Josephson surface plasmons in spatially confined cuprate superconductors

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The Josephson plasma wave (JPW) is a characteristic low-energy excitation in high-$T_c$ cuprate superconductors, arising from interlayer tunneling of the superfluid density along the $c$-axis. In this work we determine the surface excitations that arise when the superconductor supporting the JPW is spatially confined in the form of a small spherical particle, leading to fully localized Josephson surface plasmons. We show that such excitations can be probed by optical scattering and electron energy-loss spectroscopy. The case of multilayered superconductors supporting a transverse-optical JPW is treated in detail. The present results generalize the theory of surface plasmons in metal nanoparticles to the case of spherical particles made of layered high-$T_c$ superconductors.

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High-$T_c$ superconductors (HTSCs), especially the cuprates, are strongly anisotropic systems whose layered structure can be viewed as a stack of conductive CuO$_2$ planes along the $c$-axis separated by insulating block layers. At variance from ordinary metals, they support two kinds of plasma excitations: the Josephson plasma wave (JPW), arising from interplane Josephson tunneling of the superfluid density, and quasi-2D (acoustic) plasmons that result from plasma oscillations of the condensate along the planes, while normal electrons are responsible for dissipation. JPWs propagate at the Josephson plasma frequency $\omega_J$, which depends on the properties of the Josephson junction formed between two consecutive CuO$_2$ planes through the interplane distance and the critical current, thus providing a good estimate of the strength of interplane Josephson coupling.

The mechanisms determining the critical temperature $T_c$ of HTSCs are not well established, yet $T_c$ is known to depend on the doping level, on apical-oxygen distance from the CuO$_2$ plane, and in multilayered superconductors (MLSCs) it increases with the number $n$ of CuO$_2$ layers in a unit cell up to at least $n = 3$. Studying the JPW may contribute to clarifying the mechanism of high-$T_c$ superconductivity, as it has been proposed that Josephson tunneling may be responsible of an increase of the superconducting condensation energy proportional to $\omega_J^2$. This is especially likely to occur in MLSCs, where inter- and intra-multilayer couplings give rise to both low-frequency (acoustic) and optical JPWs. The frequency of the latter is in the mid-infrared range, it is of the same order of the superconducting gap, and it correlates with the transition temperature $T_c$.

In addition to bulk JPWs, the study of surface JPWs propagating on planar dielectric–superconductor interfaces or in superconductor slabs has been carried out. Surface modes in planar geometry cannot be directly excited by light, although they could be probed with the attenuated-total-reflection method. On the other hand, small particles made of HTSCs present resonances in the optical spectrum, whose characteristic frequencies can be related to the Josephson frequency. They are used to probe the Josephson coupling strength by the sphere resonance method, which is commonly analyzed in the dipole approximation and in the framework of a simple Drude model.

The concept of spatial confinement in low-dimensional systems has led to a number of new ideas and achievements in several areas of nanophysics including semiconductors, photonics, and plasmonics. In particular, confined surface plasmons are widely exploited in the context of usual metallic nanoparticles. In this Letter, we generalize the concept to the field of high-$T_c$ cuprate superconductors. We consider a spherical particle made of a MLSC, which is modeled as a macroscopic medium with an anisotropic dielectric tensor. The quasi-static limit, we derive a general analytic formula which provides the eigenmodes corresponding to Josephson surface plasmons (JSPs), fully confined at the particle, in the whole frequency range and for any multipolar symmetry. They represent the generalization of the well-known surface plasmons of a metallic nanoparticle, but their spectrum is far more complex, due to the presence of longitudinal and transverse JPWs, quasi-2D plasmons, and mutual couplings among them. We then show that our results can be applied to the calculation of optical extinction and electron energy-loss spectra, the latter revealing the excitation of surface plasmons of any multipolar symmetry.

