Quasiparticle BEC

David McKay

December 2006

Abstract

Bose-Einstein condensation (BEC) is a broken symmetry state in which there is macroscopic occupation of the ground state of the system. In this paper, I will detail several recent observations of quasiparticle BEC in solids. The advantage of quasiparticle BEC is the low mass of these quasiparticles, resulting in BEC at temperatures on the order of 10 Kelvin. Examples of possible systems for quasiparticle BEC include magnons (quantized spin-waves) in magnetic materials, excitons (electron-hole pairs) in semiconductors and polaritons (photon-exciton entangled states) in microcavities.

Introduction

The quantum statistics of identical particle systems has many profound implications on the physical world. One of those implications is on the phase transition known as Bose-Einstein Condensation (BEC), which causes a system to macroscopically occupy a single quantum state. Theoretically, BEC has been around for the greater part of the last century. It was first postulated by Einstein in 1924 when, together with Bose, he formulated the statistics governing bosons, which are particles with integer quanta of angular momentum. However, the topic of BEC has exploded in experimental physics due to the experimental realization of condensation in dilute, trapped alkalis in 1995 by groups at MIT and the University of Colorado at Boulder[1].

Despite the almost overwhelming success of cold atom experiments in studying BEC, there are certain limitations that remain. The largest limitation is that due to the extremely low temperatures required to reach BEC with dilute alkali gases (∼100nK), standard cyrogenic techniques are useless. Therefore, cold atoms experiments require a large and complicated experimental setup to trap and cool to these temperatures. One potential system that can achieve BEC at temperatures accessible to liquid Helium type cyrogenic setups are quasiparticles in solids. Quasiparticles are excitations of the many-body system, which behave as bosons and therefore can undergo a BEC transition. The advantage of looking at condensation of quasiparticles, is that they have an effective mass which is much lower than the mass of an alkali atom. For example, the effective mass of polaritons is 10^9 times lighter than a Rubidium atom and can therefore allow condensation at 19K[2]. However, the challenge in creating quasiparticle BEC is that quasiparticles decay and decohere easily. Decay is especially a problem, given that BEC is a process described by a system in thermal equilibrium.

In the following paper, I will address BEC with three types of quasiparticles; excitons, polaritons and magnons. I will first explain in more detail what these quasiparticle states are and under which situations we may consider them to display bosonic behaviour. Then, I will briefly discuss the theory of Bose-Einstein condensation of weakly interacting particles in quasi-equilibrium. In the final section, I will address experiments with quasiparticle BEC.

Theory

Quasiparticles

The first question to answer is what is a quasiparticle? As Snoke and Moskalenko point out[3], the use of the word is not universal. However, in this paper it is used to refer to an elementary excitation in a many-body system. The three quasiparticles I will look at are excitons, polaritons and magnons.

Excitons

Excitons are a fundamental excitation of the many-electron quantum ground state in a semiconductor crystal. When a photon with energy greater than the bandgap is incident on the crystal it may be absorbed creating a hole in the valence band and an electron in the conduction band. Due to Coulomb attraction, this electron and hole may form a hydrogen like atom. Excitons can come in a variety of sizes. One extreme are Frenkel excitons, which span one lattice site and the other extreme are Wannier excitons which span many lattice sites[3]. In general, experiments involving excitons deal with the Wannier type. The operator that creates excitons is written as:

$$c^{\dagger} = \frac{1}{\sqrt{2V}} \sum_{q,\sigma} \phi(q) a^{\dagger}_{q,\sigma} b^{\dagger}_{-q,-\sigma} \tag{1}$$

where $a^{\dagger}$ is the free electron creation operator, $b^{\dagger}$ is the free hole creation operator and $\phi(q)$ is the exciton wavefunction. Excitons are bosons by virtue of the addition of angular momentum. Since the hole and electron are both spin $1/2$ the exciton forms a spin 0 singlet and spin 1 triplet, both of which are bosons. However, the underlying fermion nature of excitons is an issue at sufficiently high exciton densities.

