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Optical and photogalvanic properties of graphene superlattices formed by periodic strain

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Graphene superlattices formed by periodic strain are considered theoretically. It is shown that an electron energy spectrum consists of minibands obtained by folding of the cone at the boundaries of the superlattice Brillouin zone with very narrow anticrossing regions. Light absorption under direct interminiband transitions is shown to be frequency dependent and dichroic. Giant dichroism of absorption is demonstrated for doped graphene superlattices. Asymmetrical graphene superlattices act as quantum ratchets allowing for the generation of photocurrents at absorption of normally incident light. The helicity-dependent photocurrent spectrum is calculated for doped superlattices with various asymmetries.

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I. INTRODUCTION

Graphene attracts a great deal of attention due to both interesting physical phenomena and device applications. Physics of graphene possesses many intriguing effects in optics, transport, and other fields.[1,2] An example is optical absorbance of graphene, which is a fundamental constant in a very wide frequency range. After focusing on pure graphene, the interest is shifted to graphene-based systems whose electronic properties can be changed by external parameters. One of the promising avenues is strain engineering of electronic band structures.[3,4] For example, gaps in an energy spectrum open in strained graphene.[5,6] In graphene, the strain is equivalent to gauge, or pseudomagnetic fields,[7,8] which suppress anomalous magnetoresistance caused by weak localization.[9]

Deformation of graphene results in periodic ripples with height $\sim 0.1\ldots 1$ nm and a period ~ 10 nm.[10,11] Such periodically strained graphene samples have D_{2h} point symmetry which allows for dichroism of light absorption. Asymmetrical strain lowers the symmetry of graphene to C_{2v} with the C_2 axis along the strain direction. In such a system a ratchet effect is possible, which means a generation of electric current under light absorption of an unbiased sample. In particular, a contribution to the photocurrent sensitive to the helicity of light normally incident on a strained graphene structure is allowed. Up until now such circular ratchet effect has been detected only in small graphene samples owing to scattering from sample edges,[12] or for oblique incidence due to the presence of a substrate.[13]

In this work we investigate electronic states and calculate dichroic absorption in periodically strained graphene samples. For asymmetrical periodic strain, we study the circular ratchet effect and calculate the circular photocurrent for various asymmetries of the strain.

II. ELECTRONIC STATES IN GRAPHENE SUPERLATTICES

The Hamiltonian for strained graphene is given by[3]

$$H(\boldsymbol{p}) = v_0 \boldsymbol{\sigma} \cdot (\boldsymbol{p} \pm \boldsymbol{\mathcal{A}}), \tag{1}$$

where $\boldsymbol{p}$ is the quasimomentum operator, $\boldsymbol{\sigma} = (\sigma_x, \sigma_y)$ is a vector of Pauli matrices, and the two signs correspond to two valleys. We consider a graphene sheet periodically strained in the armchair (y) direction:

$$\mathcal{A}_x(y+d) = \mathcal{A}_x(y),$$

where d is the period. The value of $\mathcal{A}_x$ is modulated with the amplitude $\gamma\hbar/d$ with the estimates[10] yielding $\gamma \sim 0.03\ldots 0.3$. Owing to the periodicity, a superlattice (SL) is formed, and the envelope function satisfies the Bloch theorem

$$\Psi(x,y+d) = \Psi(x,y)e^{ikd},$$

where k is the wave vector for motion along the y direction.

We consider the Kronig-Penney model where strain is a piecewise-constant periodic function of y. The wave functions within the regions with the fixed value of the strain $\mathcal{A}_x = \gamma_i\hbar/d$ can be cast as

$$\Psi_i(x,y) = e^{iqx}\left[C_i\begin{pmatrix}1\\ e^{i\varphi_i}\end{pmatrix}e^{ik_iy} + D_i\begin{pmatrix}1\\ e^{-i\varphi_i}\end{pmatrix}e^{-ik_iy}\right]. \tag{2}$$

