Relaxing Graphene Plasmon Excitation Constraints Through the Use of an Epsilon-Near-Zero Substrate

Vinicius T. Alvarenga1,2 · Dario A. Bahamon1,2 · Nuno M. R. Peres3,4 · Christiano J. S. de Matos1,2

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Abstract
Graphene plasmons have attracted significant attention due to their tunability, potentially long propagation lengths and ultracompact wavelengths. However, the latter characteristic imposes challenges to light-plasmon coupling in practical applications, generally requiring sophisticated coupling setups, extremely high doping levels and/or graphene nanostructuring close to the resolution limit of current lithography techniques. Here, we propose and theoretically demonstrate a method for alleviating such a technological strain through the use of a practical substrate whose low and negative dielectric function naturally enlarges the graphene polariton wavelength to more manageable levels. We consider silicon carbide (SiC), as it exhibits a dielectric function whose real part is between −1 and 0, while its imaginary part remains lower than 0.05, in the 951 to 970 cm\(^{-1}\) mid-infrared spectral range. Our calculations show hybridization with the substrate’s phonon polariton, resulting in a polariton wavelength that is an order of magnitude longer than obtained with a silicon dioxide substrate, while the propagation length increases by the same amount.

Keywords Plasmonics · Graphene · Epsilon-near-zero · Nanophotonics

Introduction
Plasmons, the collective oscillation of free electrons in a conductive material, have attracted great attention in the field of nanophotonics due to its high confinement of electromagnetic energy, achieving subdiffractional spatial extensions [1, 2], which allows for the construction of photonic devices at the nanometer scale. The applications range from biosensing [3–6] to enhanced spectroscopy techniques such as Surface-Enhanced Raman spectroscopy (SERS) [7–9], Surface-Enhanced Infrared Absorption Spectroscopy (SEIRA) [10], Tip-Enhanced Raman Spectroscopy (TERS) [11–14] and Scattering-type Scanning Near-field Optical Microscopy (SNOM) [15–17, 17–19].

Graphene, a two-dimensional material consisting of a single layer of carbon atoms in a hexagonal lattice [20], has emerged as a promising material for plasmonic applications due to its high electronic mobility and null bandgap, which allows for high Fermi level tunability and significant changes on the optical conductivity [21, 22]. Since the plasmon dispersion is dependent on the electronic density, graphene plasmons can be actively controlled more easily than those in conventional conductors [23]. Also, graphene can be transferred to a range of dielectric surfaces, allowing for the integration of graphene-based plasmonic devices with photonic devices, such as silicon-based waveguides [24].

The plasmon wavevector in graphene tends to be up to 2 orders of magnitude larger than that of free-space radiation in the mid-infrared range. However, efficient radiation-plasmon coupling, for the excitation of surface plasmon polaritons (SPPs), requires circumventing the large wavevector mismatch. For bridging such a high mismatch, finding a suitable high-index dielectric material for prism coupling [25, 26] becomes impractical, while diffraction grating coupling has required structures with
periodicity of tens of nanometers [27–29], posing a strain on the device fabrication step. Other approaches require the control of nanometric gaps and very thin materials [30, 31] Alternatively, localized surface plasmons (LSPs) have been excited in graphene patterned into nanostructures such as nanoribbons or nanodisks [32–34]. Again, due to the large wavevector mismatch, the features in these structures need to be in the tens to hundreds of nanometers range, for operation in the mid-infrared [23, 34, 35]. Even though such structures have been fabricated and used to excite graphene plasmons, the fabrication of such devices is challenging due to their size being close to the spatial resolution limit of current nanofabrication techniques [36].

The plasmon dispersion in graphene is dependent on the dielectric function of the adjacent media. Therefore, the choice of the substrate plays a key role in the wavevector matching for plasmon excitation. In particular, a decrease in the magnitude of the plasmon wavevector is expected for a substrate with a low permittivity [1, 24]. The plasmon wavevector is proportional to the sum of the dielectric functions of the substrate and superstrate. If we consider the latter to be air, meaning a dielectric function equal to 1, the optimal choice of substrate would be one with dielectric function of −1. However, a common drawback of materials with a negative dielectric function is that the imaginary part of the dielectric function cannot be neglected, leading to plasmon losses.

