Quasiparticle scattering

In the last chapter we have encountered the plasmon as a quasiparticle describing a collective excitation of the free electron gas. In this chapter we will see, how the plasmon and other quasiparticles can be detected via scattering with photons, electrons, or neutrons. But let us start with a brief review on the most important quasiparticle, the phonon.

Excitations of vibrations in the lattice can be described by phonons. The complex patterns of vibrations can then be treated as a superposition of these elementary excitations. slide The dependency of wave vector and frequency is given by the phonon dispersion relation. The standard model system to describe phonons is the linear diatomic chain. (slide). Note that a meaningful wave vector can only be defined inside the first Brillouin zone as otherwise the wavelength does get shorter than the unit cell, which is not meaningful as the crystal lattice is discrete. slide The energy states follow from the dispersion relation according to: $E_n(q) = \hbar\omega(q)\left(n+\frac{1}{2}\right)$ As an example, the phonon dispersion of the fcc lattice is shown (slide) for the case of Si.

There are two classifications used to describe phonons: longitudinal / transversal and acoustical / optical. For longitudinal phonons, the displacement of the atoms occurs parallel to the direction of the wave, whereas for transversal phonons this displacement is perpendicular to the wave vector. For acoustic phonons, the atoms inside the unit cell move all in the same direction, whereas for optical phonons, these atoms vibrate against each other. When the charges in a crystal are not evenly distributed (e.g. in an ionic crystal), an optical phonon induces an oscillating dipole moment, which allows coupling of these optical phonons to photons. As light is a transversal wave, only coupling to transversal phonons is possible. We have encountered these phonons already in the last chapter as the vibrational oscillators in the Lorentz-model.
A possible way to describe this interaction is in the quasiparticle picture. Here, momentum and energy have to be conserved during absorption of a photon by a phonon. The comparison of the dispersion relation of phonons and photons slide show, that this is only possible for the optical branch of the phonon dispersion curve.

We will now turn to the basics of *inelastic* scattering, which is a means to detect quasi-particles. To start general, we do not yet specify wether the scattered particle is a photon, electron, or neutron. Let us look at the conservation laws for scattering $\boxed{\text{slide}}$.

First, we have conservation of momentum. Here, we have to remember that due to the broken translational symmetry of space induced by the crystal, momentum is only conserved up to additions of reciprocal lattice vectors.

We have:
$$\underbrace{\vec{Q}}_{\text{momentum transfer}} = \underbrace{\vec{q}}_{\text{momentum of phonon}} + \underbrace{\vec{G}}_{\text{reciprocal lattice vector}} = \underbrace{\vec{k} - \vec{k}'}_{\text{incident and scattered wave}}$$

In addition, we have conservation of energy:
$$E = E' + E_{Phonon},$$

which ca be used to calculate E_{Phonon}.

For the case of electrons or neutrons, we have $E = \frac{\hbar^2 k^2}{2m}$ and hence
$$E_{Phonon} = \hbar\omega_{ph} = \frac{\hbar^2}{2m}\left(k^2 - k'^2\right).$$

For photons, we have:
$$E_{Phonon} = \hbar\omega_{ph} = \hbar(\omega - \omega').$$

Furthermore, spin is conserved.

Let us now look at scattering of photons, which is known as Raman scattering around (slide) the visible part of the spectrum. These photons have a very small wave vector:

$$k_0 = \frac{2\pi}{\lambda} \approx \frac{2\pi}{4000\,\text{Å}} \approx 1\cdot 10^{3}\,\text{Å}^{-1}$$

The edge of the Brillouin zone (≙ maximum wave vector of the phonon) is found at $\frac{\pi}{a} = \frac{\pi}{2\,\text{Å}} \approx 1{,}5\,\text{Å}^{-1}$. Therefore, visible light is only sensitive to phonons at the very center of the Brillouin zone. Raman spectroscopy can also detect other quasi particles, as the plasmon encountered in the previous chapter.