Here q is the wave vector for free motion along the x direction, k_i are determined by the energy E via $k_i = \sqrt{(E/\hbar v_0)^2 - (q-\gamma_i/d)^2}$, and $\tan\varphi_i = \pm k_i/(q-\gamma_i/d)$, where the upper and lower signs should be taken for conduction and valence-band states, respectively. The dispersion equation has the form

$$\cos kd = (T_{11} + T_{22})/2, \tag{3}$$

where T is the transfer matrix. For the periodic structure with N stripes in each period it is equal to a product $T = T_{N-1}T_{N-2}\cdots T_1T_0$, where T_i is the transfer matrix for the ith strained graphene stripe of width l_i. It is defined as $\Psi_i(x,y+l_i) = T_i\Psi_i(x,y)$ and is given by

$$T_i = \frac{1}{\sin\varphi_i}\begin{bmatrix}\sin(\varphi_i - k_il_i) & \sin k_il_i\\ -\sin k_il_i & \sin(\varphi_i + k_il_i)\end{bmatrix}. \tag{4}$$

For a graphene SL with a period formed by two stripes, the dispersion equation has the form[14]

$$\cos kd = \cos k_1l_1\cos k_2l_2 + \sin k_1l_1\sin k_2l_2\frac{\cos\varphi_1\cos\varphi_2 - 1}{\sin\varphi_1\sin\varphi_2}. \tag{5}$$

The energy spectrum represents a result of folding of the Dirac cone within the one-dimensional Brillouin zone of the SL. The cone is shifted from $q = 0$ by a value $q_0 = (\gamma_1l_1 + \gamma_2l_2)/d^2$. The spectrum is given by conduction minibands $E_n(k,q)$ and

