Conference on Modern Concepts and New Materials for Thermoelectricity

11 - 15 March 2019
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# Magnetic-Field-Induced Metallicity in Ultraquantum Graphite


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In collaboration with:


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**Robson R. da Silva (Campinas, Brazil)**

# Landau level quantization and possible superconducting instabilities in highly oriented pyrolitic graphite

Y. K., V. V. Lemanov, S. Moehlecke, J. H. S. Torres

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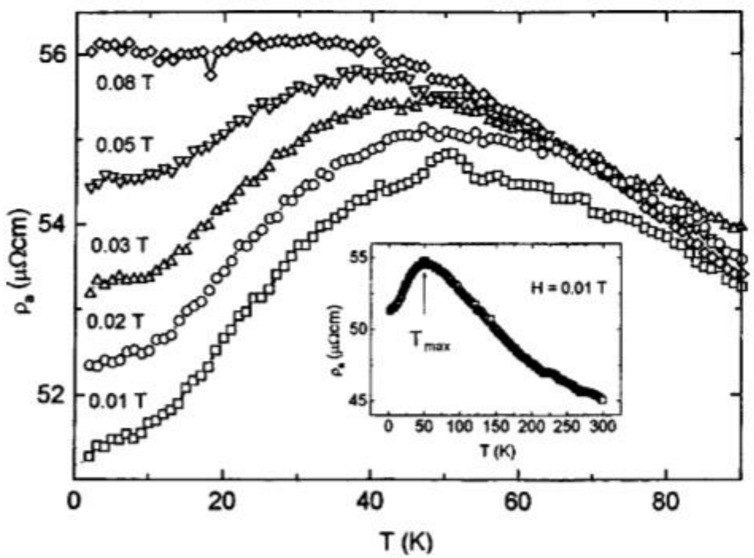
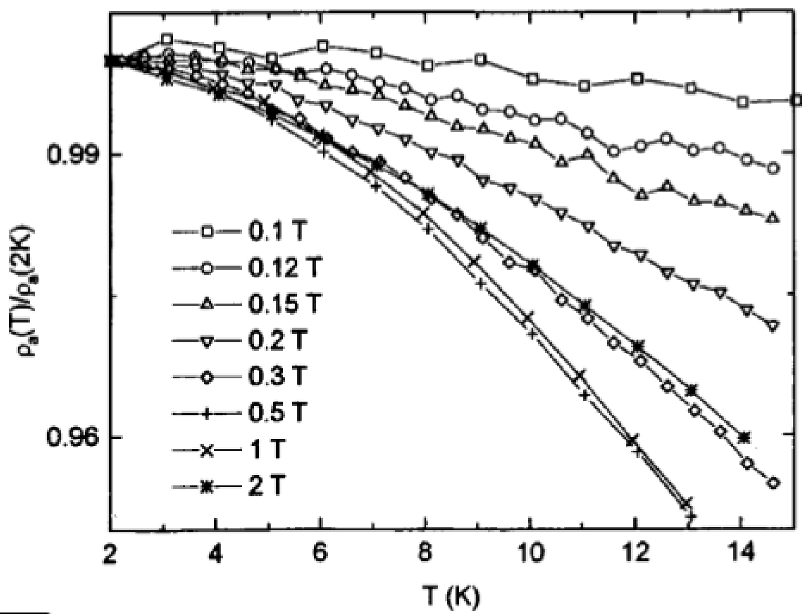
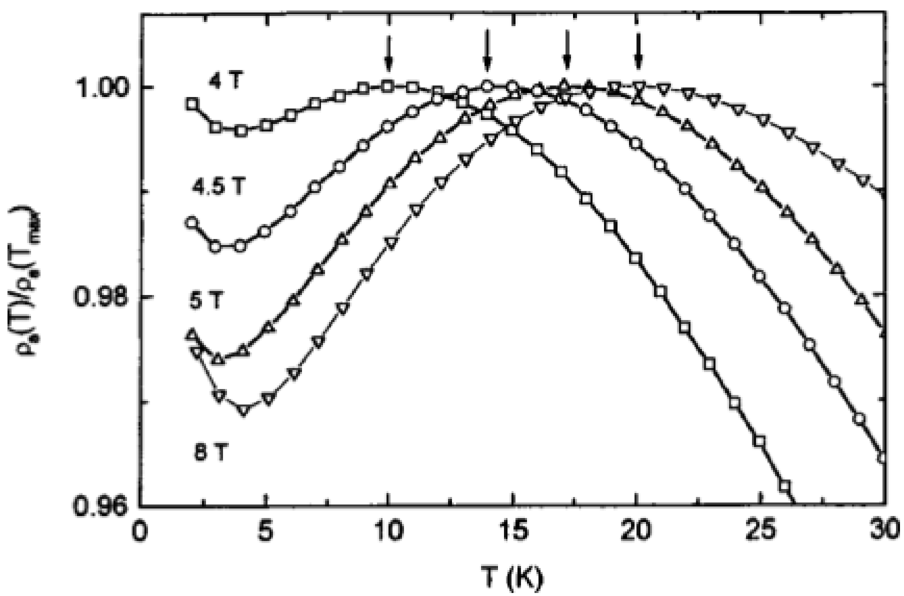
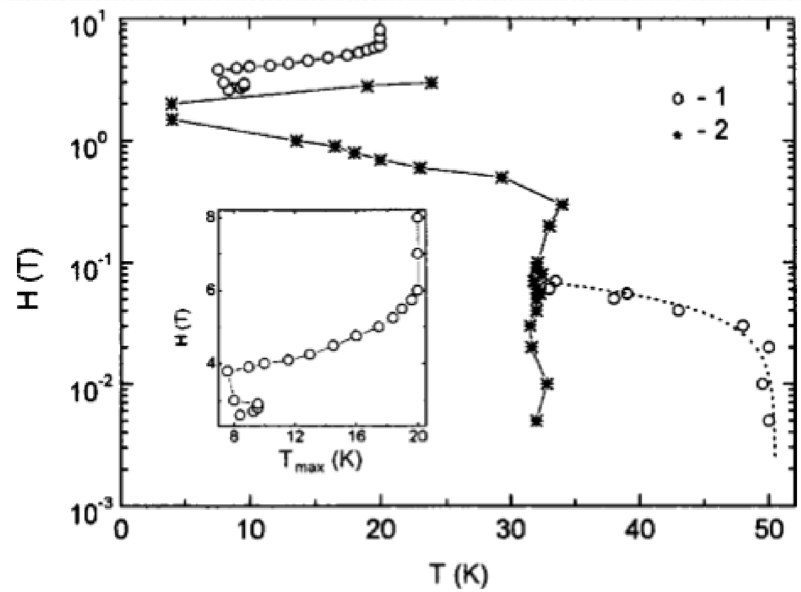
9

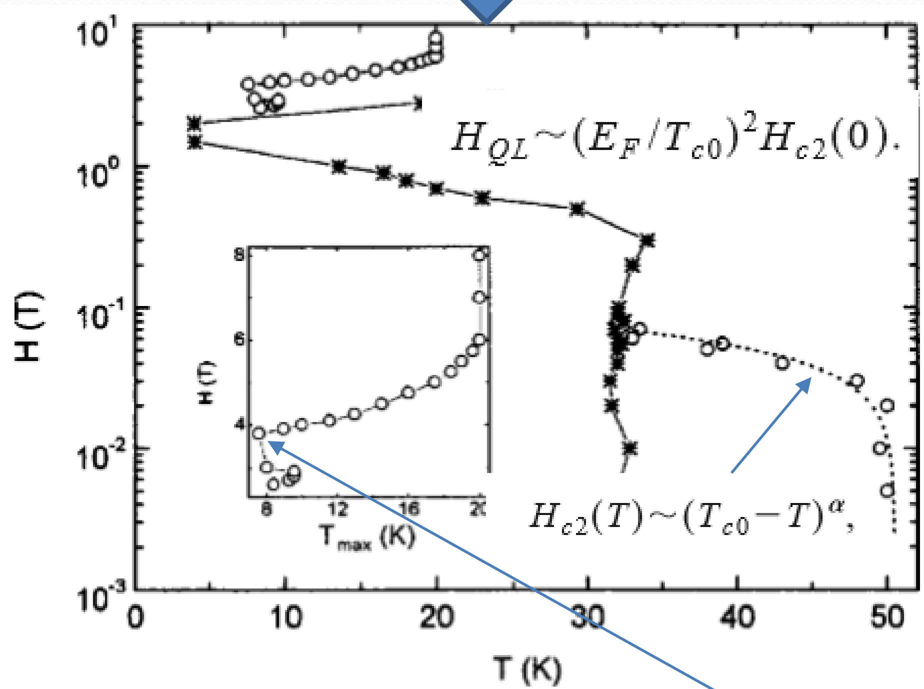


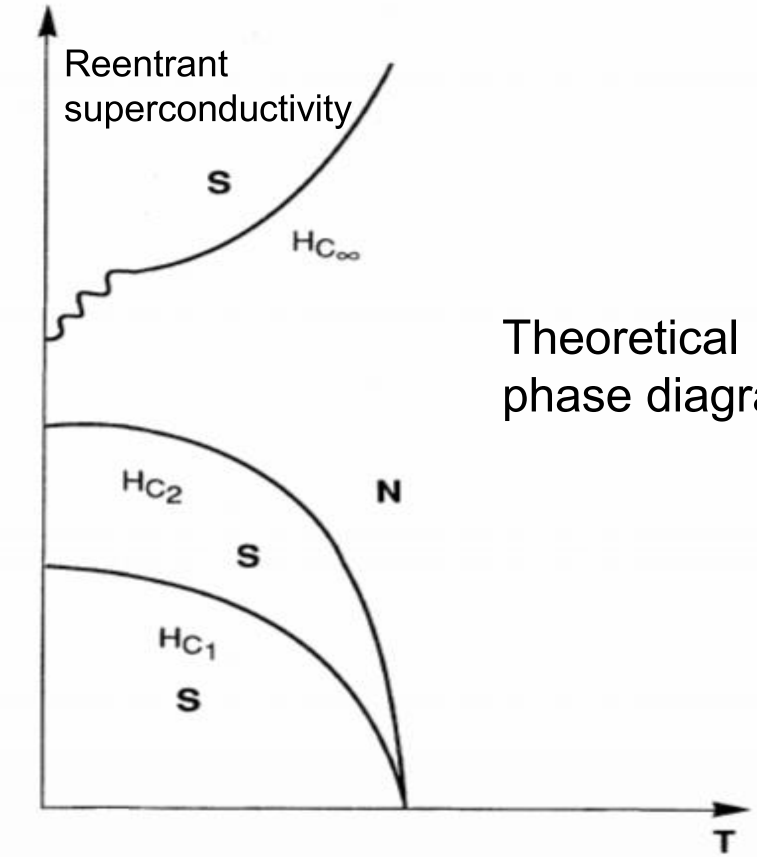



**GRAPHITE ´ 1999: Our experimental phase diagram**




**Taking, $T_{c0}$ = 50 K, $H_{c2}$ (0) = 0.1 T, and $E_F$ = 0.024 eV, one gets $H_{QL}$ ~ 2 T**




Theoretical phase diagram

M. Rasolt & Z. Tesanovic, RMP ´ 1992

## Superconducting correlations in quantizing field (both 2D and 3D ES):

$$T_{SC}^{3D} \sim \Omega \exp[-2\pi l^2/N_1(0)V]; \; H > H_{QL}$$

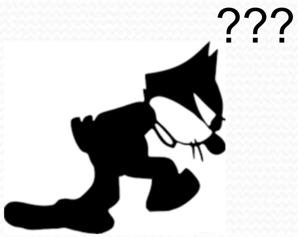
$2\pi l^2/N_1(0) \sim H^{-2}$, $l = (\hbar c/eH)^{1/2}$, **V is the attractive interaction,**

**$\Omega$ is the energy cutoff on V;**

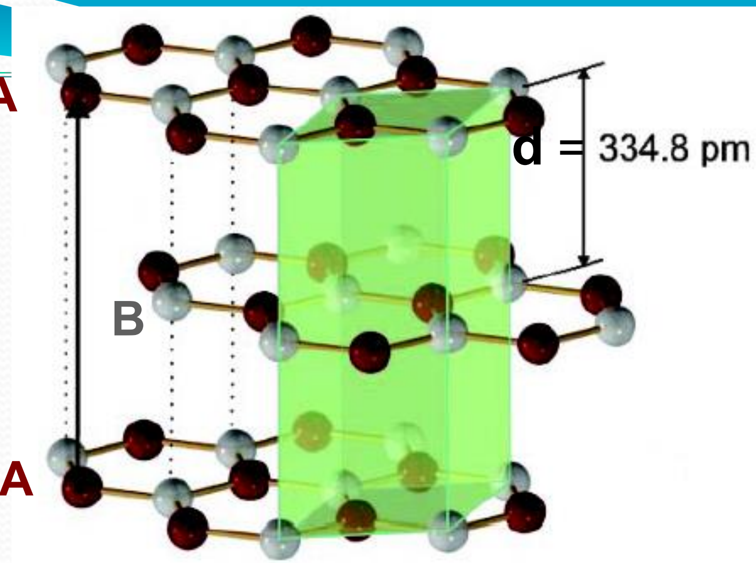
M. Rasolt and Z. Tesanovic, Rev. Mod. Phys.'1992

$T_{SC}^{2D} \sim \hbar eHVN(0)/m^*c$ [A. H. MacDonald et al.'1993]

**$T_{max}(H)$ qualitatively agrees with theoretical predictions for $T_{SC}(H)$**



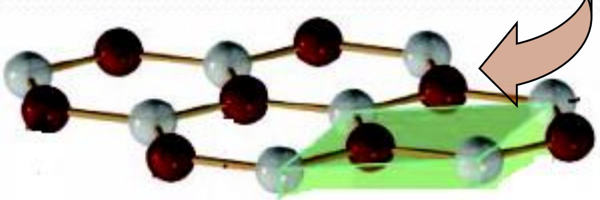
The crystal structure of graphite: layers of honeycomb lattices of carbon atoms

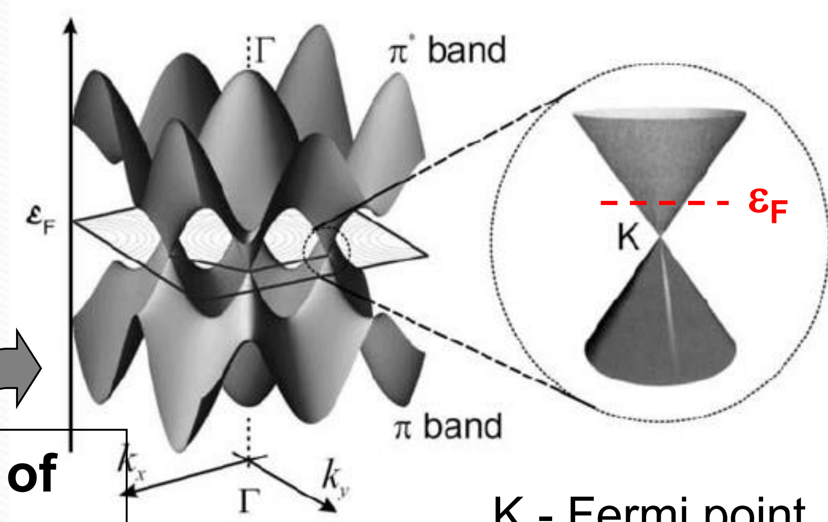



$n_e \geq n_h \sim 10^{18}\ cm^{-3}$,
$n_{2D} = n_{3D}(c/2) \sim 10^{11}\ cm^{-2}$
$m^*_e \approx m^*_h \approx 0.05 m_0$
$\mu \sim 10^6\ cm^2/Vs$

