Multiphoton intersubband transitions in an armchair graphene nanoribbon

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Abstract. We present an analytical approach to the problem of the interband transitions in an armchair graphene nanoribbon (AGNR), exposed to the time-periodic electric field of strong light wave, polarized parallel to the ribbon axis. The two-dimensional Dirac equation for the massless electron subject to the ribbon confinement is employed. In the resonant approximation the probability of the transitions between the valence and conduction size-quantized subbands are calculated in an explicit form. We trace the dependencies of the Rabi frequency for these transitions on the ribbon width and electric field strength for both the multiphoton-assisted and tunneling regimes relevant to the fast oscillating and practically constant electric field, respectively. Estimates of the expected experimental values for the typically employed AGNR and laser technique facilities show that the Rabi oscillations can be observed under laboratory conditions. The data, corresponding to the intersubband tunneling, makes the AGNR a 1D condensed matter analog, in which the quantum electrodynamic vacuum decay can be detected by the employment of the attainable electric fields.

This work relates to a subject of interest in the mid of the last century, when the decay of the quantum electrodynamic (QED) vacuum in the presence of a classical static electric field $F$ has been comprehensively studied [1]. This process, accompanied by the electron-positron pair production (pp), implies the condition $F \geq F_c^{(v)}$ with the breakdown electric field $F_c^{(v)} \sim 10^{18}$ V/m. Clearly, the latter static field remains unattainable for modern experimental techniques [2]. In the presence of a time-dependent periodic electric field with the magnitude $F_0$ and frequency $\omega$ the tunneling mechanism of the pp, associated to a dc field, is replaced by the photon assisted one [3]. However, even for the extreme fields $F_0 \approx F_c^{(v)}$ this mechanism becomes ineffective for optical frequencies, which are significantly smaller than that of gamma rays [4].

The emergence of graphene, being ideal 2D crystal of carbon atoms, and arranged in a honeycomb lattice [5], allows us to consider graphene as the condensed matter counterpart for the QED vacuum. The reason for this is the ultra-relativistic [6] and relativistic dispersion laws for the gapless and gapped graphene, respectively. Theoretical estimates show that for the gapped graphene layer with the realistic bandgap of about 0.3 eV [7] the critical electric

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field $F^{(g)} \sim 10^8$ V/m can possibly be achieved under laboratory conditions. This makes gapped graphene a suitable environment for the experimental test of the pp in the electrically biased vacuum. From the latter standpoint, armchair graphene nanoribbon (AGNR)-quasi-1D graphene strips, possess in some sense an advantage over 2D graphene layer. In order to open the bandgap in pristine gapless graphene some material processing is needed, that in turn alters the original electronic properties of graphene. The size of this bandgap is then characteristic of a specific sample. In contrast to 2D graphene, in the graphene nanoribbon the bandgap is defined not by the material processing step, but only by the ribbon width. At the same time the basic material properties of graphene, constituting the nanoribbon, remain unchanged. If necessary the bandgap energy can be measured optically. In the vicinity of the bandgap the optical absorption in the quasi-1D AGNR demonstrates the easily detectable spectral singularity in contrast to the 2D graphene, manifesting the weakly pronounced step-like feature. Since analytical studies of the dynamically assisted effects in AGNR are not widely addressed in the literature yet, we determine in the present work an explicit form for the probability of the interband transitions in the AGNR, exposed to the periodic electric field of a strong light wave. The dependencies of the Rabi frequency for these transitions on the ribbon width and the electric field strength are analyzed.

We consider the AGNR with width $d$ placed on the $x - y$ plane and bounded by the lines $x = \pm d/2$. The polarization of the electric field $F(t) = F_0 \cos \omega t$ with the magnitude $F_0$ and frequency $\omega$ is chosen to be parallel to the ribbon $y$-axis. The energy spectrum of a free electron in semiconductor-like gapped AGNR is a sequence of 1D subbands, marked by indices $N_j = 0, \pm 1, \pm 2, \ldots$, $j = e, h$ for the electron ($e$) and hole ($h$) bands, respectively. Since the optical transitions, induced by the $y$-polarized electric field, are allowed between the subbands with equal indices $N_e = N_h = N$ [8], these subbands read [9]

$$\pm E_N(k) = \sqrt{\epsilon_N^2 + \hbar^2 v_F^2 k^2}; \quad \epsilon_N = \left| N - \frac{1}{3} \frac{\pi \hbar v_F}{d} \right|; \quad N = 0, \pm 1, \pm 2, \ldots, \quad (1)$$

where $\epsilon_N$ and $\hbar v_F k$ are the size-quantized energies and longitudinal momentum, respectively, $v_F = 10^6$ m/s is the graphene Fermi velocity.

