Hadean diamonds in zircon from Jack Hills, Western Australia

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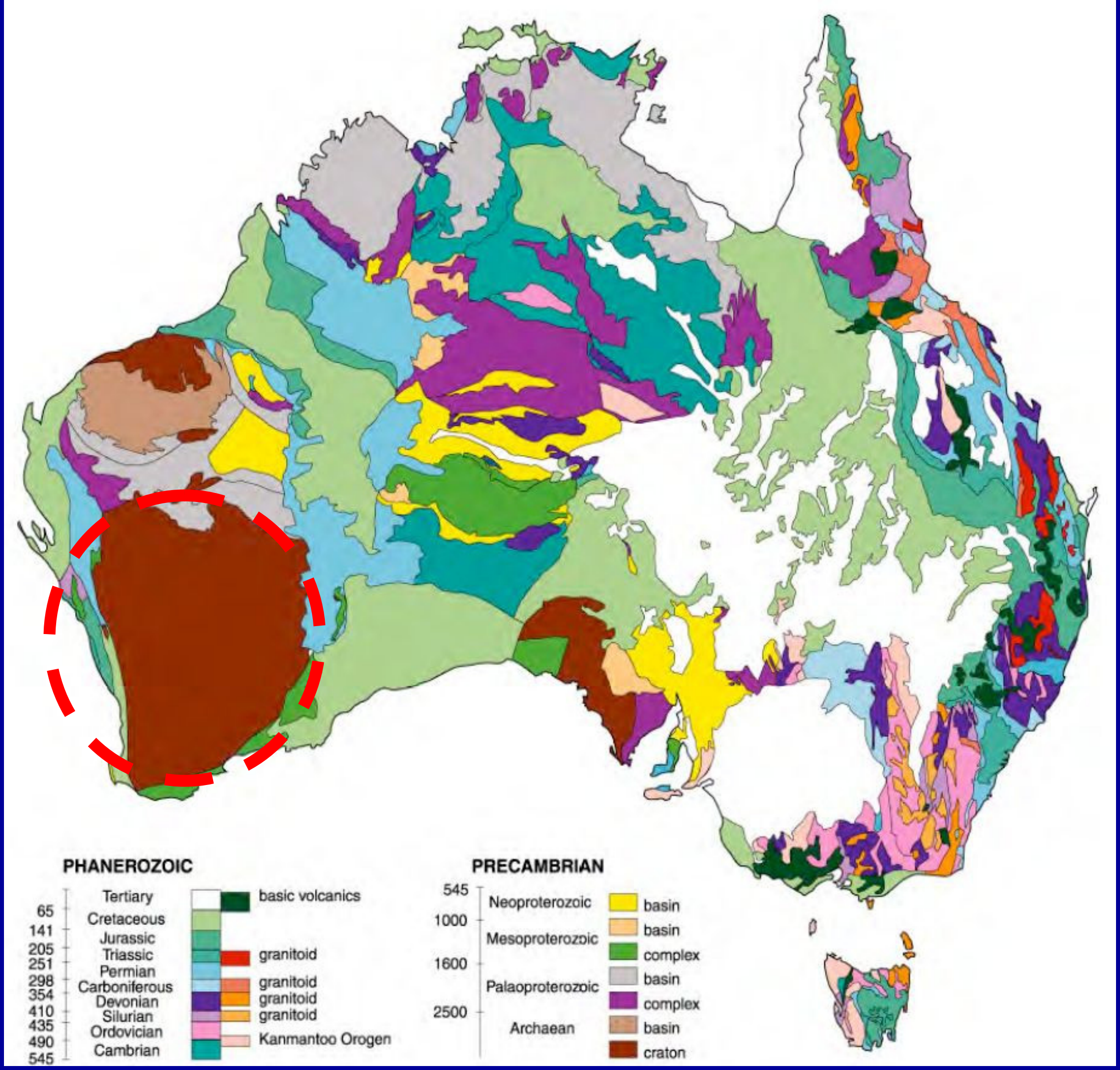
Yilgarn craton