**Dirac-like cone spectrum:**
**$E(p) = \pm vp$; $v \sim t_{||} a/\hbar$, $v \cong 10^6$ m/s**
$t_{||} = 3.16$ eV

The single-atom thick graphitic plane is called "graphene"





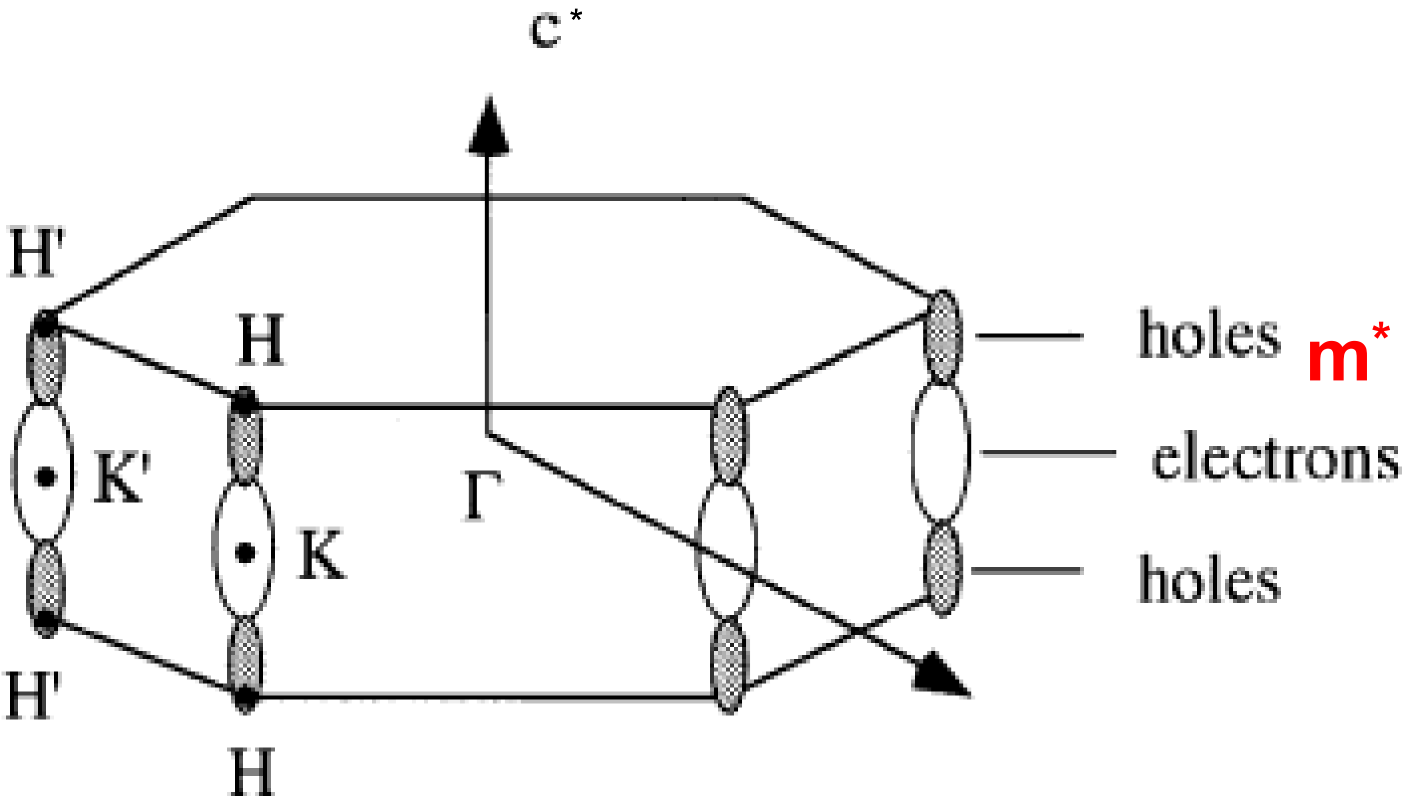

under doping

**Band structure of one graphitic layer**

K - Fermi point

According to Slonczewski-Weiss-McClure model (1955-1957), the interlayer hopping leads to $p_z$-spectrum dispersion with opening of cigar-like Fermi surface pockets elongated along the corner edge HKH of the graphite 3D Brilloin zone.

$t_\perp \approx 390$ meV



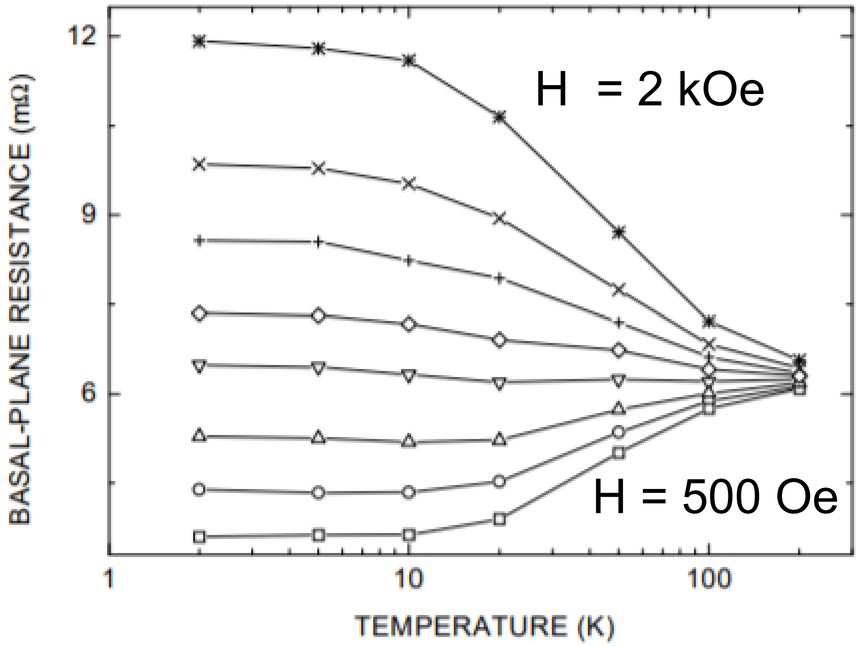

$m^* \sim 0.05 m_0$

$t_{\parallel} \approx 3.16$ eV (in-plane nearest-neighbor hopping energy)

$t_{\perp} = 5$ meV ... 0,39 eV ?!
(the c-axis hopping energy)

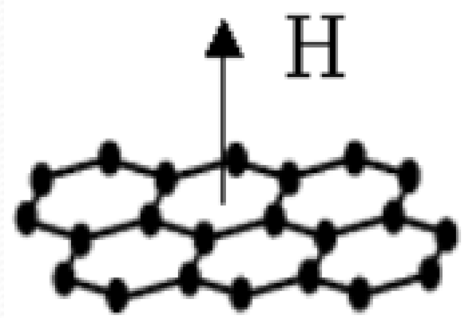
**$t_{\perp} \approx 5$ meV** **has been obtained from de Haas-van Alphen oscillations analysis by R. R. Haering & P. R. Wallace, J. Phys. Chem. Solids 3, 253 (1957) :**

**"We are thus led to the conclusion that graphite is essentially two-dimensional in structure …"**




H. Kempa, Y.K., F. Mrowka et al (2000)

Very similar to 2D superconductors



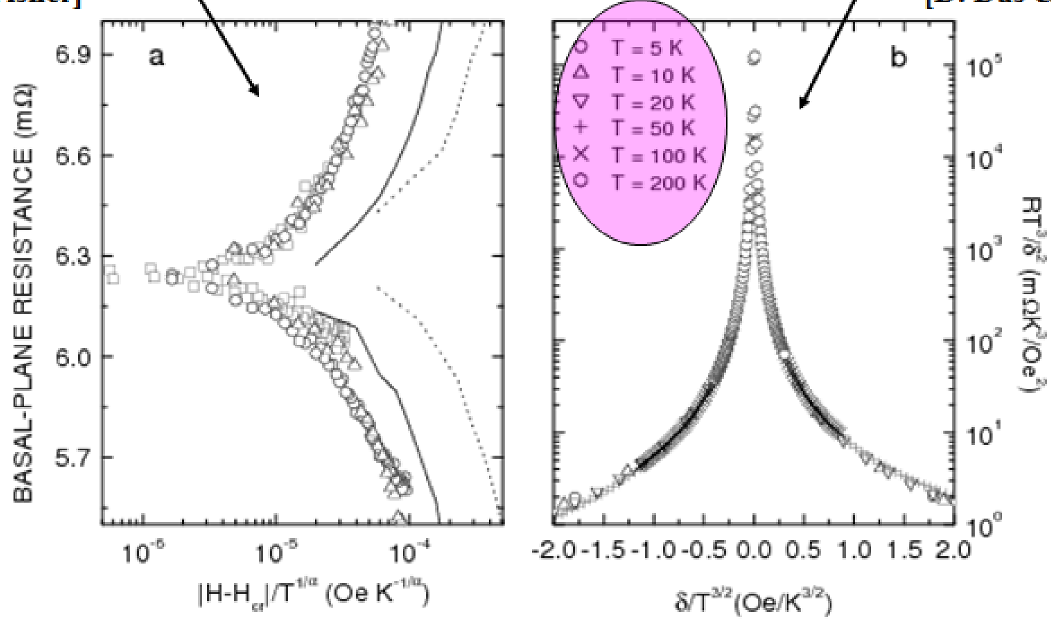

$R(\delta, T) = R_{cr} f(|\,\delta\,| / T^{1/z\nu})$

[M. P. A. Fisher]

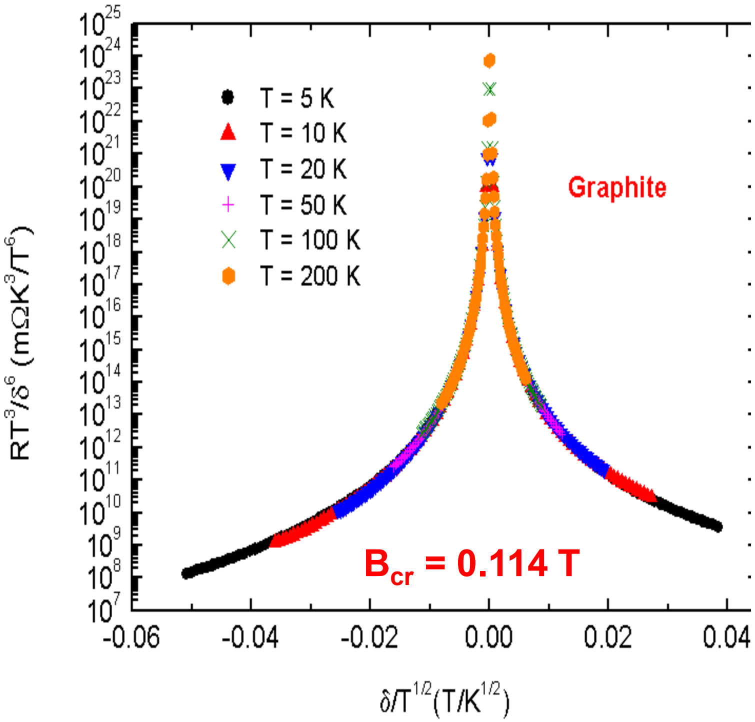
**GRAPHITE**

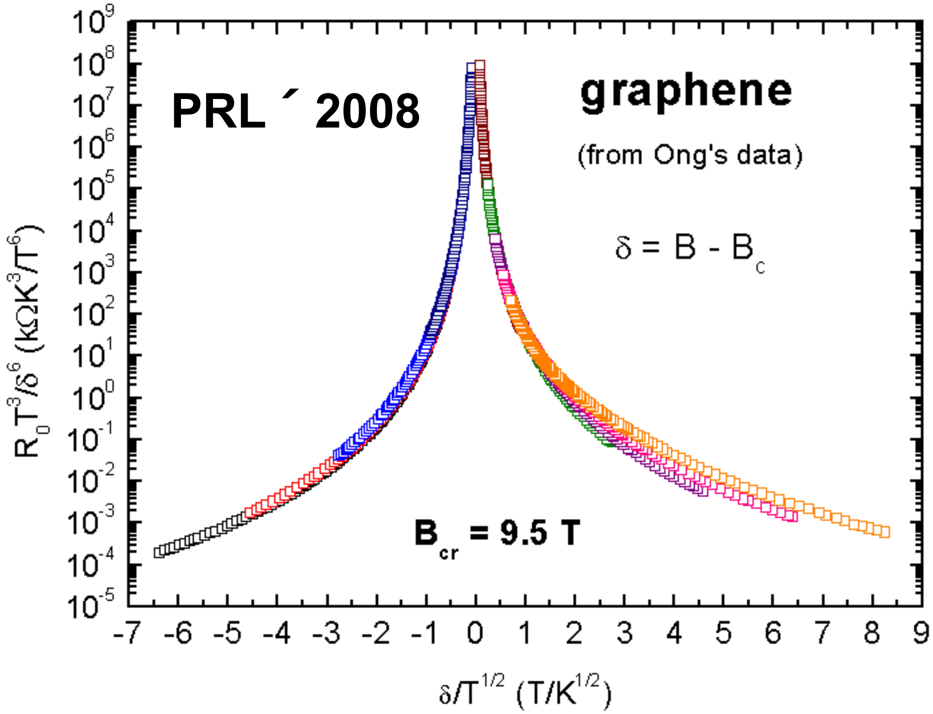
$R\left[\frac{T^{1/\nu z}}{\delta}\right]^{\nu(z+2)} = f(\delta / T^{1/\nu z})$

[D. Das & S. Doniach]



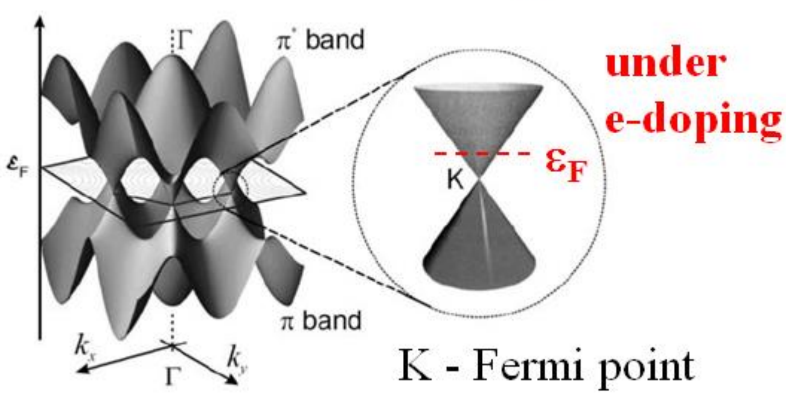
# Scaling analysis: graphite and graphene behave similarly




