Isotopic fingerprints of gold-containing luminescence centers in ^{28}Si

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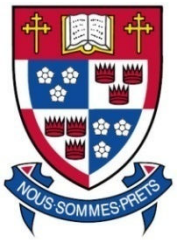

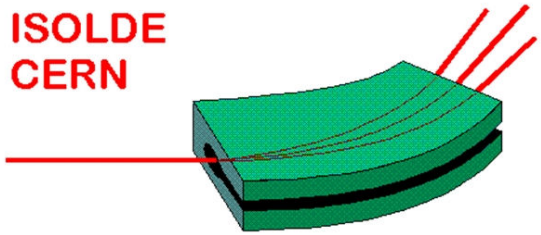

The problem(s) with the kg

- Since 1889, kg defined as mass of International Prototype Kilogram.

- Can be damaged…or worse
- Not well-defined (can accumulate foreign material)
- Ages (at an unknown rate)… 50μg in ~ 100 years(?)

Avogadro project

New definition of the kilogram

Count the number of atoms in a crystal of ^{28}Si (i.e. related to N_A)

Desired accuracy: 2.10^{-8}

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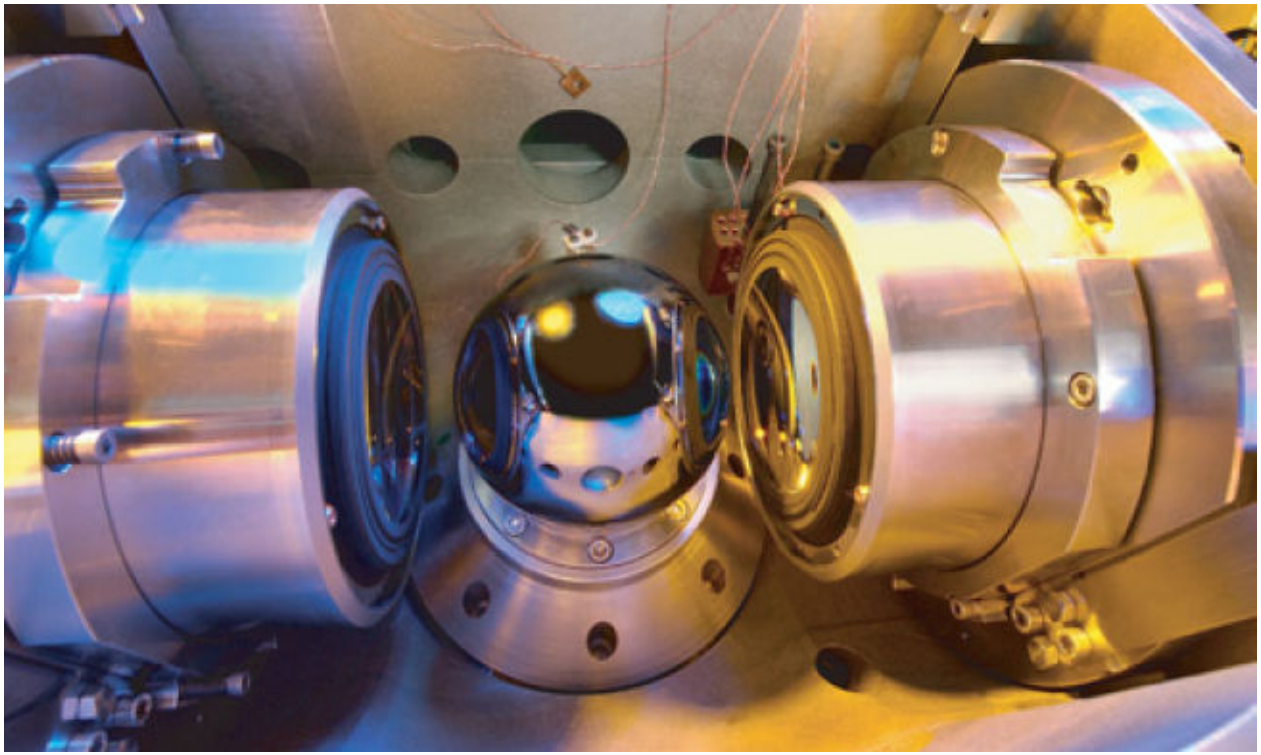
Growth of crystals has been “scaled up” but still expensive: > 1M€ for 1kg

