Hydrogen interaction with graphite: negative ion formation and more…

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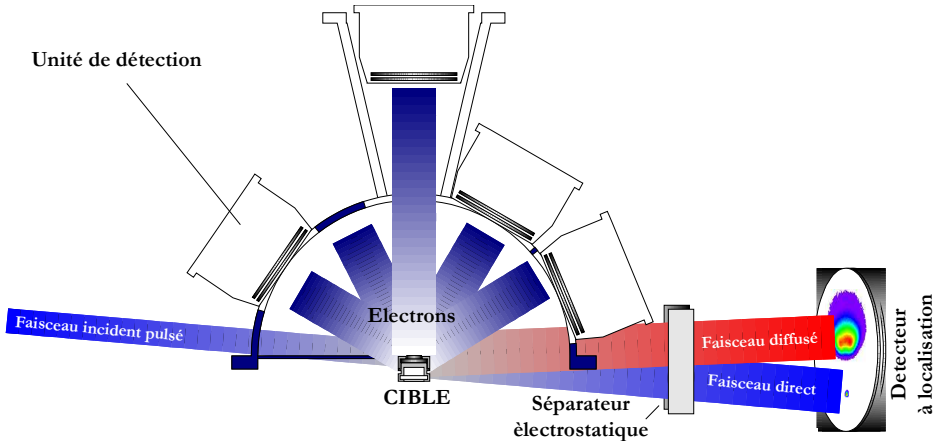
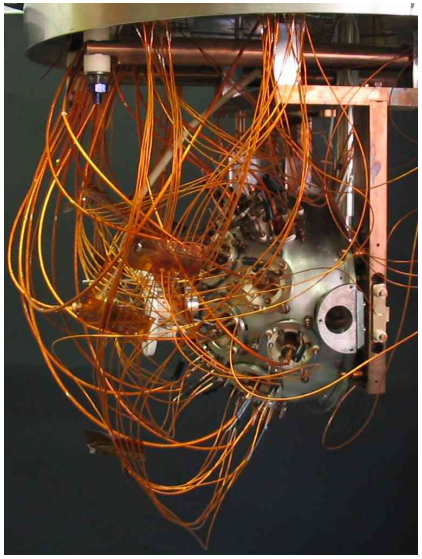
Motivation

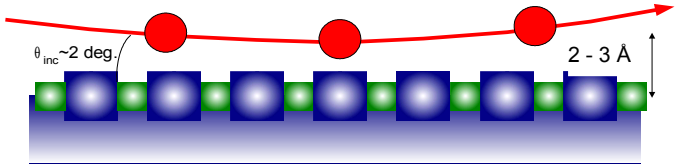
1- Negative ion production
- neutral beam injection for ITER
- low energy atom detection in space
- ion implantation

2- properties of carbonaceous materials
- electronic properties
- reactivity with hydrogen

Grazing incidence scattering







$$200 < E_0 < 10000\ eV$$

$$E_{\perp} = E_0 \sin^2(\psi)$$

$$20\ meV < E_{\perp} < 10\ eV$$

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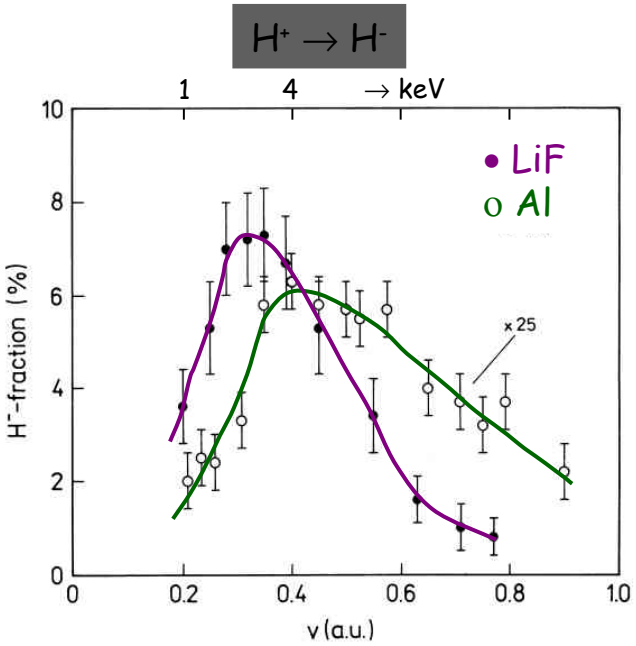
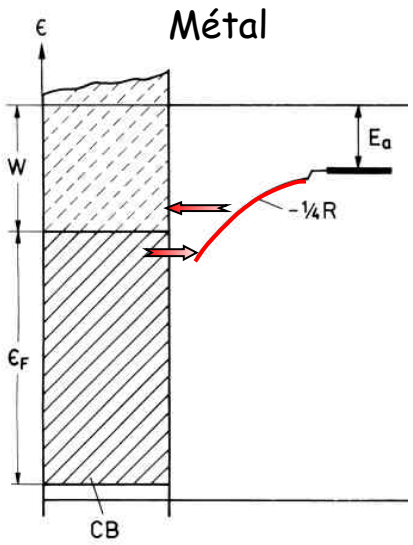
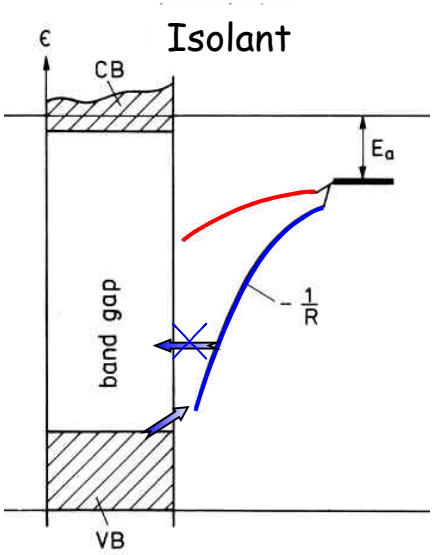
Negative ion formation: metals vs insulators

