Quantum Phase Transitions

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Abstract

While classical phase transitions are driven by thermal fluctuations, quantum phase transitions originate from the Heisenberg uncertainty principle and are driven by fluctuations of a purely quantum mechanical nature. In this worksheet, we begin with a brief presentation of the theory of classical phase transitions, highlighting the roles of temperature and entropy. Next, we introduce the theory of quantum phase transitions and contrast them with classical phase transitions. We briefly introduce ideas like level-crossing, critical exponents, and universality in the context of quantum phase transitions. Finally, we provide examples of one of the first well-understood quantum phase transitions discovered in the 1960s: the ferromagnetic to paramagnetic phase transition in $LiHoF_4$. After working through this worksheet, the reader should be able to explain the fundamental differences between classical and quantum phase transitions and understand why they are important to physics, chemistry, and the development of new quantum technologies.

I. Introduction

The field of statistical mechanics began with Boltzmann in the 1870s, who famously developed the first theories of entropy, the kinetic behavior of gases, and hydrodynamics. In the next few years, Gibbs, Maxwell, and Einstein carried on Boltzmann's work, developing much of the field of statistical mechanics by the end of the century. In 1900, quantum mechanics was born when Planck used statistical mechanics to realize that the ultraviolet catastrophe could only be resolved by quantizing the energy levels available to a blackbody [1]. Over the next several decades, great leaps were made in both fields, but it would be some time before phase transitions of a quantum mechanical nature were discovered.

In 1933, Ehrenfest became the first to propose a distinction between first and second order phase transitions, based on whether a discontinuity occurred in the free energy function or its higher derivatives [2]. Soon after, Rutgers used Ehrenfest's theory to identify a classical phase transition from the normal to superconductor phase in metals. Over the next decade, Ehrenfest's paradigm was used to understand transitions from ferromagnetism to paramagnetism and conductor to superconductor, all while theories of Fermi liquids, localization, and BCS superconductors were being proposed (but not yet connected to quantum phase transitions) [3, 4, 5].

In the 1960s, Kadanoff and Widom's studies of scaling and the renormalization group led to a new classification of phase transitions: first-order and continuous. Finally, with a fully developed theory of classical critical phenomena, the idea of quantum phase transitions was introduced in the 1970s and studied in model spin systems like the transverse Ising model [6]. During this time period, scientists were most interested in systems with minimal electron correlation, like the metal-insulator transition and the Anderson transition of non-interacting electrons. In the past two decades, interest in quantum phase transitions has once again resurged with a renewed focus on systems with correlated electrons, like cuprate superconductors, organic conductors, and, most recently, entangled qubits for quantum computers [7].

Quantum phase transitions are fascinating because they lie at the nexus of statistical mechanics and quantum mechanics, the two foundational fields of chemical physics. Although field theories are needed to treat the mathematical details of phase transitions, their fundamental physics is conceptually simple and elegant. Quantum phase transitions are exciting, because they allow us to unlock brand new, useful phases of matter simply by tuning the right control parameter. Many innovations in materials science, physics, and chemistry, like superconductors,semiconductors, and Bose Einstein condensates, cannot be fully understood without a comprehensive theory of quantum phase transitions.

In this worksheet, we will start with an introduction of classical phase transitions, which will allow us to later draw illustrative analogies to quantum phase transitions and highlight the differences between the two phenomena. Next, we will present examples of theoretical and experimentally-observed quantum phase transitions, focusing on the transition from the ferromagnetic to paramagnetic phase in $LiHoF_4$.

II. Theory

Classical Phase Transitions: A Balancing Act

In introductory chemistry, we learn that when a liquid is changing into a gas or a solid, it is undergoing a phase transition. We define each of the phases of matter available to the system by their macroscopic properties: what range of motion they have, what volume they take up, and, most importantly, how ordered the molecules are. To transition between each of these phases, external control parameters, like temperature, are adjusted.

