Far-field interaction of focused relativistic electron beams in electron energy loss spectroscopy of nanoscopic platelets

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A quantum mechanical scattering theory for relativistic, highly focused electron beams near nanoscopic platelets is presented, revealing a new excitation mechanism due to the electron wave scattering from the platelet edges. Radiative electromagnetic excitations within the light cone are shown to arise, allowed by the breakdown of momentum conservation along the beam axis in the inelastic scattering process. Calculated for metallic (silver and gold) and insulating (SiO\textsubscript{2} and MgO) nanoplatelets, new radiative features are revealed above the main surface plasmon-polariton peak, and dramatic enhancements in the electron energy loss probability at gaps of the 'classical' spectra, are found. The corresponding radiation should be detectable in the vacuum far-field zone, with e-beams exploited as sensitive ‘tip-detectors’ of electronically excited nanostructures.

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I. INTRODUCTION

A powerful technique for investigating electromagnetic (EM) field distribution around nanostructures is provided by very fast (relativistic) electron beams (e-beams), with typical lateral resolution on an atomic scale, available in scanning transmission electron microscopes (STEM). As discussed previously, when the e-beam is restricted to the vacuum near a selected nanoparticle, its EM interaction with surface plasmons or surface plasmon-polaritons (SPPs) is reminiscent of the near-field interaction of subwavelength optical probes. Several works have recently studied realizations of Cherenkov radiation excitation within various dielectric media by e-beams moving in near-field vacuum zones. In all the latter works the energy loss processes were described within a simple classical model in which the fast electron was assumed to move with a constant velocity along a straight line trajectory near a finite dielectric medium such that the energy loss intensity could be obtained from the force exerted on the electron due to its self-induced electric field through the nearby dielectric medium. The great simplification achieved by this approach amounts to reducing the full scattering problem at hand to a problem of finding the EM field induced by the e-beam in the vacuum around the dielectric medium. The resulting EM field could include Cherenkov-like radiative components around the e-beam which were restricted, however, to propagation within the interior of the dielectric medium.

In this paper we present a quantum mechanical theory for the inelastic scattering of a relativistically highly focused e-beam traveling near nanoparticles in a ‘non-touching’ aloof configuration. We show that the electron wave scattering by nanoparticle edges along the beam axis switches on Cherenkov-like radiation channels which extend into the vacuum away from the nanoparticle. The resulting far-field coupling between the electron and the nanoparticle is found to dramatically enhance various radiative channels in the loss spectrum.

To illustrate our main points we consider here a simple model (see Fig.1) where the e-beam is propagatated in the vacuum along a wide face of a rectangular nanoplatelet (oriented, e.g., in the \(x-y\) plane), and a surface or guided wave induced by the electron is propagated with a wave number \(k_x\) along the beam axis. The spatially sensitive nature of the corresponding electron energy loss process arises from the exponential dependence, \(e^{-2K^*b}\), of the EM interaction between the e-beam and the platelet on the impact parameter \(b\). The extinction coefficient, \(K^* = \sqrt{K^2 - (\omega/c)^2}\), with \(K^2 = k_x^2 + k_y^2\), determines the tail of the evanescent field in the vacuum for values
of $k$ outside the light-cone, i.e. for $K > \omega/c$. Inside the light-cone, i.e. for $K < \omega/c$, $K^*$ is purely imaginary and the corresponding interaction becomes spatially oscillating, allowing the electron to exchange photons with the particle far away into the vacuum. This striking mechanism has been overlooked in the recent literature of STEM-electron energy loss spectroscopy (EELS), since the excitation by an electron moving in the vacuum with a classical velocity $v$, has been restricted to a constant longitudinal wavenumber $k_x = \omega/v > \omega/c$, implying EM coupling to the nanoparticle which is restricted to the evanescent tail near the surface.

Our model calculation is applied to two types of nanoscopic platelets, conducting platelets made of silver or gold, and dielectric platelets made of insulators such as silica or magnesia. For both types of nanoparticles we find significant loss signals in the low energy range of the spectrum, where the electron-hole excitation probability is either zero (for the insulator) or very small (for the metals), exhibiting far-field (radiative) characteristics. In particular, specific SPP modes of the silver platelet, which penetrate into the light cone, can be excited by the external e-beam, leading to new features in the EEL spectrum which decay weakly with the plane-platelet distance.

II. MODEL AND FORMULATION

Following Ref. 4, the focused e-beam is described here as a one-dimensional wave, propagating along the $x$-axis, while in the transverse ($y-z$) directions it is described by a wave-packet localized within a smoothly converging cross section along the beam axis, whose shape is assumed to be squared for the sake of simplicity. The corresponding Green’s function for the noninteracting focused e-beam may be therefore written in the general form:

$$G_e^{(0)} (\vec{r}, \vec{r}'; t) = \frac{1}{2L} \sum_{p_x} \sum_{q_{tr}} e^{ip_x(x-x')} \sum_{q_{tr}} e^{i\varepsilon_{p_xq_{tr}}t} \left. \chi_{\vec{q}_{tr}} (y,z;x) \chi_{\vec{q}_{tr}}^* (y',z';x') \right| \frac{\epsilon_{p_xq_{tr}} - \epsilon_{p_xq_{tr}} - \epsilon_{\alpha_f} - \epsilon_{\alpha_f}}{\hbar^2}$$

where $p_x = hq_x$ is the longitudinal (along the beam axis) electron momentum, $\varepsilon_{p_xq_{tr}} = \sqrt{p_x^2c^2 + m^2c^4 + c^2p_{tr}^2}$, its total relativistic energy eigenvalue, with $m_0$ the electron rest mass, $p_{tr} = hq_{tr}$ (with $q_{tr} = (q_y, q_z)$) its transverse momentum, and $e^{iq_{tr}x} \chi_{\vec{q}_{tr}} (y,z;x)$ the corresponding e-beam eigenfunction (see Appendix A).