Image Potential : $V \sim -1/(4.R)$

Coulomb Potential (Madelung) : $V \sim -1/(R)$

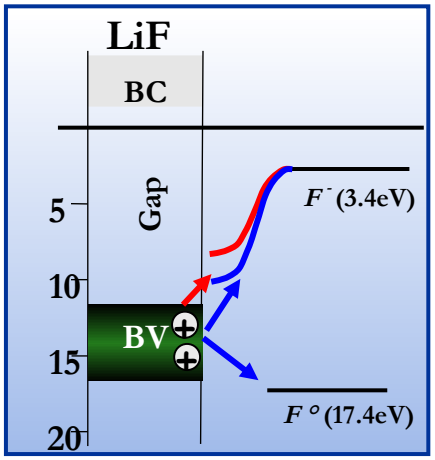




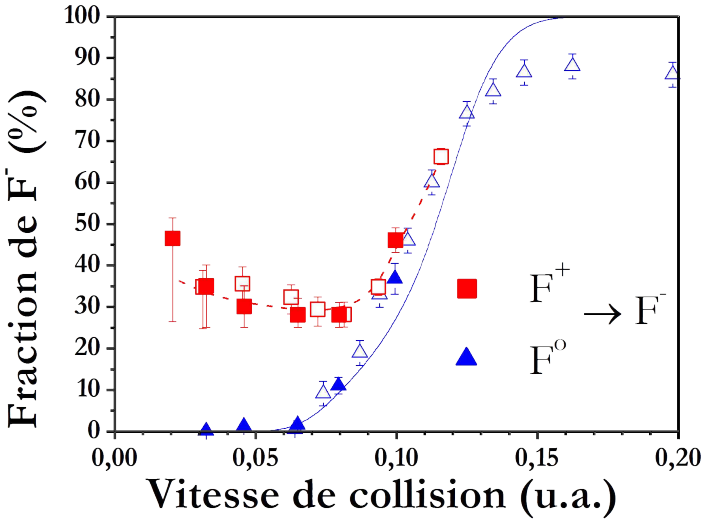


H. Winter, Progress in Surface Science **63**, 177, 2000

Negative ion formation: simultaneous double electron capture



P. Roncin et al., Phys. Rev. Lett. **89**, 43201, 2002



At low energy, it is easier to capture two electrons rather than one !!

Systemes where SDEC has been observed :

- F^+ on LiF(001) ⇒ fraction of F^- ~ 40 %
- O^+ on NaCl(001) ⇒ fraction of O^- ~ 7 %
- H^+ on NaCl(001) ⇒ fraction of H^- ~ 1 %

