Weak localization and Raman Study of Graphene Antidot Lattices obtained by Crystallographically Anisotropic Etching

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Abstract

We report about the crystallographically anisotropic etching of exfoliated graphene on SiO_2 substrates by applying a high-temperature etching mechanism (800°C and argon atmosphere) that was demonstrated to eliminate carbon atoms located on armchair sites, thus leading to zigzag edges [1]. Before exposing samples to this carbothermal reaction, they were patterned with circular antidots (diameter d ~ 40nm) by electron-beam lithography and reactive-ion etching. In the subsequent carbothermal etching step, the predefined holes evolved into larger hexagonal antidots (d ~ 100-150nm), as shown in the inset of Fig.1. We will discuss a sample preconditioning procedure which is essential to etch single-layer graphene in an anisotropic manner, however not for bi- and multilayer samples.

We investigated a set of single-layer graphene samples, patterned with square lattices of different constants a of hexagonal antidots with different diameters d, and compare them to graphene patterned with lattices of circular holes, investigated previously [2,3]. First, we compare samples by analyzing the weak localization peak in electron transport at low temperatures. By fits of the data to the model put forward in [4], we extract the phase coherence length and the lengths for inter- and intravalley scattering. The intervalley scattering length L_i increases linearly with the length a-d, the spacing between neighboring holes, as shown in Fig.1. The process of intervalley scattering is related to the presence of armchair edges in graphene. The large scatter in the observed L_i's for different hexagonal antidot samples points to differences in the ratio of the number of armchair to zigzag sites. The scatter must therefore stem from variations in the quality of the anisotropic etching process. The samples with the highest L_i's for corresponding values of a-d are those with hexagonal antidots. For this reason our interpretation is that our anisotropic etching process creates hexagonal holes with edges along the zigzag orientation. We think that the edges of hexagonal antidot samples with lower L_i values must be rough on an atomic scale, since this creates intervalley scattering. This, however, cannot by judged adequately by SEM imaging.

Second, the set of antidot samples was characterized by Raman spectroscopy, focusing on the D and G peak. The D-Peak is related to intervalley scattering and therefore to the presence of armchair edges. In a systematic study, we compare the intensity ratios between the D and the G peak for samples patterned with different antidot lattice constants and diameters. We expect the D peak to be proportional to the length of the circumference and to the number N of antidots within the laser spot, and, therefore, to Nd. The G peak is expected to be proportional to the area $N(a^2 - \pi d^2/4)$ (cf. inset Fig.2). The data shown in Fig.2 show a reasonable correlation between the patterned antidot lattice structure and the D/G intensity ratio for samples with circular antidots. For the hexagonal antidots there is again considerable scatter in the data. However, the smallest D/G intensity ratios are obtained for samples with hexagonal antidots. The interpretation of the weak localization data is consistent with the Raman data.

Furthermore, we analyze the correlation between Raman and weak localization measurements with regard to the intervalley scattering mechanism. In Fig.3 the values for the D/G intensity ratios of different antidot samples are plotted against the intervalley scattering times L_i, acquired for each sample. The plot shows a clear correlation between the D/G intensity ratio and L_i.

In addition to the above mentioned studies of graphene antidot lattices, we demonstrate the influence of the anisotropic high-temperature etching reaction on the properties of graphene by showing a series of Raman spectra, acquired between consecutive sample preparation steps.

References

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Figures

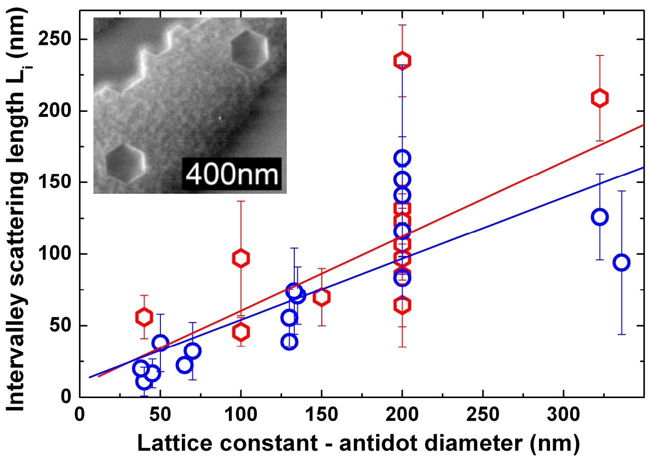

Figure 1: The intervalley scattering lengths deduced from weak localization measurements at temperatures of 1.6K for samples patterned with circular antidots and hexagonal holes obtained by high-temperature etching are depicted by the blue circles and the red hexagons, respectively. The blue and the red line show linear fits to blue and the red data points, respectively. The inset shows a SEM image of graphene that was patterned with a quadratic lattice of antidots and anisotropically etched in the high-temperature etching reaction.

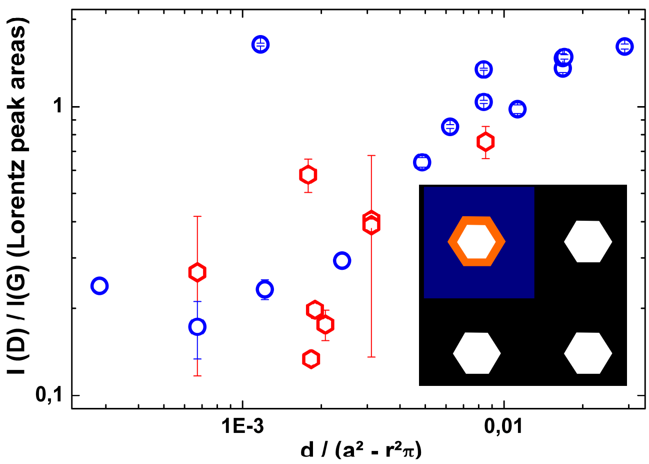

Figure 2: The D/G peak intensity ratios deduced from the peak areas in Raman spectra for samples patterned with circular antidots and hexagonal holes obtained by high-temperature etching are depicted by the blue circles and the red hexagons, respectively. The inset shows in orange and dark blue colour the areas of the Raman active zones for the D and the G peak within each unit cell of the antidot lattice, respectively.

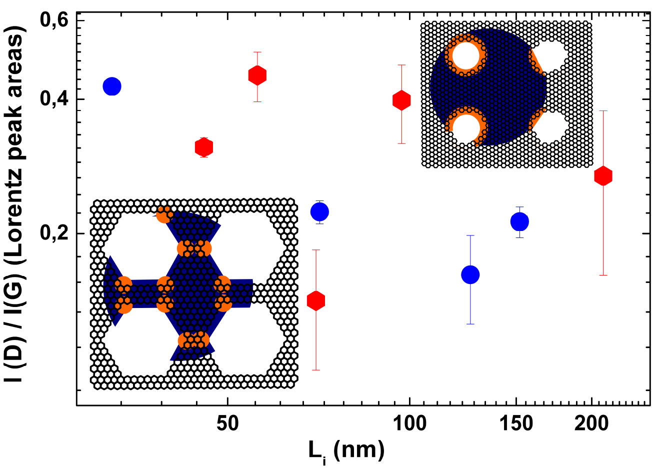

Figure 3: The D/G peak intensity ratios for samples with circular and hexagonal antidots correlates with the corresponding intervalley scattering lengths L_i acquired for specific samples (blue circles and red hexagons, respectively). The insets show the different Raman active zones for the D and the G peak for circular and hexagonal holes in orange and dark blue colour, respectively.