As we adjust our control parameter, we reach coexistence lines where two phases can exist simultaneously. To firmly push the system into one phase or the other, we must put in some latent heat to break intermolecular interactions. Phase transitions involving latent heat are called **first-order phase transitions**, and they exhibit abrupt discontinuities in their properties at a critical temperature. In some cases, at high temperatures, we can also observe **continuous phase transitions**, where a liquid becomes a gas without any latent heat, coexistence phase, or discontinuities. Typically, we draw phase diagrams to delineate the combinations of pressure and temperature that correspond to each possible phase of the system.

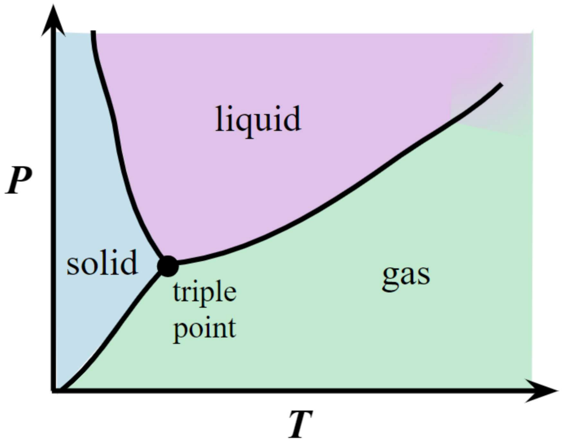

Figure 1: Classical Phase Diagram. *Depicted above is an example of a pressure-temperature phase diagram. When held at constant pressure, the system becomes more disordered as temperature increases, transitioning from a solid at low temperatures to a liquid and, finally, a gas at high temperatures. Each of the lines represents all of the pressure and temperature conditions at which adjacent phases of matter can coexist. At the triple point, all three phases of matter can exist*

simultaneously. Beyond the critical point in the liquid-gas regime, liquids can undergo a continuous phase transition to a gas.

On the microscopic level, changing the external control parameter of a system adjusts the relative importance of enthalpy and entropy. According to thermodynamics, all systems minimize their free energy $F = U - TS$ at equilibrium. To do this, they must strike a careful balance between minimizing enthalpy and maximizing entropy, which are often in competition.

For example, water has the lowest internal energy when its molecules are close together and interacting through hydrogen bonding. However, water's entropy is maximized when the number of available microstates is the greatest, which occurs when particles can move freely and have minimal interactions. We see this delicate balance lead to a phase transition as we increase the temperature of a liquid. The entropy term has a greater negative contribution to the free energy at higher temperatures, so the system will favor a phase with more disorder to minimize F, even if it means increasing the system's internal energy. On the phase diagram above, we see that at high temperatures, a liquid prefers to be a gas instead of a liquid, because gases are more disordered and generally have higher entropy [8, 9].

From this microscopic picture, we can conclude that classical phase transitions occur in any system where there is a tension between maximizing entropy and minimizing enthalpy that can be adjusted by an external control parameter. The phases available to a system are often characterized by their level of disorder, as first introduced by Landau. A measure of a system's disorder that can be used to identify a particular phase is called an order parameter [10, 11].

For example, in a system of spins, a common order parameter is magnetization. In the disordered phase, magnetization is low because spins are randomly oriented. In the ordered phase, magnetization is large because all spins are oriented in a single direction. Although built on simple principles, it turns out that this picture of phase transitions describes a lot of important physics and chemistry, from simple phenomena like boiling or freezing water to more complex examples like macromolecular self-assembly and the ferromagnetic to a paramagnetic transition in materials. Importantly, all of these phase transitions are **thermal transitions**, because they are driven by modulating the importance of entropy [12].

Quantum Phase Transitions: A Different Kind of Balancing Act

In contrast to classical phase transitions, quantum phase transitions occur at absolute zero temperature. However, according to the third law of thermodynamics, there is no entropy at absolute zero, because there is only one microstate for the system to occupy: its ground state, which is a perfect crystal. Clearly, there can be no thermal transitions without entropy. So how do quantum phase transitions occur and what drives them, if not temperature?