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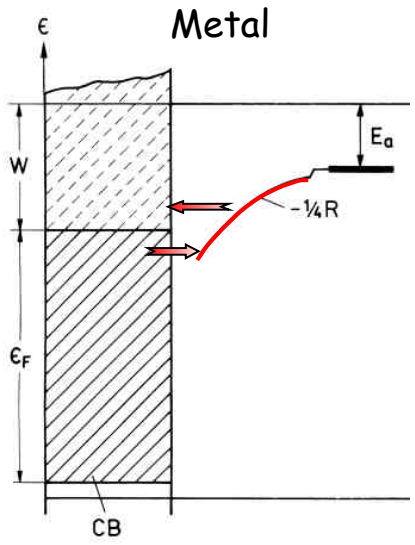
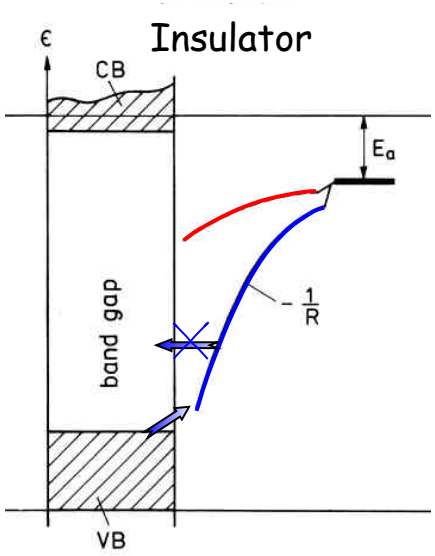
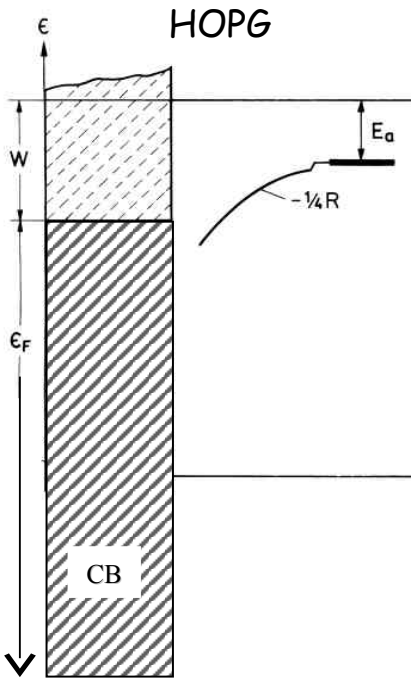
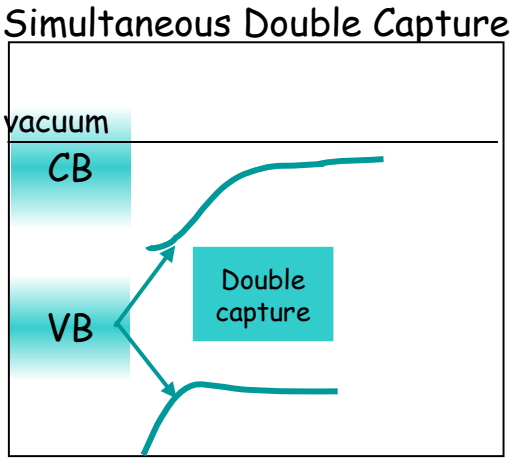
Question: what to expect from carbonaceous materials







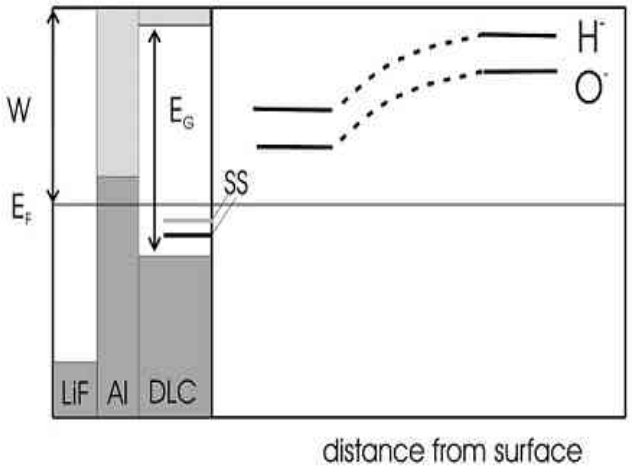


Graphite HOPG

- semi-metal (conductor)
- work-function ~ 4.6 eV
- deep valence band

Question: role of band gap in Diamond (LC)

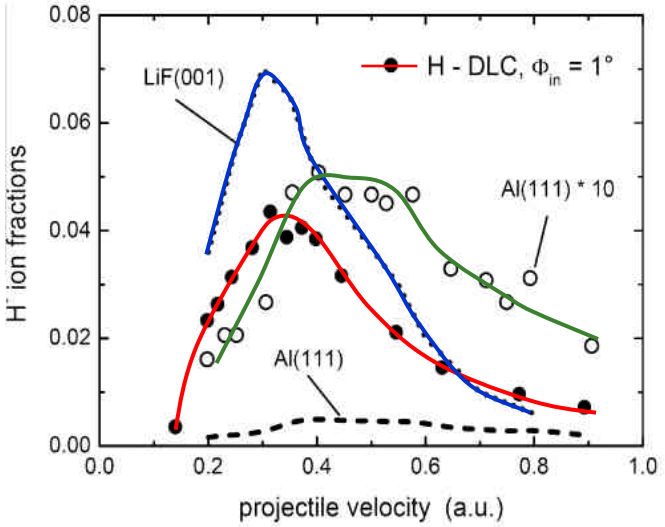
J. Lienemann et al. (2011)



Diamond :

- band gap ~ 5.5 eV
- deep valence band
- negative electron affinity

J. Lienemann et al. (2011)

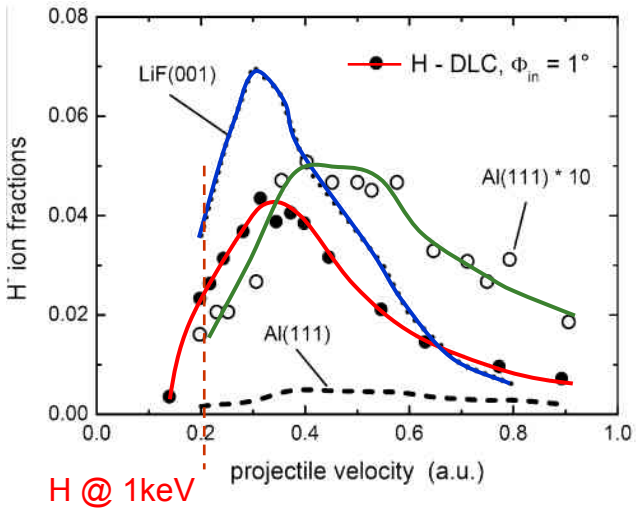


Mechanism:

