1. INTRODUCTION

Graphene as a single sheet of carbon atoms in a hexagonal lattice has shown abundant new physics and potential applications in carbon-based nanoelectronic devices because of its exceptional electrical and optical properties [1–5]. It has the highest known electrical and thermal conductivity, as well as the highest stiffness and strength [6]. The ability to modify the electronic structure of graphene is needed to make a graphene-based device or circuit. Possible tools for achieving this goal include patterning, electric field effects, and chemical doping [7,8]. Recently, it has even been proposed that strain can be used to achieve various essential elements for all-graphene electronics [9]. The strain effect in graphene provides a new way to manipulate electron transport without external fields. Graphene is structurally more tolerant than silicon to external effects such as strain. Graphene, as the strongest material ever measured, can also support strain well beyond the linear regime, without being bent or wrinkled. Very high strains can be easily exerted on graphene before the mechanical failure [10–14]. The effect of strain on graphene has been studied both experimentally and theoretically [15–19]. The mechanical strain often has unexpected effects on the electronic properties of carbon nanomaterials [20–26]. Graphene offers a new opportunity to discover such interesting nano- mechanical properties in two dimensions [1]. Mechanically strained graphene may introduce a new environment to study the novel physical phenomena [27–29]. The strain may be applied naturally or intentionally to graphene.

In many applications, graphene lays or grows on a substrate. Growth of graphene on substrates like SiO\textsubscript{2} or SiC with a different lattice constant usually introduces strain due to surface corrugations or lattice mismatch [30,31], which can be detected by Raman spectroscopy [32]. Even in the absence of a mismatch, the strain still occurs along the edges and exhibits some interesting quantum properties [33,34]. Bending the substrates may induce a uniaxial strain on it [35]. Using tension to control the physical properties of graphene has attracted an ever-increasing interest [36–45].

From a technological point of view, understanding how strain affects graphene’s electronic and optical properties is of paramount importance. Since the electronic states near the Fermi level are directly related to optical and transport properties of graphene, most researchers have focused on the electronic structure of graphene near the Dirac points [46–53]. Moreover, the density state may exhibit one or more Van Hove singularities. The wave vectors at which Van Hove singularities occur are often referred to as critical points of the Brillouin zone [54]. In bulk solids, the presence of Van Hove singularities in the density of states will result in strong optical absorption peaks as well as Raman signals. However, in graphene systems, few empirical studies show the existence of Van Hove singularities [55,56]. The existence of Van Hove singularities in graphene structures may be influenced by strain [57]. Motivated by this, we want to investigate Van Hove singularities in strained graphene. In this paper, we present a method for measuring the frequency of Van Hove singularities, based on Fresnel equations [58] and phase-shifting interferometry [59]. Researchers have used different experimental setups for phase-shifting interferometry [60–64]. Here, we introduce a method that may also be used to investigate the Van Hove singularities of the other two-dimensional materials, such as MoS\textsubscript{2}, transition metal dichalcogenides (TMDs),...
and Silicene. However, the frequency position of Van Hove singularities of these materials differs from that of the graphene.

In this method, a linearly polarized light beam passes through a phase-sensitive internal reflection device, consisting of a semi-cylindrical prism, two half-wave plates, and two quarter-wave plates with proper azimuth angles. The output beam enters a Mach–Zehnder interferometer with two acousto-optic modulators separately placed in two arms. Eventually, the two linearly orthogonally polarizations of each output beam of the interferometer interfere with each other as they pass through two analyzers, respectively. The ultimate phase difference between the two interference signals is associated with the azimuth angle of the fast axis of the half-wave plates, yielding the sensitivity-tunable functionality.

2. THEORETICAL CALCULATION AND MODEL

Let us consider a single flat sheet of monolayer graphene deposited on a substrate of refractive index \( n_s \), which is capped by a high-index prism of refractive index \( n_p \). A laser beam is incident upon the base of the prism at the angle of incidence \( \theta \) [see Fig. 1(a)]. The reflection and transmission coefficients of the structure are determined by satisfying the continuity of the tangential component of the electric fields caused by the induced surface current in graphene as

\[
\begin{align*}
\text{r}_i &= \frac{n_p \cos \theta - n_s \cos \theta' - \eta_0 \sigma}{n_p \cos \theta + n_s \cos \theta' + \eta_0 \sigma}, \\
\text{r}_p &= \frac{n_s \cos \theta - n_p \cos \theta' + \eta_0 \sigma \cos \theta + \eta_0 \sigma}{n_s \cos \theta + n_p \cos \theta' + \eta_0 \sigma \cos \theta + \eta_0 \sigma}, \\
\text{t}_i &= \frac{2n_p \sin \theta}{n_p \cos \theta + n_s \cos \theta' + \eta_0 \sigma}, \\
\text{r}_p &= \frac{2n_s \sin \theta}{n_s \cos \theta + n_p \cos \theta' + \eta_0 \sigma}. 
\end{align*}
\]  

Fig. 1. (a) Geometry of the problem with monolayer graphene deposited on a substrate. Light is incident from a high-index prism at the incidence angle \( \theta \). (b) The graphene honeycomb lattice with its zigzag and armchair directions, and the tension direction. Here, \( \theta \) and \( \phi \) denote the angles that the applied strain makes with the zigzag path and the tangential electric field, respectively.

