Reflection EELS as a probe of the bulk plasmon continuum in layered materials

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A periodic arrangement of 2D conducting planes is known to host a (bulk) plasmon dispersion that interpolates between the typical, gapped behavior of 3D metals and a gapless, acoustic regime as a function of the out-of-plane wavevector. In a semi-infinite geometry, the configuration relevant to Reflection Electron Energy Loss Spectroscopy (R-EELS), such a system is known to host a surface plasmon that ceases to propagate below a cutoff wavevector. As the f-sum rule requires a finite response whether or not there exist sharp excitations, we demonstrate that what remains in the surface loss function — the material response probed by R-EELS — is the contribution from the (bulk) plasmon of the infinite system. In particular, we provide a one-to-one mapping between the plasmon continuum and the spectral weight in the surface loss function. In light of this result, we suggest that R-EELS be considered a long wavelength probe of the plasmon continuum in layered materials.

Introduction

In the mid 1970s, A. L. Fetter applied a hydrodynamic analysis to a periodic system of conducting planes, each hosting a 2D electron gas, and obtained a rather unique plasmon continuum. The plasmon — the long wavelength oscillation of the many-electron charge density — radically changes in character depending on whether adjacent conducting planes of the system are oscillating in-phase or out-of-phase. While the in-phase oscillation (see Fig. 1a) corresponds to a gapped (optical) mode reminiscent of plasmons in simple 3D metals, the out-of-phase oscillation (see Fig. 1b) corresponds to a gapless (acoustic) mode that disperses linearly with in-plane wavevector. Between the extremes of in-phase and out-of-phase oscillation lies an acoustic-to-optical plasmon continuum (see Fig. 2).

Due to the severe conduction anisotropy in the high-$T_C$ cuprate superconductors $\text{La}_2-x\text{Ce}_x\text{CuO}_4$ and $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ [6, 7], as well as the hole-doped compounds $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) and $\text{Bi}_2\text{Sr}_{1.6}\text{La}_{0.4}\text{CuO}_{6+\delta}$ (Bi-2201) [8, 9]. Shared among these RIXS studies is a peak in the scattered intensity whose (planar) wavevector dispersion changes appreciably as the out-of-plane wavevector is tuned toward an out-of-phase oscillation between adjacent copper oxide layers. By identifying this peak with the plasmon, RIXS data provides strong evidence of a nearly-acoustic mode in doped cuprates coincident with the out-of-plane, acoustic plasmon dispersion of the Fetter model. This analogy to the Fetter model can be completed by tracking the evolution toward the optical plasmon dispersion when the out-of-plane wavevector corresponds to in-phase oscillation; however, the scattering geometry and suppression of charge excitations at large out-of-plane wavevector in the RIXS cross section make this connection difficult to establish concretely [10].

Rather than following the (3D) wavevector dispersion
of charge density excitations, we instead consider an experimental probe that lacks translation symmetry along a chosen axis: Reflection Electron Energy Loss Spectroscopy (R-EELS). When the reflection surface is perpendicular to the out-of-plane (layering) direction, the explicitly broken translation symmetry renders R-EELS, in principle, susceptible to all out-of-plane wavevector contributions— a point previously made in [11,12]. Our main result is that R-EELS does, in fact, probe the bulk plasmon continuum of the Fetter model in the long wavelength limit. As a consequence, layered materials should generically be expected to host a broad surface response at long wavelengths that reflects their underlying bulk plasmon continuum.

While the doped cuprates provide motivation for this investigation, these compounds are notoriously complex. Because of its relative simplicity and close correspondence to the plasmon behavior seen in RIXS, we will instead focus on the Fetter model of layered conducting planes. Though the aforementioned studies [6–9] contain more involved computational analyses to fit their experimental curves, the qualitative plasmon behavior (or treatment of the Coulomb interaction) often mirrors the Fetter result [1]. One could argue that the Fetter model might represent a pathological limit since it neglects inter-plane conduction; however, the inclusion of inter-layer electron hopping results in a plasmon dispersion that continuously evolves from the Fetter result to a more general anisotropic dispersion [13]. This is to say that our results apply inasmuch as the Fetter model is capable of describing any particular (strongly) anisotropic system.

I. The semi-infinite Fetter model

In our implementation of the Fetter model, we consider a system of conducting planes separated by (insulating) dielectric layers — i.e., a semi-infinite, single-layer superlattice (see Fig. 3). The conducting planes are characterized by a long wavelength, 2D Drude conductivity \( \sigma \) at frequency \( \omega \) through the standard relation

\[
\sigma(\omega) = \frac{ine^2}{m\omega(1 + i/\omega\tau)},
\]

where \( n \) is the 2D (planar) electron density, \( e \) the electron charge, \( m \) the electron mass, and \( \tau \) corresponds to a relaxation time associated with electron scattering. The layer periodicity of the 2D conducting planes (or the inter-layer distance) is denoted by \( a \) and we model the insulating, dielectric regions through a dielectric constant \( \epsilon \). The cutoff wavevector \( q^* \),

\[
q^*a := \ln \left( \frac{e + 1}{\epsilon - 1} \right),
\]

marks the lower bound for surface plasmon propagation at a given \( \epsilon > 1 \) [13,14,15].

