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Nonlocality induced Cherenkov threshold

Hao Hu,^{1,†} Xiao Lin,^{2,*†} Jingjing Zhang,³ Dongjue Liu,¹ Patrice Genevet,⁴ Baile Zhang,^{2,5,*}
and Yu Luo^{1,*}

¹*School of Electrical and Electronic Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798, Singapore*

²*Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore*

³*State Key Laboratory of Millimeter Waves, Southeast University, Nanjing 210096, China*

⁴*CNRS, CRHEA, Universite Cote d'Azur, rue Bernard Gregory, 06560, Sophia Antipolis, Valbonne, France*

⁵*Centre for Disruptive Photonic Technologies, Nanyang Technological University, Singapore 637371, Singapore*

*Corresponding author. E-mail: xiaolinbnwj@ntu.edu.sg; blzhang@ntu.edu.sg; luoyu@ntu.edu.sg

[†]These authors contributed equally to this work

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Abstract: Cherenkov radiation is generally believed to be threshold-free in hyperbolic metamaterials owing to the extremely large photonic density of states in classical local framework. While recent advances in nonlocal and quantum effects extend our understanding of light-matter interactions in metallic nanostructures, the influence of nonlocality on threshold-free Cherenkov radiation still remains unknown. This work theoretically demonstrates that nonlocality provides a new degree of freedom to flexibly engineer Cherenkov thresholds in metallodielectric layered structures. The nonlocality results in a nonzero Cherenkov threshold, whose value is dependent on the sizes of unit cells and constituent materials. Particularly, the lower-bound velocity cutoff is comparable to the electron Fermi velocity. Surprisingly, this lower-bound threshold can be significantly smaller than the classically predicted one if the metallic nanostructure has an elliptical isofrequency contour and works around the epsilon-near-zero frequency. Such a counterintuitive phenomenon relies on the excitation of longitudinal modes in the nonlocal framework, which, however, vanish in the local approximation.

1. Introduction

Cherenkov radiation was firstly discovered in experiments by P. A. Cherenkov in 1934^[1], under the supervision of S. I. Vavilov, and later theoretically interpreted by I. Frank and I. Tamm in 1937^[2]. In Frank and Tamm's theory, Cherenkov radiation arises when the particle velocity exceeds a finite velocity threshold known as Cherenkov threshold v_{th} . It is determined by the phase velocity of light in the surrounding medium, i.e., $v_{\text{th}} = c/n$, where c is the light speed in free space and n is the refractive index of background medium. It is generally believed in the early twentieth century that photon emission from a uniformly moving charge is prohibited. The discovery of Cherenkov radiation changed such a belief. Thus, understanding the threshold behaviors of Cherenkov radiation is of paramount importance to the development of particle physics^[3-10].

Cherenkov threshold is also an important parameter to many practical applications including elementary particle detectors, dosimetry, medical imaging and therapy, etc^[11-15]. As a typical example, the threshold determines the sensitivity of Cherenkov detectors for particle identifications over a wide momentum range. To be specific, a higher sensitivity of the detector requires a higher value of Cherenkov threshold (i.e., $v_{\text{th}} \rightarrow c$), and hence a smaller refractive index of the background material. Thus, the design of Cherenkov detectors generally demands materials with the refractive index close to unity (e.g., gases, aerogels) in order to identify charged particles in the multi-giga-electron-volt (GeV) range^[16-18].

The capability to flexibly engineer the Cherenkov threshold facilitates many applications mentioned above. For instance, decreasing the Cherenkov threshold enables the on-chip applications of low-energy free electrons in integrated light sources^[19-23]. However, owing to the limited value of the refractive index attainable with natural occurring materials (e.g., $n < 4$ for transparent dielectrics at the visible regime), the kinetic energy of swift electrons used in these light sources is generally above 1 mega-eV (MeV). To address this problem, artificial metallic nanostructures, including plasmonic waveguides and hyperbolic metamaterials, have been proposed^[24-30]. With a proper design, effective refractive indices of

eigenmodes in these structures go to infinity (i.e., $n \rightarrow \infty$) under the classical local approximation. Consequently, Cherenkov radiation can be made threshold-free under the local approximation, i.e. $v_{\text{th}} = c/n \rightarrow 0$. Following such theoretical proposals, a recent experiment reported the lowest Cherenkov threshold to date, i.e., $v_{\text{th}} = 0.03c$ ^[31], in hyperbolic metamaterials.