•2.8-3.1 Ga

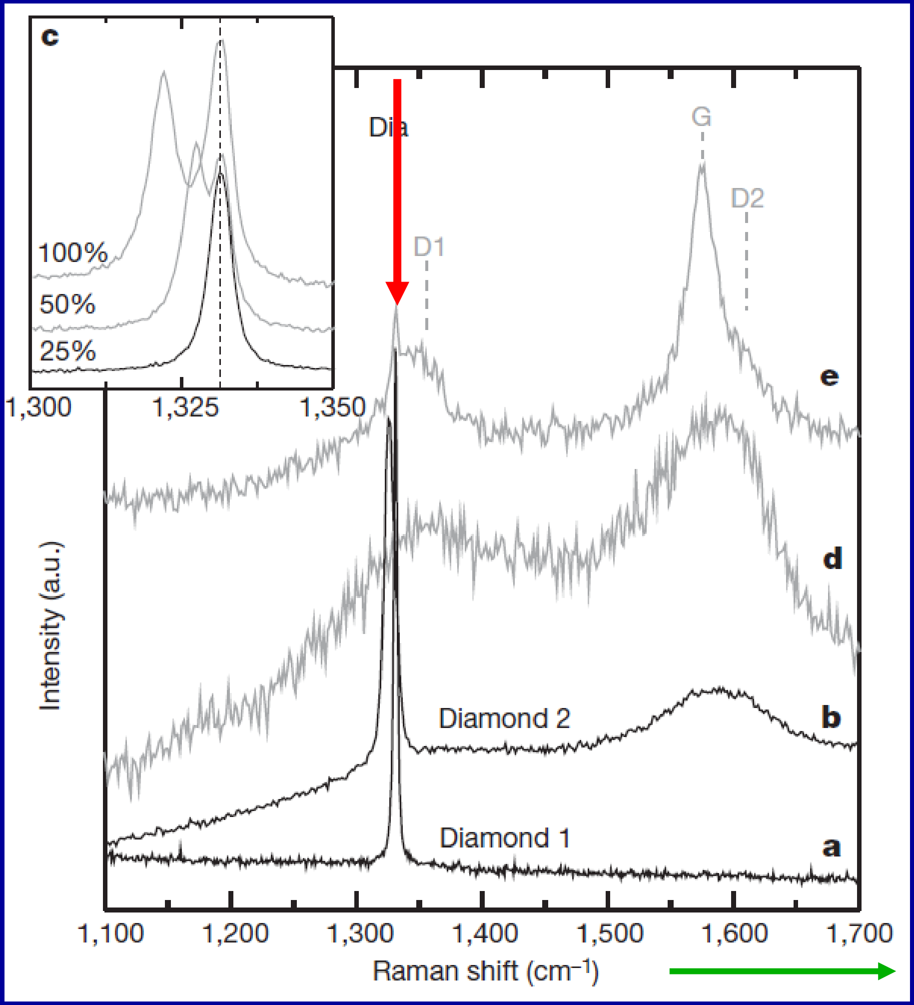


Old zircon

- Zircon obtained from Jack Hills
  - conglomerate from the site of initial discovery of old zircon
- Analysis of mineral inclusions in 1,000 randomly chosen zircon grains
- U-Pb ages
  - Determined by a sensitive high-resolution ion microprobe (SHRIMP II)
  - More than **3,900 Myr-old**
- Optical microscope analysis
  - Inclusions larger than 1 μm in half of zircons
    - Apatite, quarts, zenotime, monazite, rutile, biotite, amphibole, K-feldspar plagioclase
      - Some of them were recognized in the previous studies
  - **Diamond inclusions** in 45 zircon grains
    - Mostly associated with graphite
    - In some case with apatite and quarts

Raman spectrum of diamond

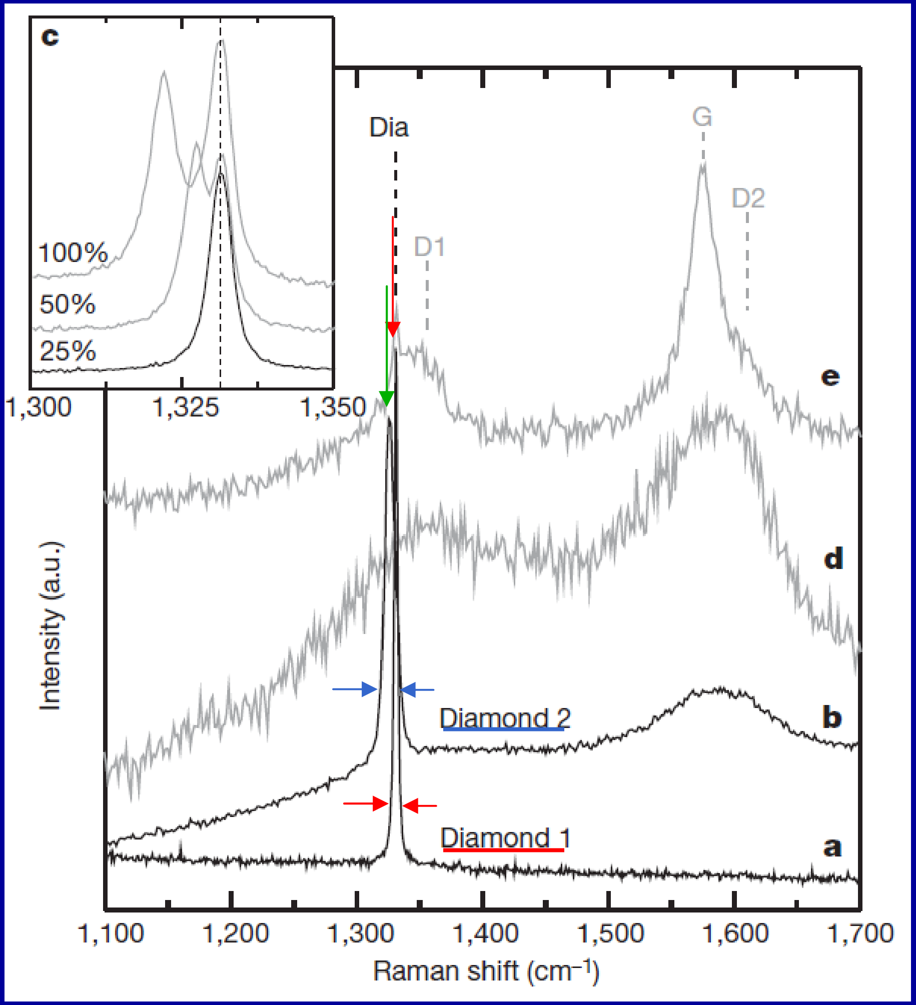
- Raman spectrum of diamond
  - A single first order band near 1,332 cm^{-1}
    - with a typical width of 1.7 cm^{-1}
    - Triply degenerate C-C stretching vibration with F_{2g} symmetry
  - a second order feature near 2600 cm^{-1}



