to external excitations. This can be achieved by coupling the systems to be entangled with the opposite sublattices [51–53], via uncompensated interfaces [Fig. 2(b)], for example. In comparison, von Neumann entropy [54] of about 1 has been measured in cold atom systems [55]. Furthermore, the high von Neumann entropy content of the squeezed-magnon makes it a non-topological, “massively entangled” excitation similar to the topological excitations hosted by some quantum spin liquids [35–37].

Quantum fluctuations in “classical” experiments.—The interaction enhancement effect [Fig. 2(a)] is rooted in high magnon-squeezing and the underlying quantum superposition of a large number of states [Eq. (9)]. It is a direct consequence of the strong quantum fluctuations in the antiferromagnetic ground state, that hosts this excitation, and is thus a quantum fluctuation effect itself. Nevertheless, this coupling enhancement is observed as an increased magnetic damping around compensation temperature in a compensated ferrimagnet [39], which mimics an AFM [19, 56]. Recently, this enhancement has been observed and exploited in a compensated ferrimagnet for an ultrastrong magnon-magnon coupling resulting in hybridization between the two enormous spin-up and -down squeezed-magnons [40]. These “classical” experiments at high temperatures may thus be considered observation of the antiferromagnetic quantum fluctuations. The high squeezing mediated enhancement [57, 58] ($\sim \sqrt{J/K}$ for our uniaxial AFM) is reproduced by the classical theory of spin dynamics [40, 56], where it is termed “exchange-enhancement”. This is understandable since the classical dynamics is captured by the quantum system being in a coherent state [48, 59, 60], which fully accounts for the average effect of these quantum fluctuations.

Generalizations.—The description in terms of squeezed Fock states developed herein is a mathematical consequence of the Bogoliubov transformation and goes beyond AFMs. It should allow a similar physical picture, and subsequent exploitation of quantum effects, in other systems such as cold atoms [61–63]. Here, we have disregarded the relatively weak spin-nonconserving interactions. Inclusion of those necessitates a 4-dimensional Bogoliubov transform [19] thereby precluding the simple two-mode squeezed Fock states description employed here. Similar complications also arise when considering AFMs lacking inversion symmetry. Nevertheless, an analogous general picture can be developed.

Conclusion.—We have developed a novel description and physical picture of antiferromagnetic ground state and excitations based on the concept of two-mode squeezed Fock states. Capitalizing on the tremendous progress in quantum optics, these fresh insights pave the way for exploiting the quantum properties of antiferromagnetic squeezed-magnons towards, potentially room temperature, quantum devices.

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