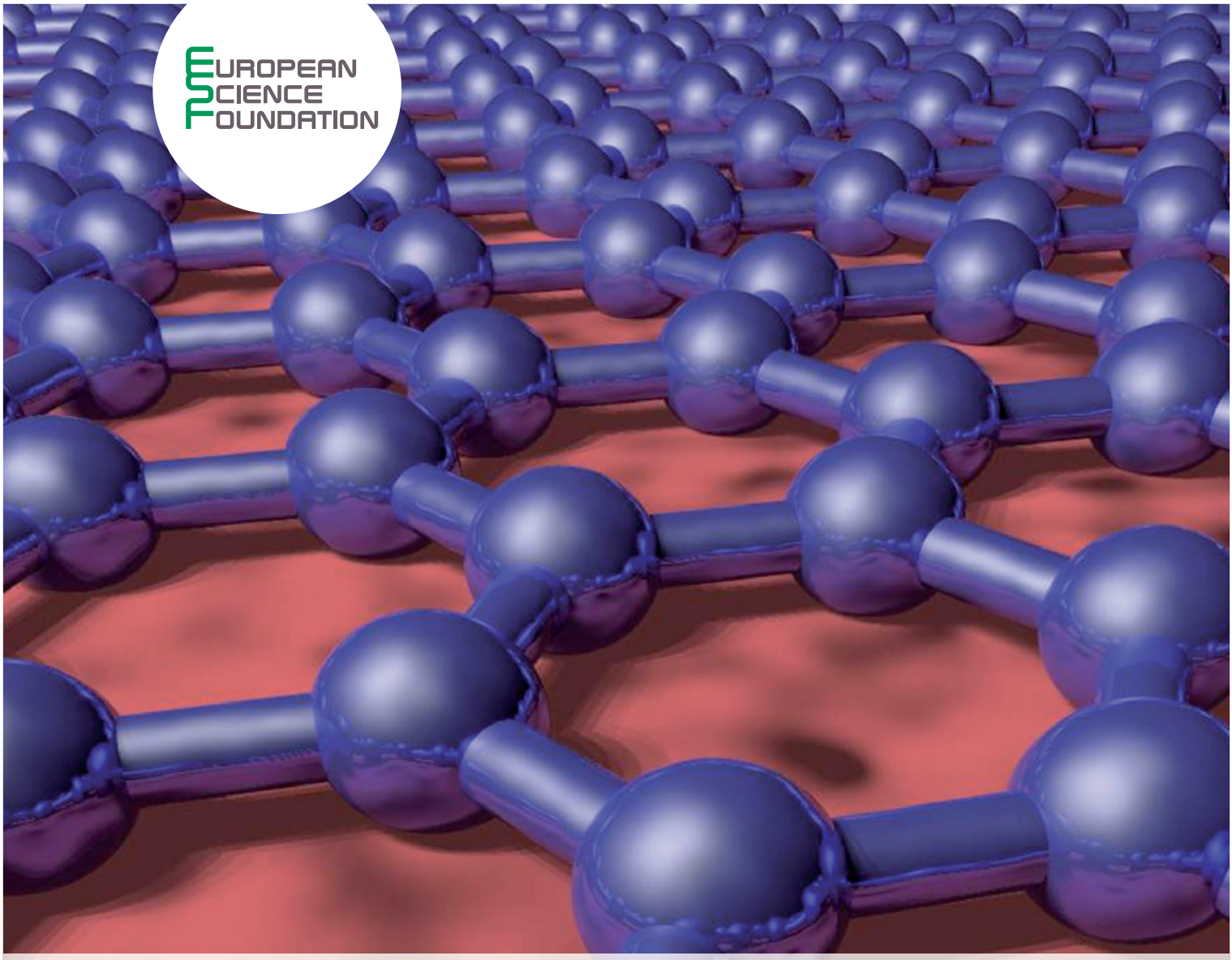
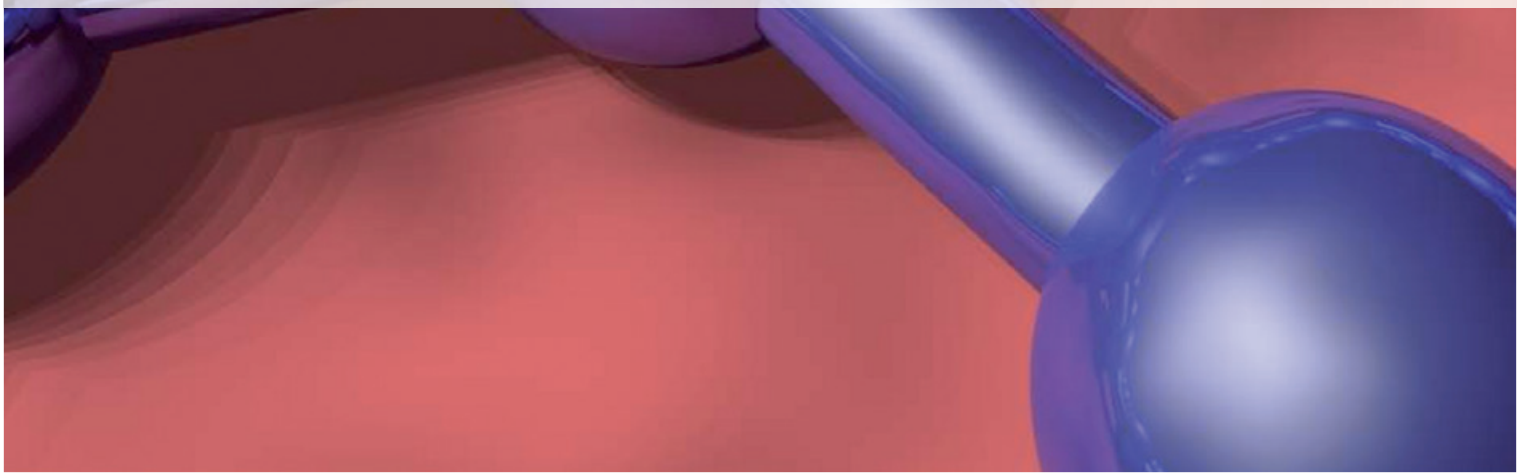


