Influence of lattice vibrations on a quantum phase transition

In the vicinity of the absolute zero of temperature, all materials, except Helium, are solid. Analogous to this, the conduction electrons in certain metallic compounds can "freeze" at low temperature into a Charge-Density Wave phase: a stationary, ordered, spatial modulation of the electron density. If the temperature is raised, the charge density wave order is destroyed (it "melts"). This is a result of thermal fluctuations, as in a classical solid-to-liquid phase transition. But at zero temperature, in spite of the absence of any thermal fluctuations, it is possible to cause the charge density wave to melt due to quantum fluctuations. Such a transition is called a quantum phase-transition. To understand the rich physics associated with this type of transition, we need to know what drives the transition.

Charge density waves (CDW's) are a particularly interesting case where the electronic phase (*i.e.* the electron-density modulation) is associated with a lattice distortion. In order to decrease the electronic energy and due to a strong interaction between the conduction electrons and the lattice, the charge density wave alters the equilibrium position of the atoms, creating a periodic lattice distortion whose repeat-length is of the order of several lattice constants.

The atoms in a solid vibrate around their equilibrium positions. The oscillation amplitude is greater the higher the temperature, but even at zero temperature the vibration amplitude is non-zero. This is the "zero-point motion", a direct consequence of the Heisenberg uncertainty principle. The potential in which lattice atoms vibrate is usually approximated by a harmonic potential, i.e. an atom's potential energy is a quadratic function of its distance from its equilibrium position. In reality, this potential is not harmonic, which explains macroscopic physical properties of materials at high temperature such as thermal expansion. But this anharmonicity can have important consequences, even at zero Kelvin, for the stability of an electronic phase coupled to the lattice.

We have studied the spectrum of the lattice vibrations (the phonon spectrum) for the chalcogenide compound Niobium Diselenide ($NbSe_2$). This compound develops a charge density wave phase below temperature 33.5 K, and also superconductivity below 7.2 K (see Fig. 1). Our experiments were done on a dedicated X-Ray beamline (ID28) at the European Synchrotron Radiation Facility, Grenoble. We measured the loss of energy of the X-ray beam, scattered by the phonons in a 100 micron size sample of $NbSe_2$. The sample was placed in a sealed capsule of Helium compressed by two diamonds, situated in a cryostat. To tune $NbSe_2$ across its quantum phase transition, we applied hydrostatic pressures up to 40 000 atmospheres (4 GPascals), at various temperatures (see Fig. 1).

We observed that the energy of a specific phonon mode, the precursor of the lattice distortion, decreases as we lower the temperature towards the phase-transition – the phonon "softens". This is because, unlike in the case of a harmonic oscillator, the oscillation frequency (or the energy of the phonon) depends on the amplitude of the oscillation, which decreases with decreasing temperature. It is this weakening of the vibrations that triggers the freezing into the CDW phase at 33.5 K for P =0 (Fig. 1). Under pressure, the amplitude of the oscillation decreases and, intuitively, we expect that the atoms experience mainly the harmonic part of the potential. However, we observe that this same phonon softens significantly even at pressures above the critical pressure.

To account for the experimental results, the phonon spectra, including the effects of anharmonicity, were calculated from first principles by our colleagues at the Pierre & Marie Curie University, Paris (UPMC). Their calculations explain the experimental results both qualitatively and quantitatively over a wide range of temperature and pressure. They show that, in the CDW phase, the lattice potential for the atomic vibrations has a "double-well" shape. Each atom is "frozen" into one of the two wells, see sketch (i) in Fig. 1 which shows the CDW-related lattice distortion.

Squeezing the crystal lattice by applying pressure smoothes out the double well (sketches ii and iii), but this shape persists over a certain pressure range outside the charge density phase. So why has the lattice distortion disappeared in that range? The reason is that, even at low temperature, the atoms feel both wells due to their large zero-point motion.

Thus, the quantum vibration of the lattice atoms is the mechanism that drives the "melting" of the charge density waves at the quantum phase transition. We note finally that, as seen in Fig. 1, the superconductivity is only marginally affected by the presence of the charge density waves.

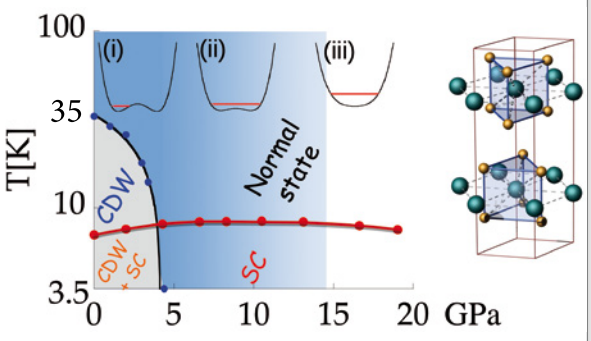

***Fig. 1 - At left**: P-T phase diagram for $NbSe_2$. Charge Density Waves are observed In the grey zone, coexisting with superconductivity (SC) below the red boundary. At 0 K, the charge density wave is stable up to 4.6 GPa pressure.*

***i, ii, iii**: Sketches of the potential energy of a lattice atom vs. its position, at T near 0 K, for three values of pressure. The red lines indicate the energy and the range of quantum motion of the atom. These atomic vibrations correspond to the specific phonon mode measured by X-Ray scattering.*

***At right**: Crystal structure of $NbSe_2$, a hexagonal lattice of Nb atoms (blue) sandwiched between surrounding Se atoms (gold).*

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FURTHER READING...

"Strong anharmonicity induces quantum melting of charge density wave in 2H-$NbSe_2$ under pressure"
M. Leroux, I. Errea, M. Le Tacon, S. M. Souliou, G. Garbarino, L. Cario, A. Bosak, F. Mauri, M. Calandra and P. Rodière
***Phys. Rev. B* 92**, 140303(R) (2015).

"Anharmonic suppression of charge density waves in 2H-NbS_2"
M. Leroux *et al.*
***Phys. Rev. B* 86**, 155125 (2012).