•2600 cm^{-1}

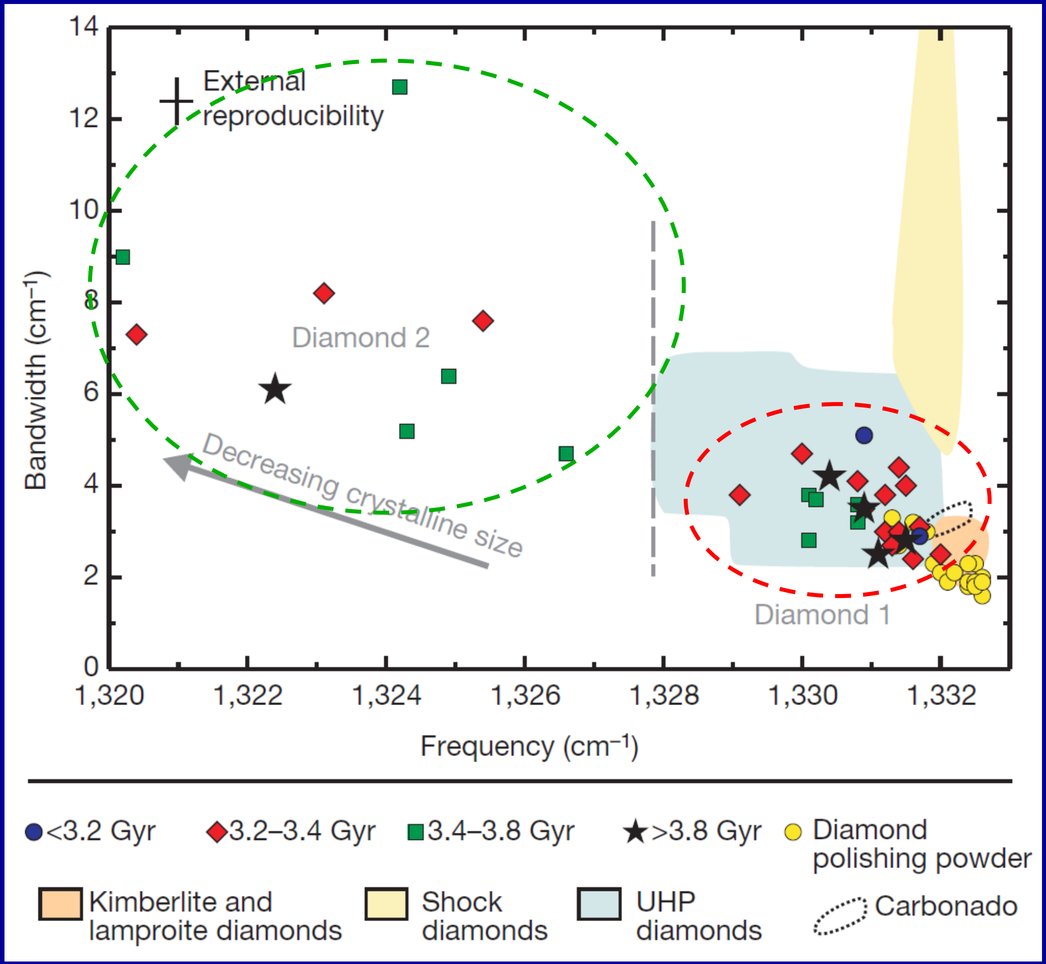
Raman spectrum of diamond

- Raman spectra of the diamond inclusions in the Jack Hills zircons
  - Diamond 1 (most of them)
    - Frequency 1,329 – 1,332 cm^{-1}
    - No second-order feature near 2600 cm^{-1}
  - Diamond 2 (some of them)
    - 1,320 – 1,327 cm^{-1}
    - A larger width compared with that of diamond 1
    - Symmetric