ESF EUROCORES Programme

Maximising the Impact of Graphene Research in Science and Innovation (EuroGRAPHENE)

Scientific Highlights



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www.esf.org/eurocores

The following national funding organisations support the EuroGRAPHENE programme:

- **Fonds zur Förderung der wissenschaftlichen Forschung in Österreich (FWF)**
  Austrian Science Fund, Austria
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  Fund for Scientific Research, Belgium
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- **Grantová agentura České republiky (GAČR)**
  Czech Science Foundation, Czech Republic
- **Eesti Teadusfond (ETF)**
  Estonian Science Foundation, Estonia
- **Suomen Akatemia/Finlands Akademi**
  Academy of Finland, Finland
- **Deutsche Forschungsgemeinschaft (DFG)**
  German Research Foundation, Germany
- **Consiglio Nazionale delle Ricerche (CNR)**
  National Research Council, Italy
- **The Dutch Foundation for Fundamental Research on Matter (FOM)**, The Netherlands
- **Ministerstwo Nauki i Szkolnictwa Wyższego (MNISW)**
  Ministry of Science and Higher Education, Poland
- **Javna agencija za raziskovalno dejavnost Republike Slovenije (ARRS)**
  Slovenian Research Agency, Slovenia
- **Vetenskapsrådet (VR)**
  Swedish Research Council, Sweden
- **Schweizerischer Nationalfonds (SNF)**
  Swiss National Science Foundation, Switzerland
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  The Scientific and Technological Research Council of Turkey, Turkey

Cover picture:
Graphene

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Introduction

The discovery of the peculiar physical properties of graphene has caused a storm of excitement, and this interest in graphene as a qualitatively new two-dimensional electronic system has boosted the rapid development of the physics of graphene and graphene-based nanostructures. The EUROCORES programme EuroGRAPHENE, which is coming to an end this year, is the first coordinated European-wide cooperation on graphene science and technology. The visionary motivation that led to the establishment of this programme was that of turning a rapidly developing field of physics, as graphene was in 2009, into a field of cross-sectional research with the aim of boosting the high- and nano-tech applications of this material with the ultimate thickness limit of one atomic layer. This vision in now a reality – we have deepened our understanding of physical properties of graphene; we have expanded research into new areas of chemical modifications of the material, and searched for methods to design its electronic properties; we have investigated its mechanical and electro-mechanical properties, broadly studying kinetic processes and optoelectronic effects in graphene; we have modelled graphene-based devices for new functional applications. As a collaborative programme, EuroGRAPHENE has built its success on diverse networking actions, open joint workshops, meetings between different teams working on focused projects, researcher exchanges – a strategy that enabled the programme to reach its ambitious targets. This vision of EuroGRAPHENE is now a reality; this programme can be considered a forerunner of larger scale future programmes and coordinated actions, the forthcoming GRAPHENE Flagship being the most prominent example. The "EuroGRAPHENE Session" of Graphene Week 2013 will report on scientific highlights achieved by this programme. This brochure provides a brief summary about EuroGRAPHENE's projects and science. Moreover, many of the topics of EuroGRAPHENE will continue to be fruitful areas of research in ongoing projects. In the following, we give a short description of work within the separate projects of EuroGRAPHENE.

Collaborative Research Projects (CRPs)

Graphene-Organic SuPramolEcular FunctionaL Composites (GOSPEL)

The aim of GOSPEL was to develop new hybrid materials based on the supramolecular interactions of graphene sheets with tailored organic molecules, either small polyaromatics or polymers, for applications in optoelectronics. The orientation of the molecules on the graphite surface can be highly specific (i.e. face-on or edge-on), including a possibility of ordered lateral arrangement of the molecules on nano-scale. Such interactions can be exploited to exfoliate low-cost graphite into graphene sheets, by mixing solutions of organic molecules with suspensions of graphite flakes.

In GOSPEL project we exploited the properties of tailored organic molecules to develop new graphene-organic hybrid materials (GOH), termed nanographenes (NG) because of their aromatic core. They have uniform shape and size, well-defined zig-zag or armchair edges and tunable optical and electronic bandgaps (Fig. 1a), and form highly ordered and stable monolayers with highly specific molecule orientation (i.e. face-on or edge-on) (Fig. 1b-d).

NG can be successfully used either as surfactants, to favour top-down exfoliation of graphite in water solutions (Fig. 1b-f) or as building blocks, exploiting the bottom-up assembly of small NG to produce graphene nanoribbons (Fig. 1g-k).