As a starting point, we discuss the dispersion relation of the bulk transverse magnetic (TM) electromagnetic modes of a cuprate superconductor. When the wavelength of radiation is much longer than the size of the unit cell, the dispersion of retarded JPWs coupled to quasi-2D plasmon modes can be cast in the form

$$\frac{k_x^2}{\varepsilon_c(\omega)} + \frac{k_z^2}{\varepsilon_{ab}(\omega)} = \frac{\omega^2}{c^2},$$

where the $c$-axis is directed as $\hat{z}$. In our model, the dielectric function $\varepsilon_{ab}(\omega) = \varepsilon_\infty (1 - \omega_p^2/\omega^2)$ is a Drude function which describes the in-plane response of the charge carri-
The lower Josephson frequency can be more than two orders of magnitude smaller than \( \omega_p \), resulting in very strong anisotropy effects, which are crucial for the electrodynamic response in the superconducting phase.

The dispersion resulting from Eq. (1) is represented in Fig. 1 for the case of a \( n = 2 \) MLSC, showing distinctive features over a large range of frequencies. On the scale of \( \omega_p \), the plasmon-polariton dispersion closely resembles that of quasi-2D acoustic plasmons in a layered electron gas with no interplane tunneling [4, 5]. At lower frequencies, the acoustic character of the quasi-2D plasmons is lost and a stop-band opens below the lower Josephson plasma frequency \( \omega_{J1} \). In MLSCs, moreover, a second stop-band is present in the dispersion at the intermediate range of frequencies between \( \omega_{J1} \) and \( \omega_{J2} \), due to the effect of the transverse-optical JPW. The stop-band can be revealed as a Reststrahl feature in the reflectance spectrum [9, 12]. Thus, the strong anisotropy combined with the presence of different Josephson plasma frequencies in MLSCs leads to highly structured optical response over several decades of frequencies.

The dispersion relation (1) naturally suggests to model high-\( T_c \) cuprate superconductors as anisotropic materials with the dielectric tensor [20]

\[
\varepsilon(\omega) = \text{diag} \left[ \varepsilon_{ab}(\omega), \varepsilon_{ab}(\omega), \varepsilon_c(\omega) \right].
\]

Transverse electric (TE) and TM electromagnetic modes play the role of ordinary and extraordinary waves, respectively. This macroscopic approach allows to calculate the spectrum of electromagnetic modes in confined geometry. Here we consider a spherical superconducting particle immersed in a material with dielectric constant \( \varepsilon_d \) and adopt the quasistatic approximation, which is valid when the particle size is smaller than the light wavelength [27]. We solve the anisotropic equation for the scalar potential \( \nabla \cdot \( \varepsilon \nabla \phi \) = 0 \) inside and outside the particle with the proper boundary conditions at the interface. This is done by means of a coordinate transformation to an ellipsoidal geometry, where the usual isotropic Laplace equation is recovered. Thus, we are led to the characteristic equation

\[
\varepsilon_c(\omega) \kappa \frac{d}{d\kappa} P_m^l(\kappa) + \varepsilon_d(\kappa + 1) P_m^l(\kappa) = 0,
\]

with \( \kappa = \left[ 1 - \varepsilon_c(\omega)/\varepsilon_{ab}(\omega) \right]^{-\frac{1}{2}} \) and \( P_m^l \) the associated Legendre polynomials.

Equation (4) is a central result of the present work. In particular, we stress that it holds for any uniaxial material: the specific properties of the solutions depend entirely on the behavior of the dielectric functions \( \varepsilon_{ab}(\omega) \) and \( \varepsilon_c(\omega) \). For the case of HTSCs, its solutions represent a spectrum of surface excitations that are localized close to the superconducting particle and which we call Josephson surface plasmons. Each surface mode is identified by the azimuthal quantum number \( l \) and the magnetic quantum number \( m \). Each \( m > 0 \) mode is degenerate with its
to a quasi-continuum of excitations \cite{28}. A large number of closely spaced JSPs gives rise to JPWs along the \( c \)-axis, as their charge oscillations are mostly related to the particle. Analogously, the two \( m = 0 \) modes at \( \omega/\omega_{11} = 0.93 \) and 9.84 can be considered as longitudinal JPWs confined along the \( c \)-axis, in agreement with the potential profile in Fig. 2(e). This is consistent with the fact that the \( m = 0 \) modes lie within the stop-bands below the Josephson frequencies \( \omega_{11} \) and \( \omega_{12} \).