The coupling of the exciton dipole moment to the vacuum field causes spontaneous emission of a photon and exciton decay[4]. This decay is a significant impediment to exciton BEC. One solution to this problem is to create excitons in layered semiconductor structures. By keeping the electron and hole separated the probability of decay is greatly reduced.

Polaritons

Figure 1 shows the spectrum of excitations in a semiconductor cystal. For energies well below the bandgap, the excitations are photon like and for energies much higher than the bandgap the excitations are exciton like. However, for the intermediate regime, the exciton and photon states become entangled. This state is known as a polariton.

Magnons

In a lattice of spin magnetic moments which are coupled, the Hamiltonian looks very similar to a system of coupled harmonic oscillators except that there is an upper bound on the value of the spin at each site. However, at low temperatures the higher spin states are thermodynamically frozen out, so the system can be modelled as a lattice of coupled oscillators. From the study of phonons we know that a system of coupled harmonic oscillators leads to delocalized quanta of energy with momenta $\vec{k}$. These quantized spin waves are known as magnons[5]. Spin waves have a repulsive interaction because of the limitation on the amount of spin at each lattice site. Because the formalism of magnons comes from the harmonic oscillator creation/annihilation operators they are bosons.

Quasiparticle BEC

The theoretical details of Bose-Einstein condensation are well covered in the literature (for example see [1]), so I will only give a brief review here. The basic result is obtained by considering the grand partition function of a non-interacting gas of bosons. When we solve for the number of particles in the excited states we discover a fundamental upper limit. If the number of particles in the system exceeds this limit then there is macroscopic occupation of the ground state. Equation 2 shows the relation between the critical density and temperature for BEC in three dimensions. Essentially, BEC occurs when the thermal de Broglie wavelength (Equation 3) is approximately the distance betweeen particles.

$$n \approx \frac{2.62}{\lambda^3} \tag{2}$$

$$\lambda = \left(\frac{2\pi\hbar^2}{mk_BT}\right)^{1/2} \tag{3}$$

A more detailed description is required if we want to look at the microscopic state of the system when there are interactions between the particles. However, because all the quasiparticles are weakly interacting, the nature of the condensate is given by the well known Bogoliubov approximation. The only difference between a BEC of excitons, polaritons or magnons is the effective mass and the effective interaction. The Hamiltonian describing the system in the Bogoliubov approximation is[1]:

$$H = \sum_q \epsilon_q a_q^\dagger a_q + \frac{1}{2}U_0 V \sum_{q1,q2,q3,q4} a_{q1}^\dagger a_{q2}^\dagger a_{q3} a_{q4} \delta_{q1+q2,q3+q4} \tag{4}$$

The parameter U_0, which characterizes the interactions between particles is $U_0 = \frac{4\pi a\hbar^2}{m}$ where a is the s-wave scattering length. When the system is condensed, equation 4, can be simplified by only retaining quadratic terms in $a_q, a_q^\dagger$. Making appropriate substituations, the Hamiltonian takes the form:

$$H = \sum_q E_q \alpha_q^\dagger \alpha_q \tag{5}$$

where the α operators create/annihilate excitations to the BEC ground state and the energy is given by:

$$E_q = \left(\epsilon_q\left(\epsilon_q + 2n_0U_0\right)\right)^{1/2} \tag{6}$$

Quasi-Equilibrium BEC

Because of the generality of BEC, quasiparticle condensation is virtually the same as for cold atoms. However, the main difference is that while atoms are stable, quasi-particles are pumped into the system and then decay. Therefore, we need to address the question of whether or not the arguments presented above can be applied to a system of quasiparticles. The basic result is that equilibrium thermodynamics will apply as long as the lifetime of the quasiparticle is much longer than the thermalization time[6]. This is a large constraint on the system and will motivate possible experiments. Another issue with pumping a system is that the experiments have to make sure that coherence of the excitations in the system is not due to the coherence of the pump source. Such a condensate is known as a driven BEC and is not an example of spontaneous condensation.