We used a combination of spectroscopy, microscopy and computational modelling to monitor the

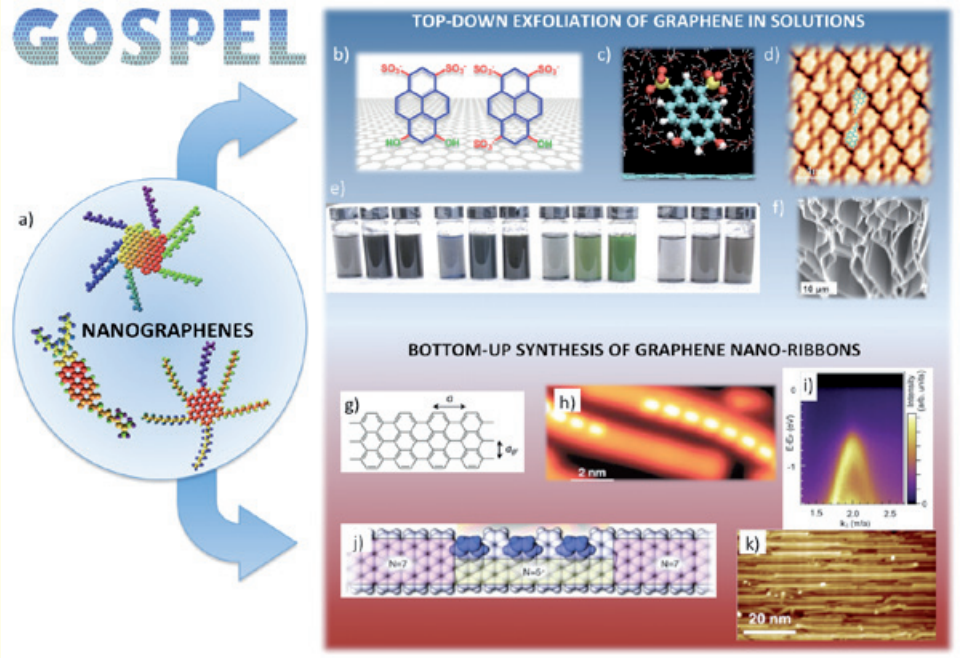

Figure 1.
a) Schematic representation of different NG. b) Structure of two NG tested for graphene exfoliation. c) Modelling of NG adsorption on graphene in water. d) STM images of a pyrene derivative self-assembled on graphene/Pt(111) e) Solutions of graphene exfoliated in water with different NG. f) SEM of partially exfoliated graphite. g-k) modelling and characterisation of graphene nanoribbons self-assembled from small NG: g,j) Structure, h,k) STM, i) ARPES.

adsorption of organic dyes on graphene and the exfoliation in water of dye-functionalised graphene sheets. We quantified the exfoliation process in solution, and assessed the graphene-NG ratio in the GOH. Using experiments and modelling (Fig.1 b-e) we found a correlation between the graphene-NG interaction energy, the molecular structure and the amount of graphene flakes solubilised. A key process is the adsorption of the NG on graphene by a "sliding" mechanism into the solvent layer, facilitating the lateral displacement of the water molecules collocated between the aromatic cores of the NG and graphene. We also monitored the effect of NG on the transfer characteristics of thin film transistors; using time-resolved photocurrent measurements we were able to detect a change of electron mobility even for very low NG coverages.

A specific highlight is the bottom-up production of graphene nanoribbons by self assembly of NG, leading to graphene nanoribbon heterojunctions with lateral dimensions below 2 nm via controllable dehydrogenation of small NG self-assembled on a Au(111) surface. Atomistic simulations revealed the microscopic mechanisms responsible for intraribbon heterojunction formation. We demonstrated the capability to selectively modify the heterojunctions by activating the dehydrogenation reaction on single units of the nanoribbons by electron injection from the tip of a scanning tunneling microscope, or to include doping heteroatoms in the edge of the nanoribbons. Angle-resolved photoelectron spectroscopy (ARPES) and scanning tunneling spectroscopy data from armchair graphene nanoribbons of width N = 7 supported on Au(111) reveal a band gap of 2.3 eV, an effective mass of 0.21 m_0 at the top of the valence band, and an energy-dependent charge carrier velocity reaching 8.2 x10^5 m/s in the linear part of the valence band.

GOSPEL partners:

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Confinement in Graphene Nanostructures (CONGRAN)

The achievement of accurate tuning of the quantum dots in graphene, which is difficult due to the disorder effects associated with the fabrication, has been in the focus of CONGRAN. Possible paths towards definition of highly tunable graphene quantum dots and coupled dots have been explored, clarifying the advantages and disadvantages of different approaches regarding the requirements for various quantum dot experiments.

CONGRAN involves the study of nanostructured single and multilayers of graphene through comprehensive experimental and theoretical approaches. The project consists of the following parts: 1) tunable etched quantum dots using local top gates, 2) top gate defined quantum dots in graphene bilayers, 3) spin effects in graphene quantum dots, and 4) graphene quantum dots at elevated temperatures. This concerted effort has resulted in novel nanostructured graphene implementation concepts.

