Journal of Luminescence 129 (2009) 1779–1781

Contents lists available at ScienceDirect

Journal of Luminescence

journal homepage: www.elsevier.com/locate/jlumin

DNA-based tunable THz oscillator

A.V. Malyshev [a,b,c,*], V.A. Malyshev [a], F. Domínguez-Adame [b]

[a] Centre for Theoretical Physics and Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands
[b] GISC, Departamento de Física de Materiales, Universidad Complutense, E-28040 Madrid, Spain
[c] Ioffe Physico-Technical Institute, 26 Politechnicheskaya str., 194021 St. Petersburg, Russia

ARTICLE INFO

Available online 2 April 2009

Keywords:
Nano-electronic devices
Field effect devices
DNA

ABSTRACT

The intrinsic helix conformation of the DNA strands is known to be the key ingredient of control of the electric current through the molecule by the perpendicular (gate) electric field. We show theoretically that Bloch oscillations in periodic systems with helical conformation are also strongly affected by such lateral field; the oscillation frequency splits into a manifold of several generally non-commensurate frequencies leading to a complicated pattern of the charge motion. For model parameters typical for the DNA the frequency of such oscillations falls in the THz domain, suggesting a possibility to design a DNA-based nano-scale source of THz radiation.

1. Introduction

The helical symmetry of the DNA is almost always neglected when modeling the charge transport through the DNA-based devices: a DNA is usually considered as a flat ladder-like sequence (periodic or stochastic) of base pairs [1–5]. Very recently the intrinsic helix conformation of the DNA strands was put forward as the key ingredient allowing for control of the electric current through the molecule by the electric field perpendicular to the DNA axis [6]. In the presence of such gate field, the helical conformation leads to an additional periodic modulation of the base pair energies, which can suppress the charge transport through the DNA. On this basis, prototypes of the single-DNA-based field effect transistor and the analogue of the Esaki diode were proposed [6].

In this contribution, we aim to further exploit the symmetry of periodic DNA molecules (such as the ploy(G)-poly(C)) and demonstrate, by means of numerical simulations in the framework of the tight-binding approximation, that (i) such systems subjected to a collinear uniform electric field can exhibit Bloch oscillations [8] and (ii) because of the helical symmetry, in the presence of the perpendicular (gate) electric field the oscillations are characterized by several frequencies rather than a single one. For model parameters typical for the DNA these frequencies fall in the THz domain, the region which is in the focus of an intense research nowadays (see Ref. [7] for a recent overview). The above-mentioned property suggests therefore that periodic DNA could be a candidate for tunable THz oscillator.

2. Model

Bloch [9] and Zener [10] argued on the theoretical grounds that a charge, moving in periodic potential and subjected to a uniform electric field E, is confined within a finite region because of the Bragg reflection. Due to the confinement, it undergoes a periodic motion which is characterized by the angular frequency $\omega_B = eEa/\hbar$ and a spatial extension $L_B = W/(eE)$, where $-e$ is the electron charge, F is the applied electric field, a is the lattice constant, and W stands for the band width (see also Ref. [11]). Here, we study such oscillations in systems with helical symmetry (bearing in mind the periodic DNA) subjected to a uniform electric field which is not collinear with the helix axis.

Charge transport in the DNA is often described in the framework of the ladder model in the tight-binding approximation [1–6]. Although very rough, the ladder model (accounting for two *linear* strands) grasps important qualitative features, such as the existence of the gap in the I–V characteristics. For the sake of simplicity, we consider only one strand (which corresponds to one band or one channel approximation) but take into account its helical symmetry. For such single helix we assume the conformation parameters of the double-stranded DNA (dsDNA) in its B form, in particular, the full-twist period of 10 base molecules. We also extend the model by including a uniform electric field **E** that is tilted by the angle θ with respect to the axis of the helix. The field has therefore both the collinear component, $E\cos\theta$, and the lateral one (the gate), $E\sin\theta$. In the case of the double helix, the picture of Bloch oscillations is somewhat more complicated as

* Corresponding author at: Centre for Theoretical Physics and Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands. Tel.: +31 50 3634784; fax: +31 50 3634947.
E-mail addresses: a.v.malyshev@rug.nl, a.malyshev@fis.ucm.es (A.V. Malyshev).