Traditionally, we imagine that a system at absolute zero is arranged in a perfect crystalline lattice. However, the Heisenberg uncertainty principle of quantum mechanics prevents us from knowing the positions and momentum of the system simultaneously, challenging this picture. In a perfect crystalline lattice, the positions in our system would be localized to exact points with absolute certainty and would, thus, necessitate infinite uncertainty in the system's momentum. An infinitely large kinetic energy would accompany this infinite uncertainty in momentum, which is not compatible with our understanding of the ground state of a system at absolute zero [6].

This paradox created by the uncertainty principle is the origin of **quantum fluctuations**. A quantum system must carefully balance its momentum uncertainty (related to a kinetic energy) and position uncertainty (related to potential energy) at absolute zero to minimize the ground state energy in accordance with the uncertainty principle. In the same way that we use temperature to adjust the relative importance of energy and entropy to drive classical phase transitions, we use an external control parameter to tune the relative importance of the kinetic and potential energies (and thus the momentum and position uncertainties) at absolute zero to drive quantum phase transitions. The external control parameters for quantum phase transitions can be pressure, magnetic field strength, or any other parameter that adjusts the coupling of the different energetic contributions [7].

As an example, let's consider a quantum system that has a coupling between energies g, which can be externally tuned. Let's imagine that the system has a Hamiltonian:

$$H = H_0 + gH_1$$

where g represents a coupling between the two independent Hamiltonians H_0 and H_1 . Additionally, let's assume that the system is on an infinite lattice and that H_0 and H_1 commute. Because the two Hamiltonians commute, they can be simultaneously diagonalized and share the same eigenfunctions, while their eigenvalues depend on the coupling parameter g.

For each value of g, we can determine the ground and first excited states of the system. As we turn our control knob and adjust g, the eigenvalues of H change. Just as we change the temperature to drive a classical phase transition by adjusting the importance of enthalpy and entropy, here we change the coupling g to drive a quantum phase transition by adjusting the importance of H_0 and H_1, which relate to the kinetic and potential energies of the system.

Since the eigenfunctions of H_0 and H_1 are independent of g, but their eigenvalues depend on g, it is possible to find a value $g = g_C$ for which the energy of the ground state and first excited state become equal to each other. As g continues to increase, the original ground state energy becomes larger than the original first excited state energy. The system adopts the lowest energy state it can, and thus a phase transition occurs that takes the system to the original first excited state. This is called a level-crossing, and it is depicted below in Figure 2. As can be seen, the ground state energy as a function of g is not analytic at the crossing point — we cannot take a derivative of a sharp corner. In fact, any point of non-analyticity in the ground state energy as a function of g suggests that a quantum phase transition has occurred.

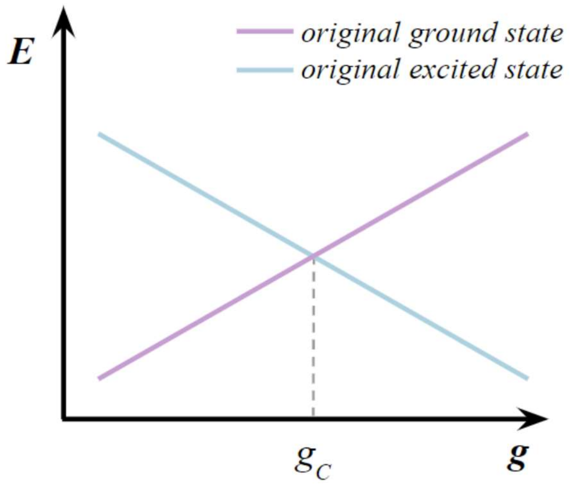

Figure 2: Level-Crossing: *Plotted are the two lowest energy eigenvalues of our system with*

$H = H_0 + gH_1$. As the coupling parameter g increases to g_C we observe a quantum phase transition, where the energies of the quantum states cross. At g_C the ground state energy of the system as a function of g is not analytical, because its first derivative has a discontinuity. As such, this is a continuous phase transition.