Dirac-like cone spectrum:

$$E(p) = \pm v_F p$$




**Before "graphene era" :**

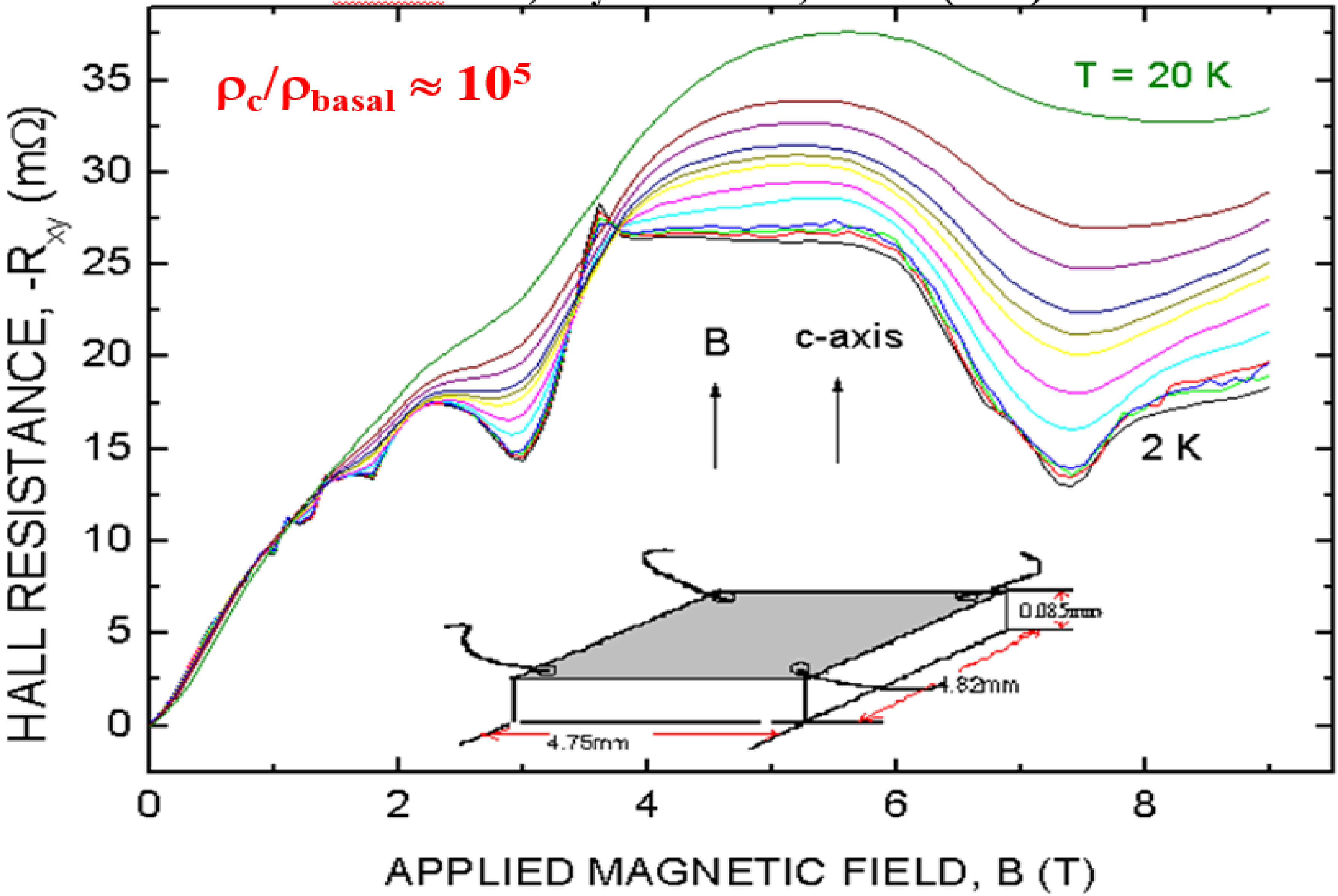
MIT in GRAPHITE:

**E. V. Gorbar, V. P. Gusynin, V. A. Miransky, I. A. Shovkovy, PRB 66, 045108 (2002).**

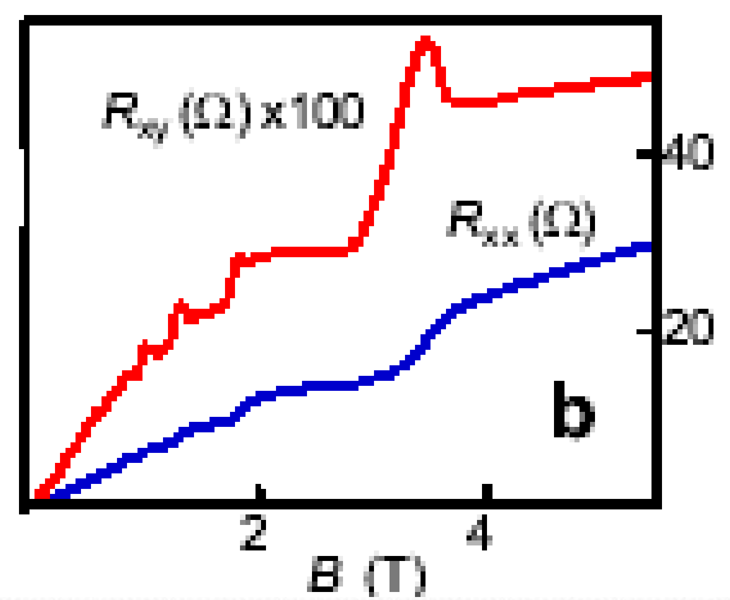
$$2\pi c n_{2D}/N_f|eH| \equiv H_c/H$$

# Finding of the Quantum Hall Effect in Graphite

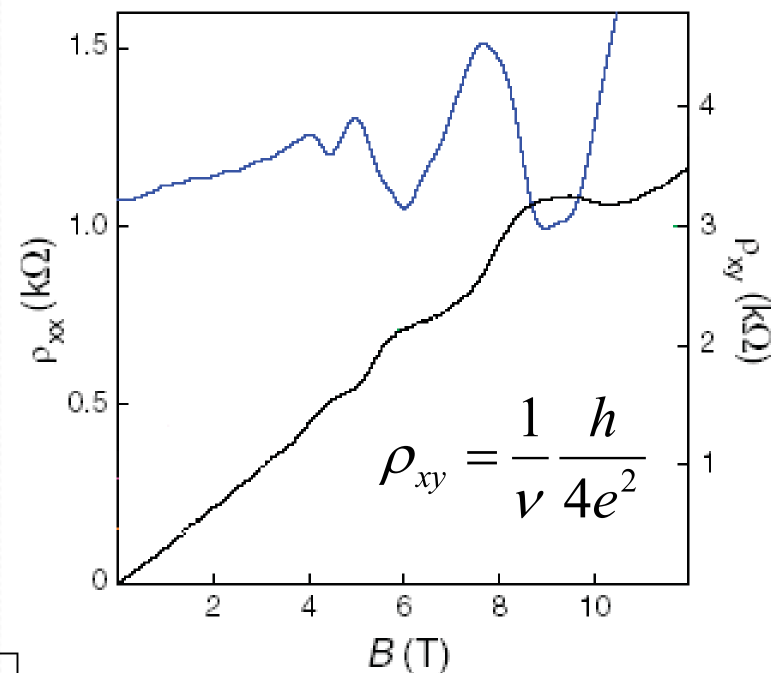
Y. Kopelevich et al., Phys. Rev. Lett. 90, 156402 (2003);
R. Ocaña et al., Phys. Rev. B 68, 165408 (2003).



The occurrence of QHE in graphite has been confirmed by other groups:
K. S. Novoselov et al., cond-mat/0410631; Science **306**, 666 (2004); T. Matsui et al., PRL **94**, 226403 (2005); and tbp; Y. Niimi et al., PRL **97**, 236804 (2006).




QHE measured for 8 μm thick HOPG sample at T = 0.3 K;
K. S. Novoselov et al., cond-mat/0410631

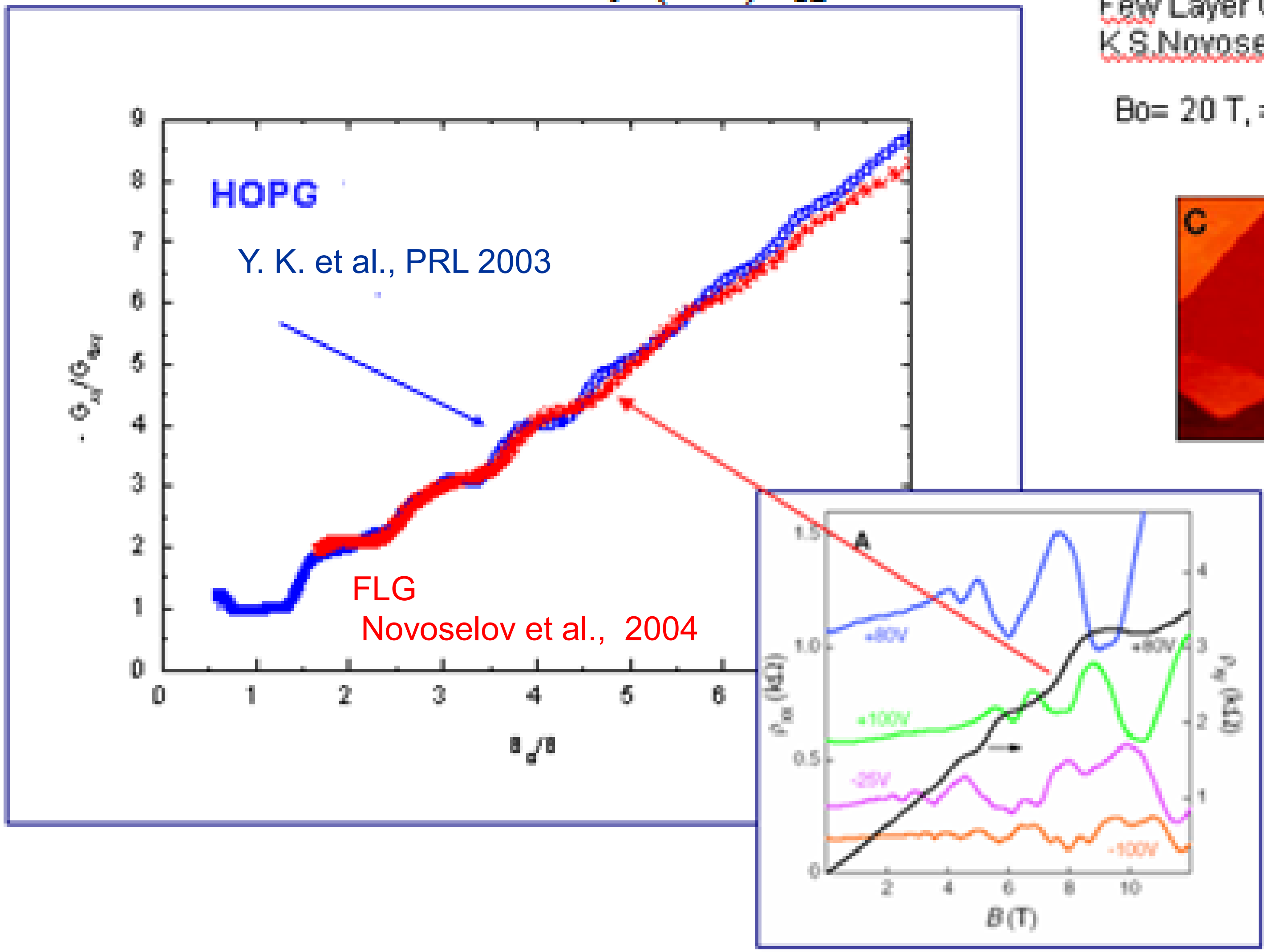



QHE measured for a few-layer thick graphite sample at T = 3 K;
K. S. Novoselov et al., Science'2004

# QHE: Graphite and multi-graphene

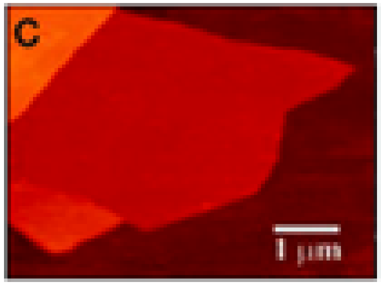
$B_0 = 4.68$ T

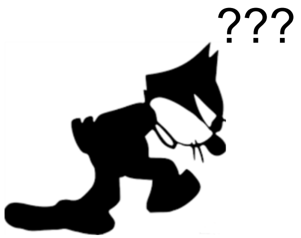
$$B_0 = (\pi\hbar/e)\, n_{2D}$$




Few Layer Graphite (FLG)
K.S.Novoselov et al., Science 2004

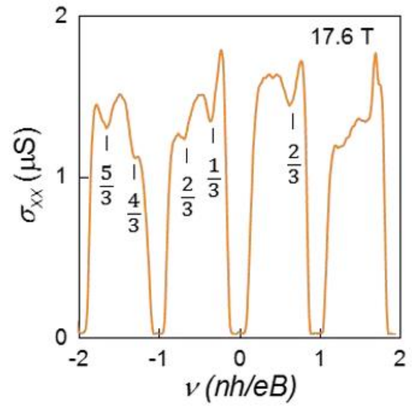
Bo= 20 T, => $n \sim 2\times10^{12}$ cm$^{-2}$