Further reducing Cherenkov threshold beyond the results in Ref. [31] has remained difficult, probably because of the elusive role of nonlocality which is generally ignored in previous theoretical modellings. Although nonlocality is known to modify dramatically the electromagnetic responses of metallic nanostructures ^[32-37], its influence on the threshold behaviors of Cherenkov radiation still remains unexplored.

Here we address this issue by systematically investigating the nonlocal effects on Cherenkov radiation in metallodielectric layered structures. These structures have been widely applied to construct hyperbolic metamaterials. We find that the interplay between the spatial dispersion from structural periodicity and nonlocal electron screening in metals can strongly modify the threshold behaviors of Cherenkov radiation. First, nonlocality always enables the Cherenkov threshold to be larger than the hydrodynamical velocity β of plasma pressure waves in metals, namely $v_{\text{th}} \geq \beta$, for metallic nanostructures including hyperbolic metamaterials. Such a finding is in sharp contrast to the previous prediction of the zero-value Cherenkov threshold in hyperbolic metamaterials under the local approximation. Second, around epsilon-near-zero frequencies, nonlocality may largely *decrease* the value of Cherenkov threshold. Such a counterintuitive finding originates from the appearance of longitudinal modes (which cannot be captured by the classical local description) inside metallodielectric layered structures. Moreover, our findings also indicate that nonlocality may provide a new degree of freedom in controlling electron-matter interactions, and it can be exploited for the flexible engineering of Cherenkov thresholds.

2. Theoretical description of Cherenkov radiation in metallodielectric layered structure

The basic structural setup is shown in **Figure 1a**. A swift electron moves with a velocity of $\bar{v} = \hat{z}v$ in free space, along a trajectory close to the top surface of the metallodielectric layered structure. The unit cell of the layered structure has a pitch of $P = d_1 + d_2$, where d_1 and d_2 are thicknesses of metal (e.g., silver) and dielectric (silicon nitride) slabs, respectively. Here we set $d_1/P = 0.4$ and $P \ll \lambda$, where $\lambda \in [0.1 \text{ } 10] \text{ } \mu\text{m}$ is the studied range of wavelength. Under the local approximation, the metallodielectric layered structure is treated as a homogeneous uniaxial material with a relative permittivity of $\bar{\bar{\epsilon}}_r = [\epsilon_{||}, \epsilon_{\perp}, \epsilon_{||}]$, where $\epsilon_{||}$ is the in-plane permittivity and $\epsilon_{\perp}$ is the out-of-plane permittivity. In particular, the structure in Figure 1a is a hyperbolic metamaterial with $\text{Re}(\epsilon_{||}) \cdot \text{Re}(\epsilon_{\perp}) < 0$ if $\lambda > 0.4 \text{ } \mu\text{m}$ [Figure S1].

When considering nonlocality from the structural periodicity, the realistic layered structure is used in the calculation by employing transfer matrix method ^[38]. When further considering nonlocality from metals, hydrodynamic model is adopted to describe the nonlocal electron screening in metals [Supplementary Section 5 and 6]. The nonlocal electron screening refers to spatial spreading of surface charge densities over boundaries of metals, originating from the quantum repulsion of electrons or the electron gas pressure ^[35, 39, 40]. The oscillation of free electron gas due to the pressure gives rise to a longitudinal response of metals. The corresponding longitudinal permittivity is wavevector-dependent, that is $\epsilon_L(\omega, k_L) = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega - \beta^2 k_L^2}$ ^[41], where ω_p is the plasma frequency, γ is the damping rate, and k_L is the wavevector of longitudinal electric fields. $\beta = \sqrt{3/5} v_F$ is the nonlocal parameter in Tomas-Fermi approximation ^[42], where v_F is the Fermi velocity. To consider the nonlocal response of metals, an additional boundary condition, i.e., the conservation of the normal component of the displacement field, is applied to solve Maxwell's equations; see details in Supplementary Section 6.