The equation, describing the electron at a position $\vec{r}(x, y)$ subject to an external time-dependent electric field, possesses the form of a Dirac equation

$$\hat{H} \tilde{\Psi}(\vec{r}, t) = i\hbar \frac{\partial \tilde{\Psi}(\vec{r}, t)}{\partial t}, \quad (2)$$

where the Hamiltonian

$$\hat{H}(\vec{k}; y, t) = \hat{H}_x(\hat{k}_x) + \hat{H}_y(\hat{k}_y) + \hat{1}(-eyF(t)); \quad \vec{k} = -i\vec{\nabla} \quad (3)$$

is formed by the Hamiltonians $\hat{H}_\alpha(\hat{k}_\alpha)$ $\alpha = x, y$ [9]

$$\hat{H}_\alpha(\hat{k}_\alpha) = \hbar v_F \left( \begin{array}{cc} -\sigma_\alpha \hat{k}_\alpha & 0 \\ 0 & \sigma^*_\alpha \hat{k}_\alpha \end{array} \right)$$

and the electric field potential $-eyF(t)$. The matrices $\hat{1}$ and $\vec{\sigma}$ are the unit and Pauli matrices, respectively.

Further, we choose the wave function $\tilde{\Psi}$, attributed to the $N$ subband, in the form

$$\tilde{\Psi}_N(\vec{r}, t) = \frac{1}{\sqrt{2}} \left[ u_A(y, t) \Phi_{NA}(x) + u_B(y, t) \Phi_{NB}(x) \right], \quad (4)$$
where $\Phi_{N\alpha(B)}$ are the fourcomponent wave functions, describing the confined $x$-states, corresponding to the A(B) superlattices (see Ref. [10] for their explicit forms and properties).

Transformation

$$u(y, t) = \frac{1}{\sqrt{2}} (\delta_x + \delta_y) \hat{U} + \exp[iq(t)y]q(t); \quad q(t) = \frac{eF_0}{\hbar} \sin \omega t + k;$$

$$\eta(t) = \begin{bmatrix} f_1(t) \exp \left[-i \int_0^t \Omega_N(\tau) d\tau \right] \\ f_2(t) \exp \left[i \int_0^t \Omega_N(\tau) d\tau \right] \end{bmatrix}; \quad \tilde{u} = \begin{bmatrix} u_A \\ u_B \end{bmatrix} \quad (5)$$

and a subsequent substitution of the wave function $\tilde{\Psi}$ (4) into eq. (2) results in the set of equations for the functions $f_{1,2}$

$$i\dot{f}_{1,2}(t) = -R_N(t) \exp \left[ \pm 2i \int_0^t \Omega_N(\tau) d\tau \right] f_{2,1}(t), \quad (6)$$

where

$$R_N(t) = \frac{\omega_N v_F e F_0 \sin \omega t}{2\hbar \Omega_N^2(t)}; \quad \Omega_N^2(t) = \omega^2_N + v_F q^2(t); \quad \Omega_N = \frac{\varepsilon_N}{h}.$$

Transforming $\tilde{u} \rightarrow \eta$ (5), involving the operator $\hat{U} + = [(\Omega_N + \omega N)1 - i\partial_s] [2\Omega_N(\Omega_N + \omega N)]^{-\frac{1}{2}}$, is analogous to the Foldy-Wouthuysen one [11]. For $F(t) = 0$ this processing separates completely the intrasubband electron and hole states, corresponding to the energies $\pm E_N(k)$ (1).

In our case the electric field term $\sim F(t)$ in the Hamiltonian (3) is split into two components $\sim \Omega_N(t)$ and $\sim R_N(t)$, governing the electron and hole intrasubband states and transitions between them, respectively.

Using the time-periodicity of the electric field, we present the exponential factors in eqs. (6) in the form $\exp \left( \pm \frac{i}{\hbar} \varepsilon_N(t) S_N(t) \right)$, $S_N(t) = S_N(t + \frac{2\pi}{\omega})$, where

$$\varepsilon_N(k) = \frac{h \omega}{\pi} \int_{-\frac{\pi}{2}}^{+\frac{\pi}{2}} \Omega_N(\tau) dt = \frac{2}{\pi} \Delta_N \frac{1}{s_N} E \sqrt{1 - s_N^2} \left( 1 + s_N^2 \frac{2h^2 v_F^2 k^2}{\Delta_N^2} \right); \quad s_N^2 = (1 + \gamma_N^2)^{-1} \quad (7)$$

is the transition energy, i.e. the quasienergy of the electron-hole pair. In this equation $\Delta_N = 2\gamma_N$, $\gamma_N = \omega \Delta_N(2e v_F F_0)^{-1}$ is the Keldysh parameter and $E(z)$ is the complete elliptic integral of the second kind [12].

Expansion of the periodic function $R_N(t)S_N(t)$ in the Fourier series

$$R_N(t)S_N(t) = \sum_{l=-\infty}^{+\infty} A_l e^{-i\omega lt}; \quad A_l(\omega) = \frac{\omega}{2\pi} \int_{-\frac{\pi}{2}}^{+\frac{\pi}{2}} R_N(t)S_N(t) e^{i\omega lt} dt \quad (8)$$

allows us to solve the set of eqs. (6) in the resonant approximation $\omega_l \ll \omega$, $\omega_l = \varepsilon_N/h - l \omega$.