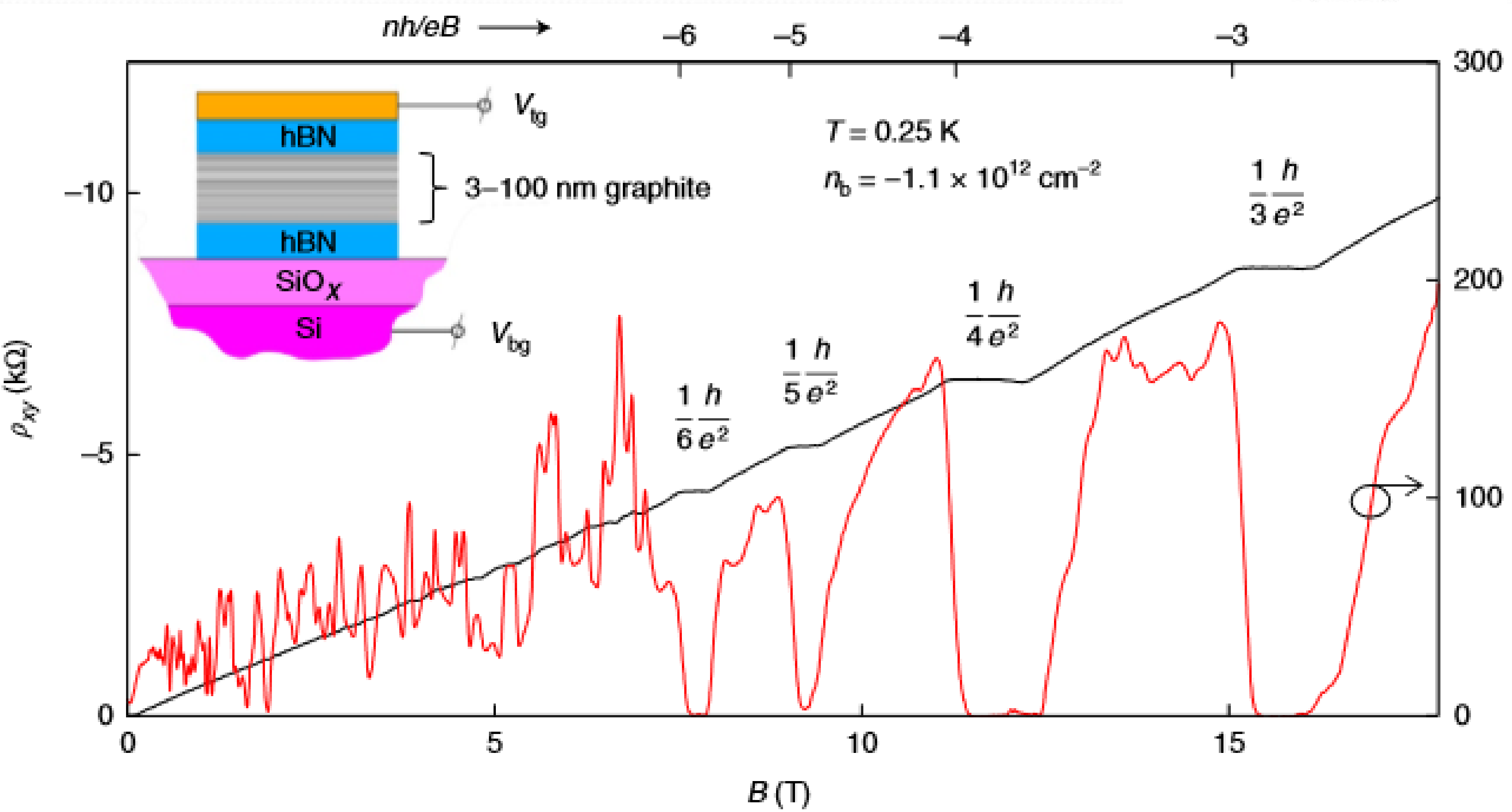


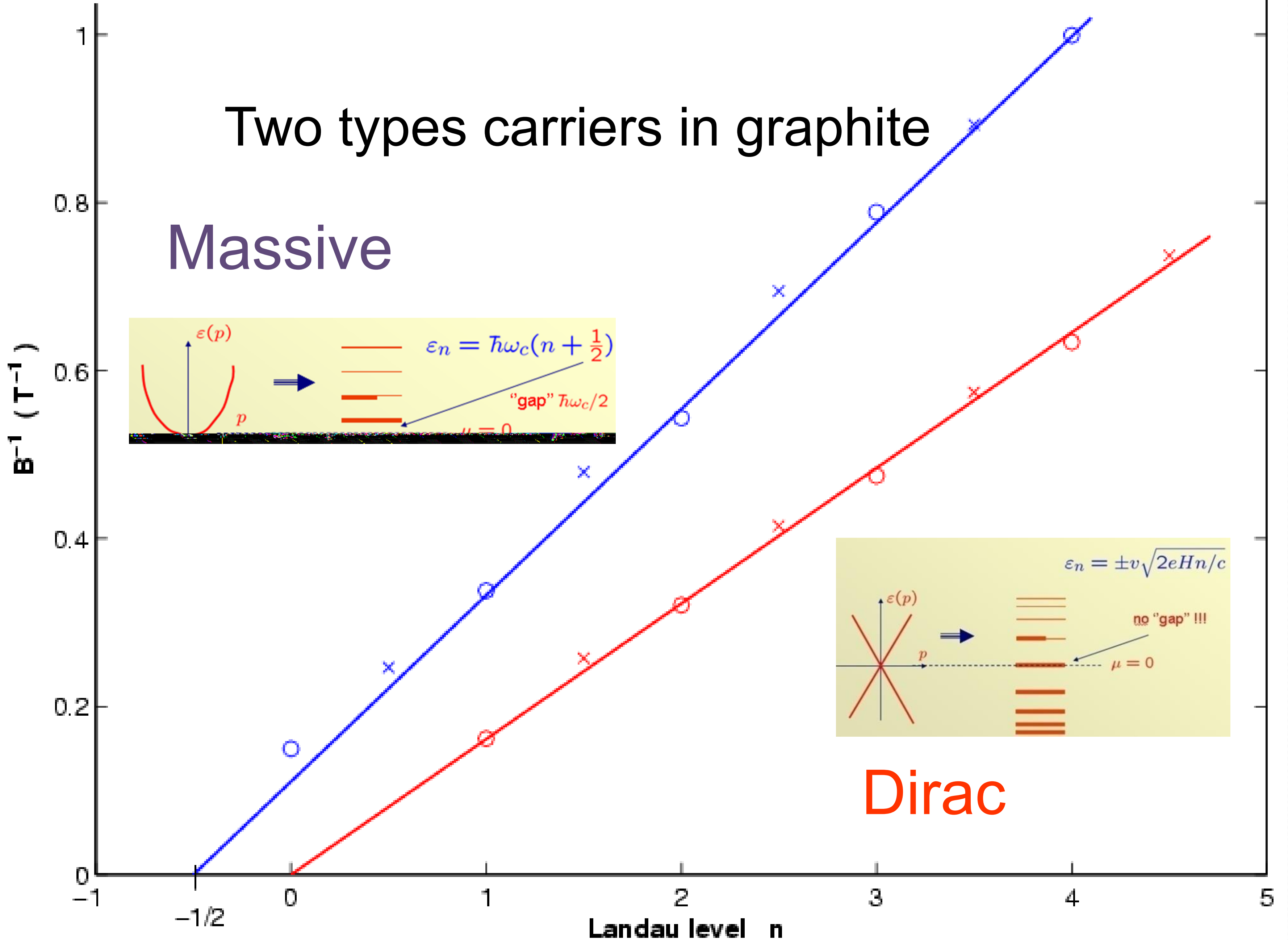
# Dimensional reduction, quantum Hall effect and layer parity in graphite films

J. Yin et al., Nature Phys. (March 2019)



I. A. Luk'yanchuk and Y. K. , PRL 93, 166402 (2004); ibid. 97, 256801 (2006)

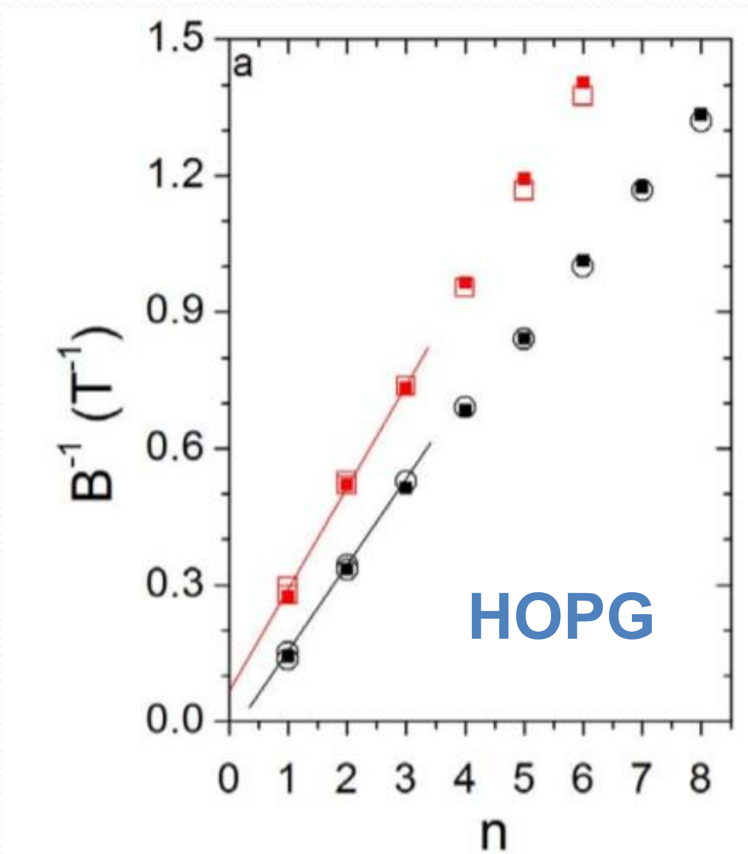


nature physics
SUPPLEMENTARY INFORMATION
DOI: 10.1038/NPHYS1437

# “Nernst effect and dimensionality in the quantum limit”

Zengwei Zhu[1,2], Huan Yang[1], Benoît Fauqué[1], Yakov Kopelevich[3] and Kamran Behnia[1]

**(2010)**




Open sumbols: $S_{xy}$ peaks

Solid symbols: minima in $R_{xx}$

**J. M. Schneider, M. Orlita, M. Potemski and D. K. Maude, Phys. Rev. Lett. 102, 166403 (2009).**

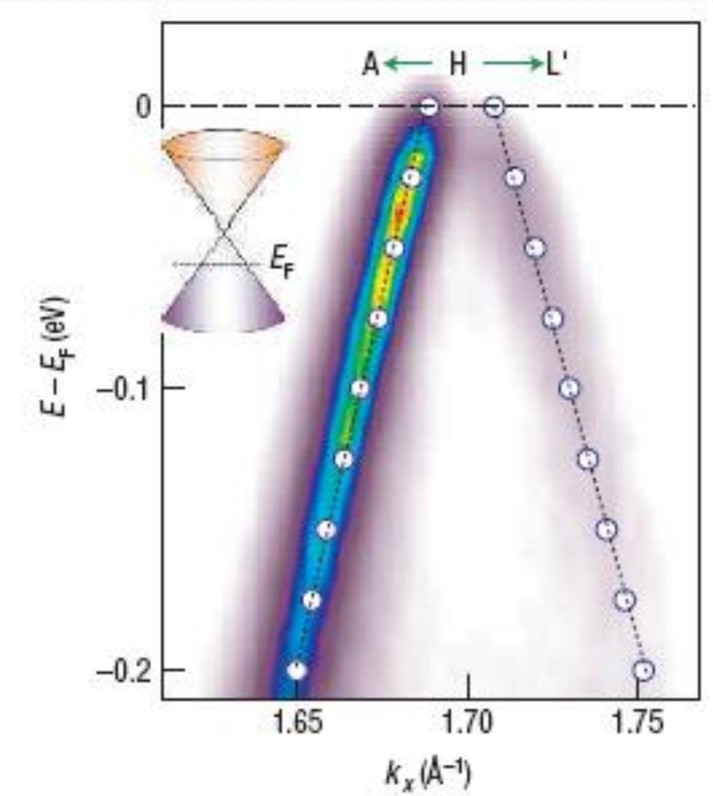
**See also I. A. Luk’yanchuk and Y.K. PRL(2010)**

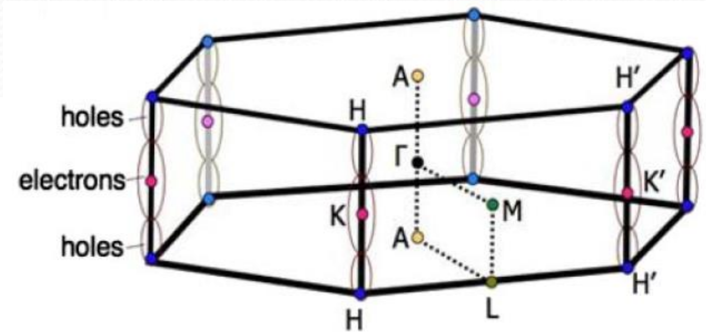
2D or not 2D ???

# Angle-resolved photoemission spectroscopy (ARPES): First direct observation of Dirac fermions in BULK GRAPHITE

**Nature Physics 2, 595 (2006)**

S. Y. ZHOU[1,2], G.-H. GWEON[1], J. GRAF[2], A. V. FEDOROV[3], C. D. SPATARU[1,4], R. D. DIEHL[5], Y. KOPELEVICH[6], D.-H. LEE[1,2], STEVEN G. LOUIE[1,2] AND A. LANZARA[1,2*]




Brilloin zone

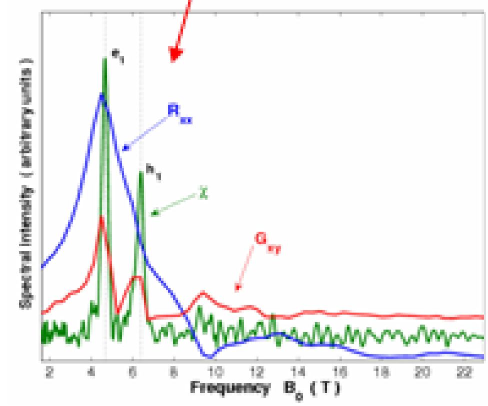
**Slonczewski-Weiss-McClure model**

Dispersion near the **H point** shows that low-energy excitations are Dirac fermions with the Dirac point slightly above $E_F$ $\Rightarrow$ **Dirac holes**.

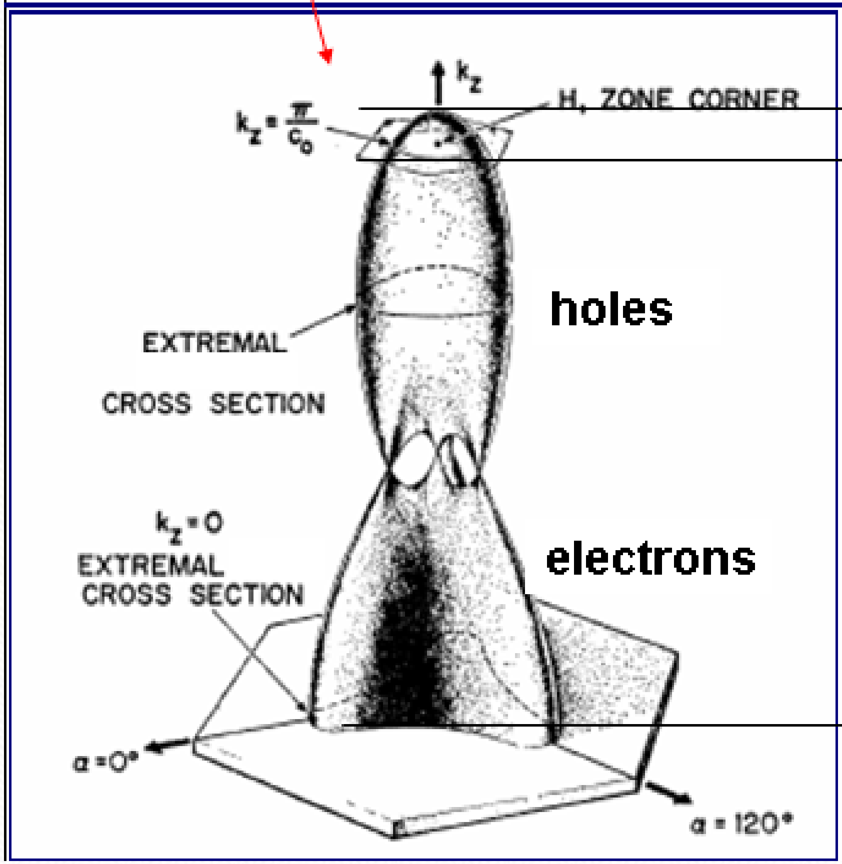
**K point:**
**massive electrons**

# Problems with band interpretation

Sh > Se ?




