Extreme ultraviolet plasmonics and Cherenkov radiation in silicon

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Silicon is widely used as the material of choice for semiconductor and insulator applications in nanoelectronics, micro-electro-mechanical systems, solar cells, and on-chip photonics. In stark contrast, in this paper, we explore silicon’s metallic properties and show that it can support propagating surface plasmons, collective charge oscillations, in the extreme ultraviolet (EUV) energy regime not possible with other plasmonic materials such as aluminum, silver, or gold. This is fundamentally different from conventional approaches, where doping semiconductors is considered necessary to observe plasmonic behavior. We experimentally map the photonic band structure of EUV surface and bulk plasmons in silicon using momentum-resolved electron energy loss spectroscopy. Our experimental observations are validated by macroscopic electrodynamic electron energy loss theory simulations as well as quantum density functional theory calculations. As an example of exploiting these EUV plasmons for applications, we propose a tunable and broadband thresholdless Cherenkov radiation source in the EUV using silicon plasmonic metamaterials. Our work can pave the way for the field of EUV plasmonics.

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1. INTRODUCTION

Silicon is the most widely used material for applications in nanoelectronics [1], photovoltaics [2], micro-electro-mechanical systems [3], and on-chip photonics [4–6]. Its dominance in industry stems from multiple factors, including the possibility of controlling its crystallinity, tailoring of its conducting properties via doping, cost-effectiveness, and availability, as well as its high purity. Although universally known for its insulating and semiconducting properties, the goal of this paper is to explore and exploit silicon’s metallic and plasmonic properties, which have remained largely ignored.

The plasmonic properties of a variety of different materials have been explored across the electromagnetic (EM) spectrum [7] [Fig. 1(a)]. This includes plasmons on graphene in the terahertz regime [8], highly doped III-V semiconductors that support plasmons in the infrared [9,10], and the universally used plasmonic materials Ag and Au in the visible [11]. Aluminum has been the most widely explored plasmonic material at ultraviolet (UV) frequencies for applications such as tunable, integratable surface plasmon (SP) sources [12,13], medical assays and biotechnology applications with fluorophores [14], as well as lensing for imaging applications and optical lithography [15]. While aluminum has shown some promise in the ≈5 eV (248 nm) regime, achieving plasmonic effects at higher energies in the deep ultraviolet (DUV) and extreme ultraviolet (EUV) is an open problem.

In this paper, we show that EUV plasmons supported by silicon can pave the way for EUV waveguides, metamaterials, and devices not possible with conventional plasmonic materials. We study the evolution of the plasmonic behavior in silicon thin films down to 60 nm and probe the photonic band structure of silicon in the EUV up to 5 times past the light line. This is made possible by probing silicon with relativistic electrons using momentum-resolved electron energy loss spectroscopy (k-EELS). Unlike the more traditional spatially resolved electron energy loss techniques [16], in our work, not only the energy but also the momentum dispersion of the EUV plasmonic excitations are mapped. We also show excellent agreement of our experimental results with first principles quantum density functional theory (DFT) calculations as well as macroscopic electrodynamic electron energy loss theory. The silicon surface plasmon polariton (SPP) is shown to have a resonance condition at approximately 11.5 eV (107 nm), more than twice as high in energy as what has been measured with aluminum for applications in the UV. Finally, we propose an EUV radiation source by exploiting the
EUV plasmonic properties of undoped silicon. Our proposed EUV source is tunable and broadband, and uses thresholdless Cherenkov radiation (TCR) in silicon plasmonic hyperbolic metamaterials (HMMs). Our work paves the way for the field of EUV plasmonic devices with silicon.

The energy scales of the SPP for silicon is between $4 \text{ eV} < E < 11.5 \text{ eV}$ ($310 \text{ nm} < \lambda < 107 \text{ nm}$) while the bulk plasmon (BP) exists at $E = 16 \text{ eV}$ ($\lambda = 77 \text{ nm}$). Even though previous work has shed light on the existence of such metallic behavior in bulk silicon [17], it is an open question whether plasmonic behavior would persist for nanoscale structures. In this work, we specifically focus on the thickness evolution of plasmonic behavior in silicon thin films that is in agreement with Drude metallic behavior. This validates that deep subwavelength excitations in the EUV regime are indeed possible for paving the way to EUV plasmonics.