Epsilon-near-zero (ENZ) materials have received considerable recent attention for a number of interesting properties, including increased nonlinear optical susceptibilities [37], which can be used to enhance nonlinear effects such as second harmonic generation [38], and wavelength enlargement, which allows light tunneling through narrow channels [39]. As in plasmonics, ENZ applications also require a low imaginary dielectric function component at the spectral range where the real dielectric function is low. As a consequence, ENZ materials are also good candidates for plasmonic applications. Indeed, they have been studied in the field of plasmonics for being capable of directly coupling SPPs to free-space light [40].

Silicon carbide is an ENZ material in the mid-infrared due to its optical phonons [41–43]. The range between the transverse optical phonon (TO), at 797 cm\(^{-1}\), and the longitudinal optical phonon (LO), at 970 cm\(^{-1}\), called the Reststrahlen band, exhibits a real negative dielectric function, with the function crossing zero at both these wavenumbers [44, 45]. In particular, a low imaginary part of epsilon is obtained near the LO phonon wavenumber. Within the Reststrahlen band, the material also supports surface phonon polaritons (SPhP) [45–48]. It has been shown that when graphene plasmons are excited within the Reststrahlen band of a SiC substrate, hybrid plasmon-phonon modes arise, which maintain the characteristics of both excitations, exhibiting a longer wavelength than a pure SPP and keeping losses low [49–53]. The use of SiC is also convenient since graphene can be epitaxially grown directly onto this substrate [54–58]. Other polar dielectrics have also been studied as substrates for graphene plasmonics due to their ENZ nature [50, 51, 53], however, ENZ substrates have not been used for increasing the graphene plasmon wavelength with the purpose of reducing the wavevector mismatch for optical excitation. In this work, we propose the use of SiC as a graphene substrate for mitigating this mismatch in the mid-infrared. Our results show an increase from hundreds of nanometers to a few micrometers in polariton wavelength on SiC when compared with a SiO\(_2\) substrate. This simplifies the polaritonic excitation while keeping the polariton modal volume 3 orders of magnitude smaller than that of free-space radiation. Although there is a loss of confinement, these polaritonic waves become easier to excite, and the fabrication of polariton-based devices becomes easier. The conditions for this trade-off can be adjusted by tailoring the size of the structures used for excitation and controlling the graphene doping.

**Methods**

To obtain the graphene plasmon dispersion, we used the condition for SPP formation [1]:

\[
\frac{\varepsilon_1}{\sqrt{\beta^2 - k_0^2\varepsilon_1}} + \frac{\varepsilon_2}{\sqrt{\beta^2 - k_0^2\varepsilon_2}} + \frac{i\sigma}{\omega\varepsilon_0} = 0, \tag{1}
\]

where \(\beta\) is the plasmon wavenumber, \(k_0\) is the free-space radiation wavenumber, \(\sigma\) is the graphene optical conductivity, \(\varepsilon_1, \varepsilon_2\) are the dielectric functions of the superstrate, substrate, respectively, and \(\varepsilon_0\) is the vacuum permittivity.

We assume graphene to be on the interface between air (\(\varepsilon_1 = 1\)) and either SiC or SiO\(_2\). We use the Drude model for the graphene conductivity [1, 24]:

\[
\sigma(\omega) = \frac{e^2E_F\tau}{\hbar^2\sqrt{\pi}(1 - i\tau\omega)}, \tag{2}
\]

where \(e\) is the electron charge, \(E_F\) is the Fermi energy and \(\tau\) is the relaxation rate. This is a good approximation when the temperature is low and electronic density is high. Also, non-local effects are neglected. These are considered important only for small nanostructures and when graphene is near metallic surfaces [61]. The value of the relaxation time is assumed to be such that \(\hbar\tau^{-1} = 2.4\) meV [62]. The dielectric function for SiO\(_2\) was taken to be given by [60].

\[
\varepsilon_{\text{SiO}_2}(\omega) = \varepsilon_{\infty,\text{SiO}_2} \sum_j \frac{\omega_j^2}{\omega_j^2 - \omega^2 - i\Gamma_j\omega_j}, \tag{3}
\]
with \( \epsilon_{\infty, \text{SiO}_2} = 2.3 \) and using the parameters shown in Table 1. The dielectric function of SiC was taken to be given by [59]

\[
\epsilon_{\text{SiC}}(\omega) = \epsilon_{\infty, \text{SiC}} \frac{\omega^2 - \omega_1^2 + i\Gamma\omega}{\omega^2 - \omega_1^2 + i\Gamma\omega},
\]

with \( \epsilon_{\infty, \text{SiC}} = 6.7, \omega_L = 120.5 \text{ meV}, \omega_T = 98.4 \text{ meV} \) and \( \Gamma = 1.4 \text{ meV} \).