1	Production of silicon tetrafluoride $Na_2{}^{nat}SiF_6 \rightarrow {}^{nat}SiF_4\uparrow + 2\ NaF\downarrow$	Centrotech St. Peterburg
2	Enrichment in centrifuges ${}^{nat}SiF_4 \rightarrow {}^{28}SiF_4$ (>99,985%)	Centrotech St. Peterburg
3	Destillation and Purification ${}^{28}SiF_4 + 2\ CaH_2 \rightarrow {}^{28}SiH_4 + 2\ CaF_2\downarrow$	IChHPS RAS Nizhni Novgorod
4	Chemical Vapour Deposition ${}^{28}SiH_4 \rightarrow {}^{28}Si$ (poly) $+ 2\ H_2$	IChHPS RAS Nizhni Novgorod
5	FZ single crystal growth ${}^{28}Si$ (poly) $\rightarrow {}^{28}Si$ (crystal)	IKZ Berlin

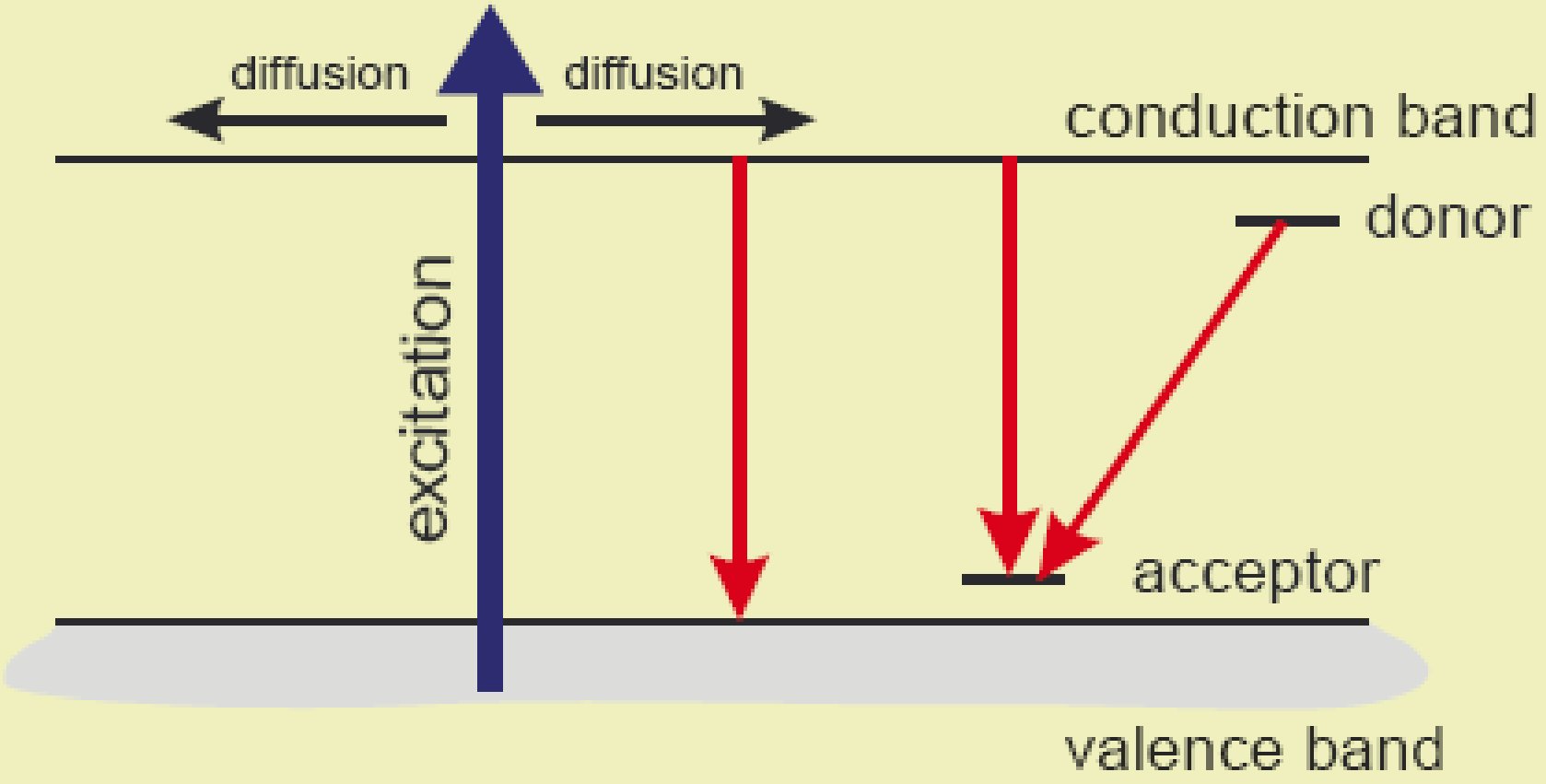


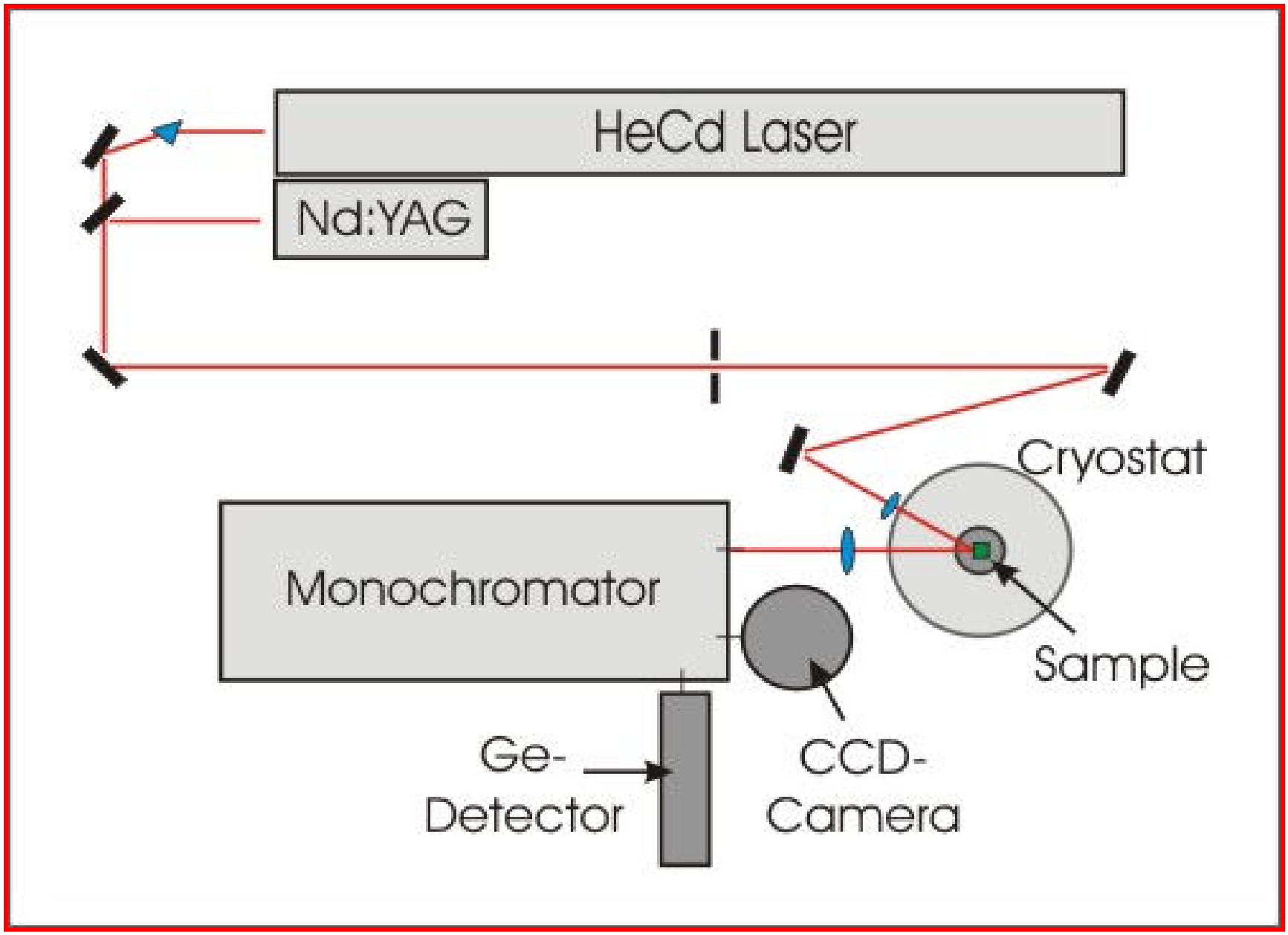
^{28}Si purity ~ 99.995%

Thus far accuracy limit of 3.10^{-7}

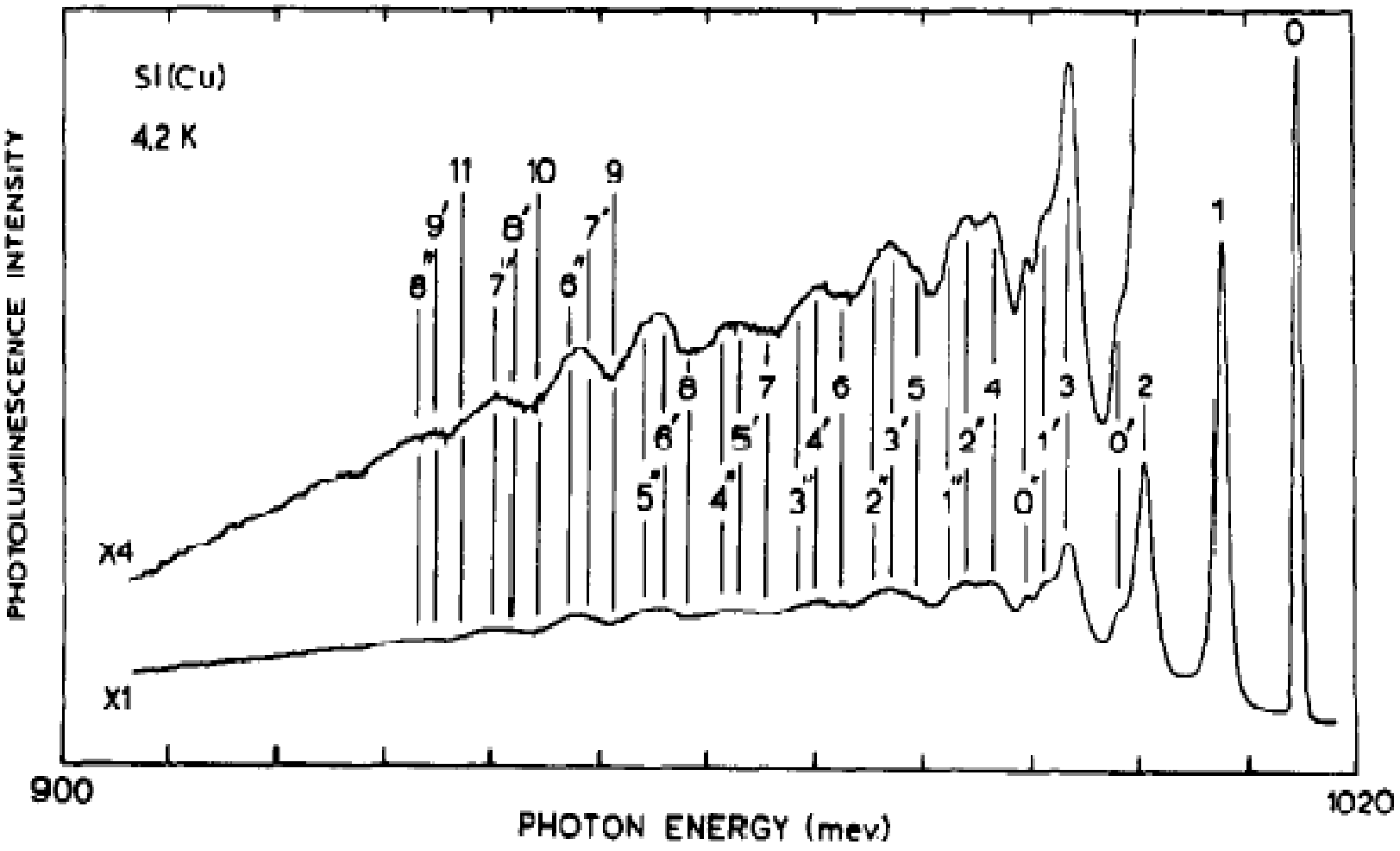
However, one can do more than just redefine the kg with this material…