The electromagnetic (EM) interaction between the e-beam and the platelet may be described by the Hamiltonian:

$$\hat{H}_{EM} (\vec{r}, \{s\}) \approx -e\Phi (\vec{r}, \{s\}) - \frac{\hbar_x}{mc} A_x (\vec{r}, \{s\})$$

where $\Phi$ and $A_x$ are the scalar and $x$-component of the EM four-vector potential respectively, $\vec{r}$ is the electron position vector, $\{s\}$ is a collective symbol for the position vectors of the platelet charges, and $m = m_0/\sqrt{1 - (v/c)^2}$ the dynamic electron mass.

To first order of the perturbation theory with respect to the EM interaction Hamiltonian, $\hat{H}_{EM}$, the probability for the e-beam to go, during the time interval $\tau$, from initial to final eigen states when the platelet initial state is the ground state is:

$$\sum_{\alpha_f} \left| K^{(1)}_{e(i-f),\alpha_0 - \alpha_f} (\tau) \right|^2 = \left( \frac{1}{\hbar} \right)^2 \sum_{\alpha_f} \left| \Delta (\varepsilon_{\alpha_0}, \varepsilon_{\alpha_f} + \varepsilon_{f}, \varepsilon_{\alpha_f}, \varepsilon_{\alpha_f}; \tau) \right|^2$$

$$= \frac{1}{2L} \int_{-L}^L dx \int_{-L}^L dy \int_{-L}^L dz \left[ \chi_{\vec{q}_{tr}} (y,z;x) \chi_{\vec{q}_{tr}}^* (y',z';x') \right] \frac{e^{-i\varepsilon_{\alpha_f}x}}{\hbar^2}$$

where the the sum is over the platelet final states $\alpha_f$, and $\Delta (\varepsilon, \varepsilon'; \tau) \equiv \frac{\exp[i\varepsilon(\varepsilon' - \varepsilon)/\hbar]}{[\epsilon(\varepsilon' - \varepsilon)/\hbar]}$. In the limit $\tau \to \infty$, $|\Delta (\varepsilon, \varepsilon'; \tau)|^2 \to 2\pi \hbar \delta (\varepsilon - \varepsilon') = 2\tau \Re \int_0^\infty dt \exp[it(\varepsilon - \varepsilon)/\hbar]$, and so the rate of change of scattering probability of the e-beam $R_{e(i-f)} \equiv \frac{4\pi}{\hbar} \sum_{\alpha_f} \left| K^{(1)}_{e(i-f),\alpha_0 - \alpha_f} (\tau) \right|^2$, $\tau \to \infty$, is given by:

$$R_{e(i-f)} = \frac{4\pi}{\hbar} \sum_{\alpha_f} \left[ \int_0^\infty dt \exp[it(\varepsilon_{f}, \varepsilon_{\alpha_f} + \varepsilon_{\alpha_0} - \varepsilon_{f}, \varepsilon_{\alpha_f}, \varepsilon_{\alpha_f})/\hbar] \right]$$

$$\left\{ \frac{1}{2L} \int_{-L}^L dx \int_{-L}^L dy \int_{-L}^L dz \chi_{\vec{q}_{tr}} (y,z;x) \chi_{\vec{q}_{tr}}^* (y',z';x') \right\}$$

$$\left\{ \int dy' \int dz' \chi_{\vec{q}_{tr}} (y',z';x') \chi_{\vec{q}_{tr}}^* (y,z;x) \right\}$$

$$\left\{ \alpha_0 \frac{e^{-i\varepsilon_{\alpha_0}x} \hat{H}_{EM} (\vec{r}, \{s\}) e^{i\varepsilon_{\alpha_0}x} \alpha_f \right\}$$

$$\left\{ \alpha_f \frac{e^{-i\varepsilon_{\alpha_f}x} \hat{H}_{EM} (\vec{r}, \{s\}) e^{i\varepsilon_{\alpha_f}x} \alpha_0 \right\}$$

where the inclusion of all terms under the real part symbol is justified by the reality of the total expression written in the last five rows within the curly brackets.

Using the relations: $e^{-i\varepsilon_{\alpha_0}x} \hat{H}_{EM} (\vec{r}, \{s\}) e^{i\varepsilon_{\alpha_0}x} = e^{i(\varepsilon_{\alpha_0} - \varepsilon_{f})x} \hat{H}_{EM} (\vec{r}, \{s\})$, with:

$$\hat{H}_{EM} (\vec{r}) \equiv (-e) \hat{\Phi} (\vec{r}) - \frac{p_x}{mc} \hat{A}_x (\vec{r})$$

the rate of change of probability for the scattering of the e-beam can be rewritten in the form:
\[ R_{e(i,f)} = \sum_{\vec{q}_{tr}, \vec{q}_{fr}} e^{-\beta \frac{(\vec{q}_{tr} \cdot \vec{q}_{fr})^2}{2m_o}} \times \left\{ \frac{4\pi}{\hbar} \text{Re} \left\{ \frac{2i}{\hbar} \int d\vec{q}^* \int d\vec{q} e^{-\beta \frac{(\vec{q}_{tr} \cdot \vec{q}_{fr})^2}{2m_o}} \right\} \right\} \]

where

\[ \Delta q_x \equiv (q^f_x - q^i_x) \approx (\omega/v) + \hbar \left[ \left( q^f_{tr} \right)^2 - (q^i_{tr})^2 \right]/2mv \]

is the longitudinal momentum transfer of the e-beam (see Appendix A). \( \vec{q}_{tr} \) and \( \vec{q}_{fr} \) the e-beam asymptotic transverse momenta, initial and final respectively, and \( \hbar \omega \equiv \left( \beta \nu_{tr} - \beta \nu_{fr} \right) \) its energy loss. Note that the width \( \beta \)-of the Gaussian distribution function, is introduced in Eq. \[ \text{(5)} \] to account for the high transverse-energy cutoff caused to our beam by the objective aperture. It is related to the length \( L \) of the region around the beam focal plane used in our model as a normalization factor for the electron wave functions.