⇒ metal-like with a gap

Results on Diamond (CVD)

Projectile E=1 keV	H- fraction (%)
H^+	**2.5 ± 0.5**
H°	**3.0 ± 0.8**
H_2^+	**1.6 ± 0.5**



Résults for H_2^+ in agreement with litterature
(Wurz P. et al. , Surf. Sci **373**, 56, 1997)
(J. Lienemann et al. NIMB, 2011)

Conclusion :

⇒ resonant neutralization of H^+

⇒ formation of H^- par capture on downshifted affinity level

⇒ survival of H^- because of band gap

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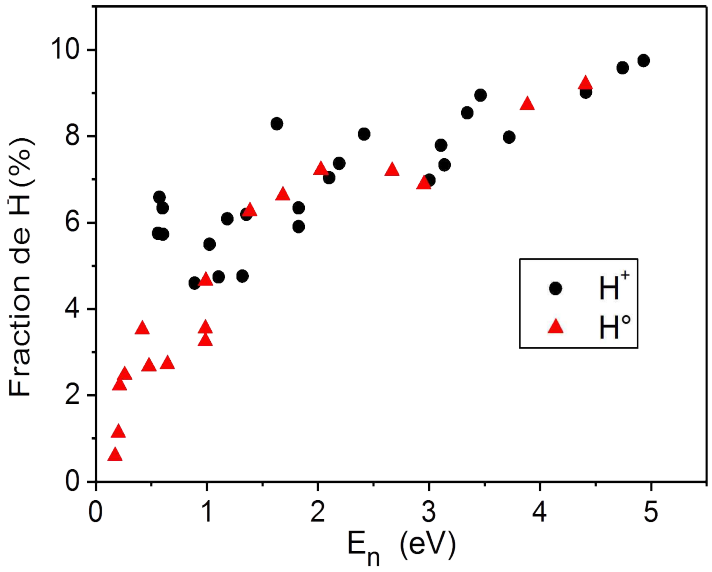
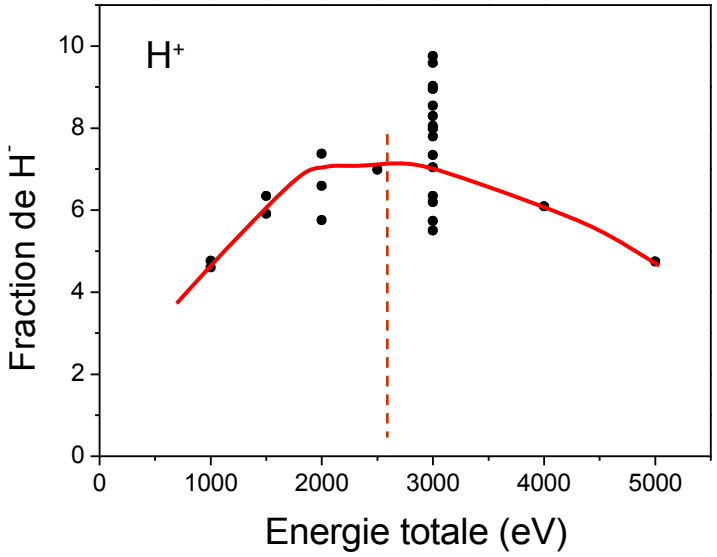
Results on graphite

H°,+ sur graphite HOPG



Energy range: 1 – 5 keV



Resonance similar to LiF and DLC

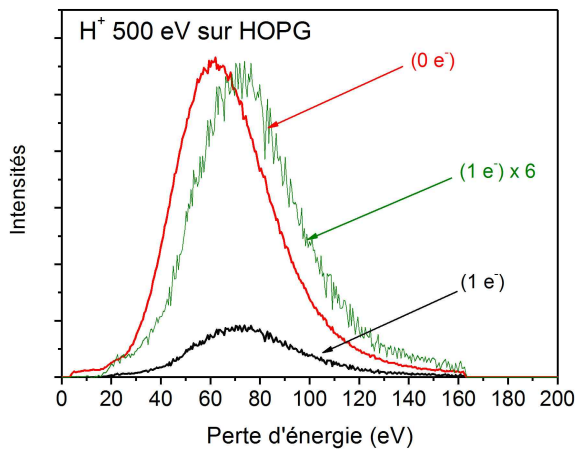
No evidence of double capture

Word record in H- production yield

⇒ Better than metals

⇒ better than insulators (LiF, diamant, etc.)