In reality, most systems do not exhibit a complete level-crossing, because level-crossing only occurs in the limit of an infinite lattice. Instead, most systems have an avoided level-crossing, in which the energy levels of the ground and excited states approach each other and then diverge again as the coupling increases. At the critical value of the coupling parameter, quantum tunneling is possible between the ground and excited states. As such, quantum phase transitions can happen at level-crossings or avoided level-crossings.

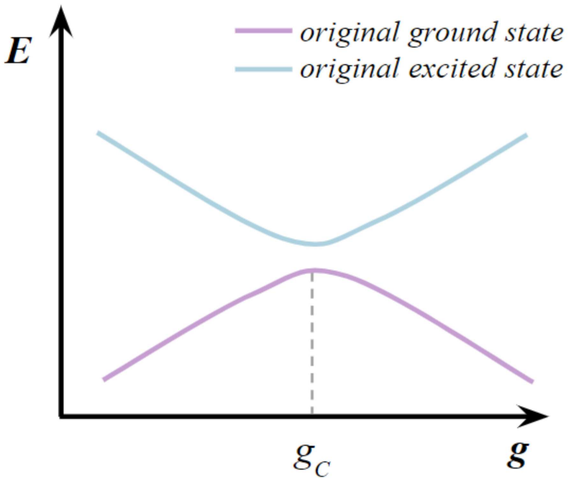

Figure 3: Avoided Level-Crossing: *In finite lattices, complete level-crossing is not observed because of size effects. However, we can still identify a continuous quantum phase transition at g_C.*

Regardless of which type of level-crossing occurs in a quantum phase transition, the same phenomena is observed near the critical point. In particular, we can define an energy scale ε , which is the energy gap between the ground and first excited states of the system. As we approach the critical value of the coupling parameter where $g - g_C = 0$, the energy scale approaches zero with a **critical exponent** $z\nu$. Energy scaling also depends on *J*, a specific parameter that sets the energy of coupling.

$$\varepsilon \sim J|g - g_C|^{z\nu}$$

Near the critical point, we can also define a length scale ξ , which characterizes the spatial correlation of fluctuations in the system's order parameter, and a time scale τ , which reflects the time correlation of fluctuations in the system's order parameter. As we approach the critical point, the length and time scales both become very large and diverge, indicating large fluctuations in these properties. The length and time scales also increase according to a critical exponent, which is related to the critical exponent that dictates energy scaling.

$$\xi \sim |g - g_C|^{-\nu}$$

$$\tau \sim |g - g_C|^{-\nu z}$$

Interestingly, the critical exponents for the energy, length, and time scales are **universal** and do not depend on the details of the system's Hamiltonian. Critical scaling and discontinuities in macroscopic quantities are a defining feature of quantum phase transitions that is shared with classical phase transitions [7,10].

Quantum phase transitions may seem like an interesting concept that is unfortunately relegated to the realm of theory. After all, it is impossible to attain absolute zero temperature, even in laboratories. However, it turns out that understanding the behavior of a system near its critical point allows us to extrapolate outwards and elucidate the system's behavior in nearby regions of phase space. Additionally, it is possible to observe quantum phase transitions at very low, non-zero temperatures, and this has been achieved experimentally in numerous systems [13, 14]. Driving quantum phase transitions at non-zero temperatures is difficult, because the frequency of quantum fluctuations, ω_f, are usually much too small to dominate the system's behavior at finite temperatures [7].

$$\hbar\omega_f << k_B T$$

As such, most phase transitions that occur at temperatures above absolute zero are thermal transitions that can be completely described by the classical theory of continuous phase transitions. However, if the system's temperature is below the critical temperature for a thermal transitions, quantum phase transitions can occur. In fact, quantum fluctuations are always occurring, no matter the temperature, which leads to a unique dynamic regime in which there is an interplay between thermal fluctuations and quantum fluctuations near absolute zero.