1) Se > Sh




2)

H: point

Dirac Spectrum

Phase volume ~0

*no Dirac Fermions*
*should be seen in experiment*

Normal Spectrum

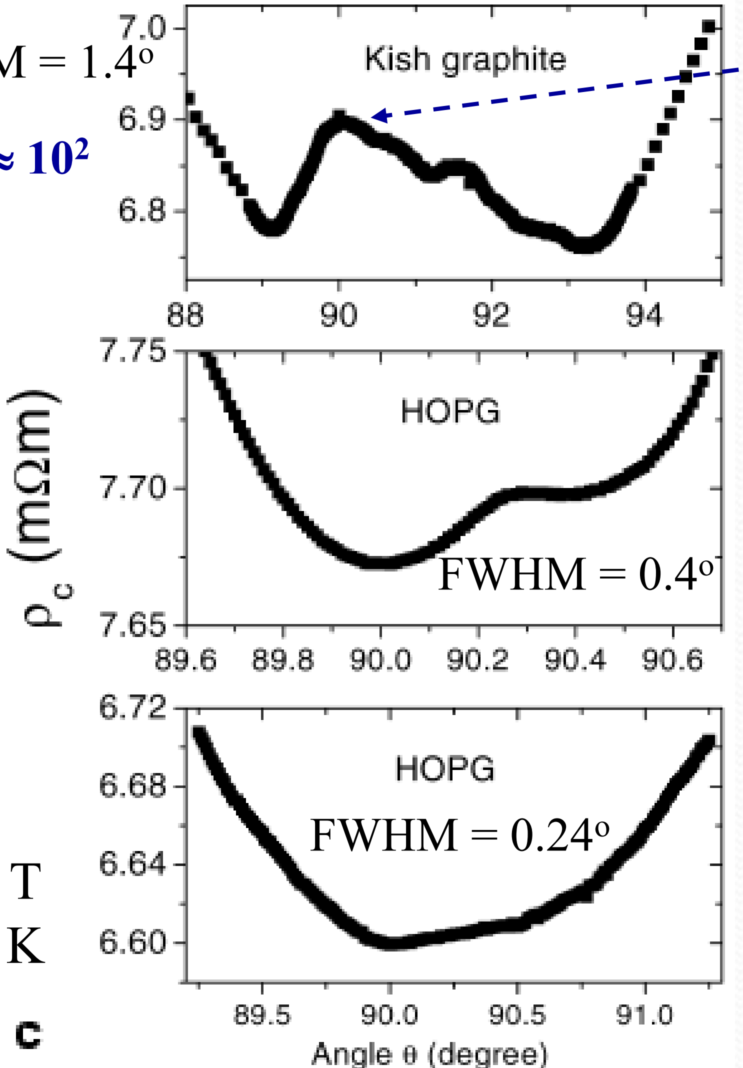
Another possibility:

Independent layers ???

# **Testing SWMC model:** INTERLAYER TRANSPORT (2D vs 3D)

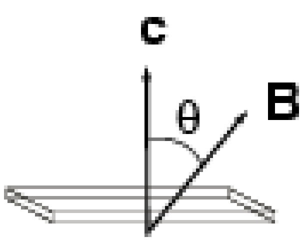
FWHM = 1.4°

$\rho_c/\rho_b \approx 10^2$




B = 9 T

T = 2 K




Resistivity peak ⇒ 3D Fermi surface
[R. H. McKenzie and P. Moses, PRL (1998)]

**No QHE was observed !**

**"Best quality" graphite samples:**
**(i) the resistivity peak is absent**
**(ii) QHE reveals itself**

$\rho_c/\rho_b \approx 10^5$

**No "resistivity peak"**
**⇒ No 3D Fermi surface ?**

Y. K., P. Esquinazi, J. H. S. Torres, R. R. da Silva, H. Kempa,
Advances in Solid State Physics **43**, 207 (2003), ed. by B. Kramer

**B. T. Kelly "PHYSICS OF GRAPHITE", APPLIED SCIENCE PUBLISHERS, LONDON and NEW JERSEY, 1981, page 294:**

**"The basic problem is that measurements of $\rho_c$ in the more perfect samples separate into two groups, one giving $\rho_c/\rho_a \sim 10^2$ and the other $\rho_c/\rho_a \sim 10^5$.**

**In general it is found that pyrolitic graphites exibit high ratios and natural single crystals and Kish graphite low ratios of $\rho_c/\rho_a$ ..."**

**"It was pointed out long ago that misorientation in the sample, short circuiting by lattice defects are likely to reduce the measured value of $\rho_c$ compared to the true value".**

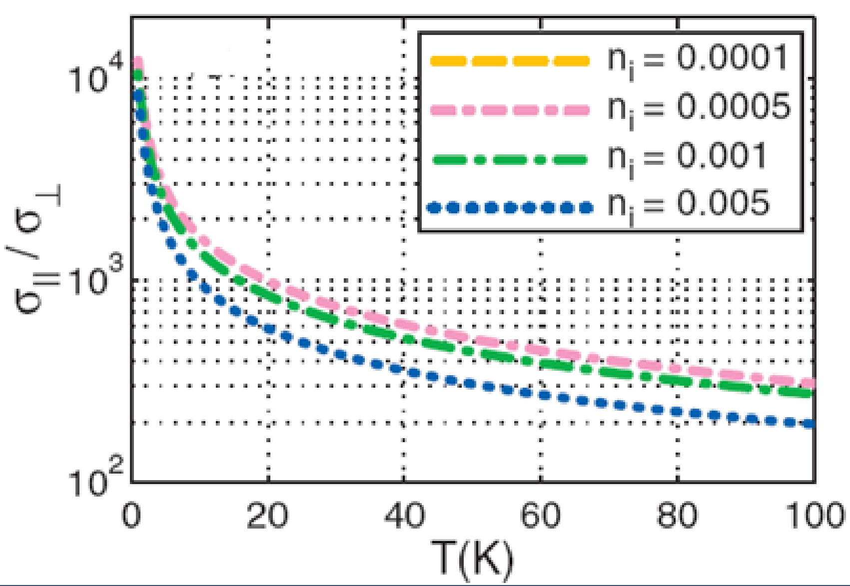
"Electron Properties of Graphene Multilayres" by J. Nilsson, A. H. Castro Neto, F. Guinea, and N. M. R. Peres, PRL **97**, 266801 (2006):

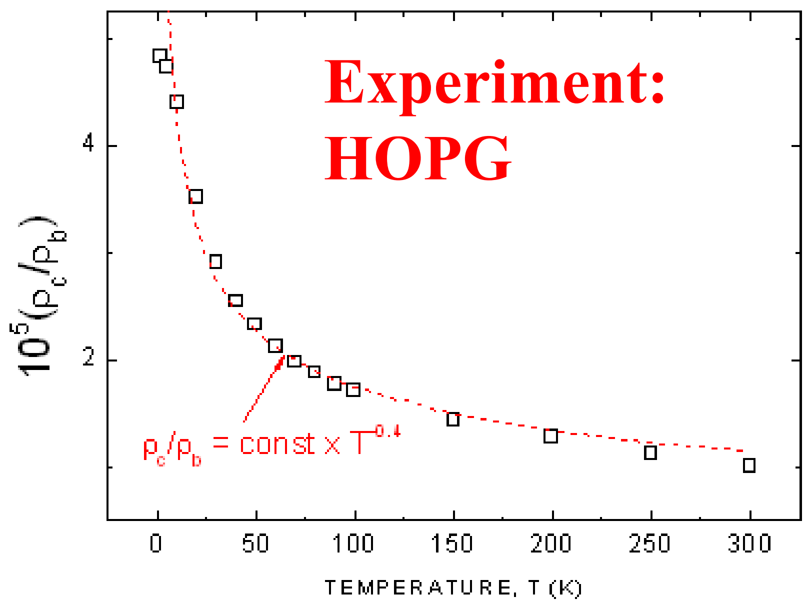
**(i) The perpendicular transport is enhanced by disorder;**

**(ii) The cleaner the system the larger the anisotropy;**

(iii) The anisotropy ($\sigma_{||}/\sigma_{\perp}$) diverges as $T^{-2/3}$

Theory:



# Theory of the Three-Dimensional Quantum Hall Effect in Graphite

B. Andrei Bernevig,[1] Taylor L. Hughes,[2] Srinivas Raghu,[2] and Daniel P. Arovas[2,3]

[1]*Princeton Center for Theoretical Physics, Princeton University, Princeton, New Jersey 08544, USA*

[2]*Department of Physics, Stanford University, Stanford, California 94305, USA*

[3]*Department of Physics, University of California at San Diego, La Jolla, California 92093, USA*

**PRL (2007)**

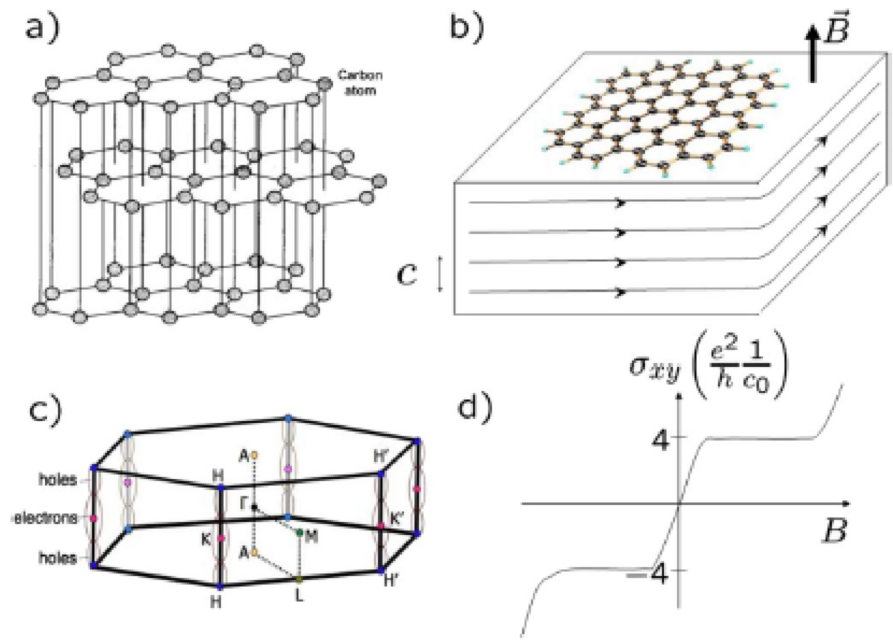
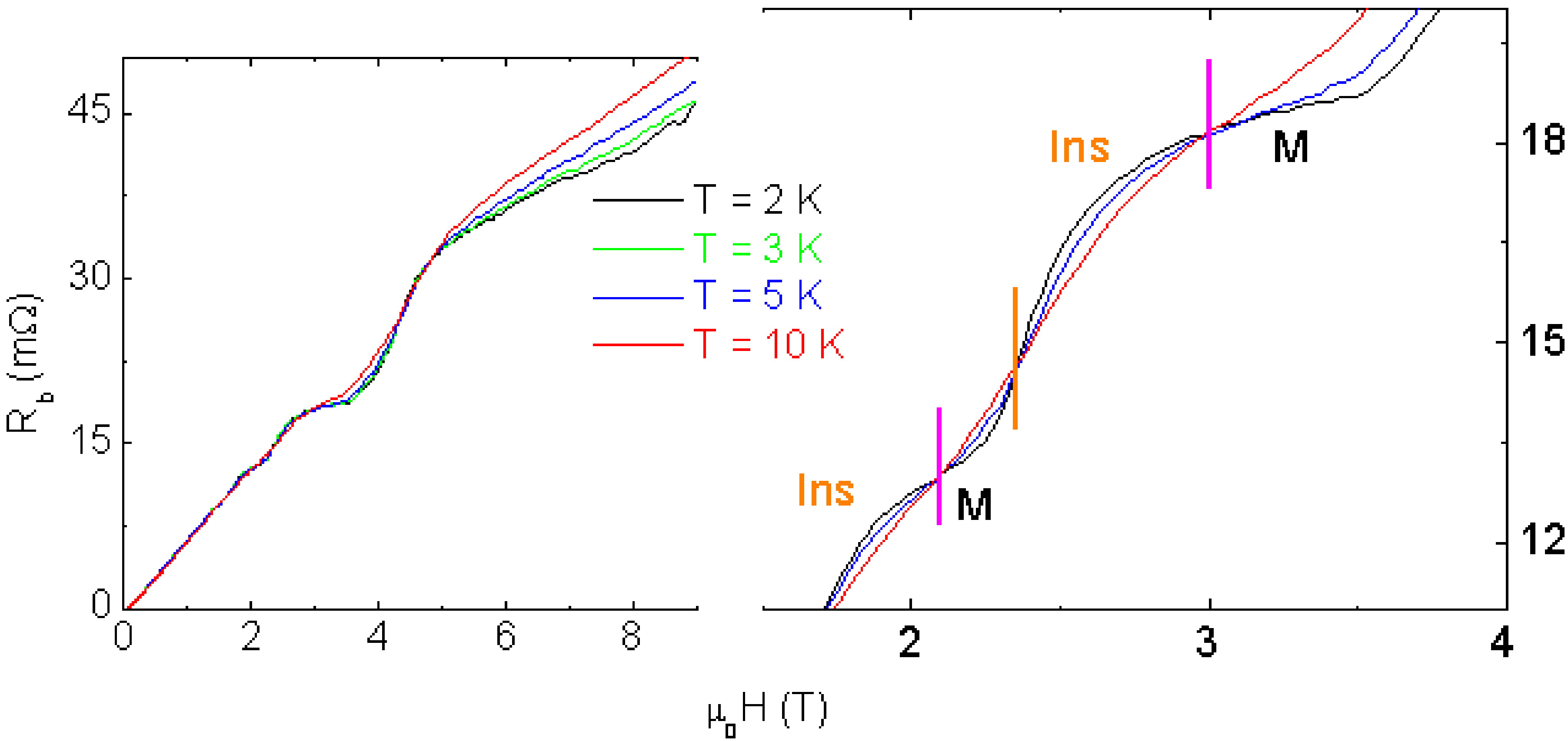


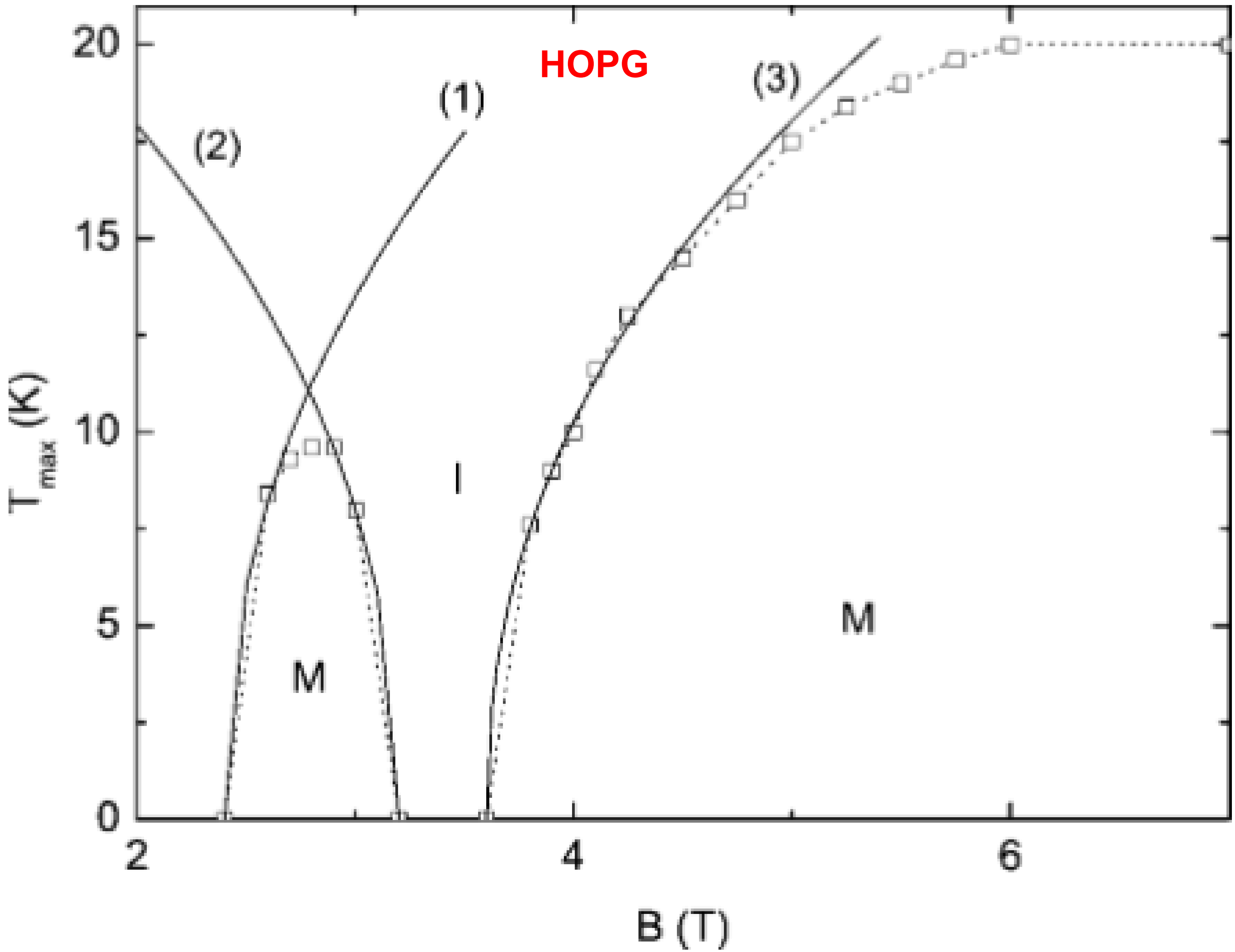

FIG. 1 (color online). (a) Graphite in Bernal stacking. (b) Under strong magnetic field, graphite is gapped in the bulk and exhibits chiral surface sheet states. (c) Idealized Brillouin zone for graphite. (d) Predicted 3D Hall conductivity, quantized in units of $1/c_0$. Only one plateau is observable in graphite.