What's special with graphite ? *Electron yiel vs energy loss*



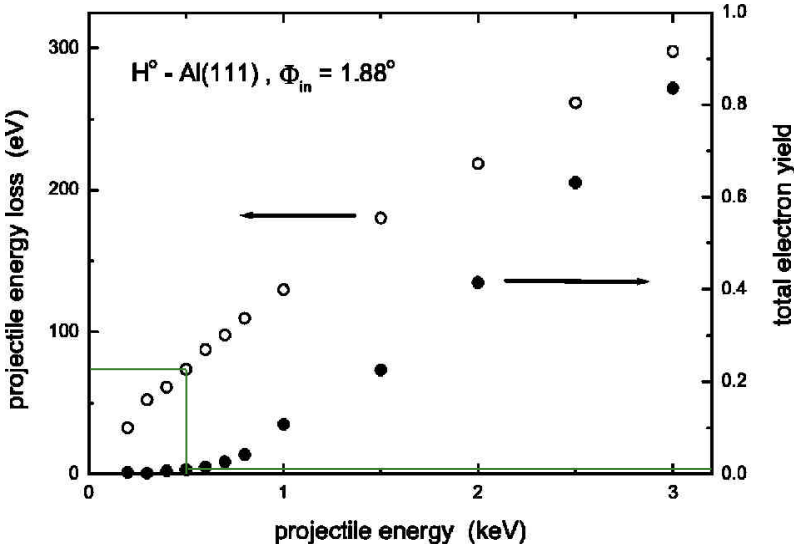
Electron yield:

- H°, ~0.15
- H^-, ~0.13

Comparaison:

H° 600 eV sur LiF(001), ~ 0.2

H° 500 eV sur Al(111), ~ 0.01



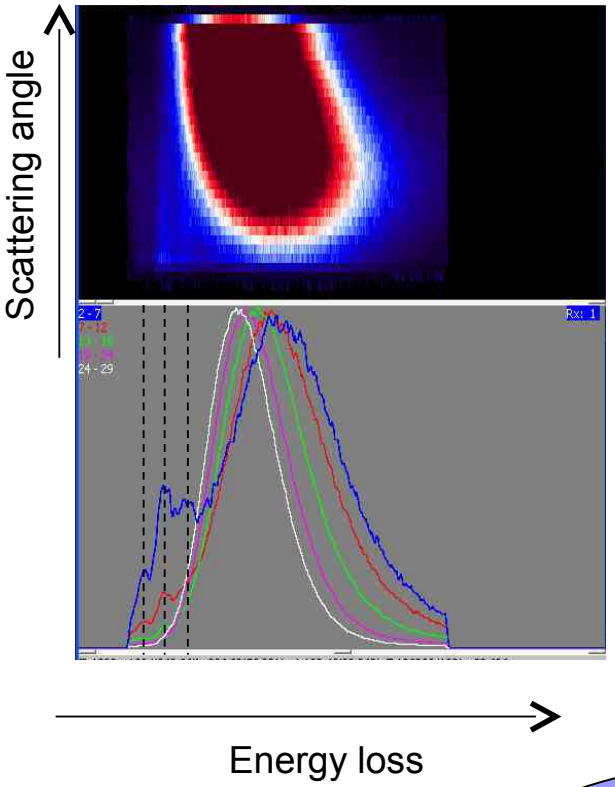
HP. Winter, S. Lederer, F. Aumayr and H. Winter

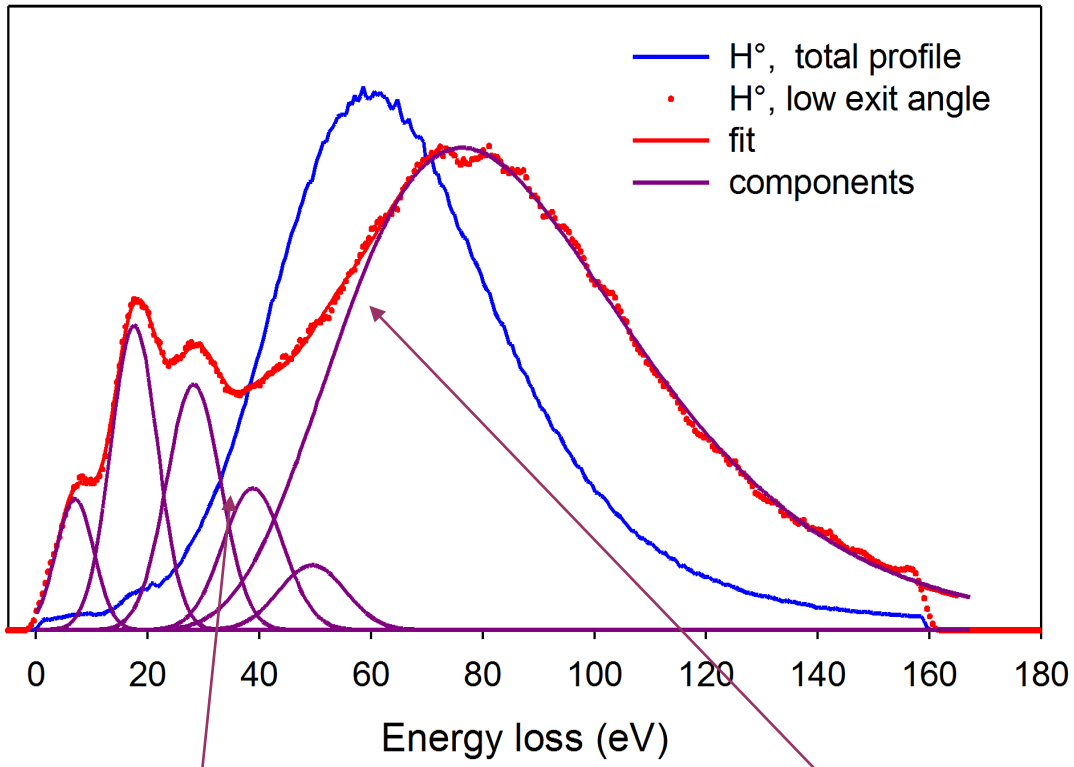
Graphite :