Experiments

The search for quasiparticle BEC experimentally has spanned more than forty years. A consistent problem plaguing quasiparticle BEC is making a definitive measurement to prove condensation has occurred. In [7], David Snoke sets out a series of criteria that would best prove condensation. These are adherence to theoretical predictions of critical density versus temperature for a weakly interacting Bose gas, a two-component distribution (for a trapped condensate), demonstration of coherence and measurement of the excitation spectrum. While these are certainly not the only criteria, they form a good set of guidelines when evaluating the following recent results.

Polaritons in Quantum Well Microcavities

One structure in which polaritons are studied is a quantum well embedded in an optical cavity. Typically, the optical cavity is created from a set of dielectric mirrors which have been deposited on a semiconductor sample, which has quantum wells in the middle (see Figure 3). Polaritons in the cavity are characterized by two properties, the energy and the momentum in the plane parallel to the mirrors. The advantage of this system is that state of the polaritons can be probed by the photons that leak from the cavity. The energy corresponds to the frequency of the emitted photons and the angle corresponds to the in plane polariton momentum. Polaritons are initially injected into the cavity in a certain state by a pump laser. Therefore one of the main concerns is that the coherence is not the result of the initial coherence of the pump laser. While this will produce a system with similar properties to a BEC, it is not a thermodynamic condensate[7]. To get around this, the cavity is pumped with a momentum and energy higher than the ground state and polariton-polariton scattering takes these states to the ground state. The imprint of the pump coherence decreases after each successive scattering event until the system can be considered incoherent[6]. Another thing to note about these microcavities is that they are two-dimensional, so the transition is technically not BEC, but the Kosterlitz-Thouless transition. For a finite system such as this, the effects are similar.

There have been many claims to polariton BEC in a microcavity setup, one of earliest in 1996[7], however, these have all been attributed to different phenomena. In the following, I will outline a recent result, which appeared in Nature in September 2006[2]. The setup of the system is illustrated in Figure 3, which is a CdTe/CdMgTe microcavity with 16 quantum wells. The cavity is pumped by a pulsed Ti:sapphire laser at 1.768 eV whereas the polariton ground state is 1.671eV. As mentioned above, this ensures that elastic scattering down to the ground state must occur to ensure the system is incoherent. The system was cooled to 5K and fixed at that temperature, so the parameter that was used to control the transition into BEC is polariton density. Several signatures of condensation were measured. First, as shown in Figure 4, the emitted photon density as a function of the emission angle was measured as the density of polaritons in the cavity was increased. However, most importantly the authors measure the classical first-order correlation function,

$$g^{(1)}(\mathbf{r}, \mathbf{r}') = \frac{\langle E^*(\mathbf{r}) E(\mathbf{r}') \rangle}{\langle E^*(\mathbf{r}) \rangle \langle E(\mathbf{r}') \rangle} \quad (7)$$

using a Michelson interferometer. This measurement confirmed long-range spatial coherence in the polariton gas, leaving little doubt that a condensate was formed.

Excitons in Bilayer Systems

Much of the early literature on the subject of quasiparticle condensation is related to the use of excitons to observe BEC. Experimentally, claims to exciton condensation in semiconductors came as early as 1975[6] and continued throughout the ensuing years. However, in each case the authors would claim that a condensate was formed due to a secondary effect, such as the appearance of superfluid transport. This could not establish the formation of a condensate due to alternative explanations for these effects. The first real evidence of exciton BEC was in a bilayer system in 2003[8]. A bilayer is created by stacking two dimensional quantum wells on top of each other. The electrons in one layer can form excitons with the holes in the other layer. The exciton lifetime increases due to the separation of the electrons and holes. However, in this experiment the authors use a novel method of obtaining exciton formation between electrons in the conduction bands between two layers in the presense of a magnetic field. In a 2D system, the magnetic field creates quantized orbits, known as Landau levels, which are highly degenerate. The field can be selected such that the number of lowest energy Landau levels is twice the number of electrons, therefore leaving half the levels empty. It turns out that the empty Landau levels behave equivalently like holes do for empty valence band states. Therefore the occupied states and empty states from adjacent layers form exciton pairs. A critical difference for this system is that the exciton pairs form in equilibrium, therefore, the problems of pumping and decay are not an issue.