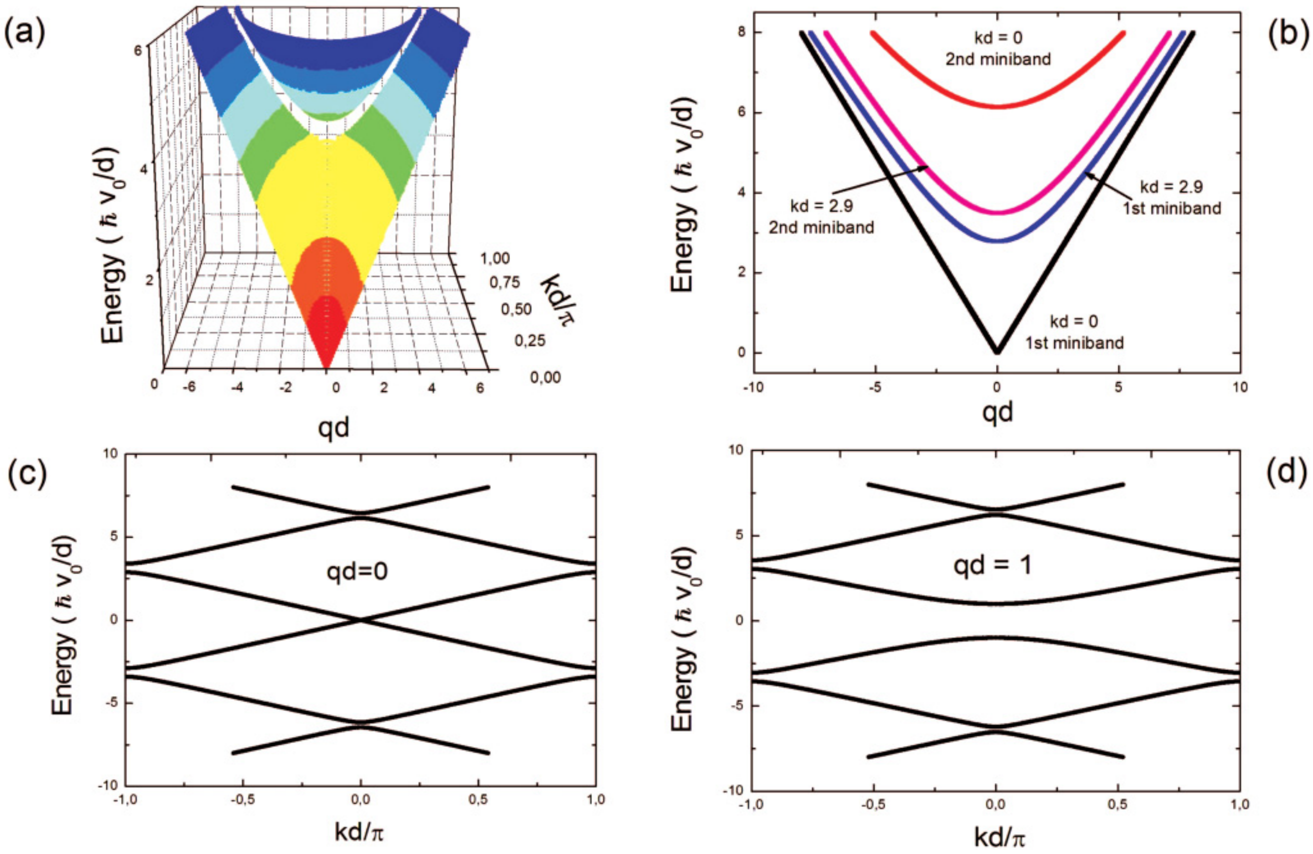

FIG. 1. (Color online) Energy spectrum in a SL with a period formed by two stripes with widths $l_1 = 0.3d$, $l_2 = 0.7d$, and the strain amplitudes $\gamma_1 = 0.7$, $\gamma_2 = -0.3$. Three-dimensional plot of the spectrum (a), energy dispersion as a function of q for four selected values of k (b), dispersion along k for $q = 0$ (c), and $q = 1/d$ (d).

valence minibands with the inverted dispersion $-E_n(k,q)$ with $n = 0,1,2\ldots$. The conduction- and valence-band cones touch each other at $q = q_0$, $k = 0$, i.e., the zero-energy state is degenerate. At the points $k = 0, \pm\pi/d$, the gaps between the minibands are opened. At small deformation $|\gamma_1 - \gamma_2| \ll 1$, the gaps are given by

$$E_{n+1} - E_n = \frac{2|\gamma_1 - \gamma_2|\hbar v_0}{n\pi d}\left|\sin\frac{n\pi l_1}{d}\right|. \tag{6}$$

At arbitrary values of the strain in the stripes, γ_i, the energy spectrum is found from numerical solution of the dispersion equation (3). Figure 1 presents the energy spectrum in the SL with the stripe widths $l_1 = 0.3d$, $l_2 = 0.7d$, and the strain amplitudes $\gamma_1 = 0.7$, $\gamma_2 = -0.3$. One can see folding of the energy spectrum at the boundaries of the Brillouin zone of the SL ($k = \pm\pi/d$), and the conical dispersion in the direction perpendicular to the strain. Figure 1 demonstrates that even the large strain with $\gamma \sim 1$ results just in small deviations from the pure conical dispersion which occur in close proximity of the anticrossing points. Therefore the energy spectrum and the electronic states can be described in the nearly-free-electron model where the strain is considered as a small perturbation. In this approach, the unperturbed energy spectrum in the nth conduction miniband is simply given by

$$E_n^{(0)}(k,q) = \hbar v_0\sqrt{q^2 + k_n^2}, \tag{7}$$

where $k_n = k - (2\pi n/d)\text{sign}\{k\}$. In the following for calculation of absorption in periodically strained graphene we will use the perturbation in the form

$$U(y) = \frac{\gamma\hbar v_0}{d}\cos\left(\frac{2\pi y}{d}\right)\sigma_x$$

assuming $\gamma \ll 1$. The energies and the wave functions in the nth miniband are given by

$$E_n = E_n^{(0)} + \sum_{m\neq n}\frac{|U_{mn}|^2}{E_n^{(0)} - E_m^{(0)}}, \tag{8}$$

$$\Psi_n = \psi_n^{(0)} + \sum_{m\neq n}\frac{U_{mn}}{E_n^{(0)} - E_m^{(0)}}\psi_m^{(0)}. \tag{9}$$

Here $\psi_n^{(0)}$ are unperturbed envelopes

$$\psi_n^{(0)} = \frac{1}{\sqrt{2}}\begin{pmatrix}1\\ \pm e^{i\varphi_n}\end{pmatrix}e^{iqx+ik_ny},$$

where the upper and lower signs are taken for the conduction and valence minibands, and $\tan\varphi_n = k_n/q$.