⇒ ≡ metal from energy loss

⇒ ≡ insulator from H- and electron yields

What's special with graphite ? *Energy loss spectrum*





Multiple structures, characteristic of an isolator-like interaction
→ H- mediated
→ localized electrons
→ Gap of 10.6 eV

log-normal component, characteristic of a metal-like interaction
→ binary collisions
→ delocalized electrons
→ no gap

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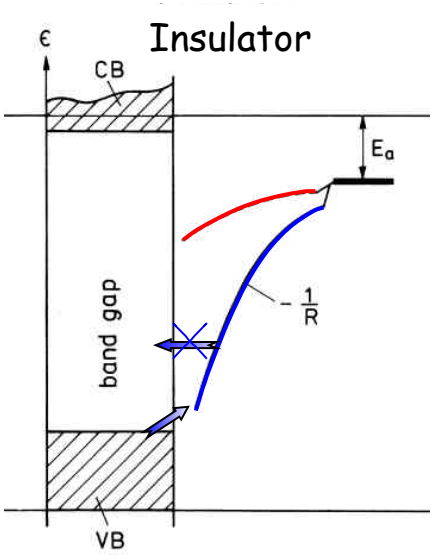
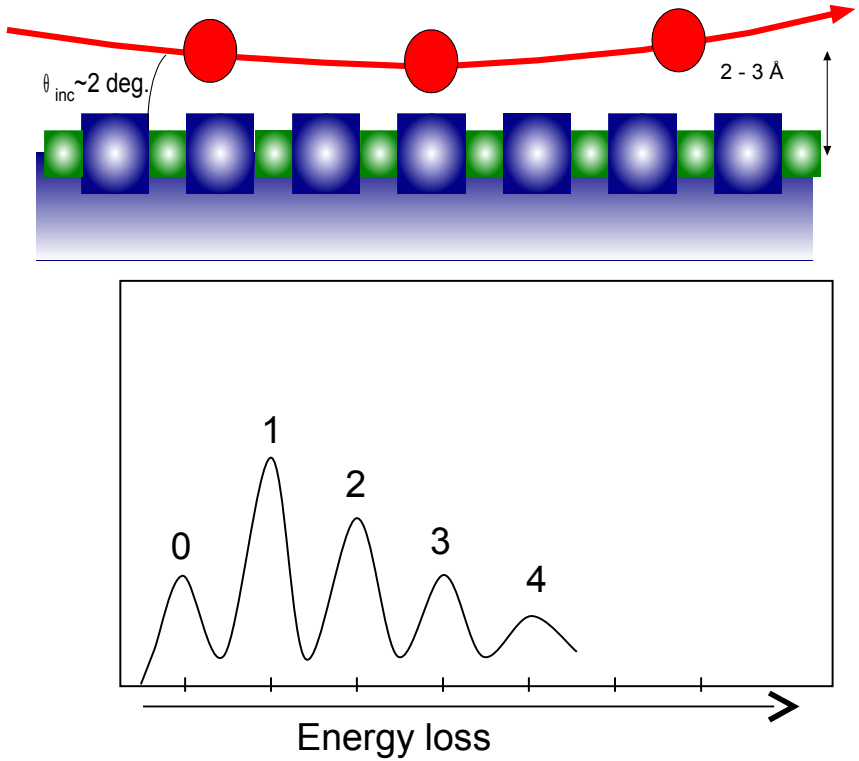
Origin of electrons