The interaction potential, \( H_{EM} (x, y, z) \), between the platelet and an external electron at \((x, y, z)\) is nearly independent of \( x \) for \( |x| \ll a^*, \) and decays to zero at least as quickly as \( 1/x^2 \) for \( |x| > a^* \) (see, e.g., Ref. \[ \text{[23]} \]). Under these circumstances the limits of the integrations over \( x \) and \( x' \) in the above expression may be set at \(-a^* \) and \( a^* \), rather than at \(-L \) and \( L \). The correlation function \( \left\langle \hat{H}_{EM}^{(i)} (x', y', z'; t) \hat{H}_{EM}^{(f)} (x, y, z; 0) \right\rangle \) can be expressed in terms of the relevant components of the 4-tensor photon Green’s function \( D_{\nu,\mu} (\vec{r}', \vec{r}; t), \nu, \mu = 0, 1, 2, 3 \) \(\leftrightarrow\) ct, \( x, y, z \), as:

\[ \left\langle \hat{H}_{EM}^{(i)} (x', y', z'; t) \hat{H}_{EM}^{(f)} (x, y, z; 0) \right\rangle = \left\{ D_{0,0} (\vec{r}', \vec{r}; t) + \frac{\nu_{tr}}{mc} D_{0,1} (\vec{r}', \vec{r}; t) \right\} \left[ \begin{array}{c} D_{1,0} (\vec{r}', \vec{r}; t) + \frac{\nu_{fr}}{mc} D_{1,1} (\vec{r}', \vec{r}; t) \\ \end{array} \right], \]

\( t > 0 \)

For the sake of simplicity we may assume translational invariance of the platelet dielectric properties in the \( x - y \) plane, that is: \( D_{\nu,\mu} (\vec{r}', \vec{r}; t) = D_{\nu,\mu} (x - x', y - y', z', z; t) \). For an impact parameter \( b \) smaller than the platelet sides along the \( x \) and \( y \) axes (i.e. \( b \ll 2a^*, 2b^* \) ) this assumption may be justified, though it is inconsistent with the breakdown of momentum conservation in the beam-platelet scattering event considered here (see a more detailed discussion below).

Substituting into the above expression for \( R_{e(i,f)} \) and rearranging the integrations we find that:

\[ (5) R_{e(i,f)} = -\frac{4\pi e^2}{\hbar} \sum_{\vec{q}_{tr}, \vec{q}_{fr}} e^{-\beta \frac{(\vec{q}_{tr} \cdot \vec{q}_{fr})^2}{2m_o}} \text{Im} \left\{ \int dk_x \int dk_y \right\} \]

where

\[ D_{f,i}^{(p)} (k_x, k_y; \nu', \mu'; z, z') = \int \frac{d^4 e^i}{2\pi} \text{Re} \left\{ \hat{H}_{EM}^{(i)} (x, y, z; t) \hat{H}_{EM}^{(f)} (x, y, z; 0) \right\} \]

\( \nu = 0, 1, 2, 3 \) \(\leftrightarrow\) ct, \( x, y, z \), \( \mu = 0, 1, 2, 3 \) \(\leftrightarrow\) ct, \( x, y, z \), \( f, i \) \(\leftrightarrow\) ct, \( x, y, z \), \( k_x, k_y \) \(\leftrightarrow\) ct, \( x, y, z \), \( \nu', \mu' \) \(\leftrightarrow\) ct, \( x, y, z \), \( z, z' \).

Now, the 4-tensor photon propagator in the vacuum (i.e. at \( z, z' \leq 0 \)) has the form:\[ \text{(7)} \]

\[ D_{\nu,\mu} (k_x, k_y; \nu', \mu'; z, z') = \frac{\delta_{\nu,\mu}}{2\pi K^*} \left[ e^{-K^* |z' - z|} - \frac{e^{-K^* |z' - z|}}{2\pi K^*} \right] \]

in which the relevant part is associated only with the second term within the square brackets (i.e. that associated with the image potential of the e-beam). Using this expression and recalling that \( 1/K^* \) is typically much larger than the beam transverse dimension, so that the extreme confinement of the e-beam wave functions \( \chi_{q_{tr},f} \) under the integrals over \( z \) and \( z' \) restrict their values to a narrow region near \( z' = z = -b \), we have:

\[ R_{e(i,f)} \approx \frac{4\pi e^2}{\hbar} \sum_{\vec{q}_{tr}, \vec{q}_{fr}} e^{-\beta \frac{(\vec{q}_{tr} \cdot \vec{q}_{fr})^2}{2m_o}} \right\} \times \text{Im} \left\{ \int dk_x \int dk_y \right\} \]

where:

\[ r_{f,i} (k_x, k_y, \omega) = r_{0,0} (k_x, k_y, \omega) + \frac{\hbar \nu}{mc} r_{1,0} (k_x, k_y, \omega) + \frac{\hbar^2 \nu}{mc^2} r_{1,1} (k_x, k_y, \omega) \]

\( \text{(8)} \)
and:

$$J\left(\frac{q}{q_{tr}}, q_{tr}; k_y, K^*; x\right) \equiv \int dz \int dy e^{-ik_0y} \chi_{q_{tr}}^{-1}(y, z; x) \chi_{q_{tr}}(y, z; x)$$  \hspace{1cm} (9)

Finally, denoting:

$$I\left(\frac{q}{q_{tr}}, q_{tr}; k_y, K^*; (\Delta q_x - k_x)\right) = \frac{1}{2l} \int_{-\pi a^*}^{\pi a^*} dx e^{i(q_{tr} - k_x)x} J\left(\frac{q}{q_{tr}}, q_{tr}; k_y, K^*; x\right)$$  \hspace{1cm} (10)

the scattering rate is rewritten as:

$$R_{e(i-f)} = \frac{2e^2}{\hbar} \int dk_x \int dk_y \text{Im} \left[ \frac{\rho_f(k_x, k_y, \omega)}{K^*} e^{-2K^*b} \right] \times \sum_{\frac{q}{q_{tr}}, \frac{q}{q_{tr}}} e^{-2q_{tr}^2\frac{a^*}{2m_e^*}} \left| I\left(\frac{q}{q_{tr}}, \frac{q}{q_{tr}}; k_y, K^*; (\Delta q_x - k_x)\right)\right|^2$$  \hspace{1cm} (11)

