Comment on "Bose-Einstein Condensation of Magnons in Cs_2CuCl_4"

In a recent Letter, Radu *et al.* present data on the specific heat of the antiferromagnet Cs_2CuCl_4 [1]. In a high magnetic field, the sample is a saturated ferromagnet. They study the (second order) transition from the ferromagnetic to the spin flop state as the magnetic field is lowered. The data are analyzed within a theory of interacting spin waves; the critical exponent which describes the variation of the transition temperature T_C with field is extracted from data and compares well with theory. We have no issues with this analysis, nor with the data. We do note that the nature of spin waves near the ferromagnet-spin flop phase boundary has been known for decades [2].

At issue is the use of the term Bose-Einstein condensation to describe this system. In statistical mechanics, this term has a clear, well-understood meaning. Bose-Einstein condensates form in a system with a *fixed particle number* as an external parameter is varied; the (true) thermodynamic chemical potential approaches the lowest excitation energy of the system from below and pins to this value when the Bose condensate exists. A key feature when the condensate is present is a coherent, macroscopic quantum phase.

The physics of the system in Ref. [1] is very different than that just described. The total number of magnons in this system is not conserved as either the external field or temperature is varied. The authors refer to the quantity μ_0 in their Eq. (7) as the "chemical potential," but this object, written as $g\mu_B(B_c - B)$, is not the true thermodynamic chemical potential of the magnon gas. Rather, it is simply a term in the spin wave Hamiltonian present when linearized about the high field state. The true chemical potential is always zero here because the magnon number is not conserved. Here one has a system wherein spin waves of a particular wave vector "go soft" as the magnetic field B passes through B_c from above. For this system, there is once again a diagonal term in a spin wave Hamiltonian whose mathematical form appears to mimic that from the true thermodynamic chemical potential for a system of particles whose number is conserved. For such systems, to generate the free energy one proceeds by diagonalizing $H - \mu N$ to find μ by requiring the number of bosons equal a fixed value. It is not uncommon to discuss the region close to, and just below, a second order phase transition accompanied by a soft mode as a state where this mode has large occupancy, in a crude but not precise analogy to Bose-Einstein condensation. In Ref. [1] the physics is that of large amplitude thermal fluctuations near the phase transition line, not that of a Bose-Einstein condensate with a coherent quantum phase.

We wish to note a recent experiment where true Bose-Einstein condensation of magnons has been observed at room temperature [3]. The authors create nonequilibrium magnons in YIG at room temperature, by parametric pumping which creates magnon pairs of equal and opposite wave vectors. These externally introduced magnons equilibrate with the preexisting thermal magnons through four magnon scattering. Once pumping ceases, the magnon number remains fixed. The magnons thus acquire a nonzero true chemical potential, measured directly in Ref. [3]. With increased pumping power, the true chemical potential μ increases, and at a critical pumping power "pins" at the minimum spin wave excitation energy $\hbar\omega_{\mathrm{min}}$. For pumping powers above the critical value, Brillouin light scattering studies of the frequency spectrum of magnons in the crystal provide explicit evidence for the presence of a magnon condensate at $\hbar\omega_{\mathrm{min}}$. Evidence for a coherent quantum phase is not presented, but this system satisfies all constraints necessary for true Bose-Einstein condensation, in contrast to the system studied in Ref. [1]. Of course, on a longer time scale, the magnon gas equilibrates with the phonons and the magnon chemical potential has returned to zero. As shown experimentally, the ratio of the four magnon relaxation rate to the magnon phonon equilibration rate is close to the relevant ratio in ultralow temperature Bose condensates of alkali atoms. The conditions realized in this experiment are thus very similar to those in the now well-documented field of Bose condensation in trapped atom gasses at ultralow temperature.

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Received 15 March 2006; published 19 January 2007
DOI: 10.1103/PhysRevLett.98.039701
PACS numbers: 75.40.−s, 03.75.Nt, 75.30.Kz, 75.45.+j

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