III. DICHROIC ABSORPTION IN GRAPHENE SUPERLATTICES

The Hamiltonian of electron-photon interaction in graphene reads

$$H_{e-\text{phot}} = -\frac{ev_0}{c}\boldsymbol{\sigma}\cdot\boldsymbol{A}, \tag{10}$$

where $\boldsymbol{A}$ is the light-wave vector potential. This yields a general expression for absorbance under direct optical transitions from an initial (i) to the final (f) miniband:

$$\eta_{fi} = 4\frac{(2\pi e v_0)^2}{c\omega}\sum_{kq}|\boldsymbol{e}\cdot\boldsymbol{\sigma}_{fi}|^2\delta[E_f(k,q)-E_i(k,q)-\hbar\omega]. \tag{11}$$

Here the factor 4 accounts for the spin and valley degeneracies, ω is the light frequency, and $\boldsymbol{e}$ is the light polarization vector.

In the following section we calculate the absorbance for interband transitions in undoped graphene SLs and for transitions between the ground and the first excited conduction minibands for doped SLs.

A. Interband absorption in undoped SLs

We consider light absorption due to interband transitions between the ground valence and conduction minibands which have opposite dispersions: $E_{v0}(k,q) = -E_{c0}(k,q)$. In the absence of strain, the interband absorbance has a universal value $\eta = \pi e^2/(\hbar c)$. For normally incident light, the D_{2h} point group of the symmetrically strained graphene sheet implies the following polarization dependence of the absorbance:

$$\eta_{c0,v0}(\omega) = \frac{\pi e^2}{\hbar c}\{1-\gamma^2[\Phi_0(\omega)+\Phi_1(\omega)\cos 2\alpha]\}. \tag{12}$$

Here α is the angle between the light polarization vector $\boldsymbol{e}$ and the x axis. We take into account in Eq. (9) only the admixture to the $c0$ and $v0$ states from the closest minibands $c1$ and $v1$, respectively. Taking into account the strain both in the energies Eq. (8) and in the optical matrix elements, we obtain the absorbance in the form of Eq. (12) with

$$\Phi_0(\omega) = \frac{\zeta^2}{2\pi^3}\int_0^{\pi/2} d\varphi\left[\frac{4P(4\zeta\sin\varphi-1)}{r(r-1)}+\zeta\frac{\partial P}{\partial\zeta}\right], \tag{13}$$

$$\Phi_1(\omega) = \frac{\zeta^2}{2\pi^3}\int_0^{\pi/2} d\varphi\left(\frac{4P}{r-1}-\zeta\frac{\partial P}{\partial\zeta}\cos 2\varphi\right). \tag{14}$$

Here $r(\varphi) = \sqrt{1+16\zeta^2-8\zeta\sin\varphi}$,

$$P = \frac{1}{r-1}\left(1+\frac{\cos 2\varphi+4\zeta\sin\varphi}{r}\right), \quad \zeta = \frac{\pi v_0}{\omega d}. \tag{15}$$

Spectral dependences of polarization-independent and polarization-dependent parts of the absorbance, $\Phi_0(\omega)$ and $\Phi_1(\omega)$, are presented in Fig. 2. One can see that the values of both parts have an order of $10^{-2}\gamma^2$. This means that, in samples with a large enough strain $\gamma \sim 1$, such modulation of absorbance can be detected experimentally.

Both absorption parts, $\Phi_0(\omega)$ and $\Phi_1(\omega)$, should tend to constants at frequencies $\omega = 0$ and $\omega = \pi v_0/d$ due to anticrossing of the energy dispersions. We do not obtain this saturation due to the oversimplified approach, Eqs. (8). However, the calculated spectra are correct everywhere beyond the narrow spectral regions $\Delta\omega \sim \gamma\pi v_0/d \ll 1$ near these two points.