III. THE 'CLASSICAL' APPROXIMATION AND BEYOND

The theory developed in the previous section can be further simplified without losing its main physical content by employing several approximations. In the long wavelengths limit discussed in Ref.\textsuperscript{12} we find that (see Appendix B):

$$\text{Im} \left[ e^{-2K^*b} \left(\frac{k}{k}, \omega\right) / K^* \right] \approx \text{Im} \left\{ \left(\frac{K^*}{k}\right) f_e + \left(\frac{v}{c}\right)^2 - \left(\omega/ck\right)^2 \right\} f_o / K^* \right) e^{-2K^*b}$$

where

$$f_e = (\varepsilon K^* - Q)^2 / D_+ D_- \text{, } f_o = (K^* - Q)^2 / D_o D_o$$

$$D_+ = \varepsilon K^* + Q \tanh (Qc^*) \text{, } D_- = \varepsilon K^* + Q \coth (Qc^*) \text{, } D_o = K^* + Q \tanh (Qc^*) \text{, } D_o = K^* + Q \coth (Qc^*)$$

Q = $\sqrt{K^2 - (v/c)^2}\varepsilon(\omega)$, and $\varepsilon(\omega)$ is the local bulk dielectric function of the platelet. In the limit of a semi-infinite medium the resulting expression reduces (see Appendix B) to the surface dielectric response function obtained in Ref.\textsuperscript{12} by using Maxwell’s equations with macroscopic boundary conditions.

The standard classical approximation for the loss function\textsuperscript{12} is obtained from Eq.\textsuperscript{11} by making the following assumptions: (1) the e-beam transverse momentum distribution function $J\left(\frac{q}{q_{tr}}, \frac{q}{q_{tr}}; k_y, K^*; x\right)$ is a constant, that is equivalent to a $\delta$-function in the corresponding real-space transverse coordinates, (2) the contribution of the transverse energy to the longitudinal momentum transfer $\Delta q_x$ (see Eq.\textsuperscript{10}) can be neglected, and (3) the effective particle size, $a^*$, appearing as an integration limit along the beam axis, is infinite. Assumption (3), in conjunction with (1), yields the conservation of longitudinal momentum, i.e. $\Delta q_x - k_x = 0$, which together with assumption (2) imposes the fixed condition $k_x = (\omega/v)$.

It is interesting to note that usually assumption (2) is not strictly satisfied since the contribution of the transverse energy to $\Delta q_x$: $\hbar \left(\frac{q_{tr}^2}{2m_e} - \frac{q_{tr}^2}{2m_e}\right) / 2mv \approx q_{tr} \Delta q_{tr} / (mv/h) \sim \pm q_{tr}^2 / q_{tr}^2$, can be as large in magnitude as $(\omega/v)$. As an example, at $h\omega \sim 10$ eV, $(\omega/v) \sim 0.05$ nm$^{-1}$, whereas the transverse beam-wavenumber uncertainty, $|\Delta q_{tr}| \sim q_{tr} \sim 2\pi/l$ (with a typical value of $l \sim 0.6$ nm for the beam radius) is 10 nm$^{-1}$, so that for $\varepsilon = 100$ keV , where $q_{tr} \sim 1500$ nm$^{-1}$, $q_{tr}^2 / q_{tr}^2 \sim 0.07$ nm$^{-1}$.

In the present paper we focus on the most interesting violation of the ‘classical’ approximation outlined above, allowing $a^*$ to be a finite length, which reflects an effective range of the actual beam-particle interaction along the beam axis. Consequently the longitudinal momentum distribution around $\Delta q_x - k_x = 0$, defined by the integral in Eq.\textsuperscript{11}, is smeared and many wavenumbers $k_x$ inside the light-cone start contributing to the loss rate, Eq.\textsuperscript{11}.

The condition for the smearing to be significant is $\pi / a^* \gtrsim (\omega/v)$, so that typically for frequencies $\omega$ in the visible range, $a^*$ should be smaller than 200 nm. Nanoplatelets of such thicknesses should dramatically enhance radiative excitations by the e-beam, previously overlooked in the literature; see e.g. Ref.\textsuperscript{12} where it was argued that recoil effects in STEM should be negligible for valence electron excitations. Recoil is only a classical remnant of the present effect and of less general appearance. In particular, it vanishes for large media, such as the porous film investigated in Ref.\textsuperscript{2}, for which (if made sufficiently thin) the quantum mechanical momentum uncertainty along the e-beam axis remains significant.

It should be stressed that, for the sake of simplicity, the platelet dielectric response is calculated by assuming its wide faces to be infinite. A fully consistent treatment of the breakdown of translation invariance is expected, however, to further enhance all radiative channels. The calculation of the kinematical factor $I\left(\frac{q}{q_{tr}}, \frac{q}{q_{tr}}; k_y, K^*; (\Delta q_x - k_x)\right)$, responsible for the longitudinal momentum uncertainty in our model, from the integral in Eq.\textsuperscript{11}, could generate artificial oscillations by the sharp cutoff of the integral at $x = \pm a^*$. To avoid such oscillations we use an equivalent Gaussian distribution function in our actual calculations. The corresponding smooth cutoff is, in fact, more realistic than that appearing in Eq.\textsuperscript{11} since it arises from the attenuation of
the e-beam-platelet interaction at $|x|$ values larger than $a^*$. In any event, the exact form of the corresponding distribution function is of no great importance for the main purpose of our present paper.

