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Generalized plasmons in layered systems

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Summary. — We study the spectrum of electromagnetic modes in layered superconductors. We include the mixing between longitudinal and transverse degrees of freedom, which has been partly overlooked in the previous literature and is crucial to describe the correct behaviour at long wavelengths. We derive a generalized plasma mode, which provides a link between the standard description of the layered plasmon and the one of the Josephson plasmon: we show that these are, respectively, the large-momentum and the low-momentum limits of our model.

1. – Introduction

Plasmons are longitudinal oscillations of the conduction electrons in isotropic metals. They constitute, along with the transverse electromagnetic (e.m.) waves propagating trough matter, the hybrid light-matter modes of metals, and can be described within the classical framework of Maxwell's equations. Theoretically, e.m. waves and plasmons arise, respectively, as transverse ($\boldsymbol{\nabla} \cdot \boldsymbol{E}_{\mathrm{T}} = 0$) and longitudinal ($\boldsymbol{\nabla} \times \boldsymbol{E}_{\mathrm{L}} = \boldsymbol{0}$) solutions obeying the following equations [1]:

$$
\begin{cases} \left(\omega^2 - \omega_{\mathrm{P}}^2 - \tilde{c}^2 |\boldsymbol{q}|^2\right) \boldsymbol{E}_{\mathrm{T}}(\boldsymbol{q}, \omega) = \boldsymbol{0}, \\ \left(\omega^2 - \omega_{\mathrm{P}}^2\right) \boldsymbol{E}_{\mathrm{L}}(\boldsymbol{q}, \omega) = \boldsymbol{0}, \end{cases} \tag{1}
$$

where $\boldsymbol{q}$ and ω are, respectively, the wavevector and the frequency of the e.m. mode. $\tilde{c} \equiv c/\sqrt{\varepsilon_{\mathrm{B}}}$ is the light velocity in the medium and $\omega_{\mathrm{P}} \equiv \sqrt{(4\pi e^2 n)/(\varepsilon_{\mathrm{B}} m)}$ is the isotropic plasma frequency, ε_{B} being the background dielectric constant. Plasmons can also arise when a metal undergoes a superconducting (SC) transition: in such case, they involve longitudinal oscillations of the superfluid electrons.

From an experimental point of view, plasmons can be studied by means of techniques such as non-linear Terahertz (THz) spectroscopy [2-5], where intense THz radiation is used to illuminate samples, or Electron Energy Loss Spectroscopy (EELS) [6, 7], where high-energy electron beams are used as an external scalar perturbation to probe longitudinal excitations. Such experimental protocols have been widely used recently to investigate layered cuprate superconductors. These systems host arrays of stacked 2D copper-oxide SC layers coupled by a weak Josephson interaction, which pushes the energetic cost of an inter-layer (*i.e.*, along the crystallographic *c*-axis) Josephson-plasma mode (JPM), whose zero-momentum value we denote by ω_{c}, down to the THz range; on the other hand, the high intra-layer (*i.e.*, along the crystallographic *ab*-planes) carrier mobility allows for the existence of a high-energy plasmon (ω_{ab} at $\boldsymbol{q} = \boldsymbol{0}$). Cuprates show then a strong anisotropy along the *c*-direction, and offer therefore a wide spectrum of excitations, ranging from few THz to the eV. EELS experiments, due to the finite electron mass and the large amount of energy carried by the beams, give access to the large-momentum and high-energy plasmon dynamics; such regime can be accounted for by a layered generalization of the isotropic plasma dispersion, $\omega_{\mathrm{L}}^2(\boldsymbol{q}) = \left(\omega_{\mathrm{ab}}^2 q_{\mathrm{ab}}^2 + \omega_{\mathrm{c}}^2 q_{\mathrm{c}}^2\right)/|\boldsymbol{q}|^2$ [8], which is the standard layered plasmon usually quoted in the literature. On the other hand, non-linear THz spectroscopy explores the dynamics of the low-momentum, low-energy JPM, whose dispersion is given by $\omega_{\mathrm{J}}^2(\boldsymbol{q}) = \omega_{\mathrm{c}}^2\left(1 + \lambda_{\mathrm{c}}^2 q_{\mathrm{ab}}^2/\left(1 + \lambda_{\mathrm{ab}}^2 q_{\mathrm{c}}^2\right)\right)$, $\lambda_{\mathrm{ab/c}} = \tilde{c}/\omega_{\mathrm{ab/c}}$ being the in-plane/out-of-plane SC penetration depth [9].

