Multiple frequency Bloch oscillations in natural superlattices

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Abstract. Numerical modelling of electron wavefunctions in a one-dimensional superlattice was conducted. The obtained series of wavefunctions were used to calculate matrix elements of dipole transitions and light emission probabilities. The transitions between distant Stark states were shown to be of high probability together with conventional transitions between neighbor states dubbed Bloch oscillations. Distant transitions have emission probabilities with the same dependence on the applied field and somewhat smaller probabilities.

1. Introduction

The phenomenon of Bloch oscillations was predicted quite a long time ago \cite{1} and it revolves around the fact that in a Stark ladder of electronic states resulting from the electric field $F$ application to a periodic structure with period $d$, the interlevel transitions are possible with a Bloch frequency

$$\omega = eFd/h$$

(1)

In a nutshell, the Bloch oscillations are produced by an electron whose movement is induced by the applied field. Since in a periodic potential the dispersion curve of an electron has a bend, where its effective mass becomes negative, the electron changes its direction of movement, and hence the oscillations.

It is thus essential for the Bloch oscillations to be manifested that an electron energy dispersion is of minizone (sine-like) type. This is true for some crystals in certain directions, but in a crystal such transitions are hardly observable. The difficulty is a consequence of too wide a minizone and hence too large an electric field needed to be applied, breaking the crystal.

However, the superlattices (SLs) as suggested by \cite{2} are attractive for this. The fact that a minizone on these artificial crystals is narrow, the needed electric fields are modest and lead to a Bloch frequency in a terahertz (THz) range. The THz radiation in various semiconductor structures received quite a bit of attention recently \cite{3,4}.

One of interesting materials with a potential in THz technology is silicon carbide (SiC). Being a natural crystal, it nevertheless exhibits some properties of a very high-quality superlattice. Its hexagonal polymorphs are well modelled by a layered structure \cite{5}. Indeed, there are recent observations of THz emission from hexagonal SiC with applied electrical bias \cite{6}.

As for the phenomenon of the Bloch oscillations in a general sense, there was previously a consensus about its harmonic nature. That is, both in quasi-classical and quantum approaches \cite{7-9} it was predicted that emission or quantum transitions are occurring only on the sole Bloch frequency. The purpose of this work is to demonstrate that emission is not confined to the sole Bloch frequency but is also possible on multiples of the Bloch frequency. The main focus of this work as to application to the real life is SiC and modelling of its properties.
2. Model

As was mentioned before, a hexagonal polymorph of SiC (of which 2H, 4H, 6H and 8H are studied in this work) can be considered to be a superlattice. Indeed, as fig. 1a,c shows, the general structure of SiC in its hexagonal form is a densely packed one. The main unit is a layer of silicon-carbon atom pairs that are arranged in triangular fashion. As the layers are stacked one upon another, there are two positions in which atoms could go in relation to the previous two layers. Thus, the layers can be denoted as A, B or C.

![Figure 1](image1.png)

**Figure 1(a-b).** (a) Densely packed hexagonal lattice; (b) the Brillouin zone in a hexagonal lattice; (c) arrangement of atomic layers in 4H-SiC.

![Figure 2](image2.png)

**Figure 2.** Band structure for different hexagonal SiC polymorphs, modelled by DFT. Fermi level denoted by dashed line.

C. Every layer has to be surrounded by layers of names different to itself to achieve dense packing (i.e. B-C-B or C-B-A). If these two layers are themselves mutually different (as in C-B-A), the layer B has
a hexagonal type of near symmetry and lower energy, thus is a quantum well. In the other case the layer is of cubic symmetry and presents a quantum barrier [10-12].

To prove that such a model is indeed plausible we have conducted energy dispersion calculations using the DFT method [13] for 2H, 4H, 6H and 8H polymorphs. The results presented in fig. 2 are to be interpreted in a following way. First, in all the polymorphs except 2H the M-L dispersion branch has the lowest energy in the valence band, which means that the electron transport models should consider this branch. Second, the nature of this branch (except in 2H) is very much like that of an ideal minizone dispersion in a SL.

We went on by selecting for 4H-, 6H- and 8H-SiC such a model potential as to resemble the one calculated by DFT as closely as possible. The results are shown in fig. 3 and table 1. Each potential period is a sum of widths of the barrier and the well and is equal to a lattice constant of a corresponding SiC polymorph in the direction perpendicular to the layers (along the line going through Si-C pair). We have also somewhat improved the findings of [5] by providing a much closer resemblance of the model dispersion to the DFT-calculated one.

