Magneto-quantum oscillations in Bi$_2$Se$_3$ nanowires

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Abstract. We report preparation of nanoribbons (crosssection ~ 250*25 nm$^2$) by focused ion beam etching of single-crystalline Bi$_2$Se$_3$ and detailed measurements of their magnetoresistance at temperatures down to 4.2 K, magnetic field up to 9 T. In a magnetic field parallel to the axis of nanowire the magnetoresistance shows up oscillations. Surprisingly, the Fourier analysis shows the presence not only of oscillations with a period corresponding to the flux quantum ($\Phi_0 = \hbar c/e$), but also oscillations with a period of 2$\Phi_0$ and 4$\Phi_0$. Possible mechanisms of the observed effect are discussed.

1. Introduction
The study of topologically non-trivial quantum states is one of the most interesting and developing directions in solid-state physics past decade. The existence of such surface electronic states was theoretically predicted [1] in materials Bi$_2$Se$_3$, Bi$_2$Te$_3$ and Sb$_2$Te$_3$ and then confirmed by several independent experiments: angle resolved photoemission spectroscopy (ARPES) [2-4], scanning tunneling microscopy STM [5], transport measurements [6-8]. However, the nature of surface states is still a subject for investigation. The reason is that the real materials contains impurities, thermal excitations and structural defects. This leads to bulk contribution in conductivity which impedes interpretation of the observed effects. One of the most efficient methods to study the nature of topologically protected surface states is low-temperature magnetotransport measurements. This method is especially efficient in quasi-one-dimensional structures where the contribution of the bulk carriers is reduced due to the large surface-to-volume ratio.

There are several works in which the magnetotransport in grown single-crystalline Bi$_2$Se$_3$ nanoribbons is studied [9-11]. Most of studied samples are whiskers of topological insulator grown by various methods: vapor–liquid–solid (VLS) growth, chemical vapor deposition (CVD), electrochemical deposition. The object of our study is Bi$_2$Se$_3$ nanowires prepared from bulk crystals by means of mechanical and focused-ion beam thinning. The group of Bhattacharyya [12,13] which
studied similar objects should be mentioned. The comparative analysis of our studies with the studies of other research groups is the subject of this article.

2. Methods
2.1. Scotch-tape exfoliation
As was mentioned above, our Bi$_2$Se$_3$ nanoribbons were fabricated from bulk crystals. We used Bi$_2$Se$_3$ crystals grown by the standard Bridgman method with a carrier concentration of $\sim 10^{19}$ cm$^{-3}$. At first, the initial bulk crystal was thinned down by the modification of classical micromechanical exfoliation technique (scotch tape method) [14,15]: a chosen high quality layered crystal of Bi$_2$Se$_3$ was attached to the substrate using epoxy glue and then was thinned down to tens nanometers with adhesive tape. The thickness of films at first was estimated optically and then measured more precisely with focused ion beam (FIB): the sample was placed at the certain angle ($\alpha$) relative to the exposition surface so that the side edge of the film could be seen; then the real thickness was calculated as $D_{\text{real}} = D_{\text{measured}} / \sin(\alpha)$. Then the area of the future nanostructure was masked and an Au film was sputtered by laser ablation (in order to make the electric contacts for the sample). After that the mask was removed and the sample was ready for FIB milling.

2.2. Fib milling
Gallium focused ion beam (SMI3050, SII NanoTechnology and Crossbeam Neon 40 EsB (Carl Zeiss)) was used to make 1D structure from the Bi$_2$Se$_3$ 2D flake. Structures have the form of nanoribbon (fig.1) with typical sizes: length of 10 µm, width of 200–500 nm, height of 10–100 nm. We tried to minimize the FIB exposition on the workspace of the sample using scanning electron microscope (SEM) to target on the sample or liquid (collodion) and solid masking as a protection from Ga ions [15-16]. The milling of the structure was made with the low ion beam currents (10 pA). The area at the distance from the structure (the edges of the flake and Au contacts) was milled with higher currents of 100 pA – 10 nA.