Experimental measurement of the silicon permittivity at high energies [18] agrees strongly with our DFT calculations under the GW approximation [19] [Fig. 1(b)] (see Supplement 1 for details). Silicon’s metallic character in the EUV is a result of the unbound nature of its valence electrons. This arises from the weak interband transitions strengths between the valence and conduction band [20, 17, 21]. In fact, this leads to a nearly freely moving sea of electrons in the valence band that can support SP excitations from the free-charge carrier oscillations. This is in contrast to the visible region of the spectrum, where prominent interband transitions lead to strongly bound electron-hole pairs between the valence and conduction band, which eliminates its metallic character [20, 22].

2. EUV PLASMONS IN SILICON MEASURED WITH k-EELS

We measure the EUV plasmonic properties of silicon with relativistic electrons and k-EELS in a transmission electron microscope (TEM). Unlike traditional electron energy loss spectroscopy techniques, where only the amount of energy loss is measured, k-EELS probes both the energy and momentum transfer of the electron. The information on momentum loss is obtained by measuring the scattering angle ($\theta$) of the electron after passing through the sample [Fig. 2(a) and Supplement 1]. Note the energy and momentum lost by the incident electron corresponds directly to the energy and momentum carried away by the excitations within the sample. Thus, the major advantage of k-EELS is the ability to map the photonic/polaritonic band structure and clearly identify photonic excitations such as Cherenkov radiation (CR), waveguide modes, and surface/BPs.

Figures 2(b)–2(d) show the measured photonic band structure as a function of thickness (200, 100, and 60 nm) for free-standing silicon films. The samples are prepared via focused ion beam milling (FIB) and mounted to a TEM grid to create free-standing structures [inset Fig. 2(f) and 2(g)]. The band structure for all three films is measured using k-EELS up to an electron scattering angle of $\theta = 30 \mu\text{rad}$ ($k_x \approx 0.1 \text{ nm}^{-1}$) at 300 keV incident energy and probed the deep near field up to 5 times past the light line.

We now explain the physical origin of the three branches seen in the band structure data in Figs. 2(b)–2(d). The dispersionless flatband at $16 \text{ eV}$ ($77 \text{ nm}$) in all three films is the BP excitation of silicon ($\omega_p^b$). The BP is a longitudinal resonance that is difficult to probe optically and occurs at the point $\epsilon_{\text{Si}} \rightarrow 0$ [Fig. 1(b)] well into the EUV. We emphasize that bulk longitudinal plasma oscillations, even for aluminum, occur in this high-energy regime. However, for waveguiding and nanoantenna applications, SPPs are necessary and do not exist in the EUV regime in the widely used plasmonic metals.

The highly dispersive band between $\approx 4–9 \text{ eV}$ for the 200 nm film and $\approx 4–11.5 \text{ eV}$ ($107–310 \text{ nm}$) in the 100 and 60 nm film is the measured SPP excitation of silicon in the EUV. Interestingly, surface excitations are stronger as compared to bulk excitations for thinner films in all electron energy loss spectroscopy measurements due to the Berggrenzungs effect [23,24]. As a result, the SP scattering intensity is large enough in the thinner 100 and 60 nm film to be probed into EUV energies. Interestingly, we note that $\omega_p^b = \omega_{\text{Si}}^b / \sqrt{2} \approx 11.5 \text{ eV}$, which is indicative that silicon is a Drude-like metal in the EUV in agreement with DFT calculations. This is in fundamental contrast to the DC semiconducting properties or transparent insulator-like optical properties of silicon at the telecommunication wavelengths.

We immediately note that the measured EUV SPP resonance energy of silicon ($\approx 11.5 \text{ eV}$) is more than double of what has been observed with aluminum, the traditional material for high-energy plasmonic applications. Furthermore, Figs. 2(e)–2(g) highlight the highly dispersive nature of the SPP (blueshifting of the peak with increasing scattering angle) for the three silicon films across the untapped $5–11.5 \text{ eV}$ range. Note that the
The dispersive properties of the SPP would be hidden in traditional electron energy loss spectroscopy techniques but is captured here by $k$-EELS. The $k$-EELS measurements prove that the SPP of silicon can be probed to an entirely new region of the spectrum as compared to other plasmonic materials opening the door for a wide range of plasmonic applications in the EUV.