Here, \( \eta_0 \) is the intrinsic impedance of free space, and \( \sigma \) is the optical conductivity of the graphene monolayer. As one knows, the optical conductivity of the graphene is the sum of the intraband conductivity (due to the intraband electron-photon scattering processes) and the interband conductivity (which originates from the direct interband electron transitions) [65]. The optical conductivity of the graphene has been derived using perturbation theory in references [66,67]. It was shown that the applied tension deforms the graphene lattice and distorts the reciprocal lattice [68]. Since the conduction and valence energy bands of the graphene vary due to the lattice deformation, the induced tension changes the optical conductivity of graphene [68]. We use the procedure explained in [65–68] to obtain \( \sigma \) under the applied tension without giving its explicit form here. Figures 2(a) and 2(b) show the real and the imaginary parts, respectively, of \( \sigma \) (in the unite of \( \sigma_0 = \frac{e^2}{\pi \hbar} \)) versus the photon energy (\( \omega \)) for applied tension with different modules at different directions. Here, we assume that the direction of the applied tension makes angles \( \theta \) and \( \phi \) with the zigzag path in the graphene honeycomb lattice and the tangential electric field of the incident beam, respectively, and \( \zeta \) indicates the strain modulus [see Fig. 1(b)]. We see some maxima at the real part of \( \sigma \), which are attributed to the Van Hove singularities at the saddle-points of the electronic band structure of the graphene in the UV band [68,69].

In unstrained graphene, there are three equivalent saddle-points in the electronic band structure, which show themselves as a single peak in the optical conductivity (see dotted lines in Fig. 2). In the strained graphene, at most two of the saddle-points may be equivalent. As a result, we see two or three peaks in \( \sigma \). The number of peaks and their position strongly depend on the modulus and direction of the applied tension. Here, we would like to present a simple procedure to obtain the effect of applied tension on the position of the Van Hove singularities.

Fig. 2. (a) Real and (b) imaginary parts of the normalized optical conductivity of the strained graphene versus photon energy \( \omega \).

Fig. 3. Schematic diagram for measuring the phase difference \( \psi \).
The schematic experimental setup for this measurement is shown in Fig. 3.

For convenience, we consider the direction of the propagated light beam as the $x$ axis and set the $x$ axis perpendicular to the plane of the paper. A linear $s$-polarized laser light after passing through the half-wave plate 1 (with the fast axis at $\frac{\alpha}{2}$ with respect to the $x$ axis), the quarter-wave plate 1 (with the slow axes at $45^\circ$ with respect to the $x$ axis), and the quarter-wave plate 2 (with its slow axes along the $x$ axis) is incident vertically on a hemi-cylindrical prism with the refractive index of $n_p$. In our considered frequency range, several laser sources such as the TA-FHG pro and SuperK EXTREME lasers may be used in the experimental setup. The light ray is refracted into the prism, and it propagates at the incidence angle $\theta$ toward the boundary surface between the prism and the graphene monolayer. For $\theta$ larger than the critical angle $\sin^{-1}\frac{n_p}{n_s}$, the light undergoes total internal reflection at the boundary. Then, the reflected light beam travels through the half-wave plate 2, whose fast axes makes angle $\alpha$ with the $x$ axis. The Jones vector of the light after the half-wave plate 2 can be written as

$$E_i = \left( \begin{array}{c} A_i e^{i\phi_i} \\ A_p e^{i\phi_p} \end{array} \right) = \frac{t t'}{\sqrt{2}} \left( \begin{array}{c} r_p \cos 2\alpha e^{-i\Delta} + ir_s \sin 2\alpha e^{i\Delta} \\ r_p \sin 2\alpha e^{-i\Delta} + ir_s \cos 2\alpha e^{i\Delta} \end{array} \right),$$

where

$$t = \frac{2}{n_p + 1}, \quad t' = \frac{2n_p}{n_p + 1}. \tag{4}$$

Here, $t$, $t'$ are the transmission coefficients at the air–prism and the prism–air interface, respectively. A 50/50 beam splitter of a Mach–Zehnder interferometer splits the reflected light beam into reflected and transmitted beams after passing through the half-wave plate 2. Those beams pass through the electro-optical modulators 1 and 2 to shift their frequencies to $\omega_1$ and $\omega_2$, respectively. Then, a polarizing beam splitter superimposes the beams. The output beams after passing through the analyzers 1 and 2 (whose transmission axis makes angle $\beta$ with the $x$ axis) are detected using the photodetectors 1 and 2 in both output ports. The intensities measured by the photodetectors 1 and 2 are

$$I_1 = \frac{1}{2} (A_i^2 \cos^2 \beta + A_p^2 \sin^2 \beta + 2A_i A_p \sin 2\beta \cos (\omega_2 - \omega_1) t + (\phi_p - \phi_i)). \tag{5}$$

$$I_2 = \frac{1}{2} (A_i^2 \cos^2 \beta + A_p^2 \sin^2 \beta + 2A_i A_p \sin 2\beta \cos (\omega_2 - \omega_1) t - (\phi_p - \phi_i)). \tag{6}$$

The intensities $I_1$ and $I_2$ are sent to a lock-in amplifier to obtain the phase difference $\psi = 2(\phi_p - \phi_i)$.