Different types of quantum dots and coupled dots were realised through edging of graphene and

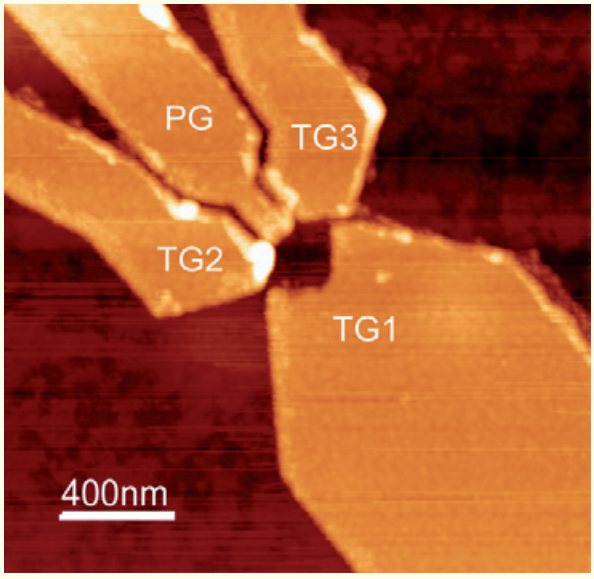

Figure 2.
Gate-defined graphene quantum dot (A.M. Goossens, S.C.M. Driessen, T.A. Baart, K. Watanabe, T. Taniguchi, and L.M.K. Vandersypen, Nano Lett. 12, 4656 (2012). TG: top gates, PG: plunger gate

by using gating of bilayer graphene. Coulomb blockade and single electron transport was observed. At low magnetic field a novel valley-related transition is predicted for a two electron quantum dot in bilayer graphene.

The electron spin relaxation in electrostatically defined nanoribbon quantum dots was investigated and it was theoretically predicted that T_1 up to seconds is possible. It was demonstrated that the transport gap in graphene ribbons (on a BN substrate) are edge related and are not determined by the substrate or the quality of the ribbon.

CONGRAN partners:

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- **Klaus Ensslin**
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Epitaxial Graphene Transistor (EPIGRAT)

EPIGRAT was aimed at synthesising and characterising epitaxial graphene layers on silicon carbide suitable for future electronics using different growth techniques, and at the development and characterisation of high frequency transistors and other devices based on these materials. Interactions of graphene with substrates have been investigated, including optimisation of graphene growth conditions and improvement of electronic mobility of carriers in graphene.

In EPIGRAT, epitaxial graphene on SiC is fabricated by the sublimation technique, optimised in the project, and by chemical vapour deposition (CVD). The latter method, evolved within the project and patented, provides key advantages over Si sublimation, because it is much less sensitive to SiC surface defects, resulting in high room temperature electron mobilities and enables the controlled synthesis of a specific number of graphene layers with a defined doping level on both Si and C polarities. Optimisation of Hydrogen intercalation into epitaxial graphene grown on the Si face led to high

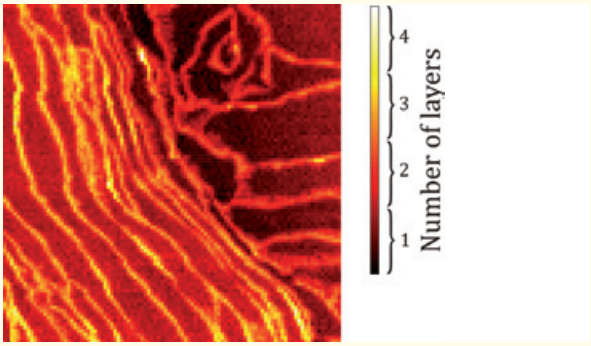

Figure 3.
Reflectivity map of graphene (see text)

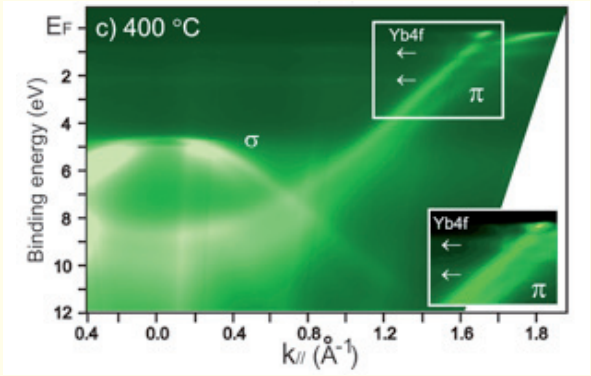

Figure 4.
Band structure along the Γ-K direction for a graphene sample after Yb deposition and heating at 400°C.

mobilities of 4000 cm^2/Vs at RT for p-type concentrations of 10^{13} cm^{-3}.

Fabrication and characterisation of field effect transistors by using epitaxial graphene on SiC substrate were studied. Fabrications were carried out by using optical and electron beam lithography techniques. Gate dependent modulation on current-voltage characteristics of graphene was observed after DC measurements. RF measurements were performed to test the performance of the devices, and it was shown that the graphene transistors can operate at ~8 GHz.

A technique to measure the velocity field characteristic versus electric field is demonstrated on as-grown and H-intercalated graphene. Van der Pauw, coplanar microbridge, and coplanar TLM structures are fabricated in order to assess the carrier mobility, carrier concentration, sheet resistance, and contact resistance of both epi-materials. These measurements are then combined with dynamic I/V measurements to extract a velocity field characteristic. The saturated electron velocity measurements indicate a value of 2.3·107cm/s for the as-grown material and 1.4·107cm/s for the H-intercalated material at 300K.