Because the excitons are not optically generated this means that there is no luminescence signal to probe, which is the common measurement made in exciton BEC experiments. Instead, the authors point to two measurements as evidence for condensation. The first measurement is the tunnelling between the two layers as a function of the applied voltage between the layers as shown in Figure 5. When the layers are far apart the tunnelling is suppressed because of Coulomb interactions between the electrons. As the layers are brought closer together, this suppression should increase. However, after the layer separation is reduced to a critical distance the tunnelling rapidly increases, suggesting a quantum phase transition. The second measurement is of the transport properties of the system. Techniques exist which allow for the measurement of current flow in each layer. An exciton flowing in one direction equals opposite currents in the adjacent layers. One consequence of exciton condensation would be the disappearance of the Hall voltage, whereas if the current is carried by independent charges in the two layers a Hall voltage would need to be present. In fact, such a decrease to zero of the Hall voltage is observed.

While this is a promising experiment that certainly pushes the boundaries of fabrication technology to create a novel bilayer state, it is clear that more investigation is required to obtain a definitive signal of condensation.

Magnons

In the following, I will discuss two experiments which achieve BEC with magnons. In the first experiment[9], the system is the magnetic insulator $TlCuCl_3$. In the $TlCuCl_3$ crystal, two Cu^{2+} ions couple to form a dimer, which is a spin 0 ground state and a spin 1 triplet excited state. The magnons in this system are delocalized spin triplet excitations in a spin 0 liquid. When there is no field a gap exists between the spin 0 and the triplet state. As a field is applied, the gap decreases due to field coupling with the $S_z = 1$ state. Eventually a temperature dependent field is reached at which the system attains long range magnetic ordering, which is interpreted as a condensation of the $S_z = 1$ magnons[10]. For $TlCuCl_3$, the critical field is $\approx$ 6T. Studies of $TlCuCl_3$ using neutron scattering show a distinct phase transition at the critical field marked by a change in the triplet level splittings (Figure 7) and the appearance of Goldstone modes (Figure 8). While these two measurements are certainly promising, a measurement confirming the coherence of the state would provide more concrete evidence of condensation.

The next experiment utilized thin films of Yttrium-iron-garnet (YIG) to condense magnons[11] at room temperature. This experiment has many similarities to the polariton experiments previously discussed in that it deals with magnons in a quasi-equilibrium state. Magnons are pumped into the system using microwaves. Since the relaxation time in the lattice for non-equilibrium magnons is 1μs whereas magnon thermalization is 100-200ns, the system satisfies the equilibrium constraints required to achieve BEC. The setup of this experiment is indicated in Figure 6. The final state of the system is measured using a laser which inelastically scatters with the magnons. This gives the occupation of the magnon states, which show a definite narrowing with increased pumping. However, it can often be hard to interprete these types of measurements and again some form of coherence test would be useful. If condensation is indeed occuring, then this system will be very important as an example of quantum coherence that can be manipulated at room temperature.

Future Work

The field of quasiparticle BEC is starting to become very active given the recent evidence of condensation that we reviewed in this paper. So what are the next steps? First, it is clear that definitive measurements can still be made of the condensed state to probe the transition and its long range coherence. Beyond this, the future of the field is in controlling the coherent state. For example, one exciting possibility for exciton experiments is trapping excitons in harmonic potentials creating in the solid using stress[7]. Also, because these experiments only need standard cyrogenic equipment and can be easily minaturized, the coherent state should be utilized for quantum information experments. One idea to start would be entangling two polariton cavities, such as what has been done with cold atom ensembles[12]. Also, like cold atom systems, quasiparticle condensates can be used to investigate other condensed matter phenomena, most notably high temperature superconductivity.