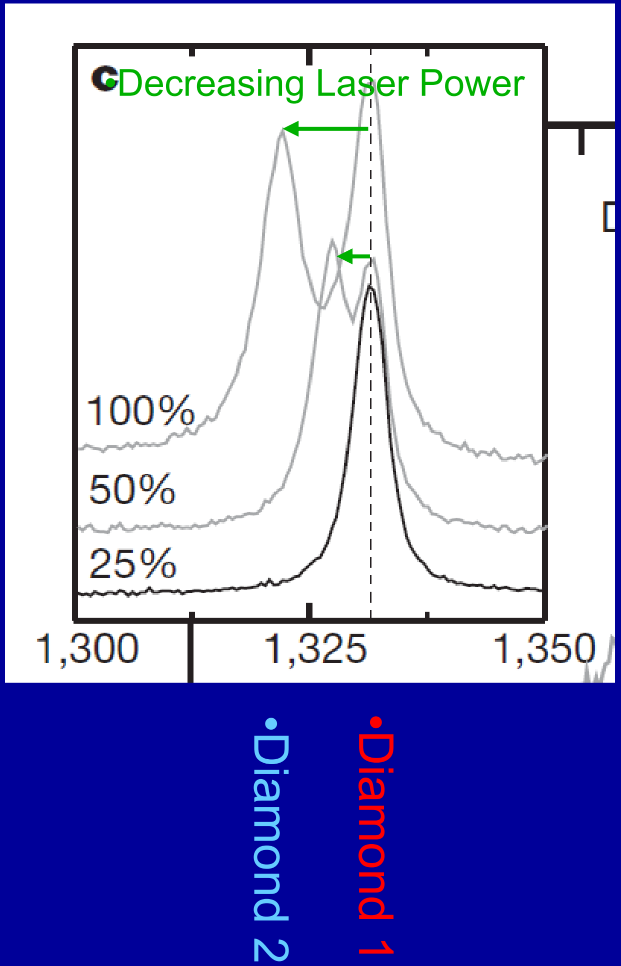
Comparison of the first-order Raman peak between diamonds 1 and 2

- Diamond 2 has larger width than diamond 1.
- Size dependent shift in frequency and band width in diamond 2