Photoluminescence




HeCd Laser
Nd:YAG
Cryostat
Monochromator
Sample
Ge-
Detector
CCD-
Camera

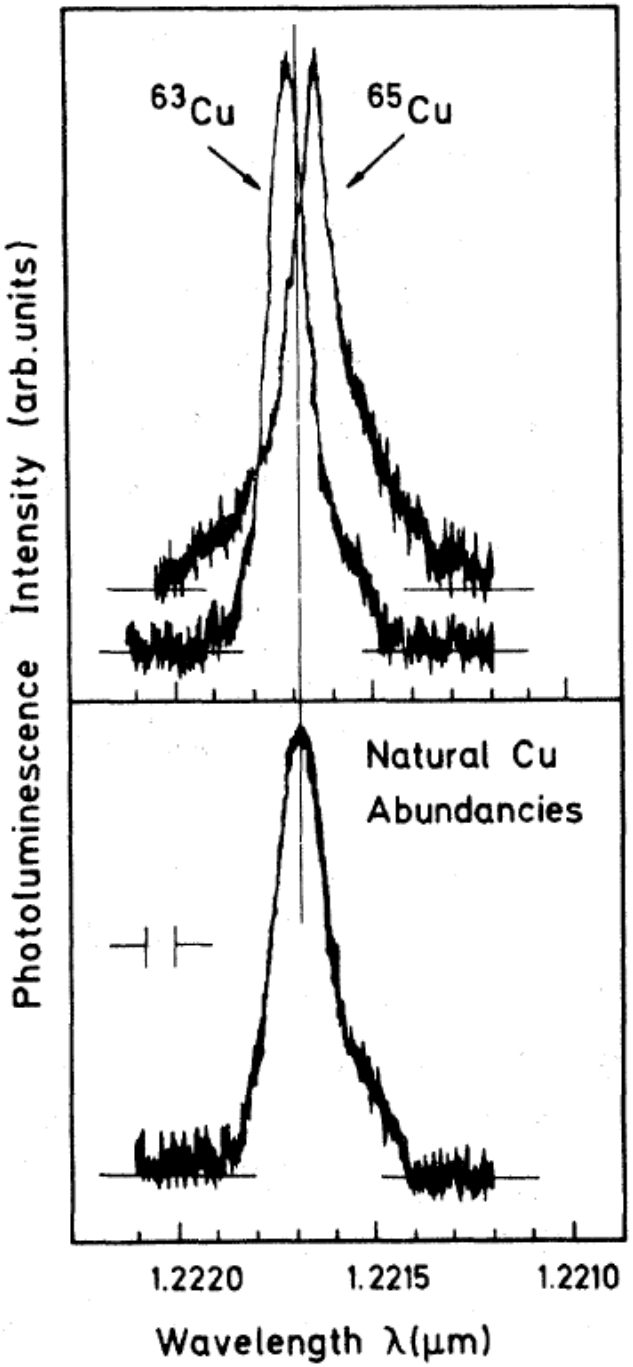
Optical spectroscopy of ^{28}Si: new information about “known” centres e.g. Cu



Cu involvement long-established via isotope shift of main line (Weber *et al* 1982)