3. Influence of nonlocal response from structural periodicity on the Cherenkov threshold

We begin with the analysis of influence of nonlocal response from the structural periodicity on the Cherenkov threshold in Figure 1. That is, we consider the case that the nonlocal response from metals is negligible, by setting the thickness of metal d_l much larger than the Tomas-Fermi screening length λ_{TF} . The value of $\lambda_{\text{TF}} \approx \frac{\beta}{\omega_p}$ is generally a few angstrom, which describes the spatial extent of surface charge densities for metals ^[13]. Here we set $d_1 = 10 \text{ nm} \gg \lambda_{\text{TF}}$ and $P = 25 \text{ nm}$ in Figure 1.

Figures 1b-e show the field distributions of free electron radiation in time domain. Under the classical local approximation, the structure in Figure 1a has $\min(v_{\text{th}}(\lambda))_{\text{local}} = 0$ in the studied range of wavelength, so-called threshold-free Cherenkov radiation in hyperbolic metamaterials; see radiation fields at two arbitrarily selected particle velocities in Figures 1b and 1d. In contrast, when under the nonlocal description, Cherenkov radiation inside the layered structure appears if $v = 0.2c$ in Figure 1c, but it disappears if $v = 0.05c$ in Figure 1e. This provides a clear evidence that $\min(v_{\text{th}}(\lambda))_{\text{nonlocal}} > 0.05c$ in the studied range of wavelength [Figure S4a]. Such a nonzero Cherenkov threshold originates from the fact that the nonlocal response from structural periodicity would prohibit the excitation of bulk Bloch modes with extremely large wavevectors. In addition, Cherenkov radiation of surface plasmons at the interface also shows up in Figures 1c and 1e, when considering the realistic structure. From Figures 1b to 1e, it is reasonable to argue that the local approximation fails to describe threshold behaviors, particularly when the free electron moves at a low speed.

4. Influence of nonlocal response from metals on the Cherenkov threshold

Next, we proceed to analyze influence of nonlocal response from metals on the Cherenkov threshold in **Figure 2**. That is, we consider the case that the nonlocal response from metals is non-negligible, by setting $d_1 \gtrsim \lambda_{\text{TF}}$. Figure 2a shows the Cherenkov threshold $v_{\text{th}}(\lambda_0)$ as a function of the pitch P of unit cell. Here we choose the wavelength $\lambda_0 = 1 \mu\text{m}$, at which the metallodielectric layered structure in Figure 1a under the local approximation is a hyperbolic metamaterial. While the nonlocal parameter β is generally around $c/300$ for

realistic metals (the value could be even one order of magnitude smaller for semiconductors), we investigate three different values of β here, i.e., 0, $c/300$ and $c/30$, to highlight the important role of nonlocal electron screening in metals on the Cherenkov threshold. If P decreases, $v_{\text{th}}(\lambda_0)$ decreases in Figure 2a. Moreover, $\lim_{P \rightarrow 0} v_{\text{th}}(\lambda_0) = \beta$. In other words, the nonlocal parameter β determines the ultimate low-bound for Cherenkov threshold in the layered structure, or we have $v_{\text{th}}(\lambda_0) \geq \beta$ for the arbitrary value of P . As such, Cherenkov threshold in hyperbolic metamaterials is always nonzero since $\beta \neq 0$ for realistic metals. The underlying mechanism is that the nonlocal response from metals regularizes the broadband singularity of photonic density of states in hyperbolic metamaterials, since the nonlocal metal is equivalent to a composite material, comprising a finite thin dielectric layer on top of a local metal ^[43]. Note that if we let $\beta = 0$, the nonlocal response of metals would be artificially neglected, and Cherenkov threshold reduces to zero if $P \rightarrow 0$ in Figure 2a.