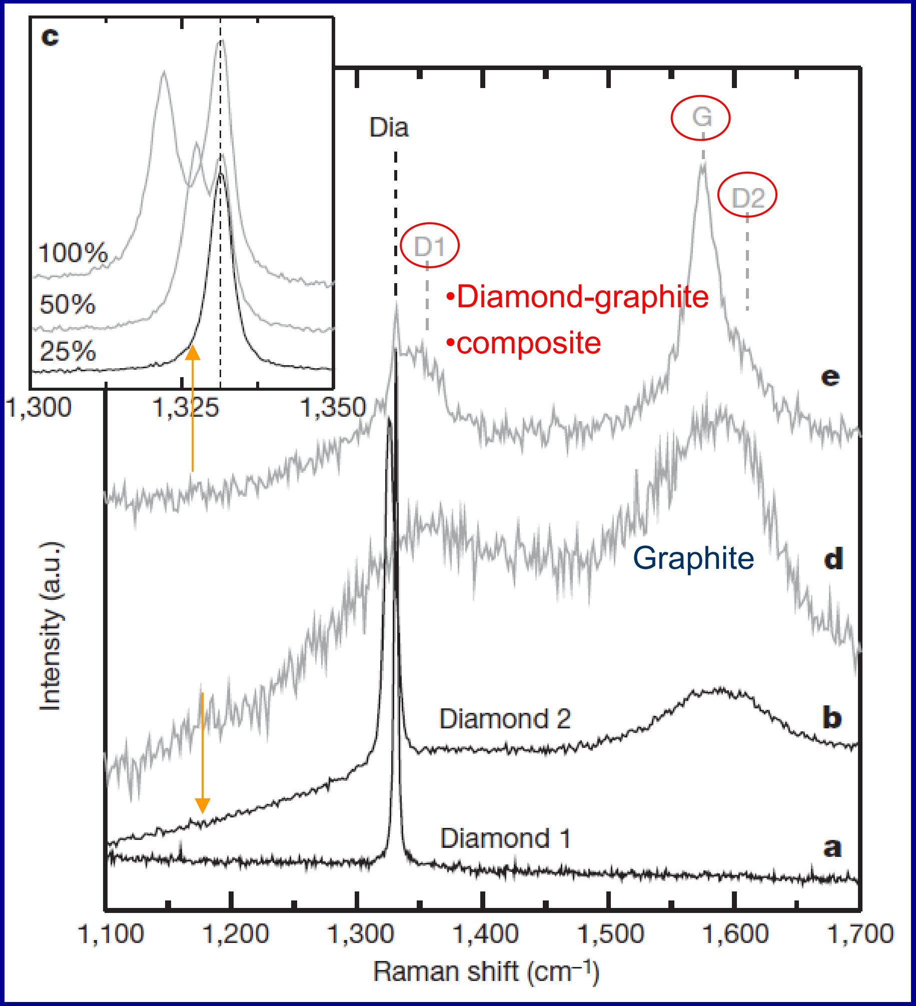
Raman spectrum of diamond

- Raman spectra of the diamond inclusions in the Jack Hills zircons
  - At the margin of some diamond
    - Overlapping spectra of diamond 1 and 2
    - Diamond 1 peak does not shift with decreasing laser power
    - Diamond 2 peak shifts towards higher wave numbers when decreasing the laser power
      - Heated by the incident laser
      - Powders of grain sizes down to 0.1 μm
        - » Size dependent frequency shift towards 1,320 cm-1
        - » Associated symmetric band broadening
      - Different thermal properties of microcrystalline diamond
    - The occurrence of polycrystalline diamond of variable grain size
      - Associated with larger diamonds in some inclusions



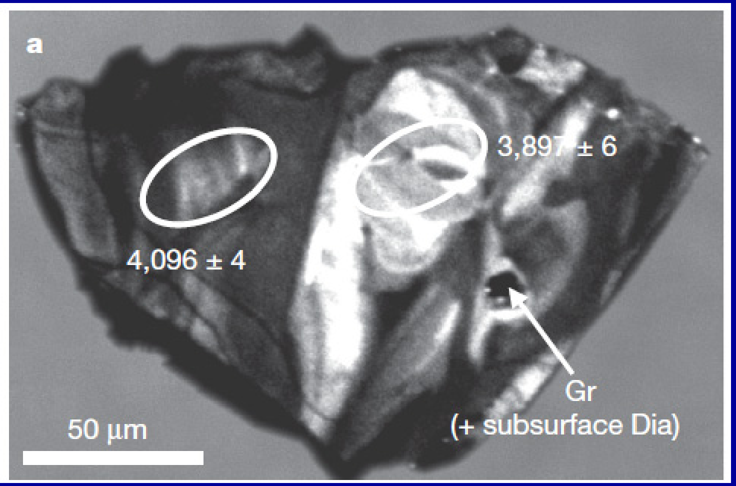
Raman spectrum of diamond

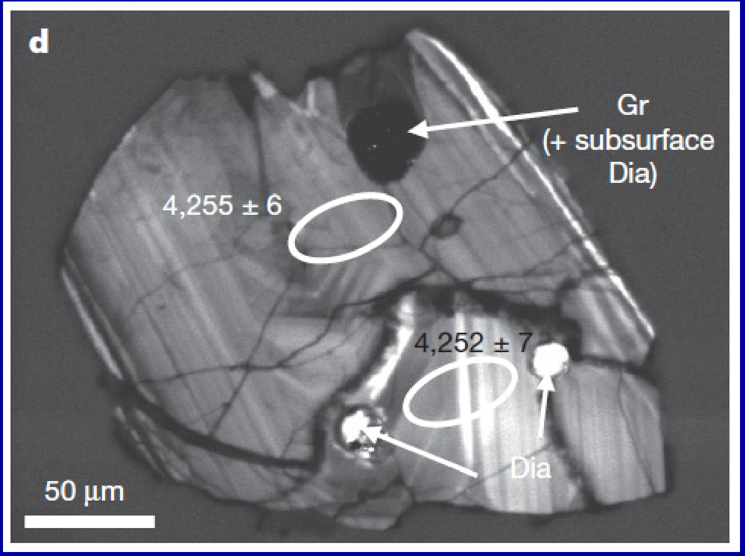
- Londsdaleite
  - Hexagonal polytype of diamond
  - Made by impact
  - A major Raman-active vibration near 1,326 cm^{-1} and additional band near 1,175 cm^{-1}
  - Not found in the present diamond
- Graphite associated with most diamond inclusions
  - Broad first-order graphite peak near 1,580 cm^{-1}
  - Disorder band (‘D1 band’) near 1,350 cm^{-1}
    - For a 532 nm excitation
  - Second order band (‘D2 band’) near 1,620 cm^{-1}