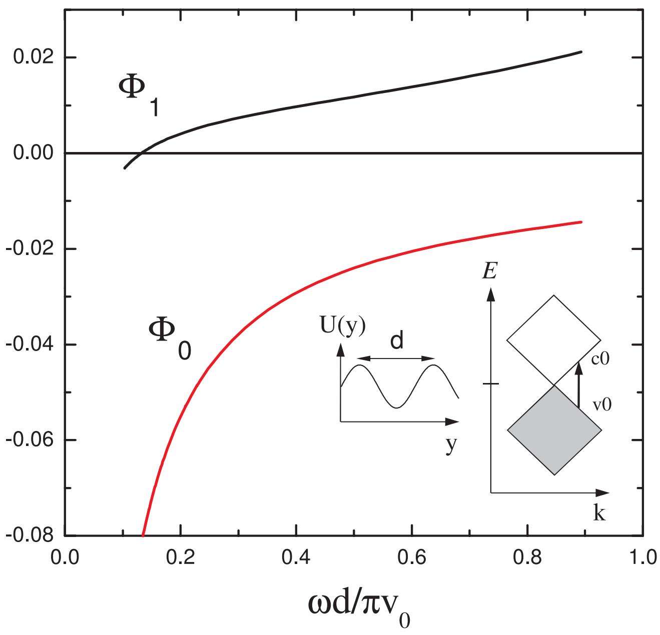

FIG. 2. (Color online) Polarization-independent (Φ_0) and polarization-dependent (Φ_1) absorbance spectra under interband transitions in the symmetrical undoped SL. Inset shows the perturbation $U(y)$ caused by periodical strain and the scheme of optical transitions.

B. Interminiband absorption in doped SLs

Let us consider absorption caused by transitions between the ground ($c0$) and the excited ($c1$) conduction minibands. The polarization dependence of the absorbance $\eta_{c1,c0}$ can be conveniently written as

$$\eta_{c1,c0} = \eta_x|e_x|^2 + \eta_y|e_y|^2, \tag{16}$$

where e_x and e_y are the light polarization vector components perpendicular and along the strain direction, respectively.

The absorbance is given by

$$\eta_{x,y} = 4\frac{(2\pi e v_0)^2}{c\omega}\sum_{kq}|\langle c1|\sigma_{x,y}|c0\rangle|^2(f_{c0}-f_{c1}) \times\delta\big[E_1^{(0)}(k,q)-E_0^{(0)}(k,q)-\hbar\omega\big], \tag{17}$$

where f_{c0} and f_{c1} are the occupations of the initial and final states. Here we set the unperturbed energies, Eq. (7), because the matrix element of interminiband transitions already contains γ. The calculation yields

$$\eta_{x,y} = \gamma^2\frac{4e^2\zeta^2}{\pi^2\hbar c}\int_0^{\pi/2} d\varphi(f_{c0}-f_{c1})\sin^2\left(\frac{\varphi-\varphi'}{2}\right) \times\frac{s(s+1)}{2\zeta\sin\varphi+1}G_{x,y}(\varphi), \tag{18}$$