Averaging the eqs. (6) over the period $2\pi/\omega$ generates the set of equations for the functions $\bar{f}_{1,2}(t)$, averaged in the vicinity of the instant time $t$

$$\ddot{\bar{f}}_1 = -i\bar{f}_2 A_1 e^{i\omega t}; \quad \ddot{\bar{f}}_2 = -i\bar{f}_1 A_1^* e^{-i\omega t}. \quad (9)$$

This set of equations coincides with that for the two-level problem with the well-known solutions [13]. Under the initial conditions $\bar{f}_1(0) = 0$, $\bar{f}_2(0) = 1$ and resonant conditions the probability of the $N$ intersubband transitions $w_N(k) = |\bar{f}_1(t)|^2$ acquires the time-oscillating form
Figure 1. The Rabi frequency $\Omega^{(R)}_{03}(0)$ versus the ribbon width $d$ and electric field $F_0$. The frequency $\Omega^{(R)}_{03}(0)$ is calculated from eq. (10), relevant to the three-photon $l = 3$ oscillations between the ground $N = 0$ subbands.

$$w_{NI} = \sin^2 \left( \frac{1}{2} \Omega^{(R)}_{NI}(k)t \right)$$

with the Rabi frequency $\Omega^{(R)}_{NI}(k) = 2|A_l(k)|$. For the momentum $k = 0$, corresponding to a maximum of the Rabi frequency, we derive from eqs. (6) and (8)

$$\Omega^{(R)}_{NI}(0) = \frac{2}{3} \omega \exp \left\{ -\frac{l}{s_N} [K(s_N) - E(s_N)] \right\} \sin^2 \frac{l\pi}{2}.$$  \hspace{1cm} (10)

In this equation $K(z)$ is the complete elliptic integral of the first kind [12]; the resonant condition $l\hbar\omega = \delta_N(0)$ (see eq. (7)) is implied. For the limiting cases of the tunnelling $\gamma_N \ll 1$ and multiphoton-assisted $\gamma_N \gg 1$ transition regimes eq. (10) reads

$$\Omega^{(R)}_{NI}(0) = \frac{2}{3} \omega \left\{ \begin{array}{ll}
\frac{1}{3} \exp \left( -\frac{\pi F_c^{(N)}}{2F_0} \right) ; & \gamma_N \ll 1 \\
\exp \left( l \left( 4\gamma_N \right)^{-1} \sin^2 \frac{l\pi}{2} \right) ; & \gamma_N \gg 1,
\end{array} \right.$$ \hspace{1cm} (11)

where $F_c^{(N)} = \Delta^2_N/8\hbar v_F e$ is the breakdown electric field, providing the significant intersubband tunnelling for the fields $F_0 \geq F_c^{(N)}$.

Eqs. (10), (11) show that the multiphoton transitions between the subband extremes are allowed only for the odd photon numbers $l$. With increasing the electric field magnitude $F_0$, driving frequency $\omega$, and ribbon width $d$ the Rabi frequency $\Omega^{(R)}_{NI}(0)$ increases for any regime, remaining much smaller than $\omega$. The dependence on the width is more pronounced as compared to that on the electric field. The Rabi frequency as a function of the ribbon width and electric field strength is depicted in Fig. 1. Noteworthy the exponential dependencies of the Rabi frequency in eq. (11) for the tunneling regime and vacuum decay probability [1] on the electric field $F_0$ are similar. This allows us to treat the Fermi sea for a graphene nanoribbon as a condensed matter analog of a Dirac sea in a QED vacuum.

In an effort to estimate the expected experimental values we choose the AGNR with width $d = 2$ nm, exposed to the light wave with the electric field magnitude $F_0 = 360$ kV/cm and frequency $\omega = 3.3 \cdot 10^{14}$ s$^{-1}$ ($\lambda = 5.4$ $\mu$m), generating the three-photon ($l = 3$) transitions between the ground ($N = 0$) subbands in the photon-assisted ($\gamma_0 = 3$) regime. This results in the Rabi frequency $\Omega^{(R)}_{03} = 2.54 \cdot 10^{12}$ s$^{-1}$ and frequency relationship $\Omega^{(R)}_{03}/\omega = 7.7 \cdot 10^{-3}$. 

In Figure 1, the Rabi frequency $\Omega^{(R)}_{03}(0)$ is plotted against the ribbon width $d$ and electric field $F_0$. The frequency $\Omega^{(R)}_{03}(0)$ is calculated from eq. (10), relevant to the three-photon $l = 3$ oscillations between the ground $N = 0$ subbands. The figure illustrates the exponential dependence of the Rabi frequency on the electrical field, with the Rabi frequency increasing for any regime. The dependence on the width is more pronounced compared to that on the electric field. The Rabi frequency as a function of the ribbon width and electric field strength is depicted in Figure 1.
The breakdown electric field $F_{c}^{(0)}$, limiting the significant intersubband tunnelling by the fields $F_{0} \geq F_{c}^{(0)}$, is $F_{c}^{(0)} = 1.68 \cdot 10^{3}$ kV/cm. Note that the vacuum related breakdown electric field $F_{c}^{(v)}$ exceeds the given above by a factor of $10^{10}$. The presented results show that the observation of the Rabi oscillations in AGNR is quite feasible under laboratory conditions.

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