Figure 2a also indicates that the nonlocality in artificial nanostructures may be exploited to engineer Cherenkov thresholds in a flexible way. If P is sufficiently small (e.g., $P < 1$ nm), the nonlocal response of hyperbolic metamaterials mainly arises from nonlocal metals, so that $v_{\text{th}}(\lambda_0) \sim \beta$. If P is sufficiently large ($P > 10$ nm), the nonlocal response of hyperbolic metamaterials is dictated by structural periodicity. As a result, a convergent tendency of $v_{\text{th}}(\lambda_0)$ emerges for different values of β , and it is possible to have $v_{\text{th}}(\lambda_0) \gg \beta$. If $1 \text{ nm} < P < 10 \text{ nm}$, $v_{\text{th}}(\lambda_0)$ is determined both by nonlocal metals and structural periodicity.

To highlight the influence of nonlocal metals on Cherenkov radiation inside hyperbolic metamaterials, Figures 2b-e show field radiations in time domain, by setting $d_1 = 0.8$ nm and $P = 2$ nm. In Figures 2b-e, $v = 0.015c$ is chosen; three different values of β are used, which are 0, $c/300$ and $c/30$, respectively, in Figures 2c-e; Figure 2b is calculated under the local approximation. Cherenkov radiation inside hyperbolic metamaterials emerges in Figures 2c and 2d since $v > \beta$, but it disappears in Figure 2e since $v < \beta$. These results numerically verify the nonzero Cherenkov threshold for realistic hyperbolic metamaterials. **Moreover, Figure 2e**

shows the absence of Cherenkov radiation of surface plasmons at the interface when $v < \beta$.

Such a result is distinct from Figure 1e. In addition, the emitted fields of Cherenkov radiation in Figures 2c and 2d are similar with that in Figure 2b, except for fields inside the triangular region highlighted in Figures 2b-2d. The difference comes from that the radiation spectrum of Cherenkov radiation is sensitive to β , since $v_{\text{th}}(\lambda)$ is dependent on both β and λ .

5. Dissipated power of a swift electron above metallodielectric layered structure

To facilitate experimental observation of the revealed nonlocal Cherenkov threshold in hyperbolic metamaterials, **Figure 3** shows the comparison of dissipated powers P_0 of the swift electron in Figure 1a under local and nonlocal descriptions. The dissipated power is equal to the integration of energy loss spectrum G of over the studied range of wavelength [Supplementary Section 7]. From Figure 3, nonlocality induced from the structural periodicity has a strong influence on the dissipated power. In contrast, the nonlocal electron screening in metals would affect the dissipated power only when the pitch of unit cell is small (e.g., $P \leq 2$ nm in Figure 3). As a clear signature for nonlocal Cherenkov threshold, if v decreases to a sufficiently small value (e.g., $v < 0.1c$ for $P = 10$ nm in Figure 3), the dissipated power dramatically decreases. The dissipated power decreases faster when the period is sufficiently large, but decreases relatively slower when the period is sufficiently small, indicating that the structural periodicity enables control of the Cherenkov threshold in the nonlocal framework. Particularly, the consideration of nonlocal response from metals enlarges the decreasing slope when $P \leq 2$. This is because the induced surface charge has a finite damping rate from the metallodielectric interface. Such a mechanism only becomes significant to reduce the photonic density of states when $P \leq 2$, thereby increasing the Cherenkov threshold. In addition, there is a large discrepancy between dissipated powers from local and nonlocal calculations, indicating the local approximation is not accurate for calculating dissipated power.

6. Influence of longitudinal modes on the Cherenkov threshold around epsilon-near-zero frequencies

Now recall the in-plane permittivity $\varepsilon_{||}$ of the effective homogeneous uniaxial material under the local approximation. Note that around frequencies of $\varepsilon_{||} \rightarrow 0$, longitudinal modes will show up with the consideration of nonlocality, in addition to transverse modes ^[37]. We then proceed to discuss the influence of longitudinal modes on the Cherenkov threshold in **Figure 4**. In Figure 4a, we let $\lambda_0 = 0.395 \mu\text{m}$, at which $\text{Re}(\varepsilon_{||}) \rightarrow 0^+$ and $\text{Re}(\varepsilon_{\perp}) > 0$ [Figure S1]. We reveal in Figure 4a that the longitudinal mode has a much lower Cherenkov threshold than the transverse mode. Such an emerging phenomenon can be understood as follows.