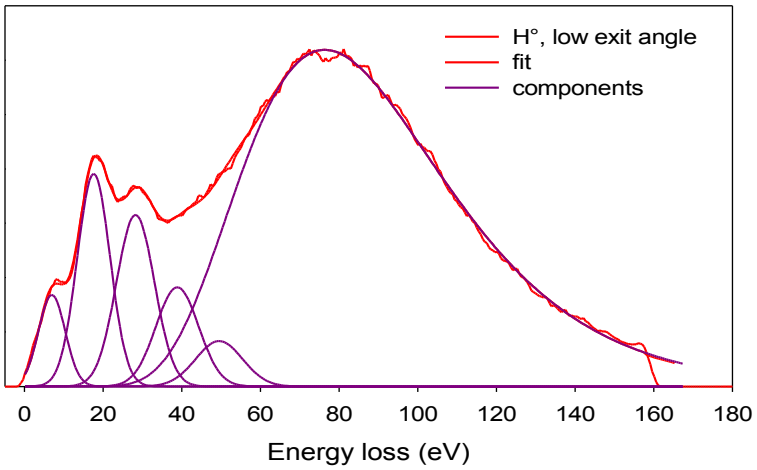


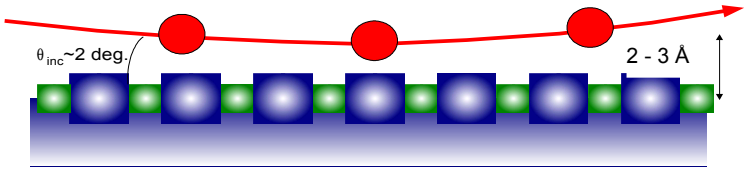


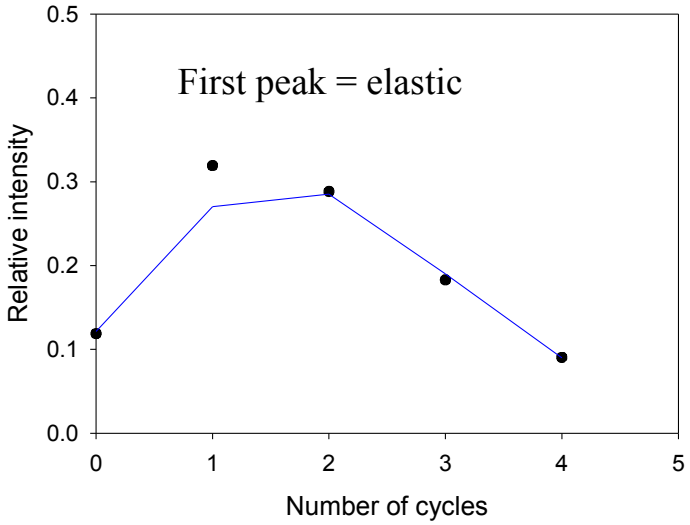
On insulators, these structures are well reproduced by a binomial distribution:
probability to extract n electrons from Ns equivalent sites…

Is it the case here ?

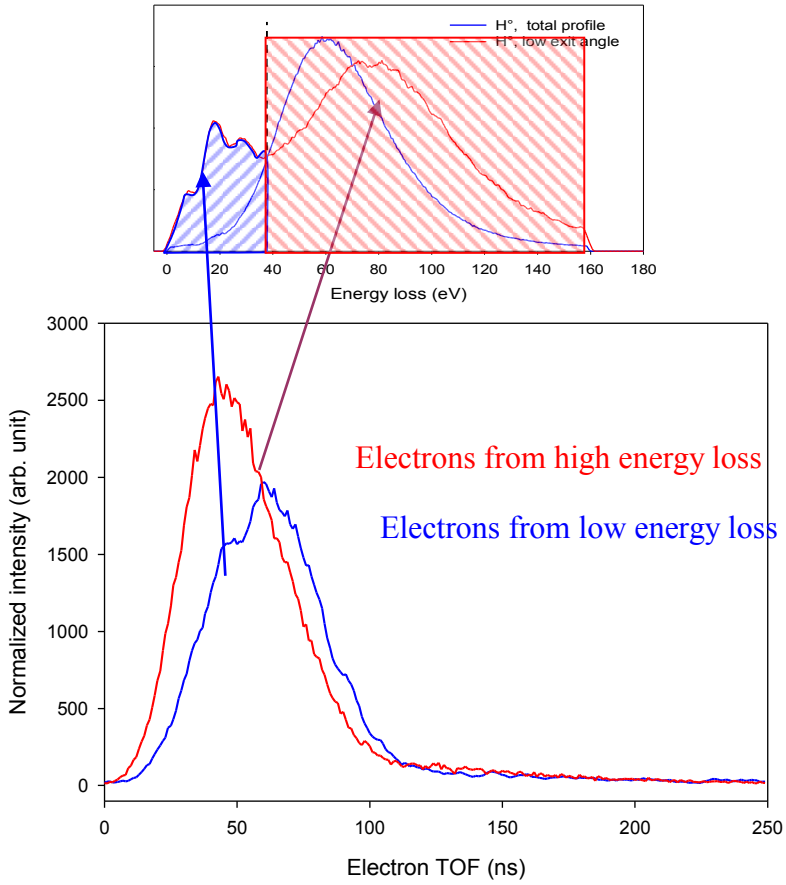
Origin of electrons







Electron emission



Two distincs populations of emitted electrons:

→ low energy loss structures are correlated to lower energy electrons

→ High energy loss is correlated to higher energy electrons

two distinct electron emission mechanisms !

