

# Non-Equilibrium Phenomena in Graphene

Submitted by Samuel Martyn Hornett to the University of Exeter as  
a thesis for the degree of Doctor of Philosophy in Physics  
April, 2013

This thesis is available for Library use on the understanding that it is copyright material and that no quotation from the thesis may be published without proper acknowledgement.

I certify that all material in this thesis which is not my own work has been identified and that no material has previously been submitted and approved for the award of a degree by this or any other University.

Samuel Martyn Hornett  
April, 2013

# Abstract

Graphene has displayed much promise as an electrical conductor and as a optical material. To date there is a large body of literature dedicated to the equilibrium properties of graphene. In this thesis the properties of graphene out of equilibrium are probed. Through combined optical and transport measurements the behaviour of hot electrons are probed at temperatures over five orders of magnitude from 50mK to 2000K. This wide range of temperatures allows access to the behaviour of quantum corrections at the lowest temperatures to the highest energy phonon modes. From ultrafast femtosecond laser pulses to steady state heating from an electric field the cooling of hot electron populations through coupling to various phonon modes in the graphene and the substrate are explored. Additionally the effect of an electric field on the weak localisation correction to the conductivity was separated from heating effects using applied magnetic fields combined with careful modelling of the heat transport properties of the graphene. Finally the desorption dynamics of oxygen bound to the surface are shown using a combination of transport and two pulse correlation technique using an ultrafast laser. Surprisingly the cooling of hot carriers in graphene at low energies shows substrate surface phonons as an important cooling mechanism, highlighting the importance of substrate choice in future graphene devices. In contrast at the very highest energy scales accessed only by photoexcitation the cooling is shown not to be influenced by the presence of a substrate, but out-of-plane phonon modes increase cooling of the hot optical phonons.

# Contents

<b>Abstract</b>	<b>2</b>
<b>Acknowledgements</b>	<b>3</b>
<b>Contents</b>	<b>3</b>
<b>List of Figures</b>	<b>6</b>
<b>Introduction</b>	<b>14</b>
<b>1 Theory</b>	<b>16</b>
1.1 Band Structure of Graphene . . . . .	16
1.1.1 Linear Regime . . . . .	21
1.1.2 Chirality . . . . .	22
1.1.3 Berry Phase . . . . .	23
1.1.4 Density of States . . . . .	23
1.2 Conductivity of two dimensional systems at low temperatures . . . . .	24
1.2.1 Electron Specific Heat Capacity . . . . .	25
1.2.2 Hall Effect . . . . .	25
1.2.3 Weak Localisation . . . . .	27
1.2.4 Universal Conductance Fluctuations . . . . .	31
1.2.5 Phonons in Graphene . . . . .	32
1.2.6 Phonon Phonon Scattering . . . . .	32
<b>2 Experimental Method</b>	<b>35</b>
2.1 Sample Fabrication . . . . .	35
2.2 Optical Contrast . . . . .	37
2.3 Raman Spectroscopy of Graphene . . . . .	38

2.3.1	Layer Determination . . . . .	41
2.4	Low Temperature Transport Measurements . . . . .	45
2.4.1	Dependence of the Resistance on carrier concentration . . . . .	46
2.5	Optical Techniques . . . . .	46
<b>3</b>	<b>Heat Dissipation Mechanisms in Graphene</b>	<b>49</b>
3.1	Introduction . . . . .	49
3.2	Samples and Measurement Technique . . . . .	50
3.3	Contact Pinning . . . . .	51
3.4	Comparison of temperature profiles with data . . . . .	53
3.5	Simple Temperature Model . . . . .	53
3.6	Heat Transport Model . . . . .	58
3.6.1	Acoustic Phonons . . . . .	62
3.6.2	Remote Optical Substrate Phonons . . . . .	64
3.7	Conclusions . . . . .	65
<b>4</b>	<b>Quantum Corrections to the Conductivity in a High Electric Field</b>	<b>66</b>
4.1	Field Dependent Weak Localisation model . . . . .	67
4.2	Averaging of the Universal Conductance Fluctuations (UCF) . . . . .	68
4.3	Sample Characterisation . . . . .	71
4.4	Magnetoconductivity . . . . .	72
4.5	Conclusions and Future Work . . . . .	75
<b>5</b>	<b>Hot Phonon Decay in Graphene</b>	<b>77</b>
5.1	Measurements of the Differential Reflection . . . . .	78
5.2	Two Temperature Model . . . . .	79
5.2.1	Excitation Pulse . . . . .	82
5.2.2	Computation of the Model . . . . .	83
5.2.3	Fitting the Data . . . . .	84
5.3	Layer Dependence of the Phonon Decay Time. . . . .	84
5.3.1	Layer Dependence of the Phonon Decay Time in Suspended samples. . . . .	85
5.4	Conclusions . . . . .	87

<b>6 Desorption of Oxygen from Graphene by Femtosecond Laser Pulses</b>	<b>89</b>
6.1 Experimental Method . . . . .	91
6.2 Laser Desorption of Oxygen . . . . .	93
6.3 Two Pulse Correlation . . . . .	96
6.4 Conclusion . . . . .	100
<b>7 Conclusions and Future Work</b>	<b>101</b>
<b>Bibliography</b>	<b>103</b>