FIG. 3. The semi-infinite model of periodic conducting planes — the semi-infinite Fetter model — considered in the text. The thick black lines denote 2D conducting planes, the dielectric constant within the insulating layers is denoted by \( \epsilon \), and the layer periodicity is given by \( a \).

is given by [1,14,15]

\[
\omega_p(q, Q) = \Omega \sqrt{\frac{qa \sinh qa}{2 \cosh qa - 2 \cos Qa}},
\]

where \( \Omega \) is the optical plasma frequency defined as

\[
\Omega^2 := \frac{ne^2}{\epsilon_0 \epsilon ma}.
\]

The \( \omega_p \) dispersion [2] is plotted in Fig. 2.

In the semi-infinite geometry (i.e., as in Fig. 3), bulk plasmons are no longer self-sustained resonances of the system due to the lack of translation symmetry in the out-of-plane direction. Instead, surface plasmons — planar charge density oscillations that decay away from the vacuum-material interface — are the long-lived excitations of the superlattice. When corrections due to the finite relaxation time \( \tau \) are negligible, the surface modes follow the dispersion relation [15,16,18]

\[
\omega_{sp}(q) = \Omega \sqrt{\frac{qae [\epsilon \cosh qa - \sinh qa]}{(\epsilon^2 - 1) \sinh qa}},
\]

whose functional form depends on the dielectric mismatch at the vacuum-material interface. The \( \omega_{sp} \) dispersion [4] is plotted in Fig. 4. Curiously, the surface plasmon ceases to exist for sufficiently long wavelengths and, in fact, is never a resonance of the system in the absence of a dielectric background [19]. The cutoff wavevector \( q^* \),

\[
q^*a := \ln \left( \frac{e + 1}{\epsilon - 1} \right),
\]

marks the lower bound for surface plasmon propagation at a given \( \epsilon > 1 \) [13,14,15].
II. Calculating the surface loss function

The lack of long-lived charge density excitations in the $q < q^*$ regime of the semi-infinite Fetter model poses an interesting question for a Reflection Electron Energy Loss Spectroscopy (R-EELS) experiment: What does R-EELS measure at wavevectors below $q^*$? Independent of sharp excitations, the spectral weight in the (dissipative) density response for $q < q^*$ is nevertheless constrained by the f-sum rule in a reflection geometry [20–23]. Put simply, R-EELS has to measure something associated with charge density excitations at long wavelengths — and a fixed amount of it. It has previously been found that, in addition to the surface plasmon, there is a finite contribution from the bulk plasmon [21, 23]; however, the bulk correction typically constitutes a weak or visually-inconspicuous shoulder in the surface loss function above the (bulk) plasma frequency. In order to quantify the redistribution of spectral weight in the $q < q^*$ regime of the semi-infinite Fetter model, we study the surface loss function

$$g(q, \omega) := -\frac{e^2}{2\epsilon_0|q|} \int_0^\infty dzd\omega' \chi''(q, \omega; z, z') e^{-|q|z} e^{-|q|z'}$$

(6)

since this dimensionless quantity captures the material response contribution to the R-EELS cross section [22, 26, 28]. In (6), $\chi''$ denotes the imaginary part of the density response function $\chi$ which has been Fourier transformed along the planar and temporal directions; i.e., we assume planar translation invariance.

As our implementation of the semi-infinite Fetter model (see Fig. 3) is made up of alternating conductive and dielectric layers, we calculate the surface loss function in (6) through a dielectric analysis of the system. In the dielectric theory of R-EELS [27, 29, 31] (and the vacuum scattering contributions in [20, 32]), one characterizes the system through local dielectric response (in the out-of-plane, or layering, direction) and derives the long-ranged effect of the Coulomb interaction by enforcing electromagnetic boundary conditions across each interface. Superlattice boundary conditions are often written in the full electrodynamic formalism in terms of the electric and magnetic field vectors [14, 15, 33]. Instead, we take the non-retarded limit $q^2 \gg \omega^2/c^2$ from the outset and work in a theory of potentials $\phi, \phi_D$ for the electric field $E = -\nabla \phi$ and the electric displacement $D = -\epsilon_0 \nabla \phi_D$, which are themselves related in the dielectric regions through $D = \epsilon_0 \epsilon E$. At each interface, we have the standard boundary conditions that the planar components of $E$ are continuous and, across a conducting plane, that the discontinuity in the $D_z$ is given by the planar charge density. By use of the continuity equation and recognizing that the finite $(q, \omega)$ components of the planar charge density are entirely due to the induced response, the discontinuity in $D_z$ can be related to the planar components of $E$ and the conductivity $\sigma$. Lastly, the (semi-)infinite system of boundary conditions can be closed by utilizing a decaying ansatz for the $\phi, \phi_D$ in the material (e.g., see [15, 18]).