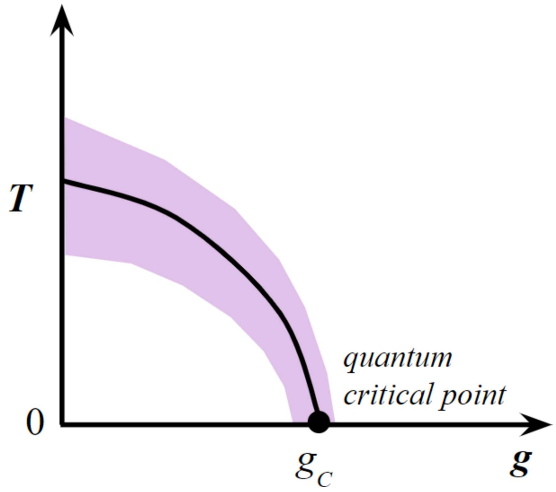

Figure 4. Phase Diagram for Quantum Phase Transition: *This phase diagram shows phase transitions that can occur near and at absolute zero temperature in a quantum system. In the shaded regime, phase transitions can typically be treated classically, because thermal fluctuations dominate over quantum fluctuations. However, at absolute zero temperature, only quantum phase transitions can occur.*

III. Applications

Superconductors, Bose-Einstein condensates, and coupled qubits are all phenomena in condensed matter physics that have garnered lots of attention in the past few decades for their potential to transform technology. However, another commonality between them is that all of these systems can exhibit quantum phase transitions near absolute zero! In this section, we will describe classical and quantum phase transitions in $LiHoF_4$ to contrast transitions driven by thermal fluctuations and quantum fluctuations.

The Ferromagnetic to Paramagnetic Transition of $LiHoF_4$

In 1996 at the University of Chicago, physicists David Bitko, Gabriel Aeppeli, and Thomas Rosenbaum (who went on to become the director of the James Franck Institute) performed experiments to study phase transitions near absolute zero temperature in the insulator lithium holmium fluoride [12, 14, 15]. Generally, $LiHoF_4$ is ferromagnetic, meaning that it becomes magnetic under the application of a magnetizing field and stays magnetic after the field is shut off. However, it is possible to drive $LiHoF_4$ into a paramagnetic phase through a classical or quantum phase transition, in which the material is magnetic under a magnetizing field but loses its magnetic properties after being removed from the field. We will walk through the details of the classical and quantum phase transitions of $LiHoF_4$ to the paramagnetic phase to highlight the differences between the two types of phase transitions using an experimentally realized example.