Despite the previous formulae being correct within their limit of validity, they do not match each other. Surely the JPM dispersion ω_{J} is not suitable at generic energy and momentum, since it is valid only around the low-momentum, low-energy JPM. On the other hand, the standard layered plasmon ω_{L} does not reduce, in the limit of small energy and momentum, to the JPM ω_{J}, which is known to be correct in such limit. The reason for such discrepancy lies in the fact that the standard layered plasmon dispersion is missing a finite contribution coming from the mixing between longitudinal and transverse degrees of freedom at small momenta, therefore it fails in this region of the momentum space. We already addressed such issue in a previous work, where we provided a complete description of plasma excitations in layered superconductors, including such mixed longitudinal-transverse contribution and thus valid at different energy and momentum scales, within the effective action formalism [10]. In this paper we show how a similar description can be achieved within the classic framework of Maxwell equations.

2. – Longitudinal-transverse mixing and generalized plasma waves in layered systems

In a superconducting medium at zero temperature, Maxwell's equations acquire a non-zero current term $4\pi\boldsymbol{J}/c$ taking into account the flow of superfluid electrons. The internal current $\boldsymbol{J}$ is related to the internal electric field $\boldsymbol{E}$ via the equation $\boldsymbol{J} = \hat{\boldsymbol{\sigma}}\boldsymbol{E}$, where the conductivity tensor $\hat{\boldsymbol{\sigma}}$ is given by the first London equation [11]. For example, in an isotropic system, conductivity reduces to a scalar, *i.e.*, $\sigma_{ij} = \sigma\delta_{ij}$, where $\sigma = ne^2/(-i\omega m)$ (as given by the isotropic London equation), n being the density of superfluid electrons, m being the isotropic electron effective mass. In such a situation, $\boldsymbol{J}$ is parallel to $\boldsymbol{E}$, hence the former can be recast as $\boldsymbol{J} = \sigma\boldsymbol{E}_{\mathrm{T}} + \sigma\boldsymbol{E}_{\mathrm{L}}$, where $\boldsymbol{E}_{\mathrm{L}} = (\hat{\boldsymbol{q}}\cdot\boldsymbol{E})\hat{\boldsymbol{q}}$ and $\boldsymbol{E}_{\mathrm{T}} = \boldsymbol{E} - \boldsymbol{E}_{\mathrm{L}}$ are, respectively, the longitudinal ($\hat{\boldsymbol{q}} \times \boldsymbol{E}_{\mathrm{L}} = \boldsymbol{0}$) and the transverse ($\hat{\boldsymbol{q}} \cdot \boldsymbol{E}_{\mathrm{T}} = 0$) components of the electric field with respect to the direction $\hat{\boldsymbol{q}} = \boldsymbol{q}/|\boldsymbol{q}|$ set by the wavevector $\boldsymbol{q}$. The previous equation for the isotropic current leads, along with Maxwell's equations, to eqs. (1), which describe the propagation of two uncoupled pure modes, *i.e.*, a purely transverse mode (the e.m. wave) and a purely longitudinal mode (the plasmon): for this reason, the isotropic e.m. modes are said to admit an exact longitudinal-transverse decomposition.

In layered systems, the stacked structure along the out-of-plane direction manifests as an anisotropy of the effective mass with respect to the crystallographic axes, *i.e.*, $m_{\rm ab} \neq m_{\rm c}$. This implies $\sigma_{\rm ab} \neq \sigma_{\rm c}$. In such a situation $\boldsymbol{J}$ is not parallel to $\boldsymbol{E}$, and a longitudinal-transverse decomposition of the e.m. modes is not achievable in general. In order to see this fact one can rotate the conductivity tensor $\hat{\boldsymbol{\sigma}}$ into the basis spanned by $\boldsymbol{E}_{\rm L}$ and $\boldsymbol{E}_{\rm T}$. Let $q_{\rm ab/c}$ be the in-plane/out-of-plane component of $\boldsymbol{q}$. Starting from $\boldsymbol{E}_{\rm L} = (\hat{\boldsymbol{q}} \cdot \boldsymbol{E})\hat{\boldsymbol{q}}$ and $\boldsymbol{E}_{\rm T} = (\hat{\boldsymbol{q}} \times \boldsymbol{E}) \times \hat{\boldsymbol{q}}$ one finds that the rotation matrix among the Cartesian basis and the $(\boldsymbol{E}_{\rm L}, \boldsymbol{E}_{\rm T})$ basis reads $\hat{\mathcal{U}}(q) = \begin{pmatrix} q_{\rm ab} & q_{\rm c} \\ -q_{\rm c} & q_{\rm ab} \end{pmatrix} / |\boldsymbol{q}|$. The conductivity tensor $\hat{\boldsymbol{\sigma}}_{\rm LT}$ in longitudinal-transverse coordinates can then be computed as:

$$\hat{\boldsymbol{\sigma}}_{\rm LT}(\boldsymbol{q}) = \hat{\mathcal{U}}(q)\hat{\boldsymbol{\sigma}}\hat{\mathcal{U}}^{\rm T}(q) = \begin{pmatrix} \sigma_{\rm L} & \sigma_{\rm mix} \\ \sigma_{\rm mix} & \sigma_{\rm T} \end{pmatrix} \tag{2}$$

where we denoted the purely longitudinal and the purely transverse elements of the anisotropic conductivity by $\sigma_{\rm L} = \left(\sigma_{\rm ab} q_{\rm ab}^2 + \sigma_{\rm c} q_{\rm c}^2\right) / |\boldsymbol{q}|^2$ and $\sigma_{\rm T} = \left(\sigma_{\rm ab} q_{\rm c}^2 + \sigma_{\rm c} q_{\rm ab}^2\right) / |\boldsymbol{q}|^2$ respectively, while $\sigma_{\rm mix} = -\left(\sigma_{\rm ab} - \sigma_{\rm c}\right) q_{\rm ab} q_{\rm c} / |\boldsymbol{q}|^2$. It is worth noting that, in the isotropic limit $\sigma_{\rm ab} = \sigma_{\rm c} = \sigma$, $\sigma_{\rm mix}$ vanishes and $\sigma_{\rm L/T} = \sigma$, so that a purely diagonal conductivity, which leads to the exact longitudinal-transverse decomposition of the isotropic e.m. modes, is recovered. In a layered system, due to $\sigma_{\rm ab} \neq \sigma_{\rm c}$, $\sigma_{\rm mix}$ is, in general, finite and the conductivity tensor is non-diagonal in the $(\boldsymbol{E}_{\rm L}, \boldsymbol{E}_{\rm T})$ basis: therefore, an exact longitudinal-transverse decomposition of the e.m. modes is not always allowed. Indeed, putting the anisotropic current $\boldsymbol{J} = \begin{pmatrix} \sigma_{\rm L} & \sigma_{\rm mix} \\ \sigma_{\rm mix} & \sigma_{\rm T} \end{pmatrix} \begin{pmatrix} \boldsymbol{E}_{\rm L} \\ \boldsymbol{E}_{\rm T} \end{pmatrix}$ into Maxwell's equations yields an eigenvalue problem whose solutions are not, in general, purely longitudinal or purely transverse: they are mixed longitudinal-transverse. The strength of such mixing will depend on the overall magnitude of the non-diagonal element $\sigma_{\rm mix}$. Since $\sigma_{\rm ab/c} = ne^2/(-i\omega m_{\rm ab/c})$, as given by the anisotropic London equation, one would find that $\sigma_{\rm mix} = -\varepsilon_{\rm B}(\omega_{\rm ab}^2 - \omega_{\rm c}^2)/(4\pi i\omega) q_{\rm ab} q_{\rm c} / |\boldsymbol{q}|^2$, $\omega_{\rm ab/c} = \sqrt{(4\pi e^2 n)/(\varepsilon_{\rm B} m_{\rm ab/c})}$ being the in-plane/out-of-plane plasma frequency. Further, one can define the crossover momentum $q_{\rm cr} \equiv \sqrt{\omega_{\rm ab}^2 - \omega_{\rm c}^2}/\tilde{c}$, so that $\sigma_{\rm mix}$ can be recast as $\sigma_{\rm mix} = c^2 q_{\rm ab} q_{\rm c}/(4\pi i\omega) q_{\rm cr}^2 / |\boldsymbol{q}|^2$: the magnitude of $\sigma_{\rm mix}$ is then set by ratio $q_{\rm cr}^2/|\boldsymbol{q}|^2$. At large momenta, *i.e.*, for wavevectors much bigger than the crossover value ($|\boldsymbol{q}| \gg q_{\rm cr}$), $\sigma_{\rm mix}$ vanishes, the off-diagonal elements of (2) can then be neglected and one finds a purely transverse and a purely longitudinal mode, the latter propagating at $\omega_{\rm L}^2 = 4\pi\omega\sigma_{\rm L}/(i\varepsilon_{\rm B}) = \left(\omega_{\rm ab}^2 q_{\rm ab}^2 + \omega_{\rm c}^2 q_{\rm c}^2\right) / |\boldsymbol{q}|^2$ (*i.e.*, the standard layered plasmon). To get an idea on the order of magnitude of the momenta for which such regime is attained one can consider the typical values of the plasma energies in layered materials, which are such that $\omega_{\rm c} \ll \omega_{\rm ab}$, with $\omega_{\rm ab} \simeq 1$ eV, $\omega_{\rm c} \simeq 10^{-3}$ eV, and the value of the light velocity $\tilde{c} \simeq 0.19\,{\rm eV}\mu{\rm m}$, thus finding that $q_{\rm cr} \simeq 5\,\mu{\rm m}^{-1}$: therefore, as soon as $|\boldsymbol{q}| \gtrsim 5\,\mu{\rm m}^{-1}$, $\sigma_{\rm mix}$ can be neglected and the standard layered plasmon is recovered. Conversely, at $|\boldsymbol{q}| \lesssim 5\,\mu{\rm m}^{-1}$, $\sigma_{\rm mix}$ is finite and can not therefore be neglected. In such case one would find two modes with mixed longitudinal-transverse character, valid at different energy and momentum scales: they consist of a quasi-transverse mode (*i.e.*, a predominantly transverse mode with a finite longitudinal projection) and a quasi-longitudinal mode (*i.e.*, a predominantly longitudinal mode with a finite transverse projection). Such generalized-plasmon solutions have already been discussed by us in a recent work [10]. Here we focus on the quasi-longitudinal one $\omega_{\rm QL}$, whose momentum dependence we show in fig. 1 at selected values of the angle η between $\boldsymbol{q}$ and the c axis. We also show, for comparison, the standard