To prove conclusively that we are observing bulk and SPs in the measured data, we conduct simulations of the macroscopic electrodynamic electron energy loss function [25] in silicon for electrons normally incident to the sample. The measured data show a strong match with the theoretical calculations [red line in Figs. 2(b)–2(d)]. The energy loss function has been shown to be analogous to the photonic density of states [26,27] and is thus an excellent quantifier for probing photonic excitations. Slight deviations at small scattering angles of the experimental SP peak from the predicted theoretical energy loss function in the 60 nm film is likely due to Ga+ implantation ($\approx 1–2$ nm) during the FIB sample fabrication process and surface oxidation. Thinner samples, which are more sensitive to surface energy loss excitations, are more affected by such impurities along the sample surface. Recorded uncertainties (error bars) in the measured EUV plasmonic resonances increase at large scattering angles due to the decrease in probability of scattering (see Supplement 1 for details).

Figure 3 shows the ratio of the bulk and surface scattering probability scaling with momentum for silicon. Theoretical predictions reveal a $k^{-3}$ dependence for BPs and $k^{-2}$ scaling for SPs [24,26] (Fig. 3). The excellent agreement between theory and experiment (ratio $\approx k^{-1}$) allows us to unambiguously separate the contributions of bulk and SPs in silicon. We emphasize that this

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**Fig. 2.** EUV plasmons and CR in silicon measured with $k$-EELS. (a) Schematic showing the key components of the $k$-EELS technique for measuring the momentum-resolved photonic band structure of silicon. The $k$-EELS experiment was performed with a Hitachi HF-3300 TEM with a GIF Tridiem in $k$-EELS mode at 300 keV incident energy with parallel illumination resulting in a quantitative energy-momentum dispersion map of the excitations in the sample (details in Supplement 1). The photonic band structure of (b) 200 nm; (c) 100 nm; and (d) 60 nm thick silicon films measured with $k$-EELS (error bars show 95% confidence interval). All three films show evidence of the BP at ($\approx 16$ eV) and the SP at ($\approx 4–11.5$ eV) in the EUV as well as CR in the visible in the ($\approx 2–4$ eV) region mapped to large scattering angles (large momentum with $k > 5 \times k_0$). A good agreement to the macroscopic electrodynamic energy loss function (red line) is seen for all three thicknesses. (e), (f), and (g) show the electron scattering probability for the three excitations as measured by $k$-EELS integrated over the indicated scattering angles for the 200, 100, and 60 nm silicon films, respectively. Insets in (f) and (g) show scanning electron microscope images of the free-standing silicon films prepared via FIB milling and mounted to the TEM grid.
high-energy (EUV) HMMs. Specifically, the multitude of applications possible with HMMs can now be expanded into the EUV, specifically the generation of a tunable, broadband, and TCR light source via electron excitation.

Interestingly, it has recently been shown that HMMs, a uniaxial metamaterial with a metallic response along one direction and a dielectric response along the orthogonal direction, can be used to eliminate the need for large velocity electrons for generating CR [35–37]. While on its own, silicon can only support conventional CR in the visible, its plasmonic properties in the DUV → EUV (Section 2) can be used to realize structures with hyperbolic behavior that generate TCR in this untapped region of the spectrum.

The novel TCR phenomena possible in HMMs can be determined by first considering the CR cone angle ($\theta_c$) in uniaxial media:

$$\tan(\theta_c) = \sqrt{\left(\frac{v_z}{c}\right)^2 - \varepsilon_{||} - \varepsilon_{\perp}},$$  \hspace{1cm} (1)$$

where $\theta_c$ is the angle between the Cherenkov wave vector ($k_x$) and the axis of the electron trajectory [Fig. 4(a)], $\varepsilon_{||}$ is the permittivity of the uniaxial structure in the planar direction, and $\varepsilon_{\perp}$ is the permittivity parallel to the c axis (see Supplement 1 for details).

In the case of an HMM, we impose the following conditions on our permittivity for the orthogonal directions of the metallic and dielectric response: $\varepsilon_{||} < 0$, $\varepsilon_{\perp} > 0$ (type I HMM) and $\varepsilon_{||} > 0$, $\varepsilon_{\perp} < 0$ (type II HMM). The CR velocity threshold with the imposed HMM permittivity conditions can be determined by requiring real values of $\theta_c$ in Eq. (1):