First, we will use the Maple Quantum Chemistry Toolbox to plot $LiHoF_4$. We obtain the geometry of $LiHoF_4$ by building the molecule in Avogadro and optimizing its geometry. Although we will only model one molecule, it is important to remember that $LiHoF_4$ exists as an ordered crystal lattice.

```
> with(QuantumChemistry) :
> mol := [["Ho",−0.4873322422, 0.1637159069, 0.2158175844], ["Li",−2.1408653684 ,
      2.5706961762 , 1.0304308439], ["F",−1.9780332663,−0.6466759198,−0.4158813579],
      ["F", 0.5037012087, 1.2140153369,−0.8762914780], ["F", 0.0122856321,
      −0.0761820676, 1.9394264582], ["F", 0.5002011921,−1.2737457920,−0.2704423940]];
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mol := [["Ho", −0.48733224, 0.16371591, 0.21581758], ["Li", −2.14086537, 2.57069618,     (3.1.1)
    1.03043084], ["F", −1.97803327, −0.64667592, −0.41588136], ["F", 0.50370121,
    1.21401534, −0.87629148], ["F", 0.01228563, −0.07618207, 1.93942646], ["F",
    0.50020119, −1.27374579, −0.27044239]]
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> PlotMolecule(mol)
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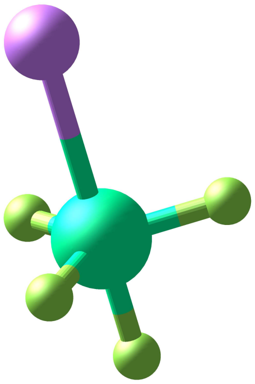

Both Li^+ and F^- have no unpaired electrons and, as such, are not magnetic. However, Ho^{3+} has an overall magnetic moment due to its spin-orbit coupling. As such, we can ignore the lithium and fluorine atoms, and image our material as a collection of nearby holmium atoms with spins. At low temperatures, adjacent holmium atoms interact through dipole-dipole interactions, and their spins will align with the crystal axis. Therefore, the holmium atoms can either be in a state with an up spin, which we will denote $|\uparrow\rangle$, or state with a down spin, which we will denote $|\downarrow\rangle$. These states have magnetic moments, M_Z, oriented along the positive or negative z-direction or crystal axis, respectively. However, because this is a quantum mechanical system, the holmium atoms can also be in a superposition of up and down states. Thus, they can also be in states $|\rightarrow\rangle = \frac{|\uparrow\rangle + |\downarrow\rangle}{\sqrt{2}}$ or $|\leftarrow\rangle = \frac{|\uparrow\rangle - |\downarrow\rangle}{\sqrt{2}}$. Similarly to the up and down states, the net magnetic moments of the superposition states, M_X, are oriented along the positive or negative x-direction, respectively, which is perpendicular to the crystal axis. From our modern day perspective, we can identify the holmium atoms as qubits! It turns out that the Heisenberg uncertainty principle requires that as we increase our certainty about M_X, we decrease our certainty about M_Z. Otherwise, if we knew exactly what M_Z was, we would violate the uncertainty principle, because we would know everything about the potential energy of the crystal. In other words, M_X and M_Z do not commute. This gives us the first ingredient we need for observing a quantum phase transition: strong quantum fluctuations in M_X and M_Z driven by the uncertainty principle!

At the time of Rosenbaum's experiments, it was already known that $LiHoF_4$ underwent a thermal transition to paramagnetism. Near absolute zero, the spins on adjacent holmium atoms prefer to adopt the same orientation to minimize energy by aligning their magnetic moments. However, as temperature is raised and entropy becomes more important, spins prefer to adopt random orientations to minimize their free energy. This results in a thermal transition from a ferromagnetic phase with completely ordered spins to a paramagnetic phase with disordered spins, which can be described by a classical Ising model. However, Rosenbaum's goal was to observe a quantum phase transition in this system. As such, he had to perform his experiments at extremely low temperatures — around 1K above absolute zero — to prevent thermal phase transitions from obscuring quantum phase transitions [15].