In this paper we have outlined the theory of weakly interacting BEC, which includes quasiparticles. Quasiparticle BEC is an attractive alternative to cold atom experiments because the critical temperature is much higher. The four experiments detailed in this paper demonstrate the observation of condensation of quasiparticles in solids. Two important advances to note are the formation of a condensate at room temperature and the first measurement of quasiparticle spatial coherence in a solid.

References

[1] Anthony J. Leggett. Bose-Einstein condensation in the alkali gases: Some fundamental concepts. *Reviews of Modern Physics*, 73:307–356, 2001.

[2] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J.M.J. Keeling, F.M. Marchetti, M.H. Szymanska, R. Andre, J.L. Staehli, V. Savona, P.B. Littlewood, B. Deveaud, and Le Si Dang. Bose-Einstein condensation of exciton polaritons. *Nature*, 443:409–414, 2006.

[3] D.W. Snoke and S.A. Moskalenko. *Bose-Einstein Condensation of Excitons and Biexcitons.* Cambridge, 2000.

[4] E. Hanamura and H. Haug. Condensation effects of excitons. *Physics Reports*, 33:209–284, 1977.

[5] J. Van Dranedonk and J.H. Van Vleck. Spin waves. *Reviews of Modern Physics*, 39:1–23, 1958.

[6] D.W. Snoke. Spontaneous Bose coherence of excitons and polaritons. *Science*, 298:1368–1372, 2002.

[7] D.W. Snoke. When should we say we have observed Bose condensation of excitons? *Physica Status Solidi (b)*, 238:389–396, 2003.

[8] J.P. Eisenstein and A.H. MacDonald. Bose-Einstein condensation of excitons in bilayer electron systems. *Nature*, 9:691–694, 2004.

[9] Ch. Ruegg, N. Cavadini, Furrer A., H.U. Gudel, K. Kramer, H. Mutka, A. Wildes, K. Habicht, and P. Vorderwisch. Bose-Einstein condensation of the triplet states in the magnetic insulator $TiCuCl_3$. *Nature*, 423:62–65, 2003.

[10] T. Nikuni, Oshikawa M., Oosawa A., and Tanaka H. Bose-Einstein condensation of dilute magnons in $TiCuCl_3$. *Physical Review Letters*, 84:5868–5871, 2000.

[11] S.O. Demokritov, V.E. Demidov, O. Dzyapko, G.A. Melkov, A.A. Serga, B. Hillebrands, and Slavin A.N. Bose-Einstein condensation of quasi-equilibrium magnons at room temperature under pumping. *Nature*, 443:430–433, 2006.

[12] C.W. Chou, H. de Riedmatten, D. Felinto, S.V. Polyakov, S.J. van Enk, and H.J. Kimble. Measurement-induced entanglement for excitation stored in remote atomic ensembles. *Nature*, 438, 2005.

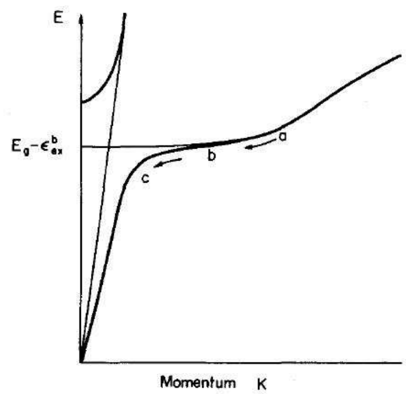

Figure 1: Excitations spectrum in a crystal. In the low k limit, the excitations are photon like and in the high k limit the excitations are exciton like. However, in the intermediate regime, the excitations are superpositions of excitons and photons (polaritons). [4]