On the one hand, for transverse modes, their isofrequency contour at λ_0 is elliptical [Figure S1], since $\text{Re}(\varepsilon_{\perp}) \cdot \text{Re}(\varepsilon_{||}) > 0$. The layered structure in Figure 1a at λ_0 is then denoted as an elliptical metamaterial below. The elliptical isofrequency contour, whether calculated under local or nonlocal descriptions [Figure S3], would generally lead to a large value of Cherenkov threshold for transverse modes; for example, we always have $v_{\text{th}}^T(\lambda_0) > 0.2c$ in Figure 4a. On the other hand, for longitudinal modes, the corresponding Cherenkov threshold $v_{\text{th}}^L(\lambda_0)$ is sensitive to nonlocality induced both by the nonlocal metals and structural periodicity in Figure 4a. In short, the features of Cherenkov threshold for longitudinal modes inside elliptical metamaterials in Figure 4a are similar with that for transverse modes inside hyperbolic metamaterials in Figure 2a.

Since $v_{\text{th}}^L(\lambda_0) < v_{\text{th}}^T(\lambda_0)$ for the arbitrary value of P in Figure 4a, it is then reasonable to argue that nonlocality would largely *decrease* the Cherenkov threshold for elliptical metamaterials at epsilon-near-zero frequencies. Such a feature is distinct from hyperbolic metamaterials, in which nonlocality would instead *increase* the Cherenkov threshold.

Figures 4b and 4c shows the field distributions of free electron radiation at λ_0 . The longitudinal mode does not show up in the local approximation [Figure 4b], while it is excited by free electrons under the nonlocal description [Figure 4c]. These results numerically verify the invalidity of local approximation in describing Cherenkov radiation inside elliptical metamaterials at epsilon-near-zero frequencies. Figures 4d and 4e show the radiation spectrum

of excited longitudinal modes around λ_0 . Figure 4d shows that the excitation of longitudinal modes is sensitive to the particle velocity v and the wavelength λ . To be specific, if v is fixed, the longitudinal mode can be efficiently excited only within a narrow range of wavelength, although they exist in a relative wide range of wavelength [Figure 4d]. Meanwhile, the longitudinal mode at a certain wavelength can be efficiently excited only for a certain range of v [Figure 4d]. Figure 4e indicates that the efficient excitation of longitudinal and transverse modes at the same wavelength requires different values of v . For example, at λ_0 , the peak for the efficient excitation of longitudinal modes appears at $v = 0.2c$, while the peak for transverse modes is at $v = 0.6c$. With a proper design, the values of these two peaks are in the same order of magnitude. These features might facilitate potential observations of Cherenkov radiation of longitudinal modes at epsilon-near-zero frequencies.

7. Conclusion

In summary, we for the first time theoretically prove the *non-existence* of threshold-free Cherenkov radiation in hyperbolic metamaterials. Instead, realistic nonlocality would always enable an ultimate lower-bound velocity cutoff (whose value is comparable to the electron Fermi velocity) inside metallodielectric layered structures, including hyperbolic and elliptical metamaterials. Moreover, we reveal that while nonlocality would increase the value of Cherenkov threshold in hyperbolic metamaterials, it would, in turn, decrease the value of Cherenkov threshold in elliptical metamaterials. In addition, although the hydrodynamic model is a very approximate approach in treating the nonlocal and quantum effects in metallic nanostructures, we emphasize that our theoretical predictions of nonlocal Cherenkov threshold is universal regardless of which quantum models used. This is because the observed threshold behaviors of Cherenkov radiation in our configuration only rely on the reduced photonic density of states and the appearance of longitudinal modes. Previous experimental results have verified that the presented approaches in this work (i.e., the hydrodynamic model and the realistic periodic structure) can precisely anticipate these two phenomena induced by nonlocality [42, 44]. In short, nonlocality could significantly re-shape Cherenkov radiation in complex media,

including the Cherenkov threshold, and it provides a valuable way to flexibly engineer the Cherenkov threshold. Our work thus provides an important theoretical guidance for many practical applications of Cherenkov radiation, such as the design of novel integrated free-electron radiation sources on chip.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