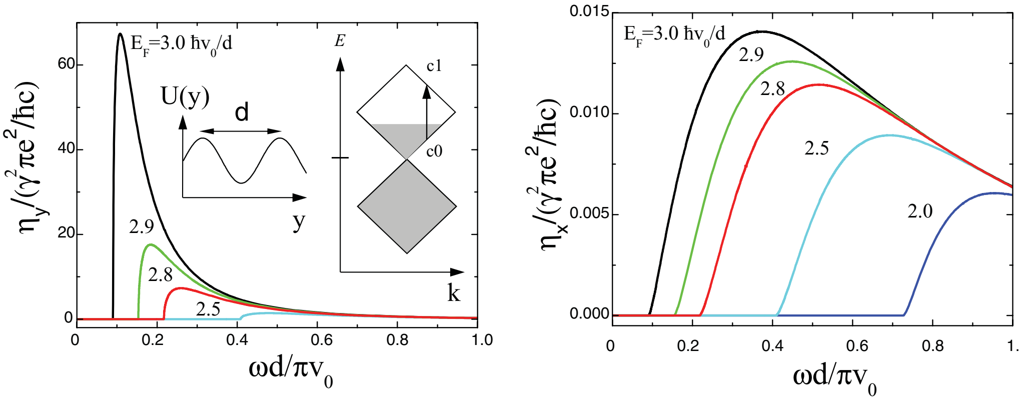

FIG. 3. (Color online) Absorbance under interminiband transitions in the doped SL. Light polarization is parallel (left panel) and perpendicular (right panel) to the strain modulation direction. Inset illustrates the scheme of direct optical transitions.

where $s(\varphi) = (2\zeta^2 - 1/2)/(1 + 2\zeta \sin\varphi)$, ζ is given by Eq. (15),

$$G_x = \sin^2(\varphi + \varphi'), \quad G_y = 4\cos^4\left(\frac{\varphi + \varphi'}{2}\right),$$

and the angle φ' is defined as follows:

$$\cos\varphi' = \frac{\cos\varphi(s+1)}{s}, \quad \sin\varphi' = \frac{s\sin\varphi - 2\zeta}{s+1}.$$

At low temperatures electrons are degenerate, and the light absorption under direct interminiband transitions is possible at $E_1 > E_F > E_0$ only, where E_F is the Fermi energy (see inset of Fig. 3). Therefore the occupation factor is given by $(f_{c0} - f_{c1}) = \theta(E_F/\hbar\omega - s)\theta(s + 1 - E_F/\hbar\omega)$, where $\theta(x)$ is the Heaviside function.

The absorbance spectra $\eta_y(\omega)$ and $\eta_x(\omega)$ are presented in Fig. 3. One can see large dichroism of interminiband absorption: $\eta_x \ll \eta_y$. This is explained by the different characters of electron motion parallel and perpendicular to the strain direction y. The light polarized along x can be weakly absorbed because of ballistic electron propagation in this direction. On the other hand, the interfaces between strained and unstrained stripes play the role of coherent scatterers, allowing for absorption of the y-polarized radiation. High absorption dichroism demonstrated here is similar to the result of Ref. 15 where grids of graphene nanoribbons were studied. In contrast, we show that even pure graphene samples can have highly dichroic absorption if they are periodically strained.

The absorbance η_x is nonzero in graphene SLs in contrast to conventional semiconductor SLs. The difference is in the electron-photon interaction. According to Eq. (10), the corresponding Hamiltonian in graphene contains the operator σ_x, while in semiconductor SLs $H_{e-\mathrm{phot}}$ is proportional to the unit operator with zero matrix elements between the states in different minibands.

Let us compare $\eta_{x,y}$ calculated above with the Drude absorbance accompanied by scattering with the characteristic time τ, $\eta_D \sim (E_F/\hbar\omega^2\tau)(e^2/\hbar c)$. For realistic parameters $E_F \sim \hbar\omega \sim 0.1$ eV, $\tau \sim 1$ ps we have $\eta_D \sim 10^{-2}e^2/(\hbar c)$, which is about an order of magnitude smaller than η_y and comparable with η_x at $\gamma \sim 1$. This demonstrates that the absorption dichroism caused by the periodic strain is observable in real samples where the Drude absorption is also present.

IV. CIRCULAR RATCHET PHOTOCURRENT

Consider a graphene SL formed by asymmetrical strain lacking the reflection operation $y \to -y$ (see insets of Fig. 4). This system has C_{2v} symmetry with the C_2 axis along the y direction. Symmetry considerations imply that absorption of normally-incident circularly polarized light is accompanied by generation of a photocurrent flowing perpendicular to the strain direction. This means that asymmetrical graphene SLs serve as quantum ratchets.