We numerically solved Eq. 1 for obtaining the in-plane wavevector \( k \), from which we can calculate both the polaron wavelength \( (\lambda_p = 2\pi/Re(\beta)) \) and the effective propagation length \( (L_{\text{eff}} = 1/2Im(\beta)) \). To better understand the plasmon-phonon interaction, we also calculated the loss function, given by the imaginary part of the p-polarized reflectivity \( r_p \) [24]:

\[
r_p = \frac{k_1\epsilon_2 - k_2\epsilon_1 + k_1\epsilon_2\sigma/\epsilon_0\omega}{k_1\epsilon_2 + k_2\epsilon_1 + k_1\epsilon_2\sigma/\epsilon_0\omega},
\]

where \( k_1 \) and \( k_2 \) are the wavevectors of light propagating in air and the substrate, respectively.

We have also calculated the reflectance and absorbance spectra for an array of graphene ribbons in order to determine the corresponding LSP resonance condition. Since the conductivity of a regular array of graphene nanoribbons is spatially periodic \( (\sigma(x + R) = \sigma(x)) \), where \( R \) is the periodicity, the reflected and transmitted fields can be expressed by a Fourier-Floquet series. Matching the boundary conditions at the interface, we arrive at the linear system of equations [24]:

\[
\frac{i}{\omega\epsilon_0} \sum_j \tilde{\sigma}_{j,n} L_{j,n}^{(1)} E_{j,n}^{(1)} + \frac{\tilde{\sigma}_n E_{n,n}^{(1)}}{\epsilon_0} = i2\epsilon_n \frac{E_{\text{inc}}^{(1)}}{\epsilon_0} \delta_{n,0}
\]

where \( \beta = k \sin \theta, k_z = k \cos \theta, \sigma_{j,n} = \sqrt{(\beta + nG)^2 - \epsilon_0\omega_0/c^2} \) and \( G = 2\pi/R \). The Kronecker delta signals that the incident field \( E_{\text{inc}}^{(1)} \) only appears for the diffraction order \( n = 0 \). The Fourier coefficients of the conductivity \( \tilde{\sigma}_j = \int_0^R \sigma e^{-iGjx} dx/R \) are calculated assuming that the width of the graphene nanoribbon in the unit cell is \( d_g \leq R \), and has the same conductivity of a graphene sheet \( (\sigma) \). Once the fields are obtained, the reflectance, transmittance and absorbance are respectively calculated as:

\[
T = \sum_n \frac{\epsilon_1 k_z}{\epsilon_2} \left| E_{x,n}^{(1)} / E_{\text{inc}}^{(1)} \right|^2
\]

\[
R = \sum_n \frac{k_z}{\epsilon_1 k_{z,n}} \left| E_{x,n}^{(2)} / E_{\text{inc}}^{(1)} \right|^2
\]

\[
A = 1 - T - R
\]

\[\begin{array}{ccc}
\omega_j & s_j & \Gamma_j \\
142 & 28.5 & 7.4 \\
133 & 88.5 & 5.4 \\
100 & 23.2 & 4 \\
57 & 56.8 & 6.2 \\
47 & 32.7 & 24.5 \\
\end{array}\]

Figure 1A, B show the dielectric functions of SiC and SiO\(_2\), respectively. For silicon carbide, the real part of the dielectric function is equal to zero at 970 cm\(^{-1}\). At 951 cm\(^{-1}\) the real dielectric function is \( -1 \) and the corresponding imaginary part is \( < 0.04 \), meaning that polaronic losses are expected to be low. In the same spectral region, SiO\(_2\) has a real dielectric function of 4 to 5, with a much higher imaginary part of around 0.4 to 0.8, meaning that the graphene plasmon wavelength is expected to be shorter and propagation losses are expected to be higher.