The schematic setup is shown in the (slide). As a monochromatic source of high intensity is needed (Raman scattering has a low probability as compared to elastic - Rayleigh - scattering) the laser is the right choice. The very large elastic signal is then filtered and the remaining light is analyzed in a spectrometer.

The atomic processes in Raman scattering can also be visualized in an energy level scheme (slide) using virtual energy levels. Besides the creation of a phonon (Stokes process), an existing photon can also be absorbed by the photon (Anti-Stokes).

Anti-Stokes scattering will only be possible if there are phonons present in the material before the light is incident. The probability for anti-Stokes scattering therefore decreases on lowering the temperature as the phonon population decreases. On the other hand, Stokes scattering does not require a phonon to be present and can therefore occur at any temperature. As the population of a given phonon level follows a Boltzmann distribution, the ratio of anti-Stokes to Stokes scattering events is given by:

$$\frac{I_{\text{anti-Stokes}}}{I_{\text{Stokes}}} = e^{-\hbar\omega_{ph}/k_B T} \qquad \boxed{\text{slide}}$$

Inelastic light scattering is a weak process. This is because we are dealing with a higher-order interaction than for linear interactions such as absorption. As an example the $\boxed{\text{slide}}$ shows a Raman spectrum of GaAs, where the TO and LO phonon are visible. In addition, a weaker peak due to the plasmon occurs.

Recently, Raman spectroscopy was extensively applied to the new material graphene, as it allows (i) to distinguish single-, bi-, and few layer graphene and (ii) allows to asses the quality (defect density) $\boxed{\text{slide}}$.

A *raman microscope* can be used to spatially resolve the vibrational properties of the sample, see the slide for a example.

For excitations very close to the Γ-point, the dispersion of these modes can be detected by Raman spectroscopy. This reveals coupling between different modes: The slide shows how the photon and the TO phonon form a polariton in GaP, leading to *avoided crossing* (anticrossing). Generally, a polariton is a coupling between a photon and a phonon/plasmon/exciton

The spatial resolution can be enhanced even further by performing Raman spectroscopy inside an STM or AFM: *Tip-enhanced Raman Spectroscopy* (TERS). The basic effect is that the local electric field can be enhanced in a gap between the surface and a metallic tip of the right shape and material (slide) The main Raman signal thus comes from the small area underneath the tip and can be related to the topography measured simultaneously. Here, vibrations of single molecules can be detected, e.g. RNA slide

In optical measurements only phonons at the center of the Brillouin zone ($\vec{q} \approx 0$) can be studied, since energy and momentum conservation can be satisfied only here on account of the great disparity between phonon and photon propagation velocities. The situation is different for neutrons. The $\boxed{\text{slide}}$ used to illustrate this was already shown in the context of diffraction. It shows the dispersion relation for photons, neutrons and electrons (and He). The region where the phonons live is the intersection of the blue bars (wavelength on the order of interatomic distances, energy in the range of thermal excitations). Of course, $\lambda = \frac{2\pi}{k}$. Only neutrons have the right energy and momenta to map the whole Brillouin zone easily. Therefore, they are the ideal probe to detect phonons. The experimental setup used here is the triple-axis-spectrometer $\boxed{\text{slide}}$. A white beam of neutrons is produced in a reactor and monochromatized using Bragg reflection. The wavelength of the incoming light is then selected by turning the monochromator around the first axis. The orientation of the sample with respect to the beam is altered using the second axis. After the scattering event, the (now reduced) energy of the neutrons is measured using again Bragg reflection around the third axis. Variation of incident energy, final energy, scattering angle and sample orientation allows to map the whole Brillouin zone. The slide shows an example.

Due to their sensitivity to the magnetic moment, inelastic neutrons are also the right probe for the detection of spin waves or magnons (slide). These are collective excitations of the electrons' spin structure in a lattice. The slide shows an example of a measured magnon dispersion curve.