The dielectric analysis provides calculational utility since the surface loss function [6] is encoded in the vacuum solution for the electric potential when a driving field is applied [22, 30, 34, 35]. To be concrete, consider a semi-infinite system whose boundary is defined by a barrier to its charge density at $z = 0$, as in our Fetter model of Fig. 3 and is embedded within a dielectric medium characterized by $\epsilon$. If a perturbing potential $\phi_{\text{ext}}$ is sourced by a charge density localized entirely within the vacuum region (implementing the negligible material penetration assumption used to construct the R-EELS cross section [27, 29]), then the material responds to a field of the form

$$\phi_{\text{ext}}(q, \omega, z > 0) = C_{\text{ext}}(q, \omega) e^{-|q|z}$$

(7)

by inducing the charge density

$$\rho_{\text{ind}}(q, \omega, z) = e^2 C_{\text{ext}}(q, \omega) \int_{z > 0} dz' \chi(q, \omega; z, z') e^{-|q|z'}$$

(8)

via the definition of the density response function $\chi$. Outside of the material (in the vacuum region $z < 0$), the induced charge density [8] sources an induced potential

$$\phi_{\text{ind}}(q, \omega, z < 0) = \frac{e^2}{2\epsilon_0|q|} \left( \frac{2}{1 + \epsilon} \right) \times C_{\text{ext}}(q, \omega) e^{+|q|z} \int_0^\infty dz'dz'' \chi(q, \omega; z', z'') e^{-|q|z'} e^{-|q|z''}.$$ 

(9)

In (9), $\phi_{\text{ind}}$ depends on $\epsilon$ through the boundary conditions across $z = 0$. Already, the induced potential in

![Graph](https://example.com/graph.png)
In order to extract the surface loss function from the induced potential \( \phi_\text{ext} \), we require an explicit form of the external field coefficient \( C_\text{ext} \) in (7). A convenient external field — \( \phi_\text{ext} \) of (7) — to consider is the classical dielectric description of R-EELS [30, 36, 37] wherein an incoming electron is modeled by a trajectory that undergos elastic reflection off the material surface \( z = 0 \) at the time \( t = 0 \). The potential \( \phi_\text{ext} \) sourced by this classical electron trajectory is

\[
\phi_\text{ext}(q, \omega, z < 0) = \frac{A(q, \omega)}{1 + \epsilon} \left( \frac{\epsilon}{1 + \epsilon} \right) A(q, \omega) e^{+|q|z} \tag{10}
\]

in the vacuum region and

\[
\phi_\text{ext}(q, \omega, z > 0) = \left( \frac{1}{1 + \epsilon} \right) A(q, \omega) e^{-|q|z} \tag{11}
\]

in the material region. In (10) and (11), dependence on \( \epsilon \) is through the boundary conditions across \( z = 0 \), the coefficient \( A \) is defined by

\[
A(q, \omega) := -\frac{2ev_z}{\epsilon_0 [q^2 v_z^2 + (\omega - q \cdot v_y)^2]} \tag{12}
\]

and the reflection trajectory of the classical electron is parameterized by its velocity components \( v_y \) and \( v_z \), which are, respectively, parallel and perpendicular to the \( z = 0 \) surface. In light of (11), we can immediately write down the material response (in vacuum) to this particular \( \phi_\text{ext} \) using (9) through the replacement \( C_\text{ext} \rightarrow A/(1 + \epsilon) \) as

\[
\phi_\text{ind}(q, \omega, z < 0) = \frac{e^2}{2\epsilon_0 |q|} \left( \frac{2}{1 + \epsilon} \right) A(q, \omega) e^{+|q|z} \tag{13}
\]

\[
\times \int_0^\infty dq' d\omega' \chi(q, \omega; q', \omega') e^{-|q'|z} e^{-|q|z} \chi(q', \omega').
\]