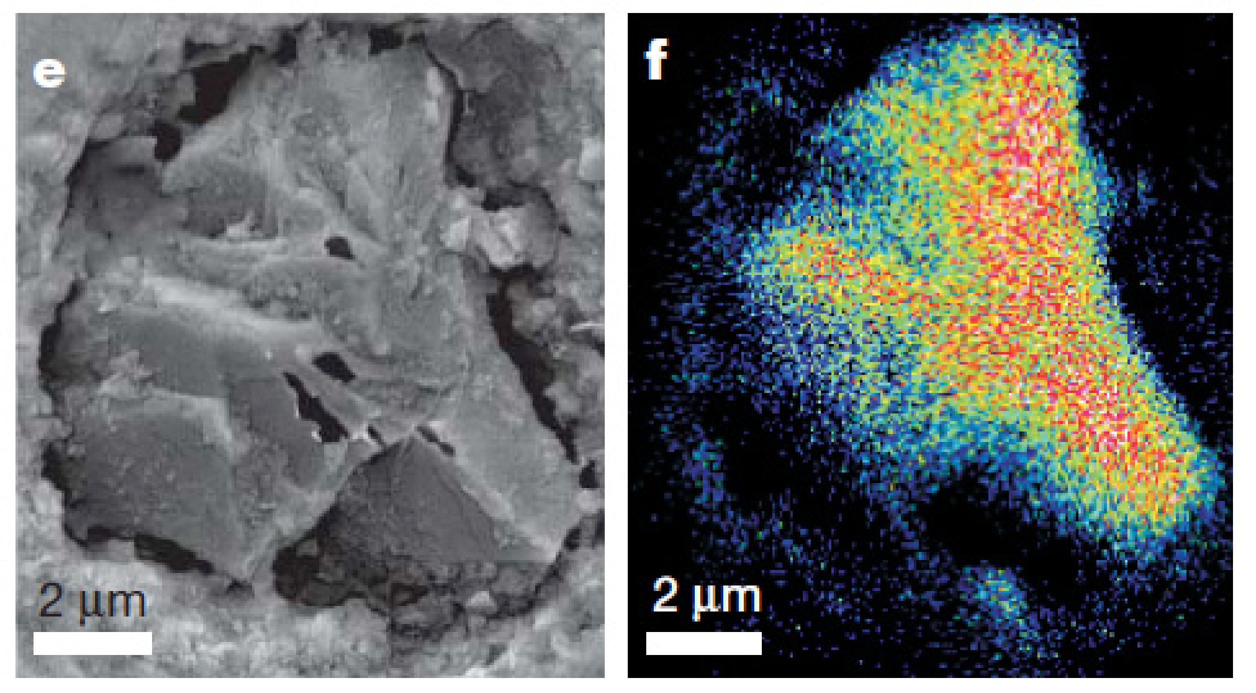
Observation of diamond

- Diamond inclusions
  - Rounded to hexagonal, oval, angular, needle-like shapes
  - Graphite surrounding diamond
    - Graphite is formed after diamond
      - Retrograde transformation
  - Bright cathodoluminescence
    - Distinguish from graphite
      - Graphite has no CL signal





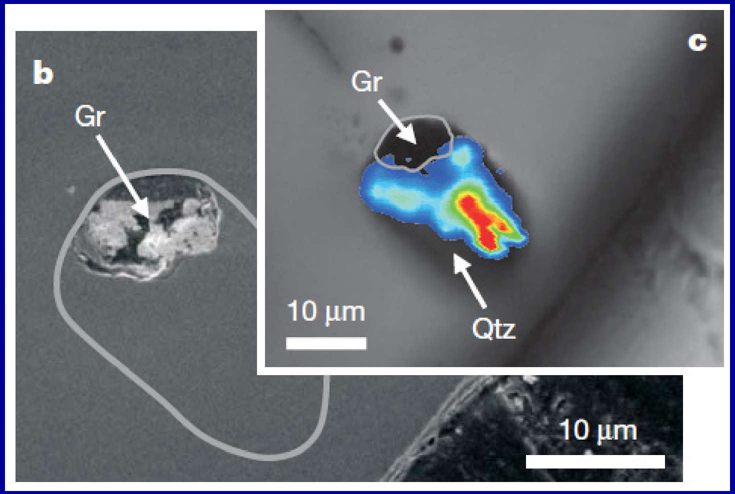
Observation of diamond



- A relatively large exposed diamond aggregate (e)
  - Originally surrounded by graphite
- X-ray analysis (f)
  - Carbon berried in a zircon

Contamination from diamond polishing powder?