The reflectivity of graphene on a substrate such as SiC in the visible region of the spectrum varies

proportionally with the number of graphene layers. Using this property, we have developed a fast high resolution micro-Raman and reflectivity mapping system, which allows recording of the reflected light simultaneously with collecting the Raman spectrum. The map in the figure represents the number of layers on a 30 x 30 μm^2 square. The reflection mapping alone is much faster than Raman mapping.

Intercalation of Yb in epitaxial graphene grown on Si-face SiC shows that Yb forms silicide at the interface, and decoupled/transformed the buffer layer into another graphene layer as manifested by the absence of buffer layer spots in the μ-LEED pattern and the appearance of an additional π band in the ARPES spectra, respectively. The Yb 4f levels located close to E_F remained core like.

EPIGRAT partners:

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Graphene-based systems for spintronics: Magnetic interactions at the graphene/3d metal interface (SpinGraph)

SpinGraph has been dealing with transport properties in graphene, such as spin-dependent transport ('spin filtering') in graphene-metal contacts sandwich layers, whereby graphene can cause a large spin anisotropy in electron transmission. When grown as a 'nano-mesh' on lattice-mismatched metals such as rhodium and iridium, graphene can provide an exciting template for the growth of size-selected ferromagnetic clusters. Surface-sensitive probes, such as scanning tunnelling microscopy and photoelectron spectroscopy, and x-ray magnetic circular dichroism and magneto-optical Kerr effect studies to characterise the magnetic properties, have been combined with theoretical studies of the interface and cluster systems, yielding an exciting progress in this field.

Even though the unique transport properties of graphene are masked when it is grown on metals, this class of systems is important, from a basic physics as well as applications-oriented point of view, as growth templates, contacts, or – in the case of graphene on ferromagnets – for spintronics applications. Graphene-metal interaction varies in strength, from almost free-standing (on Pt and Ir) to strongly reacted on Ni and Co. These latter systems deserve particular interest since they could provide a spin filter because of the overlap of minority spin states with the "Dirac" states in graphene at the Fermi level. We use x-ray photoemission and –absorption to study the valence level electronic structure and magnetic interactions of graphene on metals, and examine the structural properties by electron diffraction and scanning tunneling microscopy. The well-known process of metal intercalation in between graphene and a magnetic or non-magnetic substrate offers opportunities to prepare novel, metastable metal-graphene arrangements, for example in a strained configuration, and to investigate ferromagnetic sandwich structures from a single atomic layer upwards. Intercalated iron films in between graphene and a nickel substrate, for example, strengthen the magnetic moment transferred from the substrate to graphene's carbon atoms (as shown in Figure 5). Such data are complemented by an in-depth study of the electronic states in the valence region, and are interpreted through state-of-the-art density functional calculations.

Graphene on metals often forms moiré structures because of the lattice mismatch between substrate

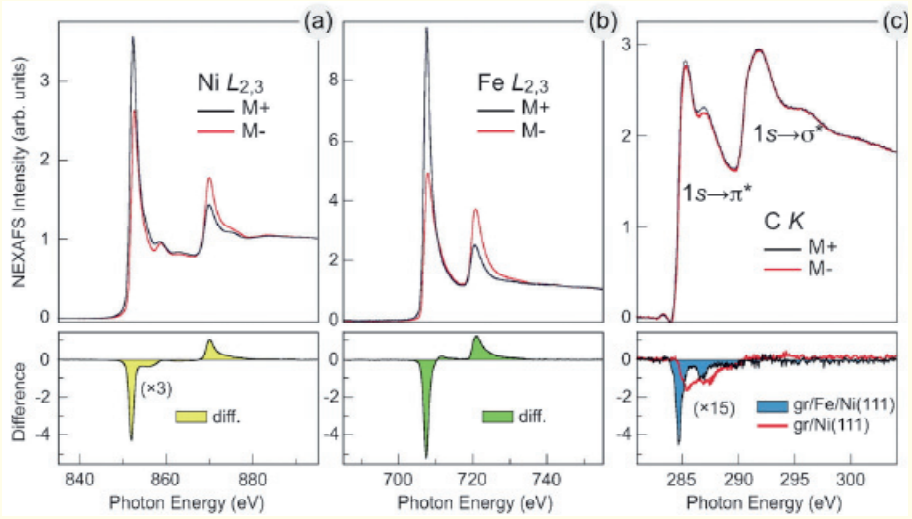

Figure 5.
XMCD spectra of the graphene/ 1 ML Fe(111)/Ni(111) system, showing the transfer of magnetic moment onto the p states of the carbon atoms in graphene.

and overlayer; these may serve to produce artificialon Ir(111), resulting in a lattice-mismatched graphene/Ni system. The intercalated Ni layer leads to a pronounced buckling of the graphene film, which enhances the interaction as reflected in angle- resolved photoemission spectroscopy, showing a clear transition from a nearly undisturbed to a strongly hybridised graphene π-band. This can be used to disentangle the two key properties which lead to the observed increased interaction, namely lattice matching and electronic interaction. Although the latter determines the strength of the hybridisation, we find an important influence of the local carbon configuration resulting from the lattice mismatch.