0022-2313/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.jlumin.2009.01.023

compared to the present case, the physics however is very similar. The Hamiltonian of our model in site representation reads

$$H = \sum_{n=1}^{N} \varepsilon_n |n\rangle\langle n| + J \sum_{n=1}^{N-1} |n+1\rangle\langle n| + h.c.. \quad (1)$$

Here, $|n\rangle$ is the state vector of the n-th base molecule along the chosen strand and the corresponding energy ε_n is given by

$$\varepsilon_n = \varepsilon_n^{(0)} - U_{\parallel} n - U_{\perp} \cos\left(\frac{2\pi n}{10} + \varphi_0\right), \quad (2)$$

where $\varepsilon_n^{(0)}$ is the site energy of the n-th base molecule at zero field. We assume all molecules to be the same, so we set the unperturbed energies $\varepsilon_n^{(0)}$ to zero from now on. The term $U_{\parallel} n$ describes the linear potential along the helix axes; the potential drop across a base molecule in the stacking direction is $U_{\parallel} = eaE\cos\theta$, where a is the nearest-neighbor distance along the helix axis, $U_{\perp} = eEr\sin\theta$ is the potential drop across the helix in the perpendicular direction, $r \approx 1$ nm being the helix radius. The phase φ_0 which determines the azimuth of the strand with respect to the field, is set to zero. The term J in Eq. (1) describes the transfer interaction between the nearest-neighbor bases; it is chosen to be positive which implies that the considered charge is a hole [12].

The parallel component of the electric field, $E\cos\theta$, yields the potential ramp along the stacking direction, which sets the frequency of the Bloch oscillations, as in the traditional case. However, Eq. (2) shows that the helix in the lateral field acquires the additional periodic modulation of the potential. This modulation leads to the modification of the electronic structure of the system: the bare energy band splits into several different minibands, which is crucial for the charge transport properties [6]. Each such miniband has its own Bloch frequency, resulting in a more complex overall picture of Bloch oscillations as we show below. The amplitude of the periodic modulation is controlled by the magnitude of the perpendicular component of the electric field, $E\sin\theta$, providing for a mechanism to alter the fundamental properties of the system.

We further solve the time-dependent Schrödinger equation (the Planck constant is set to unity)

$$i\dot{\psi}_n = \varepsilon_n \psi_n + J(\psi_{n+1} + \psi_{n-1}), \quad (3)$$

for an electron wave packet ψ_n being initially a narrow Gaussian centered at the lattice site n_0

$$\psi_n(0) = A\exp\left[-\frac{(n-n_0)^2}{2}\right], \quad (4)$$

where A is the normalization constant. The solution of Eq. (3) can be expressed in terms of the eigenvalues λ_ν and eigenfunctions $\varphi_{\nu n}$ of the Hamiltonian (1) as follows:

$$\psi_n(t) = \sum_{\nu=1}^{N}\sum_{m=1}^{N} e^{-i\lambda_\nu t}\varphi_{\nu n}\varphi_{\nu m}\psi_m(0), \quad (5)$$

where the eigenfunctions $\varphi_{\nu n}$ are chosen to be real. The quantities we use to characterize the dynamics of the electron wave packet are the mean position of the packet (centroid)

$$x(t) = \sum_{n=1}^{N} (n-n_0)\,|\psi_n(t)|^2, \quad (6a)$$

and its Fourier transform

$$f(\omega) = \frac{1}{2\pi}\int_0^{\infty} dt\, e^{i\omega t} x(t). \quad (6b)$$

3. Results and discussions

In all simulations we used a single helix of $N = 101$ bases. The hopping integral was chosen to be $J = 0.27$ eV [6], the value that was used to reproduce the experimental I–V characteristic of a single dry dsDNA trapped between two contacts [15]. The initial wave packet was placed in the middle of the helix, $n_0 = 51$. We note here that shorter chains could give rise to additional confinement of the charge due to the boundaries, which would lead to additional motion quantization and result in frequency splitting and a more complicated oscillatory motion.