– Can be ruled out
  - Some diamond inclusion are completely enclosed in zircon

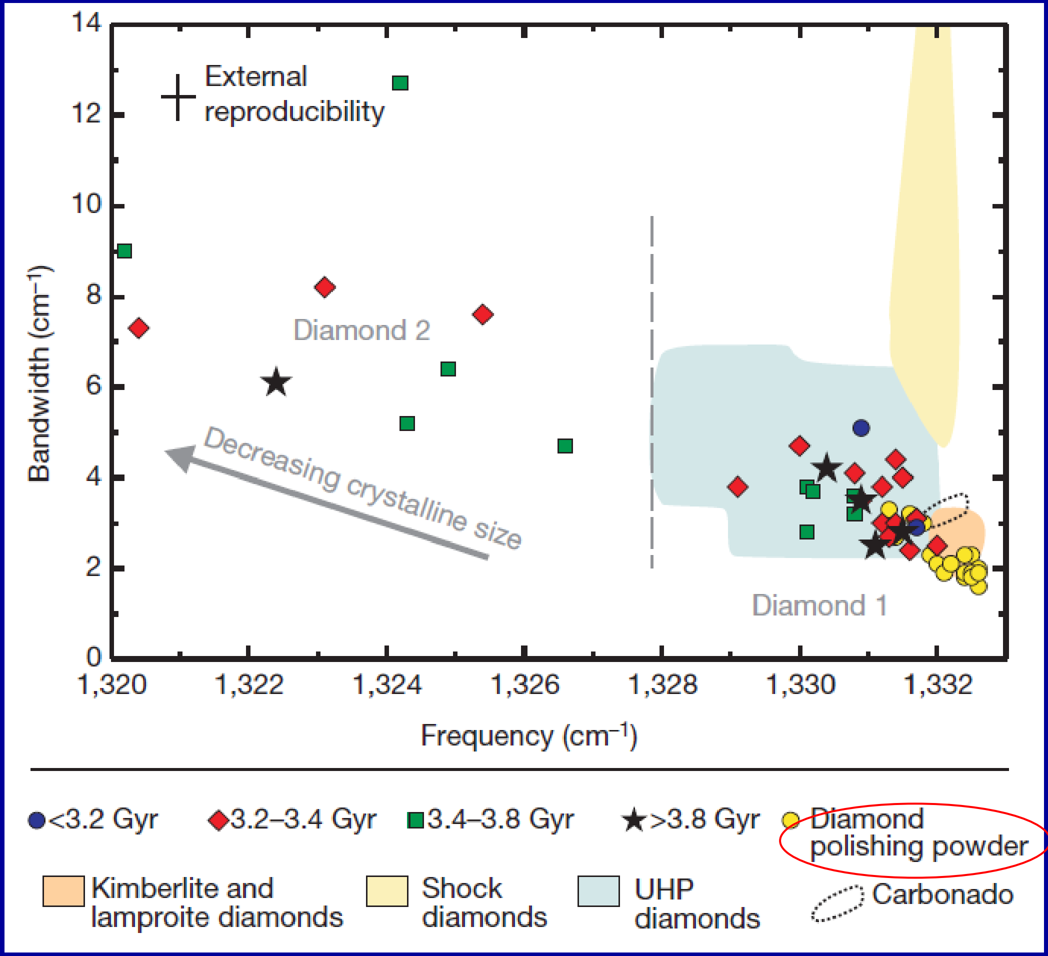


b: Backscattered electron (BSE) image of diamond inclusion. The bright BSE intensity inner region is exposed to the surface.

c: intensity distribution of the first-order Raman band of diamond near 1,332 cm^{-1}.

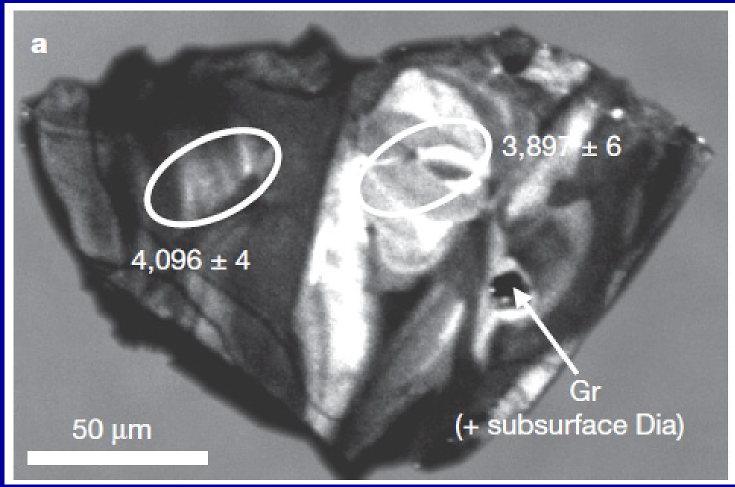
Contamination from diamond polishing powder?

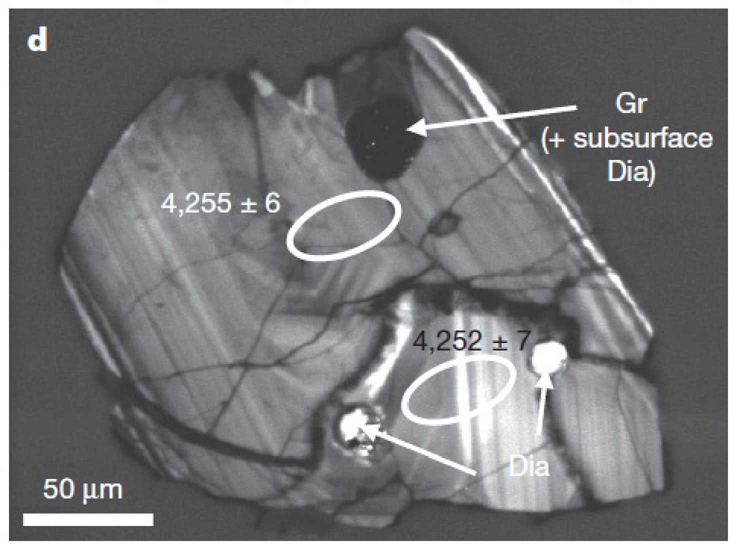
- Can be ruled out
  - Most diamonds reveal variable spectral features that clearly distinguish them from those of the diamond particles in the polishing powder
  - The size of diamond powders are larger than the diamond particles in the polishing materials
    - Inclusion 6-70 μm
    - Polishing < 12 μm
  - Associated with graphite powder