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Figure 1. Influence of nonlocal response from structural periodicity on Cherenkov threshold.

a) Structural schematic. A swift free electron has a velocity of $\bar{v} = \hat{z}v$. b-e) Field distributions of free electron radiation in time domain. The realistic layered structure is considered in (c) and (e), while it is treated as a homogeneous uniaxial material in (b) and (d) according to the classical local description. Cherenkov threshold in (a) is wavelength-dependent as $v_{th}(\lambda)$. Cherenkov radiation appears if $v = v_1 > \min(v_{th}(\lambda))$ in (c) but disappears if $v = v_2 < \min(v_{th}(\lambda))$ in (e). Other parameters are: the pitch of unit cell $P = d_1 + d_2 = 25$ nm; for all studied cases here and below, $d_1/P = 0.4$, and the studied range of wavelength in free space is $\lambda \in [0.1 \text{ } 10] \text{ } \mu\text{m}$.

Figure 2. Influence of nonlocal response from the nonlocal electron screening in metals on the Cherenkov threshold. a) Cherenkov threshold $v_{th}(\lambda_0)$ as a function of the pitch P of unit cell, where $\lambda_0 = 1 \mu\text{m}$. β is the nonlocal parameter for metals. For realistic metals, $\beta \neq 0$. The metallodielectric layered structure in Figure 1a under the local approximation is the hyperbolic metamaterial at λ_0 . Due to nonlocal metals, we always have $v_{th}(\lambda_0) \geq \beta$. b-e) Field distributions of free electron radiation in time domain. The realistic layered structure in Figure 1a is adopted for (c) to (e), while it is replaced by an effective homogeneous uniaxial material for (b) according to the local approximation. Cherenkov radiation appears if $v_3 > \beta$ in (c) and (d) but disappears if $v_3 < \beta$ in (e). Other parameters are $P = 2 \text{ nm}$, and $v = v_3 = 0.015c$ in (b) to (e).

Figure 3. Influence of nonlocality on dissipated powers of a swift electron moving above metallodielectric layered structures. For comparison, metallodielectric layered structure in Figure 1a is treated by three different ways. For the red dashed line, the layered structure is treated as an effective homogeneous uniaxial medium, according to the classical local description. For the solid lines, the nonlocal response from the structural periodicity is considered by setting the nonlocal parameter $\beta = 0$. For the data indicated by hollow symbols, the nonlocal responses both from the structural periodicity and from the nonlocal metal (e.g., $\beta/c = 1/300$) are taken into consideration. When d_1/P is fixed, the dissipated power calculated from the classical local description is independent of P . The used collision frequency here is $\gamma = 0.032$ eV for the metal.

Figure 4. Influence of longitudinal modes on the Cherenkov threshold around epsilon-near-zero frequencies. Around wavelength $\lambda_0 = 0.395 \mu\text{m}$, the metallodielectric layered structure in Figure 1a under the local approximation has an elliptical isofrequency contour and the in-plane permittivity close to zero. The realistic layered structure in Figure 1a supports both transverse and longitudinal modes around λ_0 . a) Cherenkov thresholds of longitudinal and transverse modes vs the pitch P of unit cell at λ_0 , with the consideration of nonlocality. The results calculated in the local approximation is the same as the red dashed line. b, c) Field distributions of free electron radiation at λ_0 . Longitudinal modes appear in (c) if nonlocality is considered, but they vanish in b) under the local description. d) Electron energy loss spectrum G as a function of wavelength λ and particle velocity v . (e) Electron energy loss spectrum G vs v at λ_0 . For (b) to (e), $P = 25$ nm.

ToC text: Cherenkov radiation is generally thought to be threshold-free in hyperbolic metamaterials. However, nonlocality in the metallodielectric layered structure results in a nontrivial Cherenkov threshold, whose value is associated with the sizes of unit cells and the constituent materials. The nonlocality thus provides a new degree of freedom to control Cherenkov thresholds and contributes to a variety of applications.

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Hao Hu, Xiao Lin, Jingjing Zhang, Dongjue Liu, Patrice Genevet, Baile Zhang*, and Yu Luo**

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