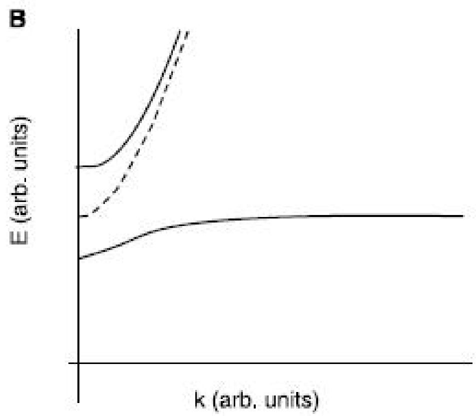

Figure 2: Dispersion curve for polaritons in a microcavity. The effect of the cavity is to change the curve from Figure 1, so that the lower mode of the cavity is in the polariton regime. [7]

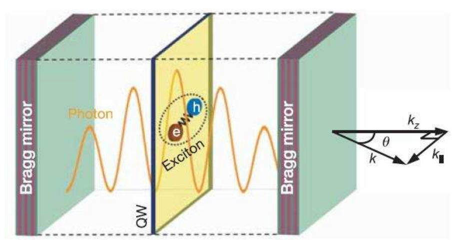

Figure 3: Quantum well microcavity for creating polariton BEC [2]

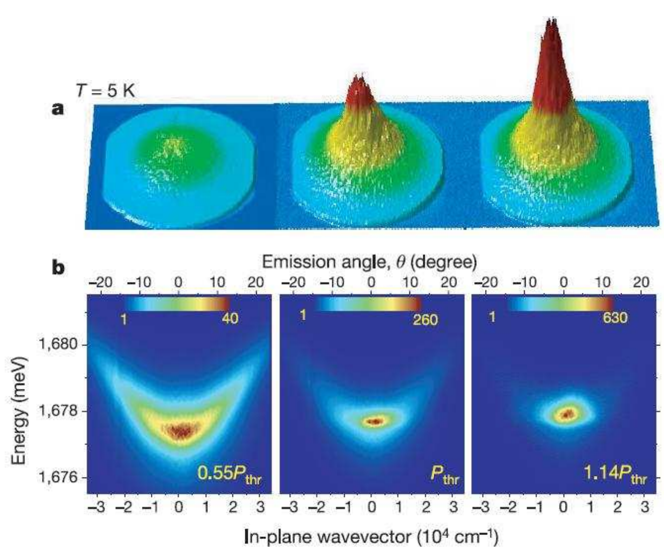

Figure 4: Far-field emission from a microcavity as the pump laser power is increased demonstrating the BEC transition [2]

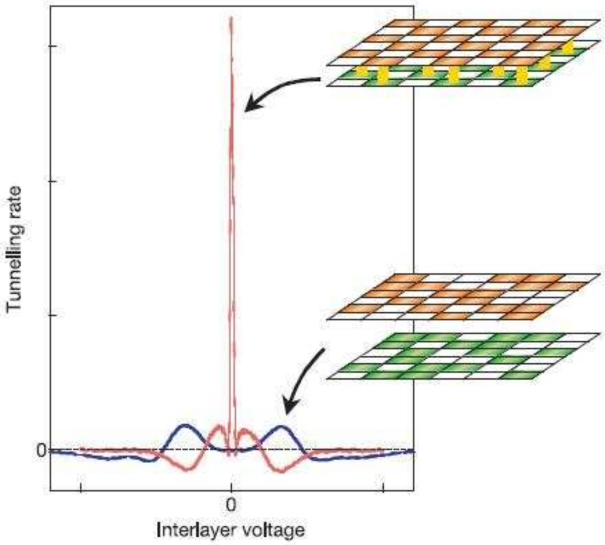

Figure 5: Tunnelling between the electron-electron bilayers as a function of voltage. Below a critical layer separation, tunnelling increases sharply at low voltage. [8]