Diamond-bearing zircon

- Highly variable internal structures in CL images
  - Typical of the whole Jack Hills population
- Oscillatory growth zones (d)
  - Typical of zircon crystals grown from a melt or fluid phase
- Irregularly curved growth zones (a)
  - Zircon re-equilibration in an aqueous fluid or melt





Diamond-bearing zircon

- Zones containing zircon shows
  - $^{207}Pb/^{206}Pb$ ages similar to the average statistical age distribution of the Jack hills zircon population
  - Pronounce peak between 3,200-3,400 Myr
  - 10 % of population being older than 3,900 Myr.
- No correlation between the presence of diamond and,
  - The age
  - Internal growth structure
  - Th/U of host zircon crystals
- Polycrystalline diamond (diamond 2) occur in all age groups
- The oldest diamond in terrestrial rocks
  - 4,094±4, 4,132±5 and 4,252±7 Myr
    - Previously know oldest terrestrial diamond
      - 3,300±200 Myr in 90-Myr-old kimberlites from Finsch and Kimberley in South Africa

$^{207}Pb^{*}/^{206}Pb$ age (Myr)
4252 ± 7
4255 ± 6
3380 ± 15
3454 ± 7
3681 ± 5
3058 ± 7
3312 ± 10
3355 ± 11
3266 ± 7
3369 ± 6
3370 ± 12
3600 ± 6
3730 ± 12
3451 ± 7
3389 ± 7
3357 ± 9
3158 ± 8
3378 ± 17
3302 ± 7
3561 ± 8
3499 ± 5
3377 ± 5
3328 ± 12

3476 ± 10
3384 ± 6
3470 ± 6
3225 ± 10
3456 ± 5
3463 ± 8
3395 ± 7
3897 ± 6
4096 ± 4
3437 ± 6
3334 ± 6
3382 ± 6
3402 ± 13
3282 ± 12
3465 ± 4
3467 ± 8
3370 ± 8
3440 ± 6
3451 ± 5
3460 ± 9
4132 ± 5
3299 ± 9
3222 ± 12
3354 ± 5
3358 ± 10
3346 ± 4
3074 ± 7
3455 ± 5

Formation of the Jack Hills diamonds?

- Compare the observed mineralogical features with other natural diamonds
  - Primitive carbonaceous chondrites
  - Meteoritic impact structures
  - Kimberlites and lamproites
    - Delivered from subcontinental lithosphere
  - Komatiites
  - Crustal rock
    - Typically sedimentary rock
    - Subjected to ultrahigh-pressure metamorphic conditions

Similarity with diamonds in UHP metamorphic rock

- The only known setting of micro-diamonds with 3-50 μ m in size
- Inclusions in zircon
- Secondary graphite
  - Slow ascent
- The same Raman spectra feature with diamond 1
  - Different from shock-related diamond
  - Different from kimberlite and lamproites
    - Characterized second-order Raman band near 2,600 cm-1

No high-pressure origin of zircon

- No high pressure minerals
  - No evidence that the observed quartz were derived from coesite
- Mineral inclusion assembly is granodioritic to granitic
- Similar with the mineral paragenesis commonly associated with carbonados
  - large polycrystalline nodules that consist of euhedral diamond crystals (Typically up to 200 μm) set in a matrix of microcrystalline diamond
  - Associated with crustal minerals such as quartz, orthoclase, xenotime and zircon
  - Metamorphism of earliest subducted lithosphere?
  - Radioactive transformation of mantle hydrocarbons?
  - Meteoritic impact on a concentrated biomass
  - Raman spectral character of carbonadoes similar to kimberlite- or lamproite diamonds
  - The present diamond is not related to carbonados

Incorporation of old diamonds in zircon

- Zircons was not formed at UHP metamorphic conditions
  - Absence of high-pressure minerals except diamond
  - With a range of primary and secondary internal structures
  - Highly variable Th/U ratios
- Diamonds and at least some of their host zircons were formed under different pressure-temperature conditions
- The diamond could have been formed during a single event before or at 4,252 Myr ago
  - The crystallization age of the oldest diamond-bearing zircon from Jack Hills zircon
  - Extensive reworking of material separated from the mantle before about 4,140 Myr ago
- Diamond fund in younger zircon grains would be a result of recycling of the older material
- Or diamond could have been incorporated in zircon at several periods by repetition of the process that resulted in diamond formation

Presence of old crust

- Many questions
  - The origin of diamonds?
  - Their incorporation in the Jack Hills zircons?
- Similarities with UHP diamonds
  - Comparison of the observed mineralogical features with those formed under known geological conditions
    - Raman spectra characteristics
    - Occurrence with graphite in zircon
- Presence of a relatively thick continental lithosphere
  - Crust-mantle interaction occurred on Earth as early as 4,250 Myr