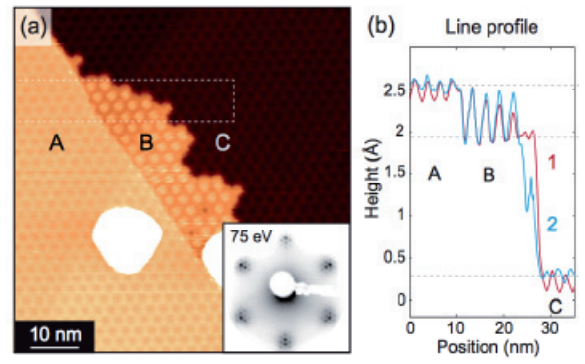

Figure 6.
STM overview of graphene on Ir(111) with a partially intercalated Ni submonolayer. Ni accumulates at step edges (B area) showing an increased moiré corrugation. These areas have reduced mean apparent heights (see line profile).

SpinGraph partners:

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- **Harald Brune**
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Graphene on SiC wafers for high performance RF transistors (Graphic-RF)

Graphic-RF was focused on transport properties of graphene and its possible applications in integrated electronics, dealing with such open questions as the possibility of growing a few high-quality layers of graphene on SiC considering different polytypes and different crystal orientations on large diameter wafers, development of novel nanocharacterisation methods and a multiscale approach to simulate the transport properties considering atomistic models as well as semi-empirical continuum models. Novel high mobility graphene field-effect devices for future high-frequency electronics applications have been demonstrated.

Graphene epitaxially grown on Silicon Carbide (GrSiC) substrates could represent a viable route towards the integration of present and future

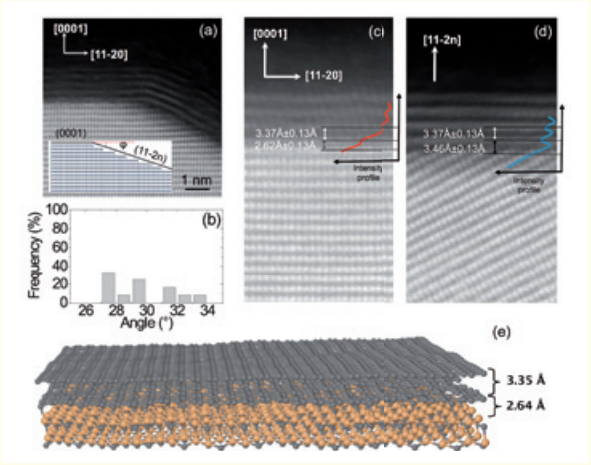

Figure 7.
(a) HAADF atomic resolution STEM image of a step facet in the EG/4H-SiC sample cross sectioned along [11-20]. (Inset) Schematic of the epitaxial graphene grown on a (11-2n) SiC (b) statistical distribution of the angle ϕ on the analysed specimens. (c) Intensity profiles orthogonal to the (0001) surfaces (d) Intensity profiles orthogonal to the (11-2n) surfaces. The carbon buffer layer, visible on the planar (0001) surface, becomes quasi-freestanding graphene on the (11-2n) surface. (e) Ab initio simulations of the (0001) interface. Si atoms are represented as large brown balls, C atoms as smaller grey-coloured balls

nanoelectronics. This intriguing system is formed by one of the most promising materials for next-generation electronic applications (Graphene) and the SiC bulk semiconductor, which has already substituted Si in many commercial applications. When the GRAPHIC-RF team started its activity within the CRP, the GrSiG was recently discovered. At that time, different challenging issues were still not fully understood and numerous questions were raised:

i) Is it really graphene and when? If it is not, how can it be transformed into graphene?
ii) Is polytypism of SiC a resource or a problem for GrSiC?
iii) How are structural properties related to the electronic/phononic characteristics and the electronic transport in GrSiC?
iv) How could it be integrated in a device manufacturing flow?
v) Do such devices improve the performance with respect to conventional ones?

Relevant advancements in GrSiC science and technology have been achieved with the fundamental contributions of the GRAPHIC-RF team. We now know that:

- GrSiC grown on the hexagonal polytypes of SiC (6H and 4H) has many properties in common with free-standing graphene, but exhibits also some peculiarities depending on the substrate orientation.
- The electronic properties of GrSIC on the Si face of 6H- and 4H-SiC strongly depend on the presence of a C buffer layer strongly bound to the substrate below the graphene layers. Atomic intercalation can turn the buffer layer into quasi-free-standing graphene. The graphene/SiC Schottky barrier can be tailored by the intercalation and vertical devices based on the engineering of such interface can be envisaged.

When grown on off-axis SiC (0001) substrates, the buffer layer delaminates at the step facets, turning into quasi-freestanding graphene (Figure 7) and exhibits different structural and electronic properties than on the basal plane. New devices can be obtained by nanoribbons grown on facets. High quality epitaxial graphene can be efficiently grown also on the cubic SiC polytype. Moreover, the C-face of SiC as well as the nonpolar (11-20) and (1-100) faces are ideal substrates for the growth of buffer-layer-free epitaxial graphene. By growing on different SiC polytype/surfaces we can fine-tune some of graphene's electronic properties in a controllable manner.

- GrSiC can be efficiently integrated in the standard device manufacturing flow of semiconductor fabs, whereas high mobility grafene field effect devices (gFETs) on such substrates have been demonstrated. GrSiC represents a material of choice for future RF applications, due to its high carrier mobility.

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Electrical and Optoelectronic Graphene Devices (ELOGRAPH)

ELOGRAPH was focused on exploration and realisation of graphene nanoribbon (GNR) devices, the direct and tuneable band-gap of which makes it attractive for future optoelectronic applications (e.g. on-chip optoelectronic data-links or infrared photo-detectors for thermoelectric energy harvesting of residual heat). The areas of activity included GNR device fabrication, optoelectronic characterisation, and numerical simulations and modelling.