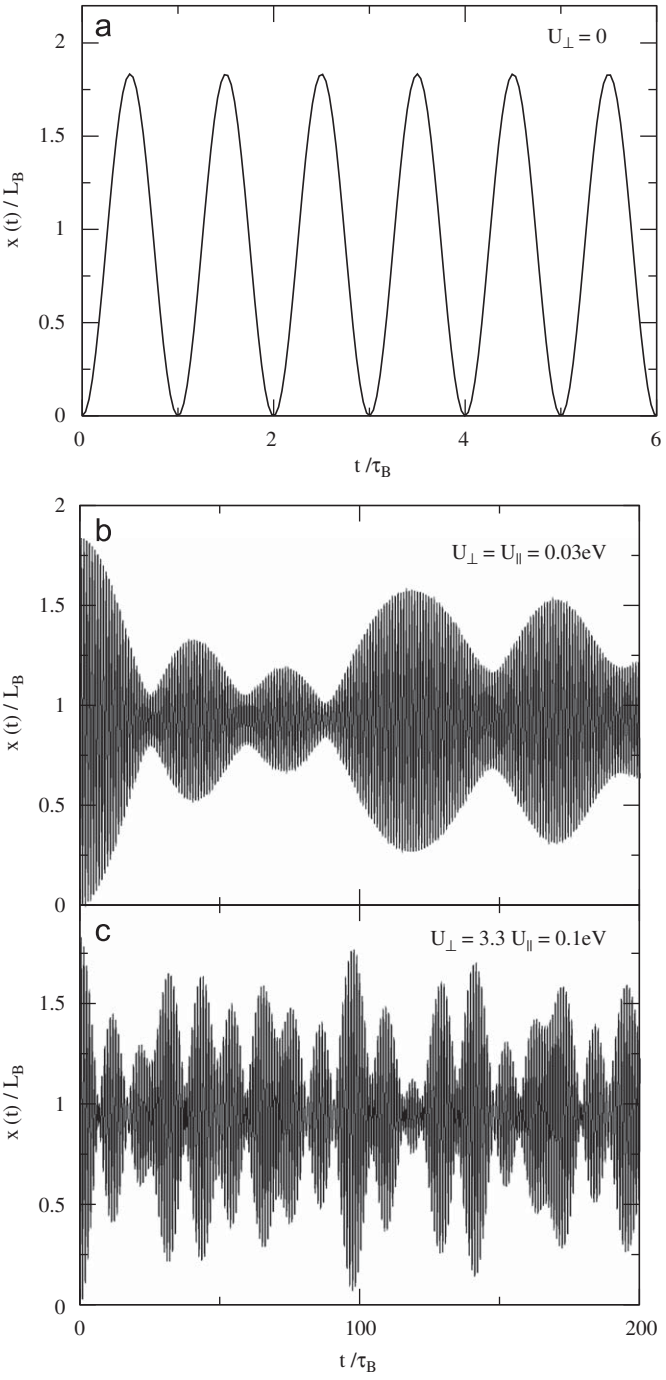

Fig. 1. Bloch oscillations of the centroid $x(t)$, Eq. (6a), calculated for a single-stranded helix of 101 bases at various magnitudes of the gate field. The corresponding gate potentials are indicated in the plots.

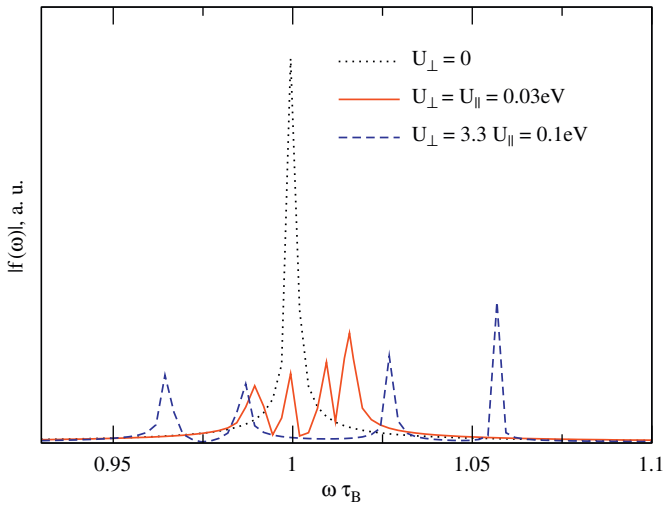

Fig. 2. Spectra of Bloch oscillations of the centroid depicted in Fig. 1.