IV. RESULTS AND DISCUSSIONS

A. Silver and Gold Nanoplatelets

As a first example we calculate the EEL function of a 100 nm long silver and gold platelets for an external 100 keV e-beam at various impact parameters (see Figs.2 and 3). To analyze the various SPP resonances one may consider the zeros of the denominator of the extraordi-

nary wave amplitude $f_e$ in Eq.(12) in the complex $K$-

plane. With the experimental optical dielectric function, 

$\varepsilon(\omega)$, for silver\textsuperscript{16} the resulting dispersion relation (inset, 

Fig.2) exhibits a rather flat branch of $\omega(\text{Re } K)$ inside 

the light-cone, which can be attributed to radiative SPP, 

seen as a mirror image of the usual non-radiative SP dis-

persion curve with respect to the light-line. The sector of 

$\omega(\text{Re } K)$ connecting the two branches across the light line 

has a vanishing negative slope, where $\text{Im } K(\omega) \propto \text{Im } \varepsilon(\omega)$ 

has a sharp peak. The sharp dip in the EEL spectrum 

just above the classical SP frequency (at 3.8 eV) reflects 

these closely related features.

At slightly higher frequencies the EEL signals exhibit 
a pronounced rise due to the enhanced SPP density of 

states associated with the flat radiative SPP branch. 

These peculiar features are missing in the loss spectra 

of the gold platelet, shown in Fig.3.

The EEL intensity in this spectral region exhibits atten-

tuation with increasing impact parameter significantly 

weaker than the corresponding attenuation of the main 

SP peak calculated in the classical limit. The radiative 

nature of the beam-particle coupling shown in Figs.(2,3) 

is even more pronounced in the low energy region below 

the main SP peak, where the classically calculated signal 
drops to very small values. Here our calculated EEL 

function exhibits a pronounced broad band with linearly 

increasing intensity for increasing frequency and almost 

no attenuation with increasing impact parameter. These 

features are due to the fact that the loss signal well bel-

low the main SP frequency is dominated by the contribu-

tion from the ordinary wave amplitude $f_o$, appearing 

in Eq.(12), which is singularly enhanced near the light line 

(where $K^* \to 0$ ), and thus reflecting the nearly pure 

(transverse) photonic nature of the excitations by 

the e-beam in this ‘classically forbidden’ region.

The results of our calculations may be compared to 
the experimental data reported in Ref.\textsuperscript{17} for silver and 

gold nano rods and ellipsoids. Fig.4 shows our calculated 
EEL spectra for three silver platelets with 

c$^* = 15$ nm and $a^* = 10, 15, 30$ nm at impact parameter $b = 10$ nm. 

The shown curves may be compared to the spectrum in 

Ref.\textsuperscript{17} obtained for a silver ellipsoid with a long half-axis

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{EEL spectra (solid lines) of a 100 keV e-beam propa-

gating parallel to the $x$-axis of a rectangular Ag platelet (with 

half sides: $a^* = 50$ nm along $x$, and $c^* = 10$ nm along $z$, see 

Fig.1) at impact parameters $b = 10, 20, 40$ nm above its wide 

$x-y$ face. The experimental optical dielectric function, $\varepsilon(\omega)$, 

for silver\textsuperscript{16} has been exploited. Dashed lines represent spec-

tra calculated by the classical theory. Inset: surface plasmon 

polariton (SPP) dispersion curves, $\omega(\text{Re } K)$, $\omega(\text{Im } K)$ in the 

complex $K$-plane for silver. The indicated values of $K$ and $\omega$ 

are normalized by $K_n = \omega_n/c$, and $\omega_n = 10$ eV, respectively.}
\end{figure}

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig3.png}
\caption{The same as Fig.2 for a platelet made of gold (Au). 

Note the absence of the sharp dips appearing just above the 

main plamon peaks in the corresponding Ag spectra (Fig.2).}
\end{figure}
FIG. 4: The same as Fig.2 for three Ag platelets with half sides along the beam axis $a^* = 10, 15, 30$ nm, and half width $c^* = 15$ nm, and with the e-beam at an impact parameter $b = 10$ nm. The dashed line represents the corresponding classical spectrum (i.e. for $a^* \to \infty$).

($\sim 30$ nm) and two short half-axes ($\sim 15$ nm) at impact parameter $\sim 10$ nm above the ellipsoid wide face.

The two lower curves, particularly those corresponding to $a^* = 10$ nm, exhibit good agreement with the relevant experimental data. Specifically, in addition to the very good agreement of the calculated main plasmon peak position ($\sim 3.45$ eV) with the experimental one, the intensities ratio ($\sim 2$) between the main plasmon peak and the high energy broad peak, and the extent and magnitude of the low energy tail shown in Fig.4, are seen to agree pretty well with the corresponding experimental results. In contrast a large intensities ratio ($\sim 8$) and a very small low energy tail characterize the classical curve shown in Fig.4, both indicating the importance of the quantum effects predicted by our theory. Note that an important feature of our calculated spectra, the large dip just above the main plasmon peak, which is missing in the experimental data, is shown to develop only at relatively large values of $a^*$ (i.e. for $a^* > 20$ nm).

B. Insulating Nanoplatelets

The situation in the forbidden energy gap region of semiconductors and insulators is in a sense an extreme case of the effect demonstrated in the low energy region of Fig.2: The EEL spectra shown in Fig.5 are calculated for an external 100 keV e-beam, propagating parallel to the $x - y$ face of a 100 nm long SiO$_2$ platelet with half thickness $c^* = 50$ nm, at different impact parameters $b$.

The spectra reveal a pronounced double-peak structure within the forbidden gap region, which does not decay with increasing $b$ values. Strictly speaking, this structure reduces to a single broad peak for platelets of widths $c^* \lesssim 10$ nm, reflecting a finite-size effect. Similarly to the situation with the silver and gold platelets well below the main SP peak, the strong radiative nature of this feature arises from the ordinary wave amplitude $f_o$, corresponding to the excitation of purely transverse EM waves, polarized within the $x - y$ plane, which totally dominates the loss signal in the forbidden gap region.