For the generic modes with \( l > 1 \) this interpretation is not valid anymore, since \( \varepsilon_c(\omega) \) and \( \varepsilon_{ab}(\omega) \) are coupled together in the characteristic equation \( \varepsilon_{ab}(\omega) \) and \( \varepsilon_{ab}(\omega) \). The \( l > 1 \) plasmonic modes do not have a specific Josephson or quasi-2D character, but they present confinement both along the optical axis and perpendicular to it, as shown by the potential plots in Fig. 2(c–d). This can be interpreted as a phenomenon of mixing of JPWs and quasi-2D plasmons, which is a peculiar feature of the multipolar surface modes of confined layered superconductors.

Measuring JSPs requires considering an external probe located outside the superconducting particle. Some quantities commonly investigated are, for instance, the extinction cross section or the electron energy loss (EEL) spectrum. The calculation of these quantities depends on the multipolar polarizabilities \( \alpha_{lm}(\omega) \), which take into account the boundary conditions at the particle surface. Several results for spheres of isotropic media can be generalized to the anisotropic case, as long as the polarizabilities \( \alpha_{lm} \) are redefined with the new expression

\[
\alpha_{lm} = \frac{\varepsilon_c(\omega) \varepsilon_{ab}(\omega) P_l^m(\kappa) P_{l+1}^m(\kappa) R^3}{\varepsilon_{ab}(\omega) \frac{d}{d\kappa} P_{l+1}^m(\kappa) + \varepsilon_d(l+1) P_l^m(\kappa) R^3},
\]

where \( \kappa \) is defined as in Eq. \( 4 \) and \( R \) is the sphere radius.

For fixed \( m \) and increasing \( l \), the JSP modes tend to satisfy the relation

\[
\sqrt{\varepsilon_{ab}(\omega) \varepsilon_c(\omega)} = \varepsilon_d,
\]

which is the characteristic equation for the (quasistatic) surface plasmons over a planar superconductor–dielectric interface \cite{29}. Thus, the solutions of Eq. \( 5 \) represent the true watersheds between the different regions of the excitation spectrum; however, as a consequence of the large anisotropy of the system, they are very close to the Josephson plasma frequencies (the relative difference to \( \omega_{11,2} \) is of the order of \( \omega_{11,2}^2/(2\omega_d^2\varepsilon_{ab}^2) \approx 10^{-5} \)). In Fig. 2, in particular, they are virtually indistinguishable from \( \omega_{11} \) and \( \omega_{12} \).

Figure 2. (color online) Quasistatic Josephson surface plasmons in a spherical particle made of a \( n = 2 \) MLSC. (a) The modal frequencies, as a function of the azimuthal quantum number \( m \) and for three values of the magnetic quantum number \( m = 0, 1, 2 \). Notice that, for better visibility, the \( \omega \)-axis has been split in three different ranges which do not have the same scale. The frequencies \( \omega_{11}, \omega_{12}, \omega_p \), and \( \omega_f \) scale as in Fig. 1 in addition, \( \varepsilon_\infty = 10 \) and \( \varepsilon_d = 1 \). (b–e) Potential maps of the modes indicated by the dashed lines. The potential is shown on a cut along the \( xz \) plane including the center of the sphere, with the superconductor \( c \)-axis oriented as \( z \).

\(-m \) counterpart; however, at variance from the case of isotropic materials, modes with the same \( l \) and different \( m \) are nondegenerate.

The spectrum for the case of \( n = 2 \) MLSCs is shown in Fig. 2(a) as a function of \( l \), and it spans a large range of frequencies up to the in-plane plasma frequency \( \omega_p \). In particular, several \( m = 0 \) modes lie entirely inside the stop-bands, as their charge oscillations are mostly related to JPWs along the \( c \)-axis. In the remaining regions of the spectrum, a large number of closely spaced JSPs gives rise to a quasi-continuum of excitations \cite{28}.
Fig. 3. (color online) The frequencies of the two \( l = 1 \), \( m = 0 \) JSPs of the same particle as in Fig. 2 versus the transverse-optical Josephson frequency \( \omega_T \). The (red) dotted lines indicate the frequencies calculated with Eq. (8).

function centered on the zeros of Eq. (4), i.e. the frequencies of the JSPs. However, due to the presence of noncondensed carriers, a certain amount of dissipation is to be expected even in HTSCs and it is responsible for linewidth broadening of the peaks.