These emitted electrons have initial binding energy around 10 eV, i.e. well below the Fermi level.

Conclusion

Graphite seems to have two faces, depending on how you look at it

⇒ insulator-like with respect to electron and H- yields

⇒ metal-like with respect to energy loss

Energy loss shows two components:

⇒ smooth peak with large straggling
(binary collisions with delocalized conduction electrons)

⇒ discrete structures with 11 eV difference
(excitation of localized electrons, below VBM)

Two electron emission mechanisms :

⇒ higher energy electrons from high energy loss

⇒ lower energy electrons from discrete energy loss structures

HELP !!

- Why so many negative ions ?

- Why don't they detach by resonant transfer to conduction band ?

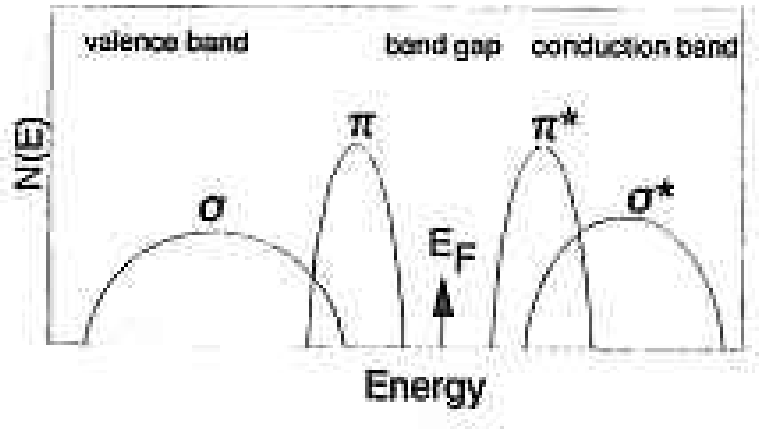
- Where do the electrons come from ? (specific bands, defects, etc)

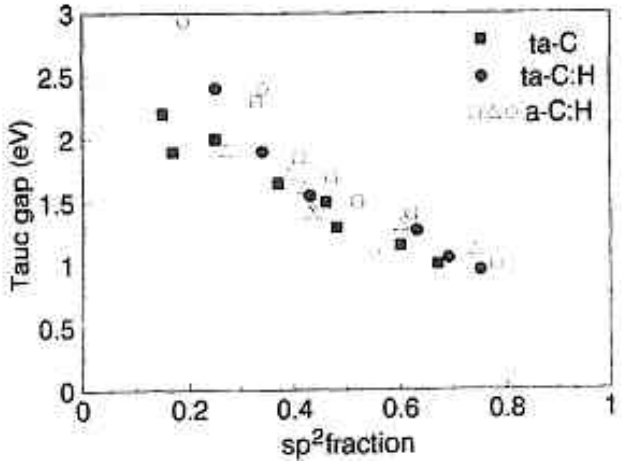
- Why graphite is so different from diamond (LC) ?

- …

Extends these studies to other carbon materials

⇒ Search for influence of sp^2/sp^3 ratio…

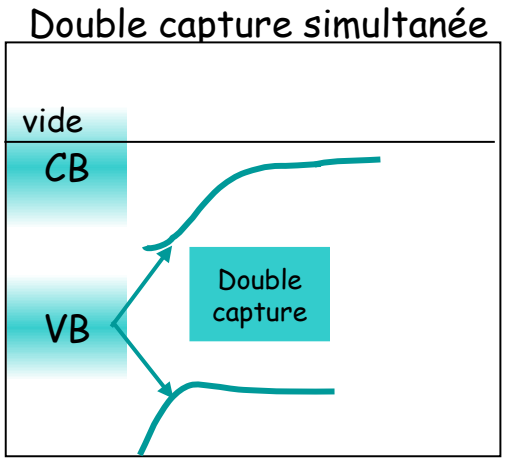


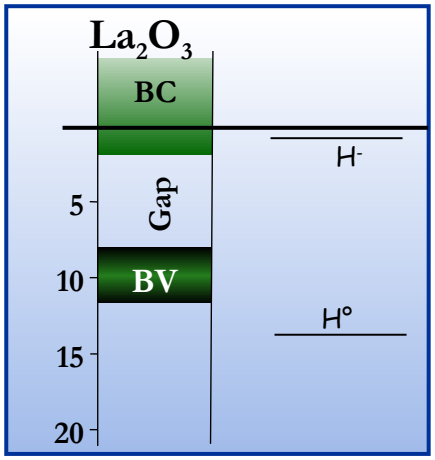


J. Robertson,
Diamond and Related Materials **6**, 212 (1997)

Exploit simultaneous double electron capture

⇒ potentially very efficient on a large energy range





Oxyde de lanthane:

- Grand gap
- bande de valence étroite
- faible affinité électronique
- caractère ionique