**Contrary to the experiment, only ONE quantum Hall plateau is predicted within a framework of SWMC model**

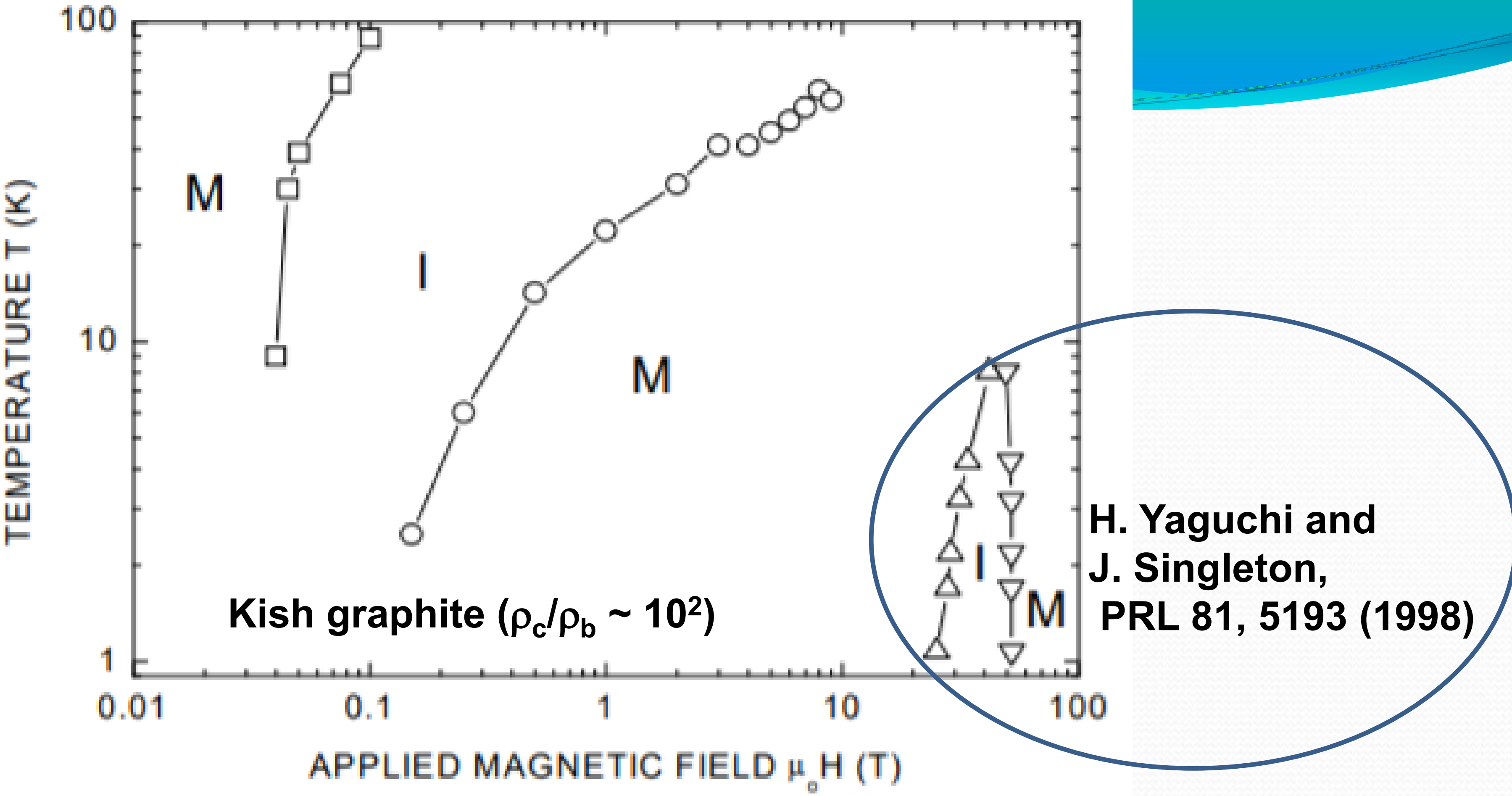
# Strongly anisotropic graphite ($\rho_c/\rho_b \sim 10^5$)




**HOPG: sequence of the field-driven insulator-metal-insulator transitions**




Y. K. , J. C. Medina Pantoja, R. R. da Silva, and S. Moehlecke
PRB 73, 165128 (2006)

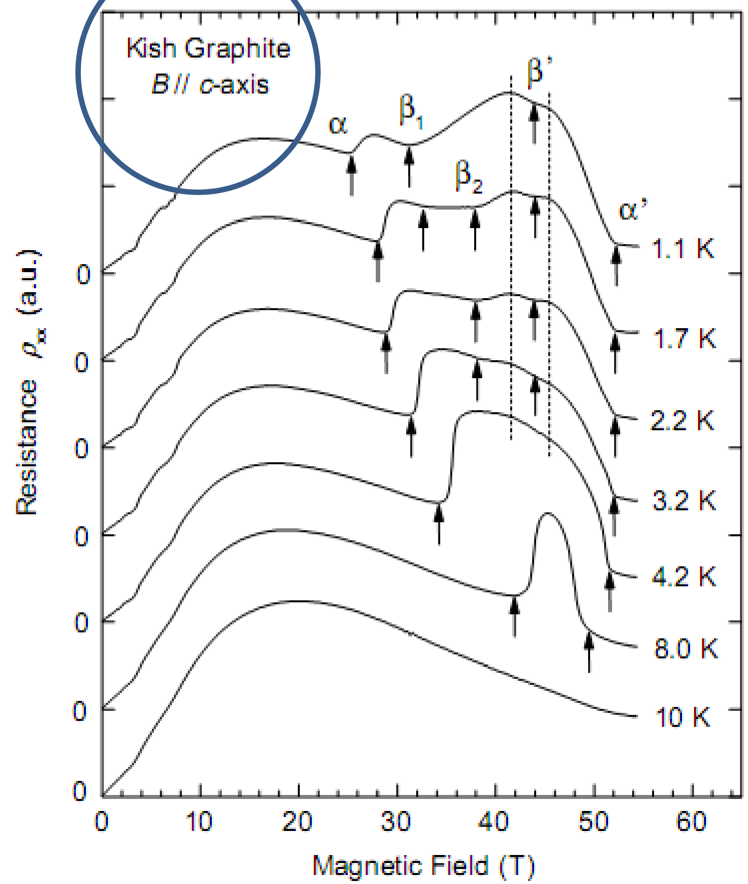



Y. K., P. Esquinazi, J. H. S. Torres et al. (2002)

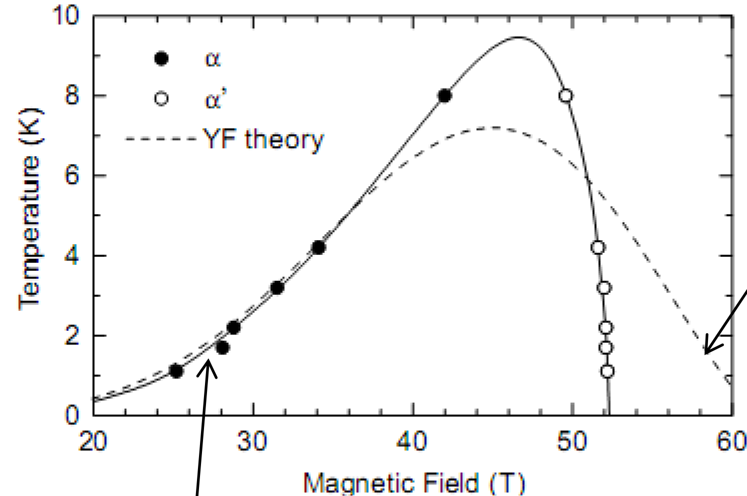
**PART II – Ultraquantum limit ($B \gg B_{QL} \sim 7$ T):**

**Another half-century puzzle**

# Kish & Natural graphite in ultra-quantum limit



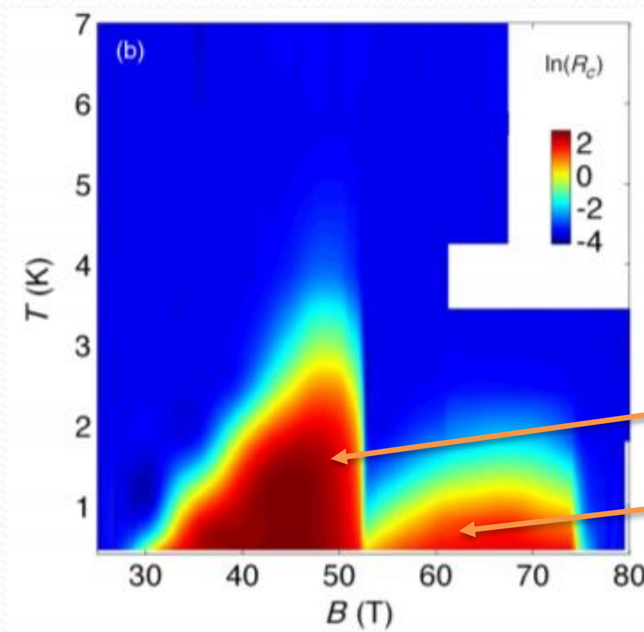

H. Yaguchi & J. Singleton, PRL 81, 5193 (1998)




Yoshika & Fukuyama (YF) 1981

BCS-type

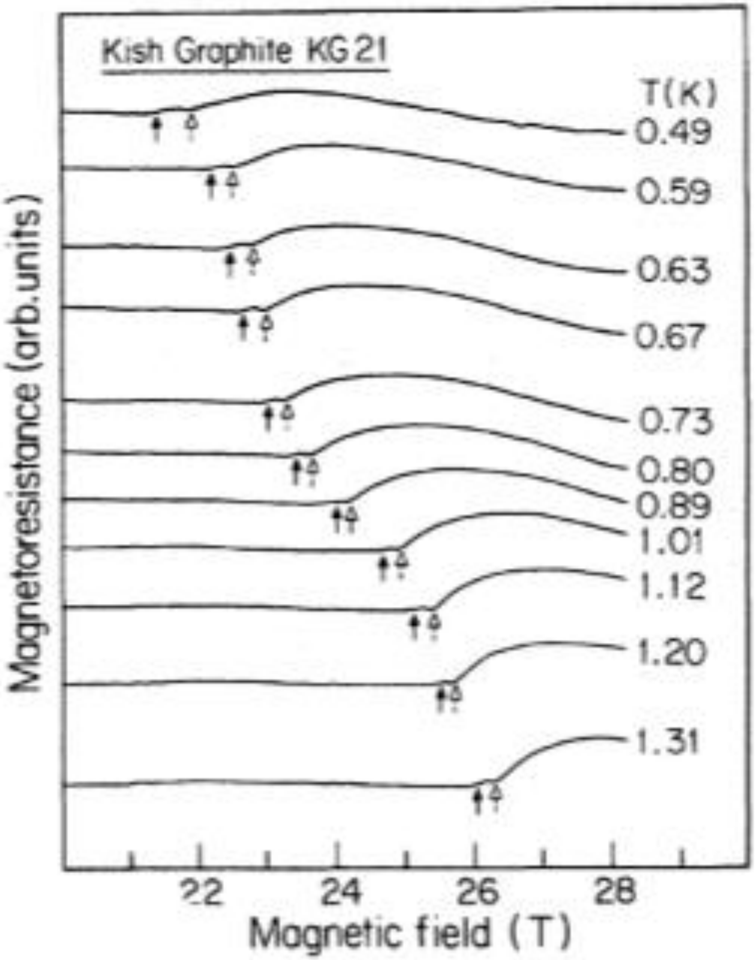
$$k_B T_c = 1.14 E_F \exp\left(-\frac{1}{N(0)V}\right)$$

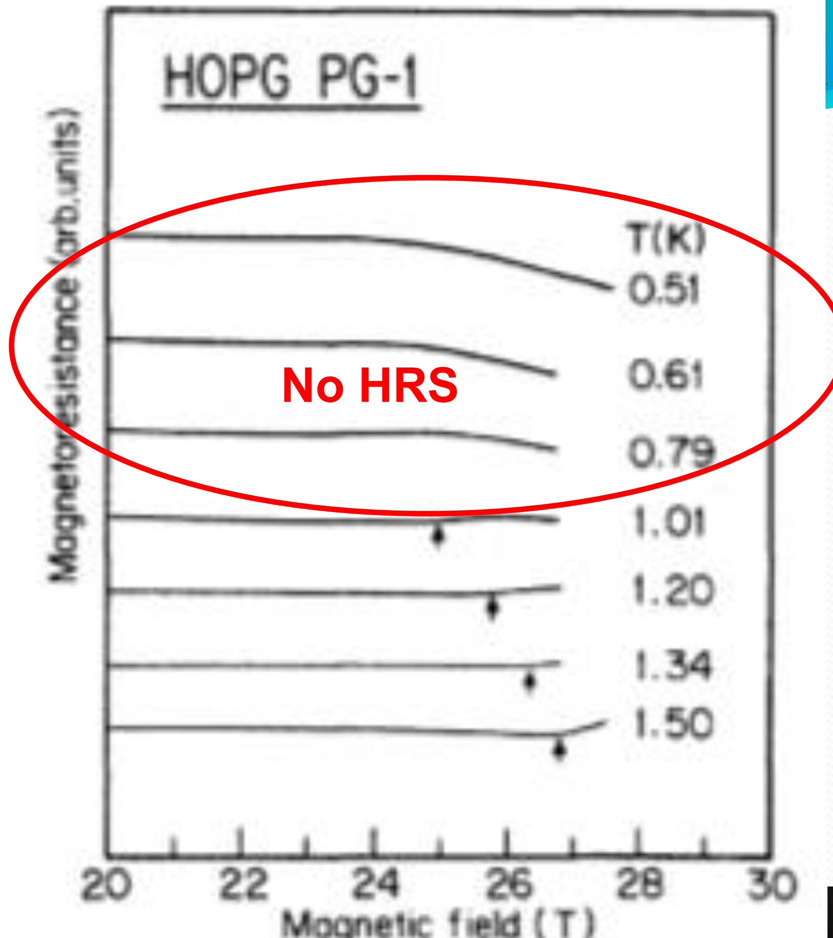



**B. Fauque et al., PRL 110, 266601 (2013)**

**High resistance states (HRS)**

**Kish Graphite**




???