Table 1 Parameters used for calculating the dielectric function of SiO\(_2\) using Eq. 3

| \( \omega_j \) (meV) | \( s_j \) (meV) | \( \Gamma_j \) (meV) |
|----------------------|----------------|-------------------|
| 142                  | 28.5           | 7.4               |
| 133                  | 88.5           | 5.4               |
| 100                  | 23.2           | 4                 |
| 57                   | 56.8           | 6.2               |
| 47                   | 32.7           | 24.5              |
Results and Discussion

Figure 2A, B respectively show the polariton wavelength and effective propagation length as functions of free-space wavenumber calculated by solving Eq. (1) for an air/SiC interface with and without graphene with a Fermi energy of 0.4 eV. We focus this analysis in the spectral region where the real dielectric function of the substrate is between 0 and −1. Without graphene this system should not, in principle, exhibit surface phonon polaritons for an excitation wavenumber beyond 951 cm$^{-1}$, as $\text{Re} [\epsilon_1 + \epsilon_2] > 0$, not allowing for polaritons. However, because of the imaginary part of $\epsilon_2$, quasibound and leaky modes [2, 24] arise for wave numbers between 951 and 970 cm$^{-1}$. Also, beyond 970 cm$^{-1}$ SiC’s dielectric function is positive and the curves correspond to a radiative mode rather than to surface polaritons [2]. The addition of graphene to the interface allows for the excitation of lower loss plasmon-phonon polaritons above 951 cm$^{-1}$, which is expressed by a shorter wavelength and a longer propagation length. For lower wavenumbers,
within the Reststrahlen band, graphene imposes only a minor change to the wavelength, which suggests that in that region the polariton would exhibit a dominant SPPhP character.

Figure 2C, D show a comparison of the polariton wavelength and propagation length using SiC and SiO₂ as the substrate. At the spectral range where the real dielectric function of SiC lies between −1 and 0, an order of magnitude increase in polariton wavelength is obtained. At 951 cm⁻¹, for example, the plasmon wavelength is 0.4 μm on SiO₂ and 5.3 μm on SiC. As for propagation length, it is 0.2 μm for SiO₂ and 7 μm for SiC. Note that for SiO₂, the propagation length is shorter than the polariton wavelength, while for SiC, the propagation length is longer. The combination of longer wavelength and propagation lengths makes polaritonic devices based on graphene on SiC simultaneously easier to fabricate and more efficient. Note that despite the polariton wavelength increase, it remains about half the light wavelength at the same frequency, allowing for subdiffractional devices to be designed. The optimal wavelength for a polaritonic device can be tailored by changing the free-space wavenumber or adjusting graphene’s Fermi level.

Figure 2E, F respectively show the polariton wavelength and propagation length for various graphene Fermi energies, ranging from 0.05 eV to 0.8 eV. As expected, λ_{sp} increases with E_F. At 951 cm⁻¹, for example, Fermi energies of 0.2, 0.4 and 0.8 eV result in plasmon wavelengths of approximately 4, 5.3 and 6.8 μm, respectively. Once again we notice that the propagation length also increases. For the same Fermi energies, we get propagation lengths of 3, 6.5 and 12.8 μm. Even for a low graphene doping such as 0.2 eV, we get a much longer polariton wavelength and propagation length, on the order of micrometers, than with a SiO₂ substrate, which results in wavelengths of hundreds of nanometers, with much higher losses.

It is important to highlight that the performance of a polaritonic material is generally addressed by the quality factor (Q = Re[β]/Im[β]) [63], that can be easily calculated from our data as Q = 4πT_{eff}/λ_{sp}. For example, with a graphene Fermi energy of 0.4 eV and at 951 cm⁻¹, for SiO₂ Q = 4π(0.2/0.4) = 6.3, while for SiC Q = 4π(7/5.3) = 16.6, indicating that SiC, as we already discussed, is a superior substrate for polariton excitation. For Fermi energies of 0.2 and 0.8 eV, Q = 9.4 and 23.6, respectively. To put these numbers in context, experiments with graphene encapsulated by hBN have found Q ≈ 25 [64] while experiments with graphene encapsulated in SiO₂ measured Q ≈ 5 [62, 65]. Another metric that can be calculated is the squeezing factor (or confinement factor) [66], defined as the ratio between the wavelength in free space and the in-plane polaritonic wavelength. At 951 cm⁻¹ for graphene (E_F = 0.4 eV) on SiC we obtain Re[β]/k₀ = 2.1. In comparison, for gold at 530 nm (i.e., close to the surface plasmon frequency), Re[β]/k₀ = 1.1. Thus, while our approach alleviates coupling difficulties that arise from the ultrahigh (> 10) squeezing factors usually obtained in graphene plasmonics, it still performs ~ 2x better than gold, in this figure of merit.