The final ingredient that we are missing to drive a quantum phase transition in this system is a control knob, or an external parameter that we can adjust to move between quantum phases of the system. Ideally, it will couple the energies from the two magnetic moments M_X and M_Z. With the set up we have discussed, we can consider the Hamiltonian for this system to be:

$$H(g) = H_X + gH_Z$$

where g is the control parameter that we will adjust to drive the system into a new phase. The Hamiltonians H_X and H_Z represent energies that arise from the magnetic moments M_X and M_Z of each holmium ion.

We can think of H_Z as a potential energy term, since the net magnetic moment of each holmium ion will induce the formation of a magnetic field. The system's energy will be lowest when the magnetic fields of adjacent holmium ions are aligned. Subsequently, we can think of H_X as a kinetic energy term, because its energy contribution will come from spins on the holmium atoms flipping between the up and down states. We can define a notion of velocity of the spin flips and think of the switch between up and down states as a form of quantum tunneling.

Rosenbaum realized that he could use a transverse magnetic field as his control parameter g, to adjust the the relative importance of the kinetic and potential energy terms. When the transverse magnetic field is turned off, the potential energy term will dominate, and the holmium atoms will all tend to be exclusively in the up state $|\uparrow\rangle$ or down state $|\downarrow\rangle$ with aligned magnetic fields. Applying a magnetic field along the +x direction of this system will increase tunneling and cause all of the holmium atoms to favor the $|\rightarrow\rangle$ state. This will destroy any order in the spins, leading $LiHoF_4$ to adopt a paramagnetic state. This system can be modeled nicely by a transverse Ising model, which is the quantum analog to the classical Ising model [7,12,14, 16].

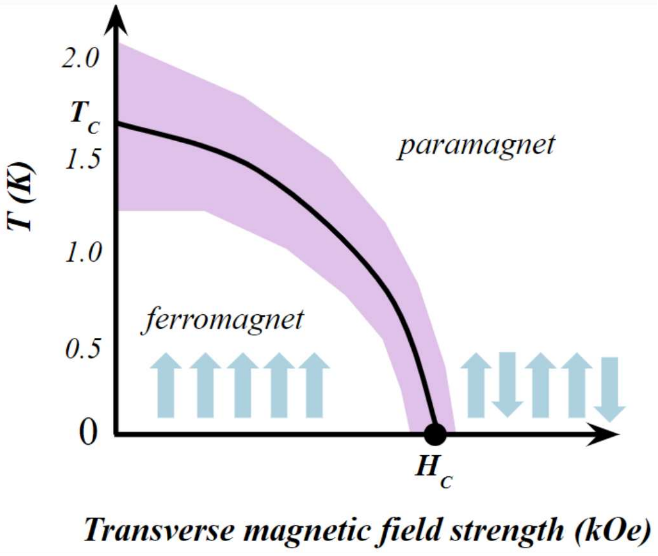

Figure 5. Phase Diagram for $LiHoF_4$: *The phase diagram above shows how adjusting temperature and transverse magnetic field strength can cause a phase transition from a ferromagnetic phase, in which spins are ordered, to a paramagnetic phase, in which spins are completely disordered. At absolute zero, the phase transition is driven by quantum fluctuations that are tuned by the strength of the external transverse magnetic field. Notice that below the critical temperature* $T_C = \sim 1.8\ K$*, quantum phase transitions are possible because thermal fluctuations are not strong enough to completely dominate over quantum fluctuations. The shape of the curve indicates that with a stronger transverse magnetic field, a lower temperature is needed to drive a thermal transition to the paramagnetic phase. In the finite temperature regime below the critical temperature, there is an interesting interplay between quantum and thermal fluctuations [10].*

Although both quantum and thermal fluctuations can drive $LiHoF_4$ from a ferromagnetic to a paramagnetic phase, this example illustrates the difference in the fundamental driving forces and control parameters of the two types of phase transitions. While quantum fluctuations in M_X and M_Z controlled by a transverse magnetic field drive the quantum phase transition, entropy controlled by temperature drives the classical phase transition. This example also highlights the similarities between the two types of phase transitions. Both require fluctuations, are modulated by an external control parameter, and result from the system's need to balance two energies that are in tension with each other in order to minimize an energy that determines the phase of the system.

Unlocking the Phase Beyond the Bose-Einstein Condensate