Once redefined the multipolar polarizabilities as in Eq. (6), the extinction cross section for an incident plane wave in the dipolar approximation is given by [21]

\[
C_{\text{ext}} = \frac{4\pi\omega}{c} \Im \left( \alpha_{10} \cos^2 \theta_E + \alpha_{11} \sin^2 \theta_E \right),
\]

where \( \theta_E \) is the polar angle of the polarization of the electric field with respect to the \( c \)-axis. For a field with \( \theta_E = 0 \), only the \( l = 1, m = 0 \) surface plasmons are excited. Indeed, scattering from small objects is dominated by the modes with dipolar symmetry, while higher-order modes will become manifest for larger particles.

These results are relevant for sphere-resonance experiments. In the usual interpretation, the absorption resonances are associated to the frequencies

\[
\omega_{\text{SR},\alpha} = \omega_{J\alpha} \sqrt{\frac{\varepsilon_{\infty}}{\varepsilon_{\infty} + 2\varepsilon_d}}, \quad \alpha = 1, 2.
\]

However, the frequencies of the dipolar \( l = 1, m = 0 \) JSPs obtained from Eq. (4) generally differ from the expressions (8) as an effect of the transverse-optical JPW. This can be seen in Fig. 3, where the JSP frequencies are plotted as a function of \( \omega_T \). In the limit \( \omega_T \to \omega_{J1} \) the lowest JSP mode tends to \( \omega_{\text{SR},1} \), while the difference is maximum for the upper JSP mode. In the limit \( \omega_T \to \omega_{J2} \), on the other hand, the situation is reversed. These results show that a careful analysis of sphere-resonance experiments should be undertaken in order that \( \omega_{J1}, \omega_{J2} \) and \( \omega_T \) are simultaneously determined.

At variance from extinction spectroscopy, EEL spectroscopy allows to excite the full spectrum of multipolar JSPs. The EEL probability function (in atomic units) for an electron with impact parameter \( b > R \) (moving outside the particle along the \( c \)-axis) includes all multipolar contributions, according to the expression [30]

\[
\frac{dP}{d\omega} = \frac{4}{\pi v^2 R^2} \sum_{l,m} M_{lm} \left( \frac{\omega R}{v} \right)^{2l} K_m \left( \frac{\omega b}{v} \right) \Im (\alpha_{lm}),
\]

where \( K_m(x) \) are the modified Bessel functions, \( v \) is the electron velocity, and \( M_{lm} = (2 - \delta_{m0})/(l+m)!/(l-m)! \). The EEL spectrum for a 300 eV electron is represented by the solid curve in Fig. 4 and compared with the extinction cross section, indicated by the dashed curve. The effect of higher-order JSPs is particularly evident in the region around \( \omega_T \) and \( \omega_{J2} \). The peak in proximity of \( \omega_{J2} \) is blue-shifted with respect to the extinction cross section, as a consequence of the increasing weight of the \( l > 1 \) higher-frequency modes (see Fig. 2); moreover, an additional peak arises just below \( \omega_T \), due to the excitation of the corresponding modes of nondipolar symmetry.

In conclusion, we have determined the frequencies of Josephson surface plasmons confined in small spherical particles made of HTSCs. The JSP spectrum contains a rich variety of dipolar and multipolar modes, which span a wide frequency range up to the in-plane plasma frequency. For the case of multilayered cuprates, spatial confinement leads to delicate coupling effects between acoustic and optical JPWs. Confined JSPs can be probed by optical absorption or by EEL spectroscopy, the latter being sensitive to modes of any symmetry. The present approach can be extended to superconducting particles of non-spherical shapes, in analogy with surface plasmons in ordinary metal nanoparticles, but with the additional effects of strong dielectric tensor anisotropy. The study
of Josephson plasmons via their size confinement is a promising new route to explore phenomena associated with interlayer tunneling in cuprates and, possibly, their relation to high-$T_c$ superconductivity.

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