The vacuum response of the material [13] provides a recipe to calculate the surface loss function within a dielectric model. Since \( A[12] \) is real (and, as a result, so is \( \phi_\text{ext} \) in vacuum), the only imaginary part of the total potential \( \phi_\text{tot} := \phi_\text{ind} + \phi_\text{ext} \) is through the material contribution in \( \phi_\text{ind} \). From the definition of the surface loss function \( \phi \), only the imaginary contribution of the material response is needed and we can extract this quantity from the total vacuum potential \( \phi \) as

\[
g(q, \omega) = -\left( \frac{1 + \epsilon^2}{2A(q, \omega)} \right) \text{Im} \left( \phi(q, \omega, z = 0) \right) \tag{14}
\]

\[
= -\left( \frac{1 + \epsilon^2}{2A(q, \omega)} \right) \text{Im} \phi_\text{ind}(q, \omega, z = 0) \tag{15}
\]

even when the \( \phi_\text{ind}, \phi_\text{ext} \) aren’t known separately. Naturally, the (total) potential \( \phi \) associated with the electric field can be obtained through a dielectric analysis of the semi-infinite Fetter model and the calculation of the surface loss function reduces to using (14) on the vacuum solution.

III. Results

Upon applying the aforementioned electromagnetic boundary conditions to our implementation of the semi-infinite Fetter model, we can extract the surface loss function \( g \) from the vacuum potential \( \phi \) through the relation given in (14). The surface loss function can be separated into two components

\[
g(q, \omega) = g_s(q, \omega) + g_b(q, \omega), \tag{16}
\]

where

\[
g_s(q, \omega) := \frac{(1 + \epsilon)^2}{4} \tag{17}
\]

\[
x \text{Im} \left( \frac{\phi \epsilon^2 q \sinh q - (2\omega^2 \epsilon^2 + q \epsilon) \sinh q}{\epsilon^2 q \sinh q - [(\epsilon^2 - 1) \omega^2 + q \epsilon] \sinh q} \right)
\]

and

\[
g_b(q, \omega) := \frac{(1 + \epsilon)^2}{4} \tag{18}
\]

\[
x \text{Im} \left( \frac{\epsilon^2 \sinh q}{{\epsilon^2}^2 q \sinh q - [(\epsilon^2 - 1) \omega^2 + q \epsilon] \sinh q} \right)
\]

are loosely associated with surface \( g_s \) and bulk \( g_b \) plasmon excitation; similar decompositions have been previously noted [16, 21, 25].

In the definition of \( g_s, g_b \), and \( q_0 \), the dimensionless parameters \( q := qa, \omega := \omega/\Omega, s := (1 + i/\omega\tau) \) have been introduced, \( \Omega \) is the optical plasma frequency (3), and the assumption \( \epsilon > 1 \) has been used. Additionally, the complex square root in the definition of \( g_b \) corresponds to the branch with positive imaginary part.

At sufficiently large \( q > q^* \), the essential behavior of the surface \( g_s \) and bulk \( g_b \) contributions to the surface loss function \( g \) can be gleaned from their identical pole structure. Notably, both \( g_s \) and \( g_b \) have the same denominator that, in the \( s \to 1 \) \( (\tau \to \infty) \) limit, can be written as

\[
\frac{1}{\epsilon^2 q \sinh q - [(\epsilon^2 - 1) \omega^2 + q \epsilon] \sinh q} = \frac{1}{[(\epsilon^2 - 1) \sinh q] \omega_p^2(q)/\Omega^2 - \omega^2, \tag{19}
\]

in terms of the surface plasma frequency \( \omega_p \), given by (4). So long as the relaxation time \( \tau \) is not short enough to radically alter the surface plasmon dispersion, \( \text{both} \) the surface \( g_s \) and bulk \( g_b \) terms peak at \( \omega = \omega_p \). When the surface plasmon is a sharp resonance of the semi-infinite Fetter model (i.e., for \( q \) sufficiently larger than \( q^* \)), both \( g_s \) and \( g_b \) appear similar in character and simply provide two contributions to the spectral weight at the surface plasma frequency \( \omega_p \). This behavior can be seen at \( qa = 4 \) in Fig. [5a] the \( g_s, g_b \) curves lie atop one another and the cutoff wavevector \( q^* a = \ln 3 \approx 1.1 \) (from [5] using \( \epsilon = 2 \) of Fig. [5]) is suitably smaller than \( qa = 4 \).

When \( q > q^* \) approaches \( q^* \), the distinction between \( g_s \) and \( g_b \) becomes apparent as the square root in the
The surface loss function $g = g_s + g_b$ of (16) at $\epsilon = 2$ and $\tau = 10/\Omega$ across several values of the in-plane wavevector $q$. At $\epsilon = 2$, the cutoff wavevector is given by $q_a = \ln 3 \approx 1.1$. At each wavevector the surface plasma frequency $\omega_{sp}$ and continuum of bulk plasma frequencies can be determined by (2) and (4), respectively. In (a), $q_a = 4$ ($q > q^*$), $\omega_{sp} \approx 1.63 \Omega$, and the plasmon continuum spans $1.39 \Omega < \omega < 1.44 \Omega$. In (b), $q_a = 1.5$ ($q > q^*$), $\omega_{sp} \approx 1.10 \Omega$, and the plasmon continuum spans $0.69 \Omega < \omega < 1.09 \Omega$. In (c), $q_a = 0.9$ ($q < q^*$), $\omega_{sp} \approx 1.04 \Omega$ is non-propagating, and the plasmon continuum spans $0.44 \Omega < \omega < 1.03 \Omega$. In (d), $q_a = 0.01$ ($q < q^*$) is the $q \rightarrow 0$ limit, $\omega_{sp} \approx 1.15 \Omega$ is non-propagating, and the plasmon continuum spans $0 < \omega < \Omega$.