### 3. RESULTS AND NUMERICAL CALCULATIONS

In this section, we describe the numerical analysis. Here, we assumed the incidence angle $\theta$ to be $52.2^\circ$, $n_p = 1.7786$, $n_{p1} = 1.4$, $\alpha \cong 25^\circ$, $\Delta \cong 25^\circ$, and $\beta \cong 0^\circ$, respectively.

Figures 4(a) and 4(b) depict the simulated results by plotting, respectively, the phase difference $\psi$ and the sensitivity $\frac{d\psi}{d\omega}$ versus photon energy ($\omega$) in the absence of the strain. As it is clear from Fig. 4(a), the phase difference $\psi$ monotonically decreases to reach its minimum at a specific frequency and then increases by increasing $\omega$. We find that the phase difference changes its behavior at the frequency in which the Van Hove singularity occurred [compare Figs. 4(a) and 2(a) (the dotted–dashed line)]. In this method, the Van Hove singularity exhibits itself as a sharp peak or deep valley in the phase difference curve. From Fig. 4(b), we find that this method has an acceptable sensitivity ($\frac{d\psi}{d\omega}$) to obtain the frequencies at which the Van Hove singularities occur.

Now we consider the phase difference curve for the case of strained graphene. We first study the case of graphene under strain in a given direction with different moduli of applied strain. Figure 5 shows the phase difference $\psi$ versus the photon energy ($\omega$) for two different moduli $\zeta = 0.14$ (the solid line) and $\zeta = 0.21$ (the dashed line) of the applied strain with the fixed direction at $\theta = 20^\circ$ and $\psi = 0$.

From Fig. 5, we see that the number of sharp deeps in the phase difference curve has increased to three due to the stress, which indicates the existence of three Van Hove singularities in the density of states of the strained graphene. It is clear from the figure that the position of the singularities changes slightly as the stress increases. The first two singularities move to lower frequencies, and the third singularity suffers from a blueshift. We also note that the number of Van Hove singularities is independent of the modulus of applied stress. To illustrate this more clearly, we have plotted the phase difference $\psi$ on the plane ($\omega, \zeta$) for the case of $\theta = 20^\circ$ and $\psi = 0$ in Fig. 6. Here, the applied stresses have eliminated the triplet degeneracy of the Van
Hove singularities. Also, the singularities move away from each other by increasing the modulus of the applied strains.

Continuing, we want to obtain the phase difference $\psi$ for the case of strained graphene at different directions of the applied strain with a given modulus. Figure 7 depicts the phase difference $\psi$ vs. the photon energy ($\omega$) for three different directions of the applied strain at Fig. 7(a) $\theta = 0^\circ$ (the solid line), $\theta = 15^\circ$ (the dashed line), $\theta = 30^\circ$ with $\varphi = 0^\circ$ (the dotted–dashed line) and (b) $\varphi = 0^\circ$ (the solid line), $\varphi = 15^\circ$, (the dashed line) $\varphi = 30^\circ$ (the dotted–dashed line) with $\theta = 30^\circ$. Here, the modulus of the applied strain ($\zeta$) is 0.14. As one knows, $\theta$ is the angle between the direction of the applied tension and the zigzag path in the graphene honeycomb lattice, and $\varphi$ indicates the angle between the directions of the applied tension and the tangential electric field of the incident beam. Figure 7 reveals that the number of the Van Hove singularities mostly depends on $\theta$ (the angles between the applied strain and the zigzag path), and it is approximately independent of $\varphi$.

In order to further investigate the effect of the stress direction on the Van Hove singularities, we plot the phase difference $\psi$ on the planes of ($\omega$, $\varphi$) for different values of $\varphi$ with the applied strain of the modulus $\zeta = 0.14$ in Fig. 9. Figure 9 reveals that the number of Van Hove singularities mostly depends on $\theta$ (the angles between the applied strain and the zigzag path), and it is approximately independent of $\varphi$.

4. CONCLUSION

An experimental method for measuring the frequency of the Van Hove singularities in the density of states of strained graphene is proposed. The phase difference of $p$- and $s$-polarized reflected light under the total internal reflection in a hemi-cylindrical prism, whose base is contacted with the strained graphene, can be measured accurately with the phase-shifting interferometry. The Van Hove singularities present themselves as some sharp dips or peaks in the phase difference spectrum. The validity of the method has been examined using the special equations derived to estimate the phase difference of the interferometric signals. We showed that the number of Van Hove singularities strongly depends on the direction of the applied strain. The direction of the applied strain and also its modulus affect the frequency locations of the Van Hove singularities. We demonstrated that the phase-shifting interferometry has an acceptable sensitivity to obtain the frequency of the Van Hove singularities.
Disclosures. The authors declare no conflicts of interest.

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