To address possible experimental fluctuations in SiC’s dielectric constant, we independently varied the parameters in Eq. (4). We found that varying α₁ and α₂ essentially shifts the frequency at which the ENZ point is obtained, thus, simply requiring that the operating frequency be accordingly shifted. A variation in Γ, which could be associated with a varying degree of disorder in the SiC crystal, on the other hand, virtually does not affect λ_{sp}, and only minimally changes L_{eff}. For example, varying Γ by ±10% results in a variation in L_{eff}, at 951 cm⁻¹, between 6.15 and 6.82 μm (for a graphene Fermi energy of 0.4 eV) in comparison with 6.5 μm as previously shown in Fig. 2F.

In order to easily visualize the wavevector mismatch between free-space radiation and the polariton wave in graphene on the two different substrates, we plot, in Fig. 3, ΔK = |λ/λ₀ − Re[β]|, where λ₀ is the free-space light wavelength, for both SiC and SiO₂ as substrates. We consider a graphene with 0.4 eV Fermi energy. A decrease of more than an order of magnitude in ΔK is obtained by using SiC. The lower wavevector mismatch between polariton and free-space radiation makes the fabrication of polaritonic devices simple. Even if we consider a graphene with a low doping of 0.2 eV, as our results show, the polariton wavelength is 4.9 μm, meaning an excitation grating, for example, with a periodicity of 2.45 μm (considering it to be half the wavelength). Such a feature size is easily fabricated with simple photolithography, rather than electron beam lithography, commonly used for conventional graphene plasmonic devices.
Since patterned graphene is one of the most convenient ways to build plasmonic devices, we now analyze the case of localized surface plasmons in microstructured graphene. We consider ribbon widths, $D$, from 0.5 to 2 $\mu$m, and fix the ribbon array periodicity at $R = 2D$. Figure 4A, B show the reflectance and absorbance spectra for various ribbon widths with a 0.4 eV Fermi energy. Since silicon carbide has a negative dielectric function below 970 cm$^{-1}$, for lower wavenumbers the material has a high reflectance, which is seen as a step in reflectance spectra. The polaritonic resonance is observed as a localized dip in reflectance and as peaks in absorbance. The resonance point can be tuned by adjusting the ribbon width and for excitation at 951 cm$^{-1}$ 2 $\mu$m ribbons are required. With $D = 1.0$ and 0.5 $\mu$m the polariton resonance is at 951 and 964 cm$^{-1}$, respectively. Figure 4C, D show the same calculations for different Fermi energies considering 1 $\mu$m wide ribbons. Fermi energies of 0.2, 0.4 and 0.8 eV present polaritonic resonances at 952, 954 and 961 cm$^{-1}$ respectively. Our calculations, therefore, show that the resonances in this spectral region can be excited using structures on the scale of micrometers, which alleviate the strain on fabrication. The absorbance peak that remains still ($\sim$ 970 cm$^{-1}$) while the doping level and period of the graphene array are modified is produced by bulk evanescent waves in the substrate (SiC). It is important to notice that this peak is present even without the excitation of the surface polaritons[67].

Figure 5 shows the calculated loss function for graphene at an air-SiC interface at various doping levels, through which the polariton dispersion relation can be visualized as bright yellow/white lines. Figure 5A–C show the polariton dispersion for graphene with a Fermi energy of 0.05, 0.2 and 0.4 eV, respectively. When graphene is added to the interface, plasmon-phonon polariton hybridization takes place upon an anti-crossing of the dispersion relations, yielding 2 polariton branches, which shift towards higher free-space wavenumbers as the graphene Fermi level increases. As a general rule, the hybridized modes behave more like an SPP for low free-space wavenumbers and more like an SPhP for higher free-space wavenumbers [24]. Figure 5D shows a zoom of Fig. 5C at the area indicated by the yellow rectangle. The dashed cyan lines represent the wavenumbers of the LO and TO SiC phonons, the dashed green line represents the point where Re$\{\varepsilon\} = -1$, which is also the point where SPhP would appear for SiC without graphene and the yellow line represents light propagation through silicon carbide. The long polariton wavelengths reported here correspond to the upper hybrid polariton branch.