The numerator of $g_b$ [18] begins to appreciably contribute. In the $s \rightarrow 1$ ($\tau \rightarrow \infty$) limit, the quartic polynomial in $\bar{\omega}$ within the square root of (18),

$$ (4\bar{\omega}^4 + \bar{q}^2 \sinh \bar{q} - 4\bar{\omega}^2 \cosh \bar{q}) , \quad (20) $$

has the zeroes

$$ \omega = \omega_p(q, Q = 0) \quad \text{and} \quad \omega = \omega_p(q, Q = \pi/a) . \quad (21) $$

In [21], $\omega_p$ is the bulk plasma frequency given by (2), and $Q$ labels the out-of-plane wavevector of the bulk plasmon in the infinite Fetter model. Even in the $s \rightarrow 1$ limit, the square root in the definition of $g_b$ [18] contributes a finite imaginary part when $\omega$ is within the bulk plasmon continuum, or $\omega_p(q, Q = \pi/a) < \omega < \omega_p(q, Q = 0)$. The bulk part ($g_b$) of the surface loss function $g$, then, is made up of two contributions: a peak at the surface plasma frequency $\omega = \omega_{sp}$ and a continuum of modes across the bulk plasmon dispersion $\omega_p(q, Q)$ [2] at fixed in-plane $q$. The contribution of both bulk and surface modes can be observed in Fig. 5(b) at $q_a = 1.5$ where the chosen value of $\epsilon = 2$ still maintains $qa > q^* a = \ln 3 \approx 1.1$ from [5].

For $q < q^*$, the surface plasmon is no longer a self-sustained resonance of the semi-infinite Fetter model; this is how $q^*$ in [5] is defined. The lack of surface plasmon propagation is a precise statement occurring at $q^*$ so long as finite-$\tau$ corrections to the surface plasmon dispersion are negligible [16, 18, 38]; however, the spectrum itself evolves continuously across $q = q^*$ [59]. Nevertheless, there is a sharp, qualitative change in the surface contribution $g_s$ across $q = q^*$. From the definition of $g_s$ [17] at finite $\tau$ (i.e., not in the $s \rightarrow 1$ limit), $g_s$ is
proportional to the frequency-independent factor

$$g_s(q, \omega) \propto [-2\epsilon \cosh qa + (1 + \epsilon^2) \sinh qa]$$,

(22)

which vanishes at the cutoff wavevector $q = q^*$ of [6]. For $q > q^*$, the multiplicative factor in (22) is positive whereas it is negative for $q < q^*$. Naturally, the total surface loss function $g = g_s + g_b$ cannot become negative as the density response function in (0) is related to a sum of transition amplitudes for charge density excitations. Instead, the sign change across $q = q^*$ represents a kind of inverse $\text{begrenzungs}$ effect [40]: the surface component $g_s$ suppresses spectral weight at the (non-propagating) surface plasma frequency arising from the pole in $g_b$ at $\omega = \omega_{sp}$ (the pole in (19)). The net effect of the suppression at $\omega = \omega_{sp}$ is to increase the relative contribution from the bulk plasmon continuum to the (total) surface loss function $g$. This behavior can be observed in Fig. 6c at $qa = 0.9 < q^*a = \ln 3 \approx 1.1$ for the chosen value $\epsilon = 2$.

In the $qa \to 0$ limit, the non-propagating surface plasmon peak at $\omega = \omega_{sp}$ becomes increasingly suppressed, leaving only a broad response across the bulk plasmon continuum $0 < \omega < \Omega$. This behavior can be observed in Fig. 5d for $qa = 0.01$ and $\epsilon = 2$; there is no appreciable feature in the surface loss function $g$ at the peak $\omega = \omega_{sp}$ visible for either $g_s$ or $g_b$. As the vertical scale due to $g_b$ and $g_s$ obscures $g$ in Fig. 5d, $g$ is plotted by itself in Fig. 6a. Curiously, the broad response across the bulk plasmon continuum is relatively insensitive to the relaxation time $\tau$ so long as the plasmon dispersion is not noticeably altered from its free electron value in (2). In Fig. 6b, the surface loss function $g$ is plotted for the same $qa \to 0$ limit ($qa = 0.01$) and dielectric constant $\epsilon = 2$, but at the long relaxation time $\tau = 1000/\Omega$; notwithstanding, the broad shape across $0 < \omega < \Omega$ remains and the only qualitative difference is the loss of curvature near $\omega = 0$, $\Omega$ when compared to $g$ at $\tau = 10/\Omega$ in Fig. 6a. The low energy, linear tail and broad peak near — but not at — the optical plasma frequency $\Omega$ appear as signatures of the underlying bulk plasmon continuum, rather than any explicit damping factor. While this statement is qualitatively general, it should be noted that the precise shape of the surface loss function in the $qa \to 0$ limit is sensitive to the dielectric constant $\epsilon$. In Fig. 7, the $qa \to 0$ surface loss function is shown at $\epsilon = 1.1, 2, 5, 10$ to demonstrate the influence of $\epsilon$ on the spectrum.