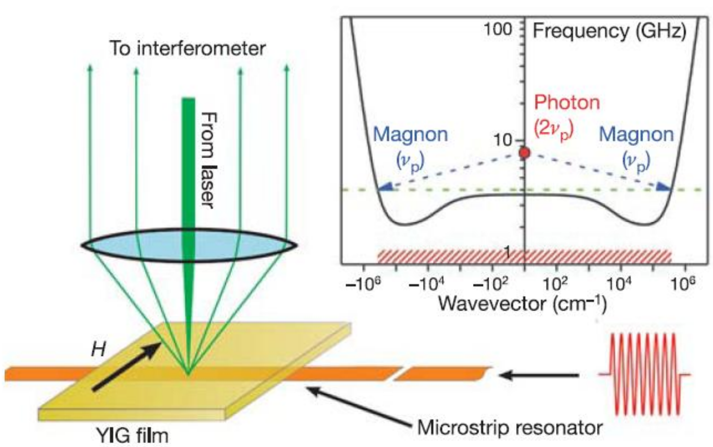

Figure 6: Experimental setup to observe condensation of magnons in a YIG film. Magnons are pumped into the system through a microstrip resonantor and are measured using Brilluoin light scattering. [11]

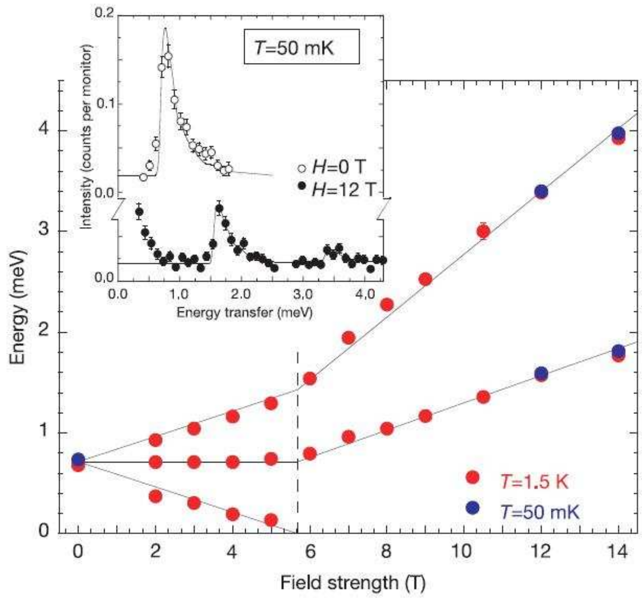

Figure 7: The spin triplet energy levels as a function of the magnetic field for $TlCuCl_3$. The inset shows how these levels were extracted from neutron scattering data. Below the critical field the levels perform Zeeman splitting as expected. However, there is a second order change in the splitting at $\approx$ 6T attributed to condensation of magnons. [9]

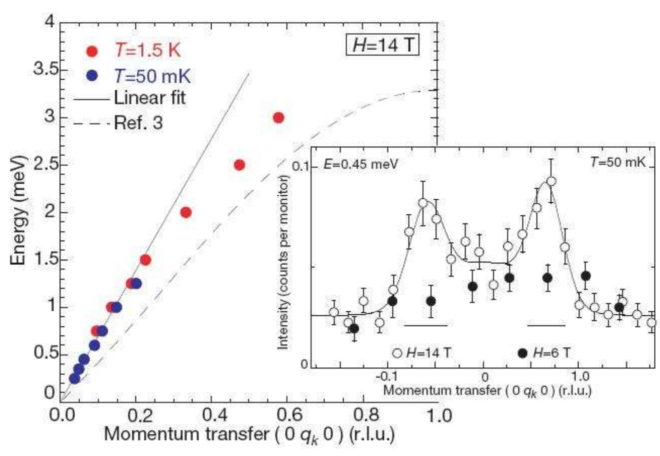

Figure 8: Dispersion relation at 14T for excitations from the ground state. A linear dispersion as k$\rightarrow$0 implies Goldstone modes, which indicate spontaneous broken symmetry. [11]