It should be noted that similar hybridization takes place near the optical phonon wavenumbers of other polar...
dielectrics used as substrates and novel physical phenomena have been reported. For example, negative refraction of coupled graphene plasmon and type-I phonon polaritons in graphene-BN heterostructures \[66\], tunable planar focusing of hyperbolic phonon polaritons in $\alpha$-MoO$_3$ \[68\] and topological transition in hybrid polaritons of graphene-$\alpha$-MoO$_3$ heterostructures \[69\]. In fact, in SiO$_2$ at 1184 cm$^{-1}$, where its dielectric function equals $-1$, we calculate that the polariton wavelength reaches a more moderate value of 1.2 $\mu$m, with a corresponding sub-wavelength propagation
length of 0.9 \( \mu m \) \((Q = 9.4)\). When compared with SiC \((Q = 16.6 \text{ at } 951 \text{ cm}^{-1})\), the shorter polariton wavelength and propagation lengths are a consequence of SiO\(_2\)'s imaginary dielectric function, which remains high, 0.6 at the 1184 \text{ cm}^{-1}, at the epsilon-near-zero point. Similar quality factors \((Q = 20)\) are also obtained for the aforementioned coupled graphene plasmon and type-I phonon polaritons [66]. Therefore, we can conclude that, although polariton wavelength increase can be achieved for other polar dielectrics, SiC is a more efficient substrate due to its simplicity and lower losses.

Although superior polariton confinement has been achieved in various 2D and very thin van der Waals materials, efficient far-field coupling requires meticulous and costly nanofabrication processes. For example, record values of confinement have been found for hyperbolic surface phonon polaritons in nanometric hBN ribbons [10] and cones [44]. The same is valid for image polaritons in graphene [70] or hBN [71], where the performance strongly depends on the gaps between graphene/hBN and the metallic plate. For metals, at any wavelength, strong confinement is accomplished with very thin insulator layers sandwiched between two metals [72]. Notwithstanding, confinement is not the only variable to be minimized; losses appear through distinct mechanisms and the rule of thumb for metal plasmonics (i.e., the better the confinement, the lower the propagation length [2]) ends up valid. The trade-off among confinement, losses and effective far-field coupling will define the feasibility of practical polaritonic devices. Here we have tackled this question, showing an approach that reduces the complexity of polaritonic devices with competitive confinement and loss performances.

**Conclusions**

We have demonstrated a method for increasing the wavelength of graphene polaritons in order to relax the conditions for matching their wavevector with that of free-space radiation. For this purpose, we choose a substrate with a real dielectric function between -1 and 0 in the mid-infrared region: silicon carbide. In the 951 to 970 \text{ cm}^{-1}\ spectral range, we observe an order of magnitude polariton wavelength increase relative to SiO\(_2\) as a substrate, which is accompanied by an order of magnitude increase in propagation length. Due to the Reststrahlen band formed by phonons in the crystal, graphene plasmons hybridize with SiC phonons, allowing for the wavelength increase, which makes the excitation of polaritons with launching gratings easier. Along the same lines, graphene structures of up to 2 \( \mu m \) can be used for localized surface plasmon excitation. These results confirm SiC as a convenient substrate for graphene plasmonics, allowing for simple and large-scale microfabrication methods to be employed, which can contribute to the more widespread use of graphene-based subdiffractive photonics in real-world applications. Our approach is different from previous works that considered graphene on SiC, but exploiting SiC as an optical dielectric layer \((\text{Re}[\varepsilon_{\text{SiC}}] > 0)\) [73].

**Author Contribution** VTA carried out all numerical calculations. DAB and CJSM proposed and supervised the work. DAB, NMRP and CJSM analyzed the data. All authors contributed to the writing of the manuscript.

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**Availability of Data and Material** All data generated and analyzed during this study are generated by the methods published in this article. The datasets are available from the corresponding author.

**Declarations**

**Conflict of Interest** The authors declare no competing interests.

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