![Figure 3. Dispersion in SiC along M-L line near the bottom of the valence band. Solid line – model potential, long dash – DFT calculation, short dash – dispersion from [5].](image)

|     | Barrier height, $eV$ | Barrier width, nm | Potential period, nm | Electron mass, $m_0$ |
|-----|----------------------|-------------------|----------------------|---------------------|
| 4H  | 1.9                  | 0.095             | 0.5                  | 1.75                |
| 6H  | 2.35                 | 0.25              | 0.75                 | 1.0                 |
| 8H  | 0.79                 | 0.25              | 1.0                  | 1.0                 |

The obtained periodic model potential can now be used to conduct calculations of quantum properties of the material under electrical bias. The calculation utilised the transfer matrix approach. The reason for this is that one cannot use DFT when an electric field is applied for the problems of large enough scale. Not only is the calculation time unrealistic, but a parasitic dipole moment often emerges and has to be corrected. The transfer matrix method, on the other hand, isn’t general and needs a one-dimensional model, but is very quick and can be generalized for the case of an applied electrical field. It helps to observe that the wavefunction and its flow (proportional to its derivative) are bound to be conserved at the boundary between two layers with different (but constant) potential energy. This leads to construction of a 2x2 transfer matrix for all the boundaries and layers. These matrices depend on the
potential energy and the energy of an electron via the electron wavevector \( k = \sqrt{2m(E - U)} / h \). Since a wavefunction must take a form of a decaying exponent outside of the structure, one can obtain an equation on \( E \). This provides a method to calculate eigen energies of a certain structure. The slope in potential energy arising from an application of an electrical field can be approximated by enough steps (a piece-wise constant function). In an agreement with quantum theory we have found out that in a biased periodic SL the energy levels are equally spaced and form a Stark ladder of translationally symmetrical states. The spatial width \( s \) of the state is proportional to the minizone width and reverse proportional to the applied field:

\[
s = \frac{W}{eF}
\]  

This is shown in fig. 4.

Figure 4(a-d). Spatial distribution of probability density (wavefunction module squared) for an electron in a SL with different bias field applied: (a) 500 kV/cm; (b) 200 kV/cm; (c) 100 kV/cm; (d) 50 kV/cm.

With energies and transfer matrices at hand we can calculate wavefunctions \( \langle f \mid \) and \( \mid i \rangle \) of any dipole transition and subsequently a dipole matrix element and the emission probability for any frequency, provided that this frequency is a difference between the energies of Stark states (that is, a multiple of Bloch frequency (1)):

\[
P = \frac{\alpha \langle f \mid |i\rangle^2 n^2 \omega^3}{\pi c^2}
\]  

For the single Bloch frequency, the matrix element can be estimated [9] to be \( s/4 \) and the estimate of the transitional probability is

\[
P = \frac{\alpha}{16\pi} \left( \frac{nWd}{\epsilon h} \right)^2 \omega
\]
3. Results
When calculated, it became evident that there exists a possibility for transitions at the frequencies of $2\omega$ and $3\omega$, together with conventional Bloch transitions at $\omega$ [14]. Importantly, the probabilities of such transitions are proportional to the applied electrical field as is the probability of Bloch oscillations. The value of the probability falls with every multiple by an order of magnitude, that is, at $2\omega$ we are getting a ten times weaker emission than at $\omega$, and the same is true for $3\omega$ versus $2\omega$. The results are summarized in fig. 5. Still, the estimate (4) holds for the single frequency transition which is shown by a dashed line and speaks for the correctness of the chosen method.

Importantly, this behaviour needs a SL with size bigger than an effective state size $s$. Otherwise, the transition is weaker due to weaker localisation but exhibits a superlinear growth against the growth of applied field (evident in fig. 1 for the part with smaller frequencies).

![Figure 5](image)

**Figure 5.** Dependence of transition probabilities for single (squares), double (circles) and triple (triangles) Bloch frequencies on frequency. The dashed line corresponds to (4).

4. Conclusions
We have studied Bloch oscillations in electrically biased and provided a theoretical proof that they might be accompanied by the emission of light on multiples of the Bloch frequency. We have also studied the energy dispersion in hexagonal SiC and provided a model for studying quantum effects in it. The advantages of the model are the speed of calculations and the sustained applicability under symmetry-breaking conditions such as an applied electric field.

Acknowledgements
The work was partially supported by the RFBR grant No. 18-32-00801.

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