The spectra shown in Fig.5 may be compared to the results reported in Ref.18 for an electron moving parallel to a 90$^\circ$ SiO$_2$ wedge at a distance of 8.5 nm (see Fig.(1)). The pronounced radiative broad band within the gap region, obtained in our calculation, dramatically contrasts the vanishing loss signal shown there in Fig.(4) for an electron beam with the same velocity ($v = 0.54c$) and nearly the same impact parameter. The lack of far-field coupling in the latter theoretical approach restricted the fast external e-beam to excitation of EM waves confined within the dielectric medium, similar to ordinary waveguide modes which can develop within a thin SiO$_2$ slab in the forbidden gap region where $\text{Re} \varepsilon(\omega) \approx 2$, and $\text{Im} \varepsilon(\omega) \to 0$. For an ideal planar geometry (as assumed in our calculation of the dielectric response func-
tion \( r(\vec{K}, \omega) \), the corresponding waveguide modes appear as extremely narrow resonances which can not be excited by an e-beam with \( \Delta q \) values outside the light cone due to the vanishingly small dielectric damping, \( \text{Im} \varepsilon(\omega) \).

Such radiation excitations become possible for the non-planar geometries studied in Refs.\(^5\)\(^\perp\)\(^8\)\(^\perp\)\(^18\), even under the rigid e-beam trajectory approximation exploited there (but only above a threshold beam energy considerably higher than 100 keV) due to the translational symmetry-broken dielectric media considered in their calculations. Yet, the corresponding Cherenkov-like channels remain fundamentally different from the ones we propose: The opening of scattering channels with wave numbers inside the light cone allows coupling of the e-beam to the continuum of EM modes which are extended into the vacuum perpendicular to the platelet wide face. The relative strength of the present radiative mechanism may be further appreciated by noting the calculated spectra near a sharp SiO\(_2\) wedge in Ref.\(^18\), where in spite of the geometrical enhancement of near-field Cherenkov coupling, beam energies far above 100 keV were needed there for ‘switching on’ such channels.

Finally, it is instructive to compare our predicted loss spectrum of an external 100 keV e-beam propagating above a MgO platelet with half-sides \( a^* = c^* = 50 \text{ nm} \) at an impact parameter \( b = 2 \text{ nm} \) (see Fig.6) to the experimental data reported in Ref.\(^19\) for a MgO smoke cube of 100 nm size. The overall agreement is good, including the occurrence, in both the calculated spectrum and the experimental data, of a broad, nonvanishing signal within the forbidden gap region, which is missing in the classically calculated spectrum. In this gap region the calculated spectrum exhibits a smooth oscillatory structure associated with the multiple reflection of the generated radiation between the two parallel faces of the platelet perpendicular to the \( z \)-axis. This finite-size effect is peculiar to the far-field radiative modes found in the present paper for platelets confined in the direction along the e-beam axis, and is different from (though related to) the extremely sharp resonances associated with the waveguide modes developed in an ‘ideal’ (i.e. wide laterally) planar dielectric thin film. Thus, the classical approach applied to such an ‘ideal’ film yields usually (i.e. except for extremely rare coincidences of the loss energy with the resonant frequencies) null loss intensity, whereas in our quantum calculations the continuous window of wavenumbers inside the light-cone removes the stringent resonant conditions and allows the appearance of a significant loss intensity in the entire gap region. It is interesting to note that the average calculated signal inside the gap region increases smoothly with increasing frequency from zero up to nearly the interband threshold where its intensity relative to the loss main peaks (at ~ 14 and 20 eV) is about 1/6. This ratio is remarkably close to the corresponding relative intensity observed experimentally in Ref.\(^19\).

![FIG. 6: EEL spectra (solid lines) of a 100 keV e-beam propagating parallel to the \( x \)-axis of rectangular MgO platelets at a distance \( b = 2 \text{ nm} \) above their wide \((x - y)\) faces. The platelets half sides along the \( x \) and \( z \) axes are \( a^* = 50 \text{ nm} \), and \( c^* = 50, 100 \text{ nm} \), respectively. The corresponding classical \((a^* \rightarrow \infty)\) results (dashed lines) are also shown. Note the finite size oscillations of the calculated loss signal inside the forbidden energy gap with a period roughly proportional to \( 1/c^* \).](image)

### V. CONCLUSION

Applying a quantum-mechanical approach to the scattering problem of highly focused relativistic e-beams near nanoplatelets, we have shown that Cherenkov-like radiation of STEM e-beams, discussed recently in the literature\(^\perp\)\(^9\)\(^\perp\)\(^10\), has a much broader scope than originally presented. Dramatic enhancements of radiative channels arise from the breakdown of momentum conservation along the e-beam axis in the inelastic process due to scattering of the electron wave by the nanoparticle edges. Further enhancements, realized due to the extreme lateral confinement of the e-beam and its associated transverse momentum uncertainty, have not been considered in detail here. The radiation predicted to be emitted from both conducting and insulating nanoplatelets can be generated at impact parameters larger than the evanescent tail of the excited surface EM modes due to the oscillatory distance dependence of the electron-platelet interaction for momentum transfers within the light-cone. Consequently, this radiation should have a significant propagation component perpendicular to its main direction along the e-beam axis.

Large deviations from the classical EEL signal are found to persist also at small impact parameters, which can be readily tested experimentally. The results of our calculations for silver platelets seem to agree pretty well...
with the experimental data reported in Ref.\textsuperscript{[17]} for silver nano ellipsoids. Furthermore, experimental observation of loss signals within the forbidden energy gap of MgO cubes of 100 nm size by Aizpurua et al.\textsuperscript{[19]} seems as well to support our main prediction.