The phenomenological expression for the photocurrent density has the form

$$j_x = \chi I P_{\mathrm{circ}}, \tag{19}$$

where $P_{\mathrm{circ}} = i(e_x e_y^* - e_x^* e_y)$ is the light circular polarization degree. Microscopically, the density of the photocurrent generated under interminiband optical transitions in doped graphene is given by

$$j_x = 2e \sum_{kq,\nu} W_{c1,c0}^{(\nu)} \left(v_x^{(1)}\tau_1 - v_x^{(0)}\tau_0\right). \tag{20}$$

Here ν enumerates the two valleys, $W_{c1,c0}^{(\nu)}$ is the interminiband optical transition rate in the νth valley, $\hbar v_x^{(n)} = \partial E_n/\partial q$, and τ_n is the momentum relaxation time in the nth miniband. The expression for χ has the following form:

$$\begin{aligned}\chi = \frac{8\pi^2 e^3 v_0^2}{\omega^2\hbar c} \sum_{kq,\nu} \left(v_x^{(1)}\tau_1 - v_x^{(0)}\tau_0\right) \mathrm{Im}\left\{M_x^{(\nu)} M_y^{(\nu)*}\right\} \\ \times (f_{c0} - f_{c1})\,\delta[E_1(k,q) - E_0(k,q) - \hbar\omega],\end{aligned} \tag{21}$$

where $M_{x,y}^{(\nu)} = \langle \nu c1|\sigma_{x,y}|\nu c0\rangle$.

Numerical calculations performed according to Eq. (21) for various asymmetrical SLs show that under the change of the strain in all stripes $\gamma_i \to a\gamma_i$ the circular photocurrent scales as

$$\chi \to a^4 \chi. \tag{22}$$

This result can be confirmed by the analysis in the free-electron approximation with a noncentrosymmetric periodic

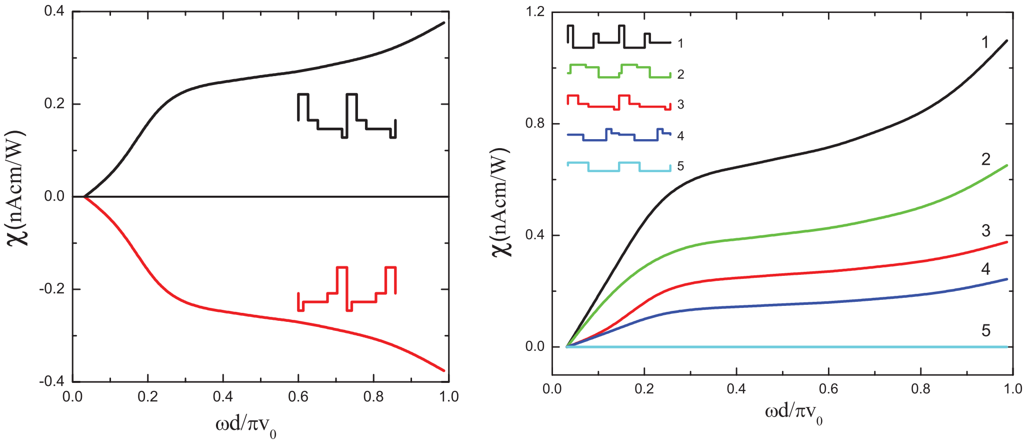

FIG. 4. (Color online) Circular-polarization-sensitive photocurrent in asymmetrical SLs. Insets show strain profiles for different SLs. The difference between the highest and lowest strain in the stripes, $|\gamma_{\max} - \gamma_{\min}| = 0.8,\ 0.45,\ 0.5,\ 0.4$, and 0.3 for the structures $1\cdots5$, respectively. The upper SL in the left panel coincides with structure 3 in the right panel. The period $d = 10$ nm, the relaxation time in the $c0$ miniband $\tau_0 = 1$ ps, and the Fermi energy $E_{\rm F} = 3\hbar v_0/d$.