$$v_z ≤ c/\sqrt{\varepsilon_{||}}$$  \hspace{1cm} Type I,  \hspace{1cm} (2)$$

$$0 ≤ v_z ≤ c$$  \hspace{1cm} Type II.  \hspace{1cm} (3)$$

We see that in the case of an HMM for the type I case, the electron velocity now has an upper limit. This is the exact opposite of a conventional isotropic dielectric, where a minimum velocity, i.e., lower limit, exists ($v_z ≥ c/\sqrt{\varepsilon}$). Furthermore, for the type II HMM, any electron velocity will generate CR (details in Supplement 1). These are the cases of TCR. Observe that if we consider a simple isotropic dielectric in Eq. (1) ($\varepsilon_{||} = \varepsilon_{\perp} ≥ 0$), we retrieve the conventional CR limit ($v_z ≥ c/\sqrt{\varepsilon}$). Additionally, note that the conditions for the type I and type II CR velocity thresholds would be flipped for an electron traveling along the $x$ direction, as was seen in [35].

Via harnessing silicon’s unique EUV plasmonic properties (Section 2), novel EUV HMMs can be designed using widely used materials in a simple geometry such as a Si/SiO$_2$ multilayer stack. Figure 4(a) shows such a Si/SiO$_2$ multilayer structure whose permittivities in the effective medium limit (homogenized with Maxwell–Garnett theory; details in Supplement 1) possess both type I and type II hyperbolic behavior from the DUV to the EUV [Fig. 4(b)]. We envision that a practical realization of this structure would consist of approximately 16–20 alternating 8–10 nm layers of SiO$_2$ and crystalline Si. We strongly emphasize that our silicon-based metamaterial design is unique and is unrelated to previous approaches exploiting doped semiconductors [9,10] or alternate plasmonic media [7]. Also note
that doped semiconductors cannot have plasmonic responses at high frequencies beyond the infrared region.

The hyperbolic regimes of the Si/SiO₂ multilayer stack give rise to the unique TCR excitation. This is clearly seen in Fig. 4(d), which shows the simulated CR fields in the dielectric and hyperbolic regimes of the Si/SiO₂ multilayer at different electron velocities. We observe in the type I regime that as the electron velocity decreases, the relative CR fields increase. This is the fundamental characteristic of TCR and is the exact opposite trend seen in the dielectric regime, where CR is supported. CR in the hyperbolic regime can be observed down to electron velocities as low as \( v_e / c \approx 0.0136 \) in the effective medium limit; however, there is a fundamental trade-off between the velocity threshold reduction and the loss in the structure (see Supplement 1).

Note that the type I regime supports TCR in the EUV (≈11–15.5 eV) for the Si/SiO₂ structure. This allows for a
of light in the medium. The threshold velocity of CR can be connected to the phase velocity of the Cherenkov wave vector. Consequently, the threshold velocity also vanishes (see Supplement 1). Consequently, the threshold velocity vanishes when \( \omega_0 / k \rightarrow 0 \). Note that the type I region has an upper threshold when \( v_z \geq c / \sqrt{\epsilon_r} \), and is suppressed at \( v_z = 0.9c \). The type II region has no velocity threshold.

Additionally, the TCR dispersion extends to larger wave vectors as the velocity decreases, as it is approaching the resonance condition described in Eq. (S1) of Supplement 1. TCR is clearly observed in the type I and type II HMM regimes, where the scattering probability increases with decreasing electron velocity. Note that the type I region has an upper threshold when \( v_z \geq c / \sqrt{\epsilon_r} \), and is suppressed at \( v_z = 0.9c \). The type II region has no velocity threshold.

### 4. CONCLUSION

In conclusion, we experimentally demonstrate the generation of EUV plasmons supported by silicon with energies twice as large as those seen with aluminum via momentum-resolved electron energy loss spectroscopy. \( k \)-EELS is the ideal tool to observe such high-energy excitations while simultaneously mapping the photonic band structure of plasmonic excitations to large wave vectors not possible with conventional electron energy loss spectroscopy techniques. Our experimental observations are rigorously validated using macroscopic electrodynamic simulations of \( k \)-EELS and also first-principles DFT. Additionally, we proposed a simple \( Si / SiO_2 \) multilayer stack with a hyperbolic isofrequency response that can generate tunable and broadband TCR in the EUV by harnessing silicon’s unique EUV plasmonic properties. This can lead to applications in EUV waveguides/metamaterials/nanowires/hybrid MEMS based on silicon, EUV light sources generated with low energy excitations, detectors for observation of nonrelativistic particles, and the development of TCR free-electron lasers [37].

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See Supplement 1 for supporting content.

### REFERENCES