The ELOGRAPH CRP combines experimental knowledge, engineering sciences and theoretical expertise within an interdisciplinary research consortium to realise and to explore the potential of graphene devices. The direct band-gap and tune-ability of the band-gap in graphene nanoribbon (GNR) devices renders graphene-devices as promising candidates for future electronic and optoelectronic applications. This CRP has focused on graphene device fabrication, its electronic characterisation, numerical simulation and modeling as well. An analytical model for line-edge roughness limited mobility of graphene nanoribbons has been developed. The role of line-edge roughness scattering on the electronic properties of graphene nanoribbons has been numerically investigated. The non-equilibrium Green function formalism, along with an atomistic tight-binding model, has been successfully employed. Using these models, one can appropriately select the geometrical parameters for optimising the performance of GNR-based electronic devices. Regarding the experimental work, a novel method to fabricate graphene transistors directly on oxidised silicon wafers, without the need to transfer graphene layers, has been demonstrated. By means of catalytic chemical vapour deposition (CCVD) the self-aligned grown monolayer graphene field-effect transistors (MoLGFETs) and bilayer graphene transistors (BiLGFETs) are realised directly on oxidized silicon substrate. Transfer-free grown MoLGFETs exhibit the expected Dirac point together with the typical low on/off-current ratios. In addition, however, the BiLGFETs possess unipolar p-type device characteristics with an extremely high on/off-current ratio up to $8 \cdot 10^7$ exceeding previously reported values by several orders of magnitude (cf. Fig. 8). A novel graphene device model based on Schottky-barrier contacts has been proposed to explain these results. Besides the excellent device characteristics, the complete CCVD fabrication process is silicon CMOS compatible. This experimental breakthrough offers a simple and low-cost integration of graphene devices for nanoelectronic applications in a hybrid silicon CMOS environment for the first time.

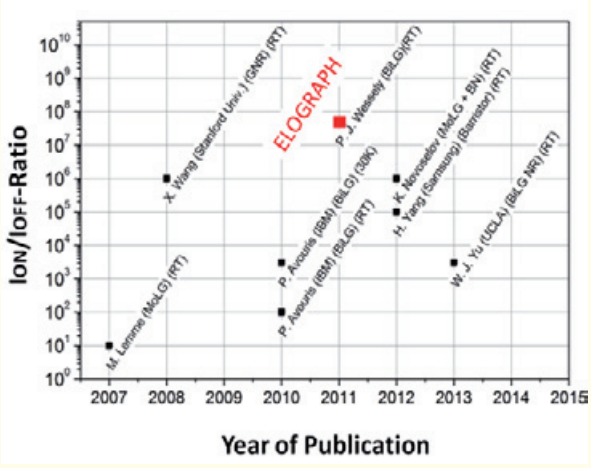

Figure 8.
I_{ON}/I_{OFF} current ratios of graphene field-effect devices as published by leading research groups in comparison with ELOGRAPH results.

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- **Thomas Zimmer**
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ENTangled spin pairs in graphene (ENTS)

ENTS explored applications of graphene in spintronics and quantum information processing. The generation of spatially separated entangled pairs of spins in hybrid systems between superconductors and graphene open novel original possibilities. Advances in this area are essential for enabling new quantum information experiments in solid-state systems.

Our goal was set to generate and manipulate spin-coherent single-electron and paired electron states in various graphene settings. A natural source of spin-entangled electron pairs is a BCS superconductor. If in contact with graphene, electron

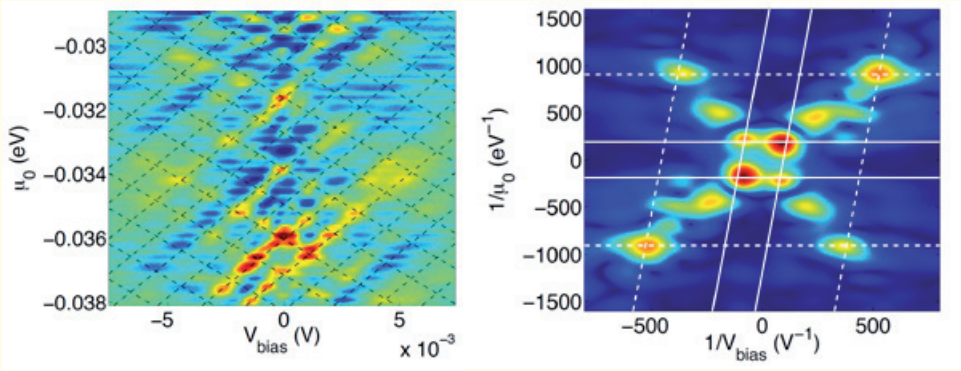

Figure 9.
Transport interferences measured on a suspended graphene sample depicted on the chemical potential vs bias voltage plane (left) and in the Fourier space (right). Both length and width quantisation are visible as indicated by the solid and dashed grids on the right.

pairs (Cooper pairs) can be injected into graphene. Utilising Coulomb interaction, the electrons of a pair can be split, thereby realising a solid-state electron entangler.