Although Einstein predicted the existence of a Bose-Einstein condensate around 1924, it took until 1995 for the first one to be experimentally created by Cornell and Wieman. Bose-Einstein condensates are important because they play a key role in superfluidity, or viscosity-free flow of particles, and superconductivity, or resistance-free flow of electrons. In fact, their discovery was so revolutionary that Cornell and Wieman were awarded the 2001 Nobel prize in physics for their creation of the first experimental Bose-Einstein condensate.

Bosons are quantum particles with integer spin which have symmetric wavefunctions. Unlike fermions, many bosons can condense into a single quantum state at low enough temperatures, exhibiting a classical phase transition. This transition is driven by the decreased importance of entropy at low temperatures, and its order parameter is the occupation of system's ground state. In their groundbreaking experiment, Cornell and Wieman created a gaseous Bose-Einstein condensate by cooling rubidium atoms to around 170 nanokelvins [17]. However, in 2002, Bloch and his collaborators realized that an additional phase of this system may be possible. Even though the rubidium atoms in the Bose-Einstein condensate occupied a single quantum state, they remained delocalized across the optical trap used to form the condensate.

In an attempt to localize a finite number of atoms in each periodic well of the trap, Bloch used a laser beam to deepen the strength of the trap's periodic potential. Upon doing so, the ultracold rubidium atoms underwent a quantum phase transition from a superfluid condensate phase to a Mott insulator phase [18]. A phase diagram depicting this transition is displayed below. In this instance, exploiting quantum fluctuations yielded a novel phase of matter with useful applications to new technologies, like the Heisenberg-limited atom interferometer [19].

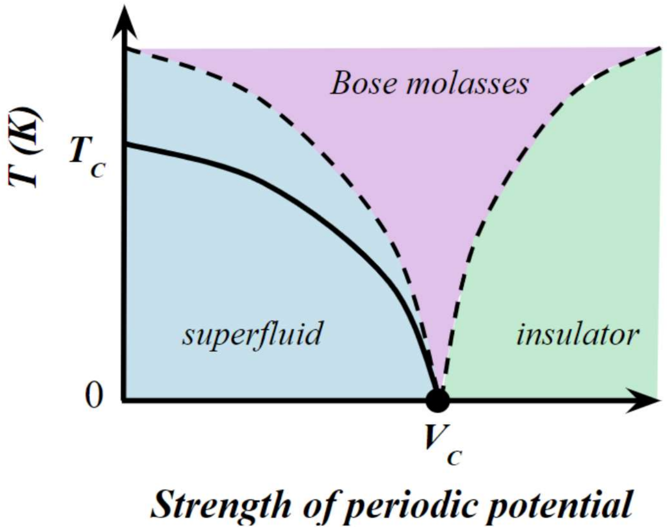

Figure 6. Phase Diagram for a Bose-Einstein condensate: *The phase diagram above shows how tuning the strength of the periodic potential in an optical trap of ultracold rubidium atoms induces a quantum phase transition. With a stronger periodic potential, a finite number of atoms become localized in their respective potential wells, as the system transitions from a condensate superfluid phase to a Mott insulator phase. In between these two phases is the Bose molasses phase, a quantum critical region where quantum fluctuations are non-negligible, and the effects of the quantum phase transition at absolute zero on the properties of the system can be observed.*

IV. Discussion

Even though quantum and classical phase transitions share a closely interwoven history, quantum phase transitions are unique, purely quantum mechanical phenomena that occur at absolute zero temperature. While entropy and thermal fluctuations drive classical phase transitions, the Heisenberg uncertainty principle and quantum fluctuations drive quantum phase transitions. Both types of phase transitions occur when an external control parameter is tuned to adjust the importance of different components of the system's energy, which drives it into a new free energy minimum or ground state. At finite temperatures, thermal transitions usually dominate over quantum transitions because thermal fluctuations are stronger than quantum fluctuations. However, below the system's critical temperature and near absolute zero, quantum phase transitions can be observed. In the past century, quantum phase transitions have been used to unlock numerous new phases of matter and play a role in countless phenomena in condensed matter, including cuprate superconductors, magnetism, the metal-insulator transition, and Bose Einstein condensates. Interest in quantum phase transitions has been renewed in the age of quantum computers, as researchers use quantum computers to perform better simulations of quantum phase transitions and understand potentially useful phase transitions in qubits [20].

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