In addition to the surface loss function having support across the (bulk) plasmon continuum, the induced potential at these energies oscillates in step with the associated plasmon. Through our dielectric analysis, we can provide a precise, one-to-one correspondence between the plasmon at out-of-plane wavevector $Q - \omega_p(q, Q)$ of [2] — and the material response at fixed $q$ across the plasmon continuum. In the dielectric theory, this out-of-plane oscillation occurs in the decay factor $\exp[-\beta a]$, which relates the potential in adjacent dielectric layers: $\phi_n = \exp[-\beta a] \phi_{n+1}$. In order to maintain a bounded solution as $n \to \infty$, we require $\text{Re} \beta \geq 0 \iff |\exp[-\beta a]| = 1$; decaying modes (surface plasmons) correspond to $|\exp[-\beta a]| < 1$ whereas wave-like modes (bulk plasmons) correspond to $|\exp[-\beta a]| = 1$ [15, 16, 11]. In the $\tau \to \infty$ limit of negligible conductivity dissipation, the decay factor across the plasmon continuum is determined solely by the out-of-plane oscillation of the (bulk) plasmon. Specifically, rearranging the plasmon dispersion $\omega_p$ in (2) as

$$\cos qa = \frac{1}{2} \left[ 2 \cosh qa - \left( \frac{\Omega}{\omega} \right)^2 qa \sinh qa \right],$$

(23)

enables us to label the bulk plasmon at fixed $(q, \omega)$ through its out-of-plane wavevector $Q$. Using the pre-
scription in \[23\], we find that \(\exp[-\beta a] = \exp[-iQa]\) at energies satisfying the plasmon dispersion \(\omega_p(q, Q)\) of \[2\]. In Fig. 8, we demonstrate this behavior by plotting the decay factor \(\exp[-\beta a]\) across the bulk plasmon continuum at \(qa = 0.5\), \(\epsilon = 2\), and in the \(\tau \to \infty\) limit. For clarity, the equivalence \(\exp[-\beta a] = \exp[-iQa]\) is maintained at arbitrary in-plane wavevector \(qa\) and dielectric constant \(\epsilon > 1\) across the bulk plasmon continuum. The surface loss function, then, is a direct probe of the (bulk) plasmon across \(\omega_p(q, Q)\) at fixed in-plane \(q\).

![Graph](attachment:image.png)

FIG. 8. Plotted above are the (a) real and (b) imaginary parts of the decay factor \(\exp[-\beta a]\) to demonstrate the equality between the out-of-plane oscillation in the electric potential and the out-of-plane wavevector \(Q\) across the plasmon continuum. The parameters used are \(qa = 0.5\), \(\epsilon = 2\), and the \(\tau \to \infty\) limit. In both (a) and (b), the two curves lie atop one another.

The surface loss function \(g\) [18] demonstrates the transition from surface plasmon propagation at large in-plane wavevectors to a long wavelength regime dominated by the bulk plasmon continuum (see Fig. 5). Instead of a sharp peak at a single frequency, the \(qa \to 0\) surface loss function is characterized by a linear tail at low energies and a broad peak across the (bulk) plasmon continuum. This shape is only weakly influenced by the intrinsic damping parameter (see Fig. 6); however, the spectrum is sensitive to the dielectric constant \(\epsilon\) (see Fig. 7). Though the bulbous peak might initially suggest a damped, residual surface plasmon at long wavelengths, the oscillatory behavior of the electric potential across the plasmon continuum (e.g., the exact correspondence \(\exp[-\beta a] = \exp[-iQa]\) demonstrated in Fig. 8) suggests that the broad spectrum is intrinsic to the plasmon itself. From this perspective, we find that the long wavelength surface loss function is purely a bulk probe of the semi-infinite Fetter model.