2.3. Measurement details
The resistance of the samples was measured using a conventional four-wire scheme, by the Kelvin method at an alternating current of a few tens of Hertz with a low-noise Lock-in Amplifier [17]. Magnetotransport measurements were carried out in a cryostat with a superconducting magnet.

3. Results and discussion
The magnetoresistance of Bi$_2$Se$_3$ nanowires was measured for several samples at temperatures 4.2 – 15 K in magnetic fields up to 9 T parallel to the axis of nanowire (Figure 2). At low magnetic fields 0-1 T
for all our samples, a strong magnetoresistance peculiarity was observed, most probably because of weak antilocalization. The observation of this effect points out high quality of our samples.

At higher magnetic fields (0.5–9T), magnetoresistance oscillations are observed. The main period of the oscillations is close to the flux quantum $\Phi_0 = \hbar c/e$ through the cross-section of the wire (Figure 2). The existence of such Aharonov-Bohm-type (AB) oscillations is in agreement with the other experiments and is a manifestation of the surface states in Bi$_2$Se$_3$ insulator. The observation of the AB-oscillations evidences that these states are robust to the Ga ions which are inevitably implanted in the sample during the FIB milling [12].

Besides the oscillations with the period 0.8 T, corresponding to $\Phi_0$ (for the sample #1) we observed unexpectedly other periods: 1.6 T, 3.2 T, corresponding to $2\Phi_0$, and $4\Phi_0$ through the cross-section of the wire. These periods are clearly seen on the fast Fourier transform (FFT) graph (fig2b). The existence of such subharmonic oscillations is rather surprising. Such oscillations were reproduced for our other samples. Their amplitude drops with the temperature increase and almost disappears above 10 K. Signatures of such oscillations may be noticed in the raw data by Peng [9] for a 120 nm-wide sample, but little attention is paid to them. Their existence was accounted for bulk contribution to conductivity.

**Figure 2.** (a) Magnetoresistance of the sample #1 for different currents through the sample (plotted with the 7 Ohm offsets). The independent traces taken at different currents—clearly show reproducibility of oscillations with the periods $\Phi_0$, $2\Phi_0$, $4\Phi_0$ and the absence of their nonlinear dependence on current. (b) Fast Fourier transform with the Hann window function (FFT) of the oscillating part of the magnetoresistance (plotted with offsets ~ 0.01 a.u.). The main periods ($\Phi_0$, $2\Phi_0$, $4\Phi_0$) are clearly seen. The period $\Phi_0/2$ (red dotted line) is absent.

We may propose the following possible reasons for the effect observed:

i) Our sample has a rectangular shape and the subharmonics could originate from reflections at its edges. In this case the trajectories of particle motion become more complex, and this can cause oscillations with long periods.

ii) The influence of implanted Ga ions on the quality of the sample can be stronger than we presume. It is possible that gallium ions damage significant area near the edge of the sample. As a result, some particles change their trajectory due to the reflection. The latter reason however, seems less likely, because random distribution of Ga ions can hardly cause sharp reflection effects.

4. Conclusions

We have studied the magnetoresistance of Bi$_2$Se$_3$ nanowires prepared by focused ion beam and found the presence of the oscillations with abnormally large periods ($2\Phi_0$, $4\Phi_0$) beside the anticipated period $\Phi_0$. We proposed two possible reasons of their existence. In order to clarify the effect, further
research is required. The observation of the AB oscillations in our Bi$_2$Se$_3$ nanoribbons proves that the topologically protected surface states propagate coherently over the nanoribbon perimeter ~1000nm even in the sample whose side walls are prepared by etching with focused beam of Ga-ions.

Acknowledgments
The work was supported by the Russian Foundation for Basic Research (no. 16-32-00927 and no. 16-29-03330).

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