perturbation. For this purpose, let us consider the perturbation of the form

$$\mathcal{A}_x = \frac{\hbar}{d}\left[\gamma_1 \cos\frac{2\pi y}{d} + \gamma_2 \cos\left(\frac{4\pi y}{d} + \beta\right) + \gamma_4 \cos\frac{8\pi y}{d}\right],$$

where $\beta \neq 0,\pi$. It turns out that, for such perturbation, nonvanishing circular photocurrent is obtained only if all three amplitudes $\gamma_1,\gamma_2,\gamma_4$ are nonzero. Indeed, for direct optical transitions between $c0$ and $c1$ minibands, the transferred momentum $2\pi/d$ is taken up by the periodic strain, so the matrix elements $M_{x,y}^{(\nu)} \propto \gamma_1$. In order to take into account the SL asymmetry, one should consider the γ_2-related contribution. However, the momentum transfer is twice larger in this case, and the correction to the matrix element can be $\delta M_{x,y}^{(\nu)} \propto \gamma_1\gamma_2$ only. The interference of these two corrections yield the $\gamma_1^2\gamma_2$ contribution to $\mathrm{Im}\{M_x^{(\nu)} M_y^{(\nu)*}\}$ which results in no electric current after summation over two valleys since they differ by signs of γ's. Moreover, such γ-odd photocurrent in each valley would mean the valley current[16] at circularly polarized excitation, which is forbidden by symmetry. Therefore such a contribution nullifies already after summation over q and k.

The only possibility to get a nonzero photocurrent is the fourth order in the strain. It can be obtained from interference of the first-order term $M_{x,y}^{(\nu)} \propto \gamma_1$ with a third-order contribution. However, it is impossible to construct the correction to the $c0 \to c1$ transition matrix element, which is $\propto \gamma_1^2\gamma_2$. The fourth order in strain probability of the optical transition is obtained as a product of $M_{x,y}^{(\nu)}$ and $\delta M_{x,y}^{(\nu)} \propto \gamma_1\gamma_2\gamma_4$. This yields the circular ratchet current, Eq. (19), with $\chi \sim \gamma_1^2\gamma_2\gamma_4$. This dependence coincides with the numerical result, Eq. (22).

Figure 4 presents results of numerical calculation of the circular ratchet photocurrent for various asymmetrical SLs. We assume that the momentum relaxation time in the first miniband is much shorter than in the ground miniband, $\tau_1 \ll \tau_0$, and disregard the corresponding contribution to the photocurrent. In calculation we take the period $d = 10$ nm, the relaxation time $\tau_0 = 1$ ps, and the Fermi energy $E_{\rm F} = 3\hbar v_0/d$.

The left panel of Fig. 4 represents the circular ratchet current for two strains with opposite asymmetry, i.e., for two SLs transforming one to another by a reflection $y \to -y$. The calculations demonstrate that the circular photocurrents are directed oppositely and have the same magnitude. The right panel of Fig. 4 shows the circular photocurrents for different strained SLs. One can see a larger photocurrent for SLs with a higher degree of asymmetry. For the symmetrical SL (structure 5) the circular photocurrent is zero.

V. CONCLUSION

High dichroic absorption in graphene SLs demonstrated above can be used in polarizers operating in infrared and terahertz ranges. Asymmetrical strain is present if graphene is placed on a substrate with surface asymmetry. We show that the strain asymmetry can be probed by photogalvanic measurements. The magnitude of the circular photocurrent is $\chi \approx 1$ nA cm/W at typical photon energy $\hbar\omega \sim \hbar v_0/d \sim 60$ meV for the period $d = 10$ nm. These estimates are close to the experimental conditions realized in Refs. 12 and 13, which allows one to expect observation of the considered effects in experiments.

To summarize, graphene SLs formed by periodic strain are considered theoretically. We show that the electron energy spectrum consists of minibands obtained by folding of the cone at the boundaries of the SL Brillouin zone with very narrow anticrossing regions. Light absorption under direct interminiband transitions is shown to be frequency dependent and dichroic. Giant dichroism of absorption is demonstrated for doped graphene SLs. Asymmetrical graphene SLs are shown to act as quantum ratchets allowing for generation of photocurrent at absorption of normally incident light. The helicity-dependent photocurrent spectrum is calculated for doped SLs with various asymmetries.

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