**IV. Discussion**

Previous studies [16, 24, 60, 61, 62, 63, 64] have demonstrated, and emphasized, that bulk charge density excitations appear in the surface loss function (i.e., the R-EELS cross section); however, the contribution of bulk excitations typically presents as either a weak, possibly imperceptible, shoulder near sharp surface excitations or a broad background. We found similar behavior in the semi-infinite Fetter model for sufficiently large (in-plane) wavevectors. At \(qa = 1.5 > q^*\) in Fig. 5, a shoulder forms across the bulk plasmon continuum before the strong peak at the surface plasma frequency. Just below the cutoff wavevector \(q < q^*\) in Fig. 5, however, the peak at the (non-propagating) surface plasmon frequency has become suppressed and the surface loss function takes on a broad shape across the bulk plasmon continuum. In the long wavelength limit \(qa \to 0\), the peak at the (non-propagating) surface plasma frequency is visible in both \(g_s\) and \(g_b\) in Fig. 6a yet, no such feature appears in the surface loss function \(g = g_s + g_b\) of Fig. 6b. In essence, the long wavelength surface loss function of the semi-infinite Fetter model probes only the bulk plasmon continuum of the infinite system.

In the absence of a propagating surface plasmon, the transfer of spectral weight to the bulk plasmon continuum is required by the f-sum rule. While we have performed a dielectric analysis, the electromagnetic response encoded within the vacuum potential [14] genuinely behaves as the many-body response function defined in [6]. In particular, the f-sum rule [25],

\[
\int_0^\infty d\omega \omega g(q, \omega) = \frac{\pi e^2|q|}{2k_0m} \int_0^\infty dz n(z)e^{-2|q|z},
\]

relates the spectral weight within the surface loss function \(g\) [6] to the out-of-plane electron density \(n(z)\) of the system. Since the semi-infinite Fetter model is made up of regularly-spaced conducting planes with (2D) electron density \(n\), the out-of-plane density is simply \(n(z) = n \sum_j \delta(z - ja)\) and the right-hand side of the f-sum rule [24] can be evaluated as a geometric series to provide

\[
\int_0^\infty d\omega \omega g(q, \omega) = \left(\frac{\pi \Omega_0^2}{4}\right) \frac{2|q|a}{1 - e^{-2|q|a}},
\]

where

\[
\Omega_0^2 := \frac{ne^2}{\epsilon_0 ma} = \epsilon \Omega^2
\]

is the unscreened optical plasma frequency.

The crucial consequence of the f-sum rule [25] is that the \(qa \to 0\) limit of the surface loss function must contain the same spectral weight as an isotropic metal with (3D) electron density \((n/a)\) [24, 25]; i.e.,

\[
\lim_{qa \to 0} \int_0^\infty d\omega \omega g(q, \omega) = \left(\frac{\pi \Omega_0^2}{4}\right).
\]

The sharp \(q \to 0\) surface plasmon in an isotropic metal exhausts the spectral weight required by the f-sum rule [25]; however, the semi-infinite Fetter model lacks this mode in the long wavelength limit. Nevertheless, the f-sum rule is indifferent to whether or not sharp excitations are present: the contribution from the bulk plasmon continuum must compensate for the lost spectral weight at the surface plasmon peak. To demonstrate how this spectral weight is redistributed across the cutoff wavevector...
\( q^* \), we can scale the surface loss function according to the f-sum rule \(^{(25)}\) as

\[
f(q, \omega) := \left[ \frac{4}{\pi \Omega^2_0} \left( 1 - e^{-2|q|a} \right) \right] \omega \gamma(q, \omega) \tag{28}
\]

so that \( f \) has unit weight: \( \int d\omega f(q, \omega) = 1 \). One should note that the re-scaling in \((28)\) slightly distorts the spectrum of \( f \) (compared to the surface loss function \( g \)) due to the multiplicative factor of \( \omega \). In Fig. 9, the dispersion of \( f \) at \( \epsilon = 2 \) and \( \tau = 10/\Omega \) is shown across several values of \( qa \) to demonstrate the rearrangement of spectral weight as the surface plasmon peak is suppressed below \( q^*a = \ln 3 \approx 1.1 \) (using \((5)\) for \( \epsilon = 2 \)).

![Graph showing redistribution of spectral weight](image)

**FIG. 9.** Plotted above is the redistribution of spectral weight from the surface plasma frequency to the (bulk) plasmon continuum in the scaled surface loss function \( f \) \(^{(28)}\). The parameters used are \( \epsilon = 2 \), \( \tau = 10/\Omega \), and the cutoff wavevector (determined through \((5)\) by \( \epsilon = 2 \)) \( q^*a = \ln 3 \approx 1.1 \). By construction, all curves have the same (unit) integrated weight. The frequency axis has been scaled by \( \Omega_0 = \sqrt{\tau} \Omega \) to demonstrate the normalization; in these units, the plasmon continuum spans \( 0 < \omega < \Omega_0/\sqrt{\tau} \) in the long wavelength limit.