Y. Iye et al., PRB 25, 5478 (1982)

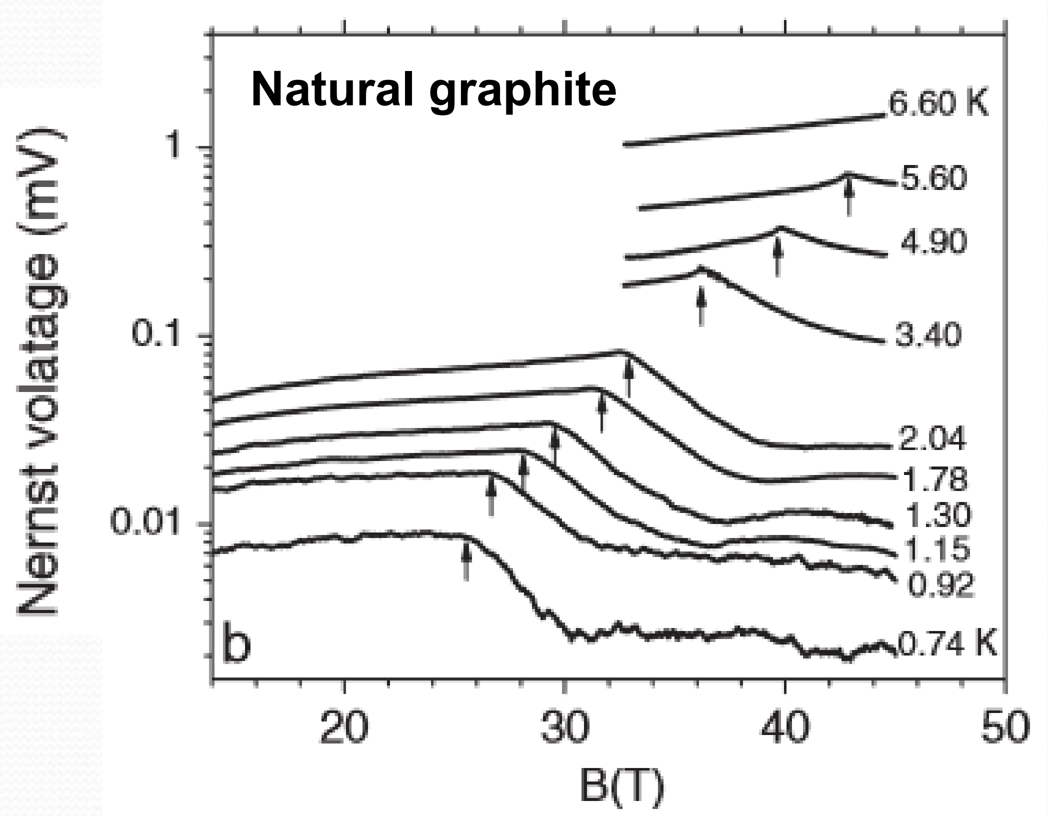
At lower temperatures ($T \lesssim 0.8$ K), no anomaly in the magnetoresistance can be identified. The negative magnetoresistance is more pronounced in this temperature range for HOPG than that observed for the Kish samples.

2D

or not

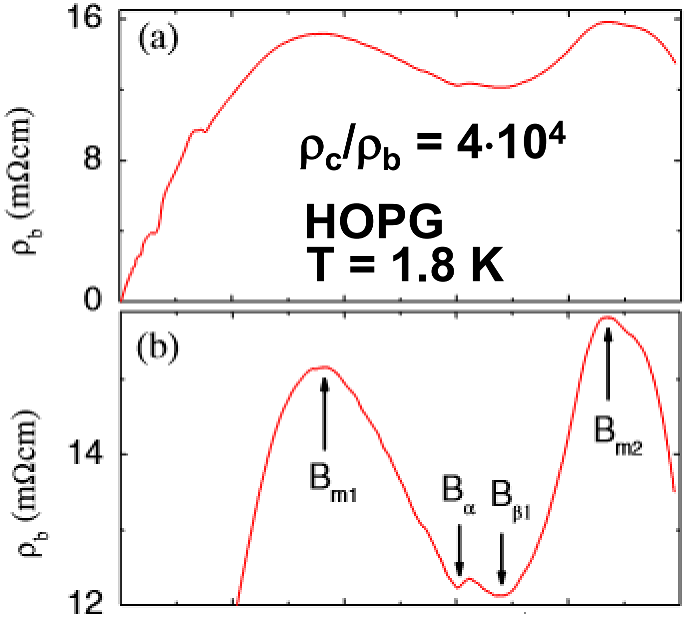
2D ?

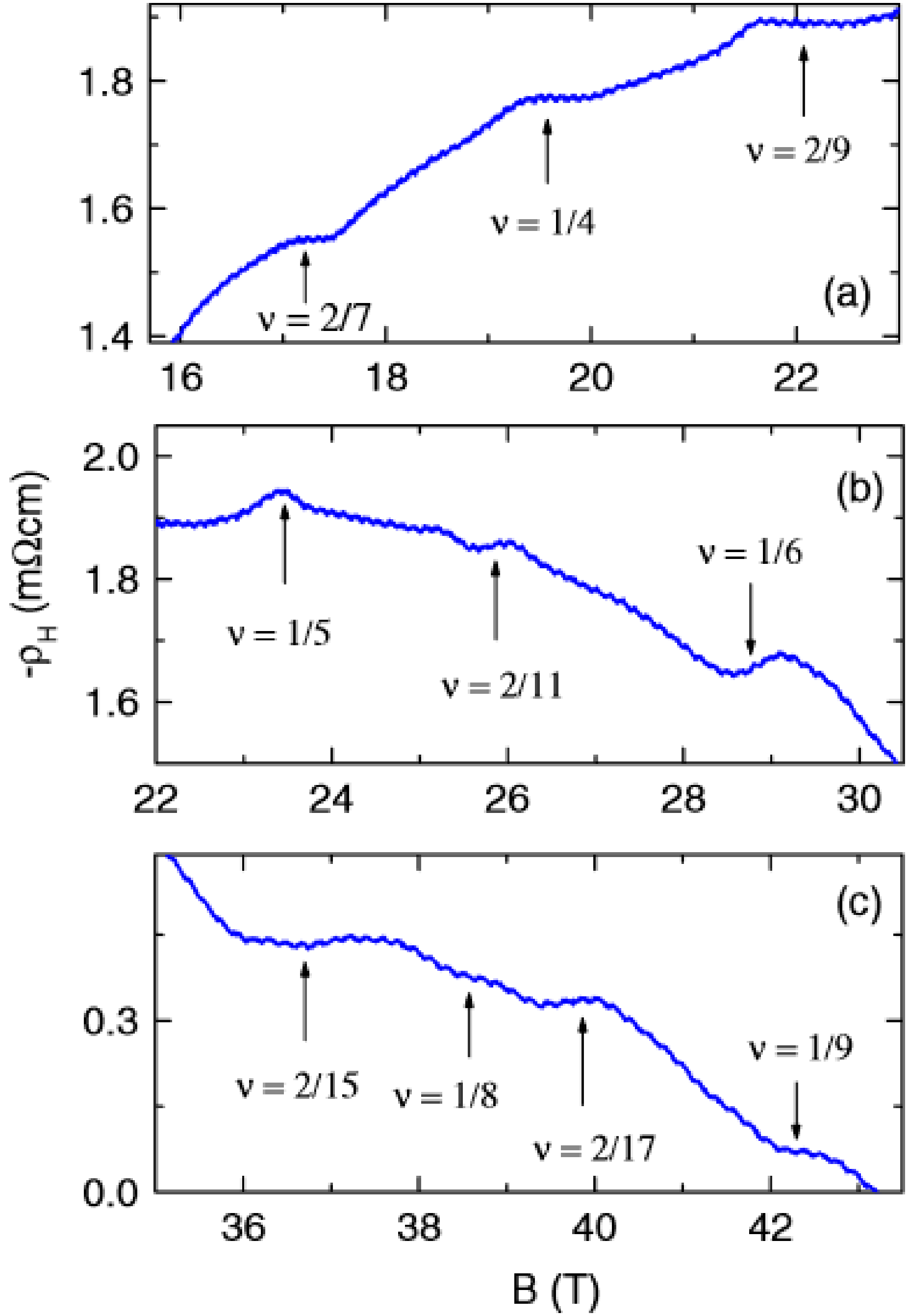
# Nernst effect measurements are also suitable for the high-resistance state studies in graphite:



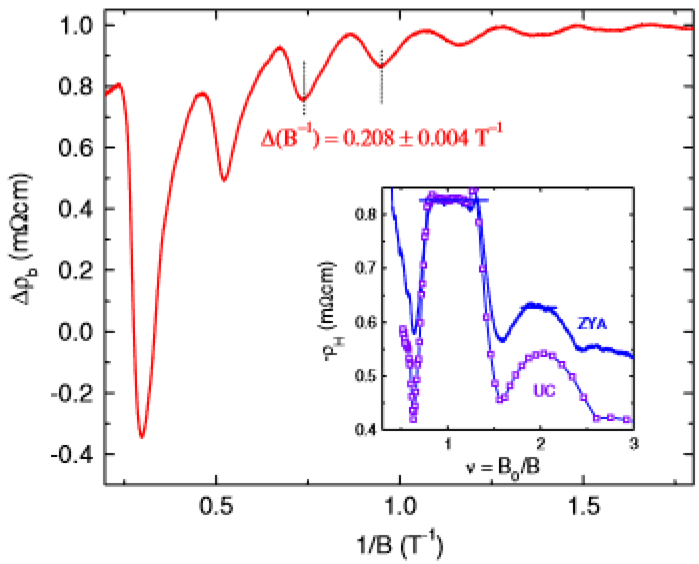

**B. Fauque et al., PRL 106, 246405 (2011)**

**Obs.: Kish & Natural graphite: $\rho_c/\rho_b \sim 10^2$**



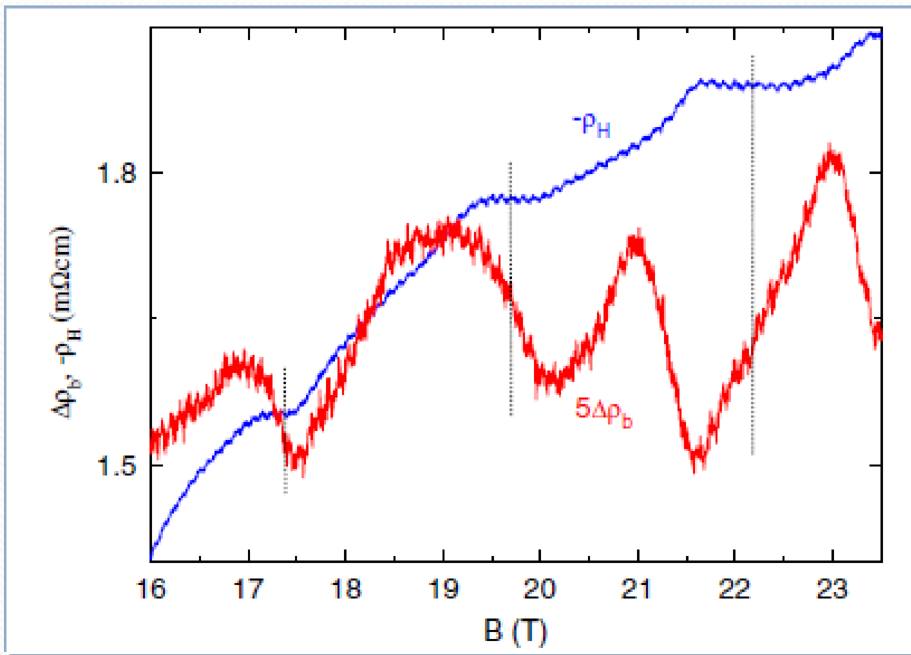




Y. K., B. Raquet, M. Goiran, et al. PRL 103, 116802 (2009).



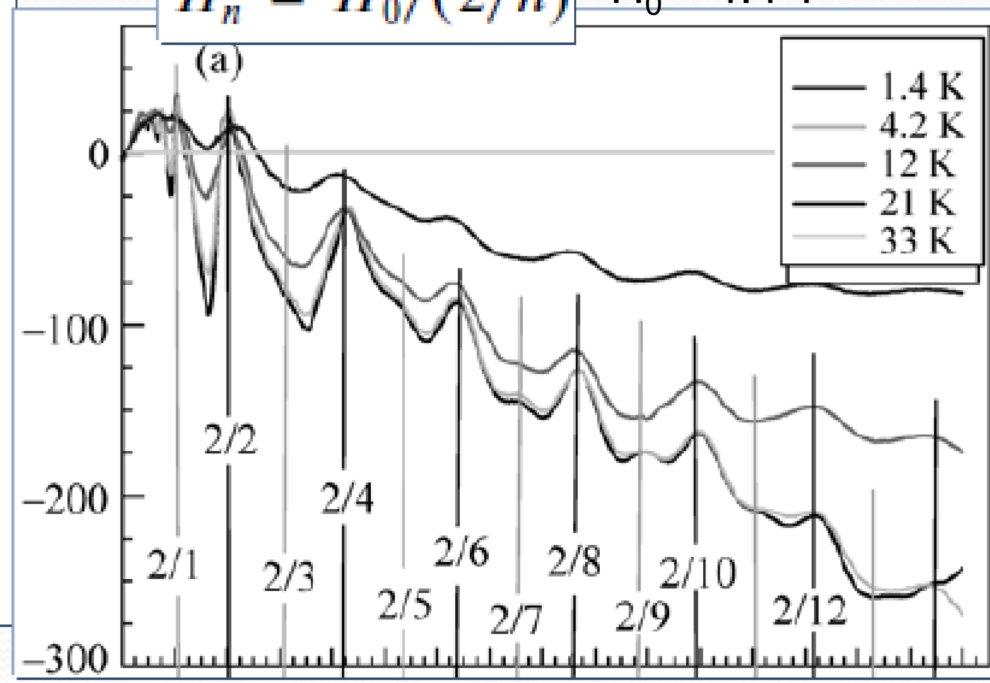
# Experiments suggest the existence of 2e-charge bosons in graphite:

Both Hall plateaus and resistance peaks correspond to the filling factors $\nu = 2/m$ (m=1, 2,3…) proposed by **Halperin [Helv. Phys. Acta 56, 75 (1983)]** for the case of bound electron pairs, i.e., 2e-charge bosons.