Two main avenues for realising the Cooper pair beam splitter were pursued: ribbons structures on substrates and quantum dots of suspended graphene. Initially, electrical transport measurements showed that, in regular ribbons, transport is based on inelastic hopping, which is detrimental for spin coherence. Techniques for making better quality ribbons were developed and were shown to work by observation of quantum Hall state in about 50 nm wide ribbons. Cooper pair splitting in the quantum Hall regime was treated theoretically and it was demonstrated that the charge 2e splits to opposite edges, which leads to characteristic features under flux modulation.

Suspended samples with multiple gates and contacts were studied. Quantum interference effects were investigated and Fabry-Pérot interferences were observed as originating from reflection at graphene boundaries, pn-interfaces, and contact interfaces. In these studies, we also analysed the relation between current-current correlations and the conductance. The correlations were clearly seen to depend on the energy level structure of the graphene sample. Gap formation and edge states in a high field were investigated and characterised in terms of conductance and, partly, shot noise.

Superconducting injectors were realised at various transparencies. The most detailed studies were performed at high transparency using Andreev interferometry. We observed a crossover in the gate-voltage-dependent Andreev conductance oscillations which exhibit scaling predicted for non-interacting electrons, except at low gate voltages. These results indicate that electron-electron interactions should be taken into account when considering the proximity effect in graphene.

To demonstrate two-particle correlations, Hanbury-Brown Twiss (HBT) type of correlation experiments were performed on graphene ribbons in cross-shaped geometry. Such HBT experiments are sensitive to exchange correlations in the central area (50 x 50 nm^2) of the graphene cross. Our results indicate that the HBT exchange effects can be tuned by the charge density in graphene. This provides a clear demonstration that interference effects can influence the results of cross correlation experiments in graphene.

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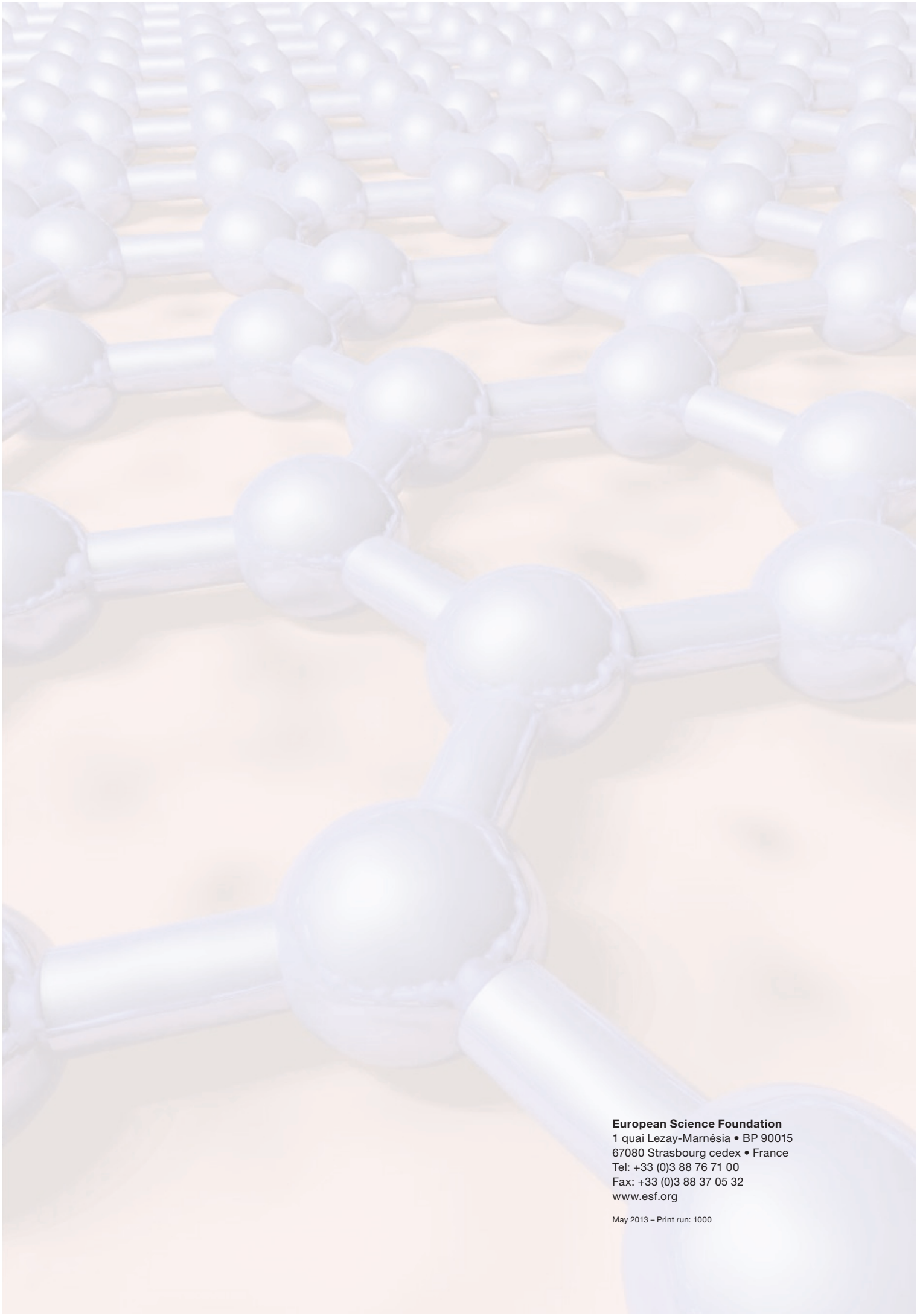

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