Before comparing our results with existing data, we should acknowledge that the model presented in section \(4\) — the semi-infinite Fetter model (see Fig. 3) — contains several simplifying approximations. The planar conductivity \( \sigma(\omega) \) \(^{(1)}\) is characterized by a constant relaxation time \( \tau \) up to — and, in the \( q > q^* \) regime, beyond — the plasma frequency; further, this conductivity is necessarily in the long wavelength (\( q \to 0 \)) limit as it lacks any dispersion. Since \( \sigma(\omega) \) is a property of the (identical) conducting planes within the system, the relevant scale setting the “long wavelength limit” is the planar Fermi wavevector \( k_F \sim O(1/l) \) where \( l \) is the planar lattice spacing. The long wavelength limit where dispersion in \( \sigma(\omega) \) can be reasonably neglected is then given by \( q \ll k_F \sim O(1/l) \). If the inter-layer distance \( a \) between conducting planes is sufficiently larger than the planar lattice spacing \( l \) (i.e., \( l \ll a \)), then the dispersion of the surface loss function \( g \) \(^{(16)}\) with \( qa \) can be tuned while simultaneously in the planar long wavelength limit of \( ql \ll 1 \). As for the insulating regions, we chose to model the dielectric layers by frequency-independent dielectric constants. Ideally, this only neglects phonon contributions at low energies so long as the band gap of the dielectric layers is sufficiently large compared to the bulk or surface plasmon energies under consideration.

With an eye toward the high-\( T_C \) cuprate superconductors, the approximations used in section \(4\) may not be as limiting as one might expect; e.g., in Bi-2212 (\( \approx 12.3 \) \( \AA \) and \( l \approx 3.2 \) \( \AA \) \(^{(49)}\)) was investigated within a dielectric model of conducting layers. In Bi-2212 \(^{(12, 47, 48)}\) and Bi-2201 \(^{(12)}\), R-EELS measurements at long wavelengths have observed a broad response — i.e., broader than the plasmon peak in Bi-2212 inferred via optical means \(^{(12, 49, 51)}\) — across energies below the (bulk) optical plasma frequency. Unfortunately, the low energies of incident electrons in these R-EELS experiments skew the spectrum at long wavelengths, rendering the precise surface loss function difficult to extract from an experimental intensity \(^{(12, 52)}\). Including the small, but finite \(^{(53)}\), out of plane conduction between copper oxide layers pushes the \( q \to 0 \) acoustic plasma frequency to a finite value \(^{(13)}\); indeed, this state of affairs appears to be supported by RIXS \(^{(6, 9)}\). Nonetheless, inasmuch as the cuprates behave as a strongly anisotropic, layered system, the qualitative features of our analysis — a broad surface loss function at long wavelengths — should apply. In particular, we expect the long wavelength surface loss function to reflect the underlying bulk plasmon continuum.

**Summary**

Reflection Electron Energy Loss Spectroscopy (R-EELS) is often understood as a surface probe whose characteristic long wavelength excitation is the surface plasmon. In the semi-infinite Fetter model (see Fig. 3), however, the extreme limit of conduction anisotropy restricts surface plasmon propagation to finite in-plane wavevectors \( q \) above a cutoff value \( q^* \). Below \( q^* \), the system lacks a surface plasmon and, therefore, has only the (bulk) plasmon as its collective, long wavelength excitation. As the plane of reflection explicitly breaks translation symmetry, the plasmon contributes a continuum of excitations due to the non-conserved out-of-plane wavevector. To understand how this continuum might be observed in R-EELS, we have employed a dielectric analysis to calculate the surface loss function — the material response probed by R-EELS — of the semi-infinite Fetter model.

Our results can largely be summarized by Fig. 3. At sufficiently large \( q > q^* \) in Figs. 5a and 5b, the surface loss function \( g \) \(^{(16)}\) is characterized by a sharp surface plasmon peak with only a weak (or, in Fig. 5a imperceptible) shoulder across the plasmon continuum. Just below \( q^* \) in Fig. 5c, the surface plasmon peak becomes actively suppressed and the plasmon continuum receives significant spectral weight. In Fig. 5d, the long
wavelength limit has successfully suppressed the non-propagating surface plasmon peak and all that remains in the surface loss function is a broad response across the plasmon continuum (see Fig. 6a or 7b). While the particular shape of the surface loss function requires a calculation, our results can be understood within the context of the sum rule [24] applied to the semi-infinite Fetter model [25]. Whether or not there exist sharp excitations in the long wavelength limit, the surface loss function must contain the same spectral weight as an otherwise ordinary metal [27]. Consequently, the long wavelength surface loss function becomes a probe of the bulk plasmon continuum in the semi-infinite Fetter model.

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