Fig. 1 displays results of our calculations of the centroid $x(t)$ for various magnitudes of the gate potential drop $U_\perp$. At zero gate potential (upper panel), the motion of the centroid represents simple harmonic oscillations with the period $\tau_B = 2\pi/U_\parallel$ and the amplitude $L_B \approx (4J/U_\parallel)a$, where $4J$ is the bandwidth, which is in full correspondence with the standard picture of Bloch oscillations in a linear chain. The helical symmetry does not affect the oscillations at $U_\perp = 0$.

On turning on the gate field, $U_\perp \neq 0$, the motion of the centroid still manifests an oscillatory behavior, however, a more complicated one as compared to simple harmonic oscillations (Fig. 1, middle and lower panels). The Fourier spectra $f(\omega)$ of the centroid plotted in Fig. 2 shed light on the situation. It demonstrates that a nonzero gate field gives rise to a splitting of the Bloch frequency into a multiplet with a frequency spacing dependent on the gate potential $U_\perp$. The resulting signals presented in Fig. 1 (middle and lower panels) are formed because of the superposition of harmonic oscillations with several different and generally non-commensurate frequencies.

Finally, for the parameters we use, which are typical for the synthetic dry DNA, the period of oscillations $\tau_B = 2\pi/U_\parallel \sim 1$ ps, i.e., the oscillation frequency falls in the THz domain.

4. Summary

In summary, we have demonstrated that the intrinsic helix conformation of the DNA strands can have strong impact on its radiation properties. The electric field along the stack direction forces the injected charge to exhibit Bloch oscillations. In a tilted electric field, however, the harmonic Bloch oscillations become a superposition of oscillations with close and generally non-commensurate frequencies which can be tuned by an external electric field. The frequency of the oscillation falls in the THz domain. This finding is important for the self-assembled DNA arrays on gold with the DNA molecules being tilted with respect to the surface [13,14]. Such arrays could provide a nano-scaled source of coherent THz radiation. Work in Madrid was supported by MEC (project MOSAICO) and BSCH-UCM (project PR34/07-15916).

References

[1] K. Iguchi, Int. J. Mod. Phys. B 11 (1997) 2405;
K. Iguchi, Int. J. Mod. Phys. B 17 (2003) 2565;
K. Iguchi, Int. J. Mod. Phys. B 18 (2004) 1845;
K. Iguchi, J. Phys. Soc. Jpn. 70 (2001) 593.
[2] H. Yamada, Int. J. Mod. Phys. B 18 (2004) 1697;
H. Yamada, Phys. Lett. A 332 (2004) 65.
[3] H. Yamada, E.B. Starikov, D. Henning, J.F.R. Archilla, Eur. Phys. J. E 17 (2005) 149.
[4] D. Klotsa, R.A. Römer, M.S. Turner, Biophys. J. 89 (2005) 2187.
[5] R. Gutiérrez, S. Mohapatra, D. Kohen, D. Porath, G. Cuniberti, Phys. Rev. B 74 (2006) 235105.
[6] A.V. Malyshev, Phys. Rev. Lett. 98 (2007) 096801.
[7] D. Dragoman, M. Dragoman, Prog. Quant. Electr. 28 (2004) 1.
[8] The possibility of Bloch oscillations in the DNA was discussed in V.D. Lakhno, N.S. Fialko, Pis'ma Zh. Éksp. Teor. Fiz. 79 (2004) 575 (JETP Lett. 79 (2004) 464), however, the helical symmetry was not taken into account.
[9] F. Bloch, Z. Phys. 52 (1928) 555.
[10] C. Zener, Proc. R. Soc. London Ser. A 145 (1934) 523.
[11] N.W. Ashcroft, N.D. Mermin, Solid State Physics, Saunders College Publishers, New York, 1976, p. 213.
[12] K. Senthilkumar, F.C. Grozema, C. Fonseca Guerra, F.M. Bickelhaupt, F.D. Lewis, Yu.A. Berlin, M.A. Ratner, L.D.A. Siebbeles, J. Am. Chem. Soc. 127 (2005) 14894.
[13] S.O. Kelley, J.K. Barton, N.M. Jackson, L. McPherson, A. Potter, E.M. Spain, M.J. Allen, M.G. Hill, Langmuir 14 (1998) 6781.
[14] S.O. Kelley, N.M. Jackson, M.G. Hill, J.K. Barton, Angew. Chem. Int. Ed. 38 (1999) 941.
[15] D. Porath, A. Bezryadin, S. de Vries, C. Dekker, Nature 403 (2000) 635.