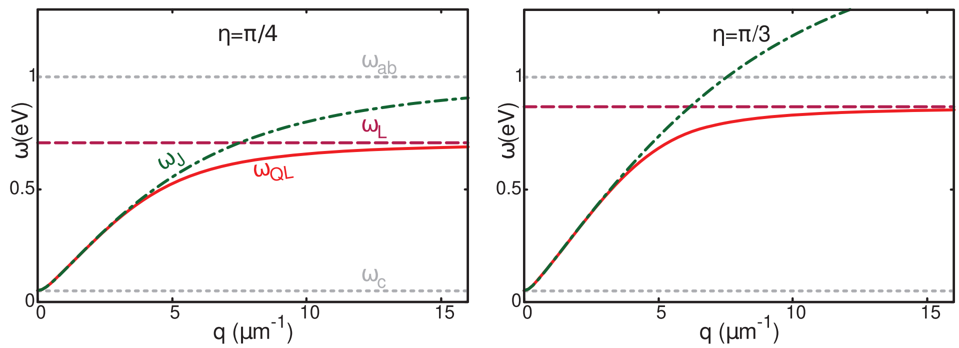

Fig. 1. – Momentum dependence of ω_{QL} (solid line) at selected value of η. We also show for comparison ω_{L} (dashed line) and ω_{J} (dot-dashed line).

layered plasmon ω_{L} and the low-energy JPM ω_{J}. Indeed, ω_{QL} interpolates between the large-momentum dispersion ω_{L} and the low-momentum, low-energy dispersion ω_{J}. As already discussed, the standard layered plasmon ω_{L} is recovered as soon as $|\boldsymbol{q}|$ is bigger than the crossover value $q_{\mathrm{cr}} \simeq 5\,\mu\mathrm{m}^{-1}$. Such behaviour is evident from fig. 1: as soon as $|\boldsymbol{q}| > 5\,\mu\mathrm{m}^{-1}$, ω_{QL} tends to the dispersion ω_{L}. On the other hand, the discrepancy between ω_{QL} and ω_{L} below the crossover value becomes crucial in order to understand the radically different description of the JPM, which is attained at low momentum and low energy, *i.e.*, for ω such that $\omega \sim \omega_{\mathrm{c}} \ll \omega_{\mathrm{ab}}$: indeed, as shown in fig. 1, the JPM dispersion ω_{J} accounts for the correct behaviour of ω_{QL} at energies around ω_{c} and small momenta, *i.e.*, in the region where the standard layered plasmon ω_{L} fails.

In conclusion, we provided a full description of plasmons in bulk layered superconductors, which we already addressed in detail in ref. [10], by solving Maxwell's equations with an anisotropic current term. By properly including the mixing between transverse and longitudinal components of the e.m. fields, mixing which is absent in isotropic systems and which has been partly overlooked in layered systems so far, we were able to identify a generalized plasma mode, valid at generic energy and momentum. Such solution fills the knowledge gap among previous results, that focused on specific regions of the energy/momentum spectrum of the plasmon. The present results, despite having been derived for the SC state, can also be generalized to the normal state, provided that the plasmon damping induced at low energy by the presence of particle-hole excitations, which is absent at low temperatures in superconductors, is properly taken into account.

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