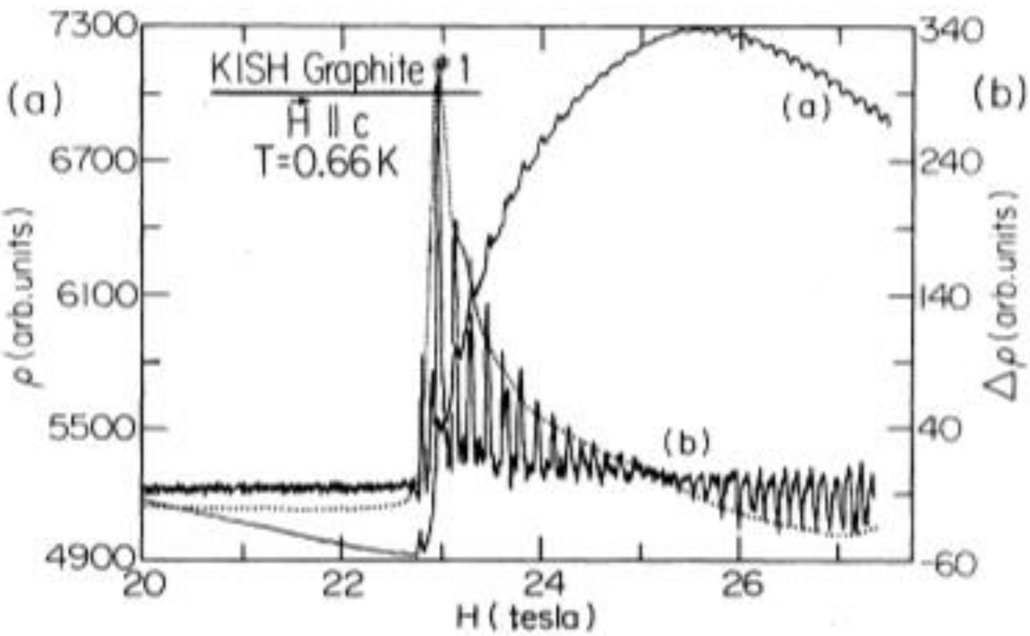



**Y. K., B. Raquet, M. Goiran, W. Escoffier, et al. PRL' 2009.**

$$H_n = H_0/(2/n)$$ $H_0$ = 4.4 T




**Yu. I. Latyshev et al., JETP Lett.' 2011**




Periodic in field oscillations in ultraquantum graphite:
G. Timp et al., PRB 28, 7393 (1983)

$$R(H) = R_{NOS}(H) + R_{OS}(H)$$

**$\Delta B$ = 0.16 (sample 1) and 0.17 T (sample 2)**

as also occurs in the Laughlin theory of the quantum Hall effect.[18] The present observations suggest that the mechanism for the oscillations periodic in $H$ might be related to the in-plane quantization of the harmonic-oscillator wave functions.

The theory of Yoshioka and Fukuyama, however, considers only the $k_z$ dependence of the electronic wave functions in describing a charge density-wave instability along the c direction. A more complete theoretical treatment of the charge-density-wave instability in graphite would have to incorporate both the inplane and z-axis dependences of the wave functions appropriate to graphite.

# Possible Observation of Phase Coexistence of the $\nu = 1/3$ Fractional Quantum Hall Liquid and a Solid

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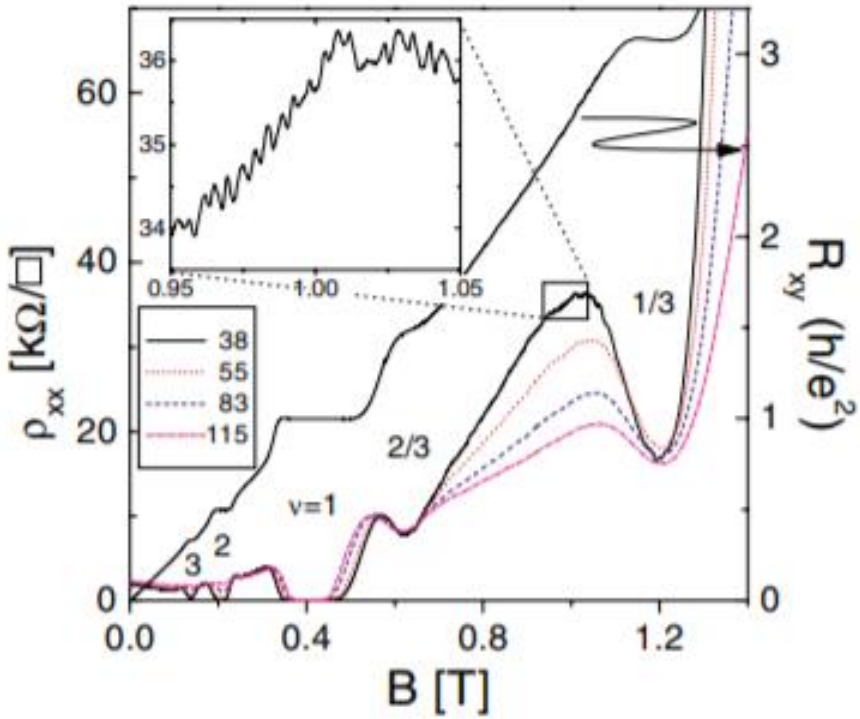
[1]Department of Electrical Engineering, Princeton University, Princeton, New Jersey 08544, USA
[2]Bell Labs, Lucent Technologies, Murray Hill, New Jersey 07974, USA




The 2DH system is formed in a 30 nm wide GaAs/AlGaAs quantum well with silicon dopants on both sides of the well.

Aharonov-Bohm oscillations:

2DH system is not homogeneous but it phase separates on the scale of 1 $\mu$m into two coexisting phases. Since the oscillations are present in the insulating phases on both sides of $\nu = 1/3$, the $\nu = 1/3$ FQH liquid and a crystalline phase are natural choices for the two phases.

In both of the insulating phases in the vicinity of the $\nu = 1/3$ filling the magnetoresistance has an unexpected oscillatory behavior with the magnetic field. These oscillations are not of the Shubnikov–de Haas type and cannot be explained by spin effects. They are most likely the consequence of the formation of a new electronic phase which is intermediate between the correlated Hall liquid and a disorder pinned solid.

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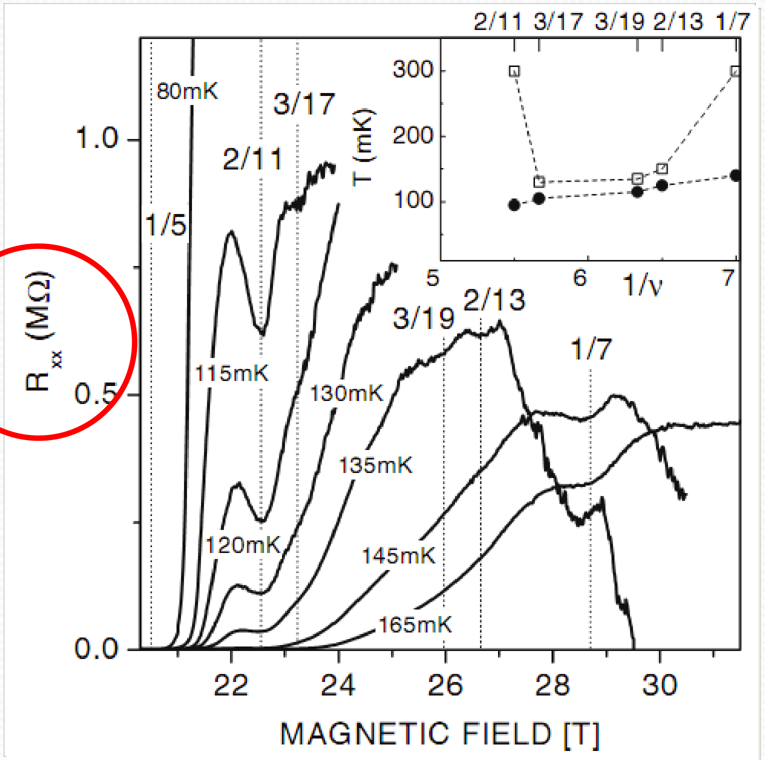
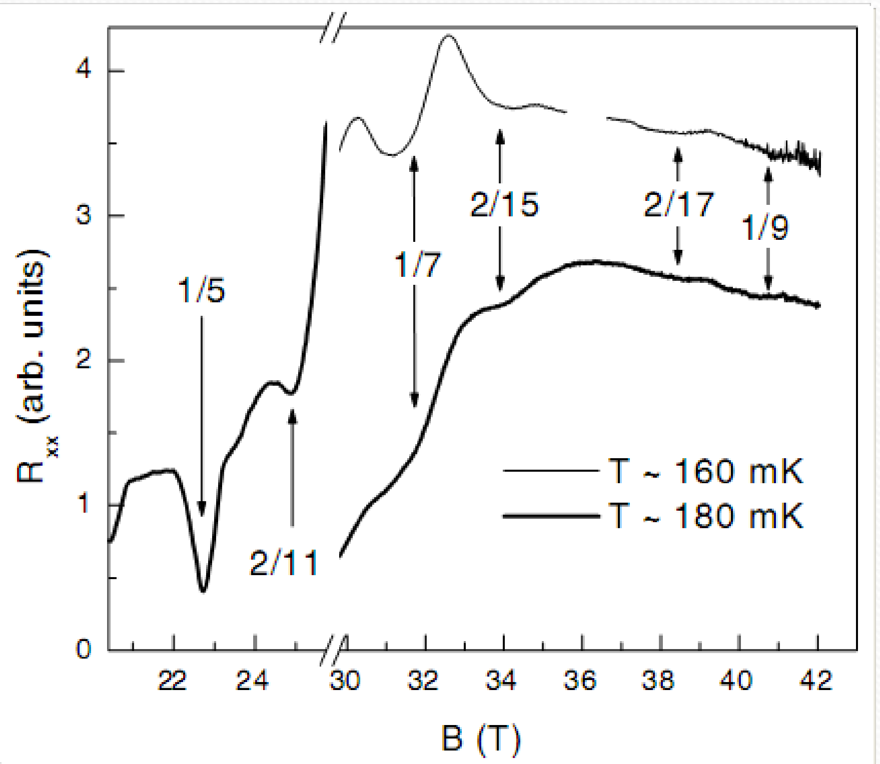
# Transition from an Electron Solid to the Sequence of Fractional Quantum Hall States at Very Low Landau Level Filling Factor

**GaAs/AlGaAs**

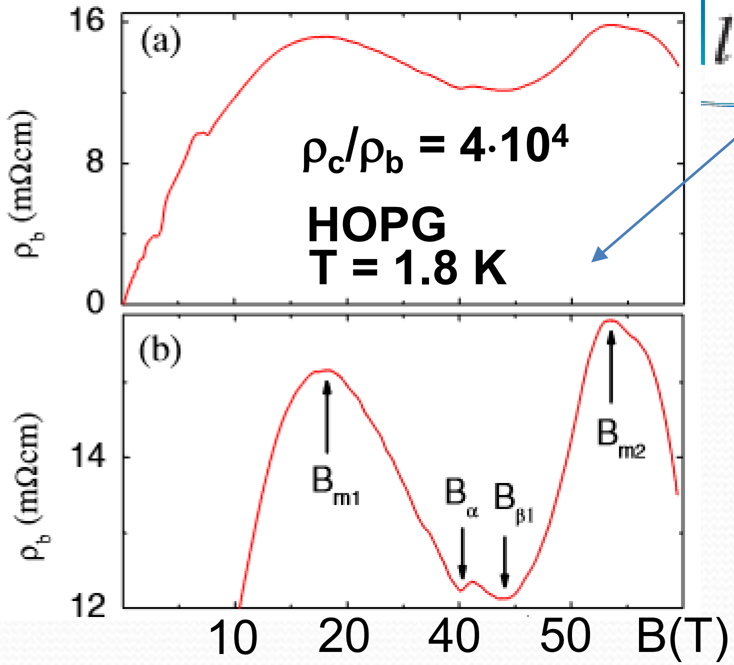
W. Pan,[1,2] H. L. Stormer,[3,4] D. C. Tsui,[1] L. N. Pfeiffer,[4] K. W. Baldwin,[4] and K. W. West[4]

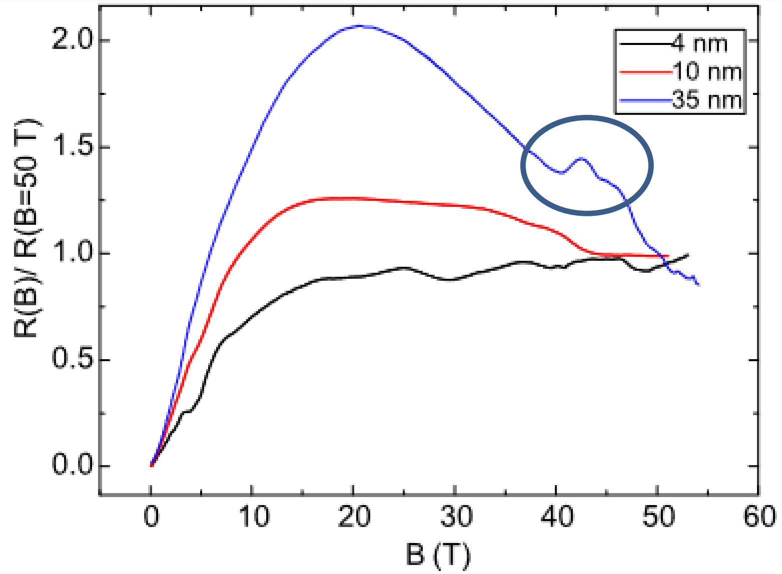



The electron density of $n \sim 1.0 \times 10^{11}$ cm$^{-2}$ and mobility of $\mu \sim 10 \times 10^6$ cm$^2$/Vsec

$l \times w \times t = 2.5 \times 2.5 \times 0.5\ \mathrm{mm}^3$




**Y. K., B. Raquet, M. Goiran, et al. PRL 103, 116802 (2009).**

**HRS vanishes in thin enough HOPG samples !**

**B.C. Camargo et al., Carbon ´ 2018**

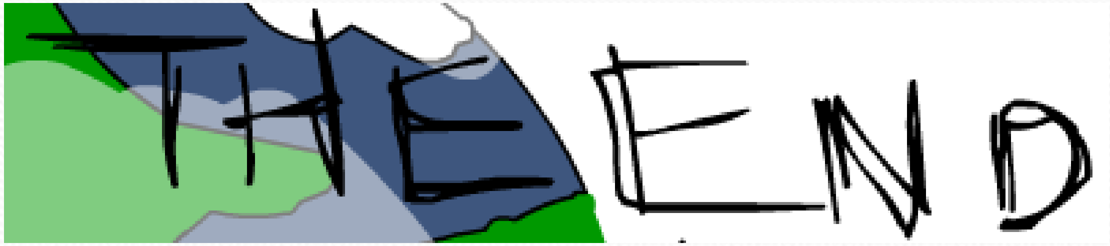
**10 μm x 5 μm x d**
**d = 4 nm, 10 nm, 35 nm**

Vanishing of the HRS in thin enough GRAPHITE samples is related to the 3D → 2D dimensional reduction,

**B.C. Camargo, Y. K. et al. (unpublished)**

**Many ground states are possible:**
**CDW, SDW, EXCITONIC INSULATOR, LUTTINGER LIQUID, electron pairing, FQHE …**

???


THE END