VI. APPENDIX A

In this appendix we specialize our general model of the focused e-beam to allow a more detailed discussion of some aspects of EELS experiments in STEM pertinent to the subject under study in this paper. We employ the relativistic Schrödinger’s wave equation:

\[
\left[ -\left( \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) - \frac{\partial^2}{\partial x^2} \right] \psi(x,y,z) = \varepsilon^2 \psi(x,y,z),
\]

subject to the boundary conditions:

\[
\psi(x,y,z) = 0, \text{ for } l(x) \geq y \geq 0 \quad (A1)
\]

and for \( l(x) - b \geq z - b \) with \( l(x) = l_0 + \alpha |x| \), \( \alpha \ll 1 \)

Due to the small converging angle \( \alpha \) one may invoke the Born-Oppenheimer approximation: \( \psi(x,y,z) = \varphi(x) \chi(y,z,x) \), in which the crossed derivatives \( \frac{\partial^2}{\partial y^2} \chi(y,z,x), \frac{\partial^2}{\partial z^2} \chi(y,z,x) \) are neglected, and the wave equation takes the approximate form:

\[
\frac{1}{\chi(y,z,x)} \left( \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) \chi(y,z,x) - \frac{1}{\varphi(x)} \frac{\partial^2}{\partial x^2} \varphi(x) = \varepsilon^2
\]

subject to the boundary conditions, Eq.(13). Solutions for the "slow" motion wave equation satisfying these boundary conditions are: \( \chi_{n_y,n_z}(y,z,x) = \frac{1}{l(x)} \sin[q_y(y)(y - l(x))] \sin[q_z(z)(z + b - l(x))] \)

The resulting equation for the "fast" motion is:

\[
\left[ \frac{d^2}{dx^2} + \nu^2_{m}(x) \right] \varphi(x) = \varepsilon^2 \varphi(x), \quad (A2)
\]

where:

\[ \nu^2_{m}(x) = q_y^2(x) + q_z^2(x) \]

Thus, to first order in perturbation theory with respect to \( \nu^2_{m}(x) \), the energy eigenvalues of an electron ‘trapped’ by the EM lenses inside the conic beam region are given by:

\[
\varepsilon_{p_x}^2 = \hbar^2 c^2 \varepsilon^2 + m_0^2 c^4
\]

where \( p_x = hq_x \) is the e-beam main (longitudinal) momentum, and \( p_{y,z} = \hbar \pi n_y,n_z/l \), with: \( n_y,n_z = 1,2,\ldots, l = \sqrt{l_0(l_0 + \alpha L)} \), its transverse momentum components in the free propagation zone outside the EM focusing domain.

This model of the e-beam is, of course, a drastic simplification of the actual focused beam in STEM. In particular the ideally reflecting boundary conditions, Eq.(13), can not be strictly realized under the smoothly varying field generated in space by the focusing EM lenses. The results of our analysis here are not expected to be very sensitive to the fine details of the momentum distribution of the beam. We may take advantage of that by eliminating the specific dependence of the transverse wave numbers on the average beam radius \( l \), and replace \( (\pi/l)(n_y,n_z) \) with the general symbol \( \bar{q}_{tr} \), such that the specialized set of eigenfunctions, \( \chi_{n_y,n_z}(y,z,x) \), may be replaced by a more general set \( \chi_{\bar{q}_{tr}}(y,z,x) \).

The relativistic asymptotic (initial and final) energies of an electron ‘trapped’ within the beam double-cone boundary are:

\[
\varepsilon^2_{f,i} = m_0^2 c^4 + \left( p_{y}^i f + \frac{\hbar^2 q_{z}^i f^2}{m_0^2 c^2} \right), \quad (A14)
\]

\[
\varepsilon^2_{f,i} = \left( \varepsilon^2_{i,f} - m_0^2 c^4 \right) / c^2 - \left( p_{y}^i f^2 + (p_{z}^i f)^2 \right), \quad (A15)
\]

The corresponding longitudinal momentum transfer is calculated from:

\[
\Delta q_x \approx (\omega/v) h \left( \varepsilon_{i,f}^2 - \varepsilon_{i,f}^2 \right) / 2mv
\]

VII. APPENDIX B

In this appendix, following the method developed in Ref.\textsuperscript{[12]}, we consider the dielectric loss function \( \Im \left[ \frac{e^{iK \cdot x} K_x}{K \cdot n} e^{-2K \cdot b} \right] \) appearing in Eq.\textsuperscript{[11]}, and show that it is proportional to \( \Re E_x (K; -b; \omega) \) - the electric field component along the e-beam axis at the beam position \( z = -b \). The latter is the key ingredient in the calculation of the power loss function in the classical limit. We shall also show in this appendix that in the long wavelengths limit discussed in Ref.\textsuperscript{[14]} the dielectric loss function reduces to the well known expression derived in Ref.\textsuperscript{[14]}.  

Our analysis starts from the expectation value of the four-vector potential \( A_{\nu} = (\varphi, -\vec{A}) \), given by:

\[
\varepsilon_{p_x}^2 = \hbar^2 c^2 \varepsilon^2 + m_0^2 c^4
\]
\[ A_{\nu} (\vec{r}, t) = \frac{1}{c} \sum_{\mu=0}^{3} \int_{0}^{\infty} dt' \int d^3r' D_{\nu,\mu} (\vec{r}, \vec{r}'; t - t') j^{ext,\mu} (\vec{r}', t') \]  

where \( j^{ext,\mu} (\vec{r}', t') \) is the external four-current density generated by the e-beam (with the components \( j^{ext,\nu} = (c \rho^{ext}, j^{ext}) \)), and \( D_{\nu,\mu} (\vec{r}, \vec{r}'; t - t') \) is the "dressed" retarded photon propagator in the Lorentz gauge, defined by the correlator:

\[ D_{\nu,\mu} (\vec{r}, \vec{r}'; t - t') = -i \left\{ \left[ \hat{A}_{\nu} (\vec{r}, t) , \hat{A}_{\mu} (\vec{r}', t') \right] \right\} \theta (t - t') \] (B2)

The "bare" four-vector potential is given by:

\[ A^{(0)} (\vec{r}, t) = \frac{1}{c} \int_{-\infty}^{\infty} dt' \int d^3r' D^{(0)}_{\nu} (\vec{r} - \vec{r}'; t - t') \times j^{ext,\nu} (\vec{r}', t') \]  

where \( D^{(0)}_{\nu,\mu} (\vec{r}, \vec{r}'; t - t') \equiv D^{(0)}_{\nu} (\vec{r} - \vec{r}'; t - t') \delta_{\nu,\mu} \), is the "bare" retarded photon propagator in the Lorentz gauge, which is given by:

\[ D^{(0)}_{\nu,\mu} (\vec{r} - \vec{r}', t - t') = \eta_{\nu} \theta (t - t') \delta (|\vec{r} - \vec{r}'| / c + t - t') / |\vec{r} - \vec{r}'|, \]

with:

\[ \eta_{\nu} = \left\{ \begin{array}{l} 1, \nu = 0 \\ -1, \nu = 1, 2, 3 \end{array} \right. \]  

The corresponding Fourier transforms with respect to the spatial coordinates parallel to the surface, with wave vector \( \vec{K} = (k_x, k_y) \) are:

\[ A_{\nu} (\vec{K}; z; \omega) = \left\{ \begin{array}{l} \frac{1}{c} \sum_{\mu=0}^{3} \int dz' D_{\nu,\mu} (\vec{K}; z, z'; \omega) \\ \times j^{ext,\mu} (\vec{K}; z'; \omega) \end{array} \right. \]  

and:

\[ D^{(0)} (\vec{K}, z, z'; \omega) = \eta_{\nu} \frac{e^{-\omega |z - z'|}}{2\pi k_e} \]

Our explicit expression for the external 4-current density associated with the e-beam is:

\[ j^{ext,\mu} (\vec{K}; z'; \omega) = \left\{ \begin{array}{c} -ce\delta (z + b) \delta (vk_z - \omega), \mu = 0 \\ -e \left( \frac{\omega}{ck_z} \right) \delta (z + b) \delta (vk_z - \omega), \mu = 1 \\ 0 \end{array} \right., \mu = 2, 3 \] (B3)

For \( z, z' \leq 0 \), i.e., both on the vacuum side of the dielectric slab, occupying the space: \( 2e^* > z > 0 \), the lateral Fourier transform of Eq. (B2) can be written in the form:

\[ D_{\nu,\mu} (\vec{K}, z, z'; \omega) = \frac{\eta_{\nu}}{2\pi k_e} \left[ \delta_{\nu,\mu} e^{-K^* |z - z'|} - \frac{r^{(odd,\nu)} (0)}{r^{(odd,\nu)} + r^{(even,\nu)} (0)} e^{K^* (z + z')} \right] \] (B4)

where the generalized reflection four-matrices for incident waves, which are either symmetric or antisymmetric with respect to the slab center, are given respectively by (see Ref. 12):
This expression should be compared to the dielectric response function:

\[
r^{f,i}(k_x, k_y, \omega) = r_{00} + \left( \frac{\hbar \Delta q_x / mc}{(v/c) K r_{01}} \right) r_{11} = \begin{cases} r_{00} + (v/c)^2 r_{11} & \text{if } 1 + TrU \leq \frac{\hbar}{v/c} \\ \infty & \text{if } 1 + TrU > \frac{\hbar}{v/c} \end{cases}
\]

which may be simplified (again due to the symmetry \(r_{10} = -r_{01}\)) and the inequality \(|\Delta q_x| \approx \omega/v \ll mc/h\) to:

\[
r^{f,i}(k_x, k_y, \omega) = r_{00} + (v/c)^2 r_{11} + (\hbar \Delta q_x / mc) [r_{01} - (v/c) r_{11}] \\
\approx r_{00} + (v/c)^2 r_{11} + (\hbar \Delta q_x / mc) [r_{01} - (v/c) r_{11}] \\
\approx r_{00} + (v/c)^2 r_{11}
\]

Consequently: \(r_{00} + (v/c)^2 r_{11} \rightarrow r_{00} + \left( \frac{\omega}{c k_x} \right)^2 r_{11} = \frac{U_{00} + (\omega/c k_x)^2 U_{11}}{2(1 + TrU)}\)

and in the long wavelength limit, where \(1 + TrU = \frac{\hbar}{v/c} + Q\) and \(\epsilon\) is the bulk optical (frequency dependent) dielectric function of the platelet, we find:

\[
U_{00} = \frac{(1 - \epsilon)}{2\epsilon} \left( 1 + \frac{(\omega/c)^2}{2K^2} \right)
\]

\[
U_{11} = \frac{(\epsilon - 1) (\omega/c)^2}{4K^2} \left[ 1 + (\epsilon - 1) \frac{K^2}{(Q + K^2)^2} \right]
\]

so that finally:

\[
\text{Re} \mathcal{E}_x \left( \frac{\omega}{c k_x} \right) = \frac{K^2(\epsilon - 1)}{K^2 + Q \left[ \left( \frac{K^2}{Q + K^2} \right)^2 + \left( \frac{1 - \epsilon}{\epsilon} K^2 \right)^2 \right]
\]

which is equivalent to surface dielectric loss function obtained in Ref. 44 by using Maxwell’s equations with macroscopic boundary conditions.

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Au

e-beam:
\( \nu/c = 0.54 \) (100keV)

\( b = 10 \text{nm} \)
Ag

e-beam:
\( v/c = 0.54(100\text{keV}) \)

EEL (arb. units)

\( \omega (\text{eV}) \)

classical

\( a^* = 30\text{nm} \)
\( a^* = 15\text{nm} \)
\( a^* = 10\text{nm} \)
SiO$_2$

e−beam:
v/c=0.54(100keV)

$\omega$ (eV)

EEL (arb. units)

b=2nm

b=8nm

$\varepsilon=100$
MgO

e−-beam:
v/c=0.54 (100 keV)

c*=50 nm

c*=100 nm