Shift is small though…

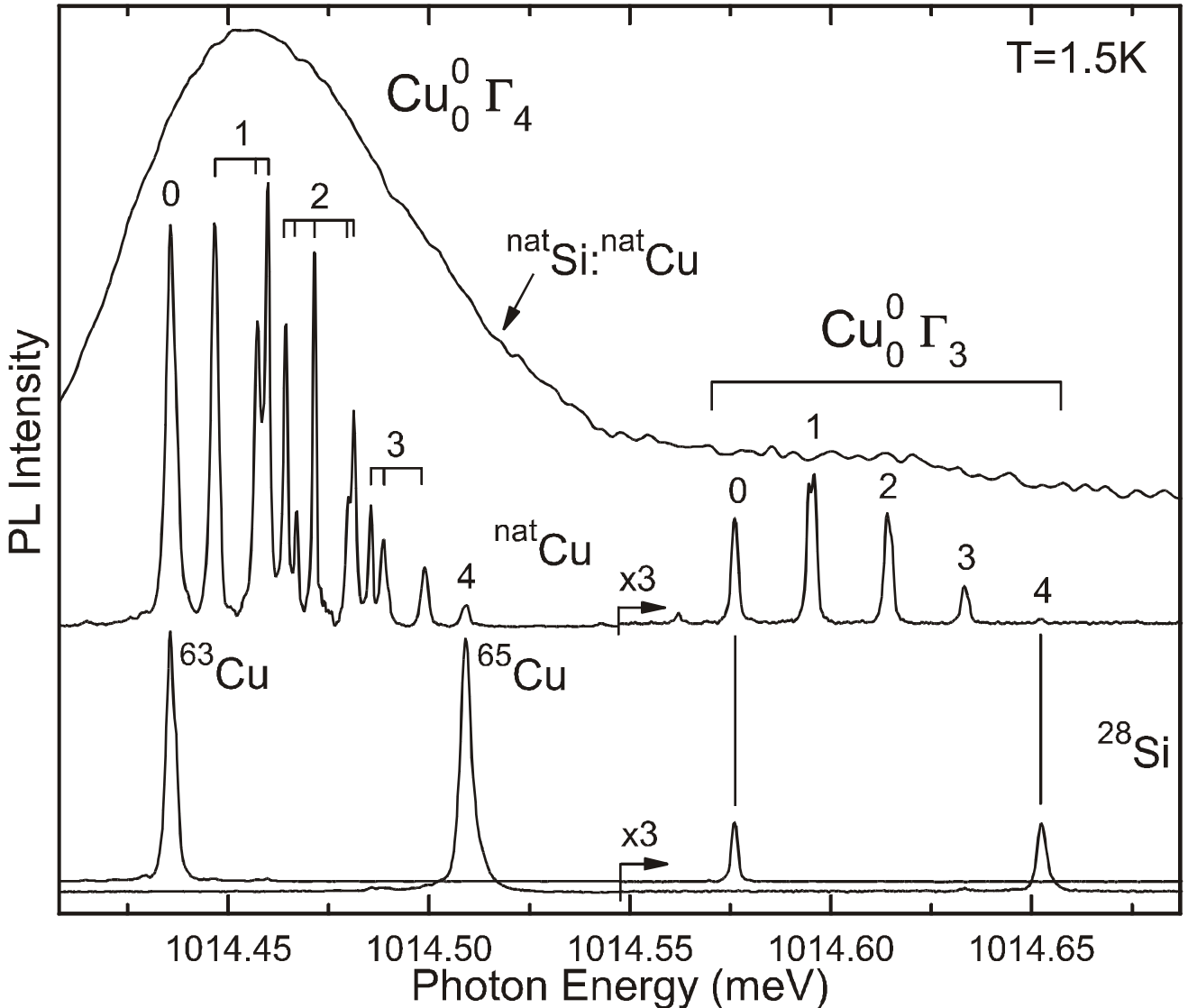
Model generally accepted as being a Cu pair.



Big surprise when same system examined in ^{28}Si

Natural Cu: ~69% ^{63}Cu + ~31% ^{65}Cu

The center **must** contain four Cu; 0, 1, 2, 3, 4 labels the number of ^{65}Cu

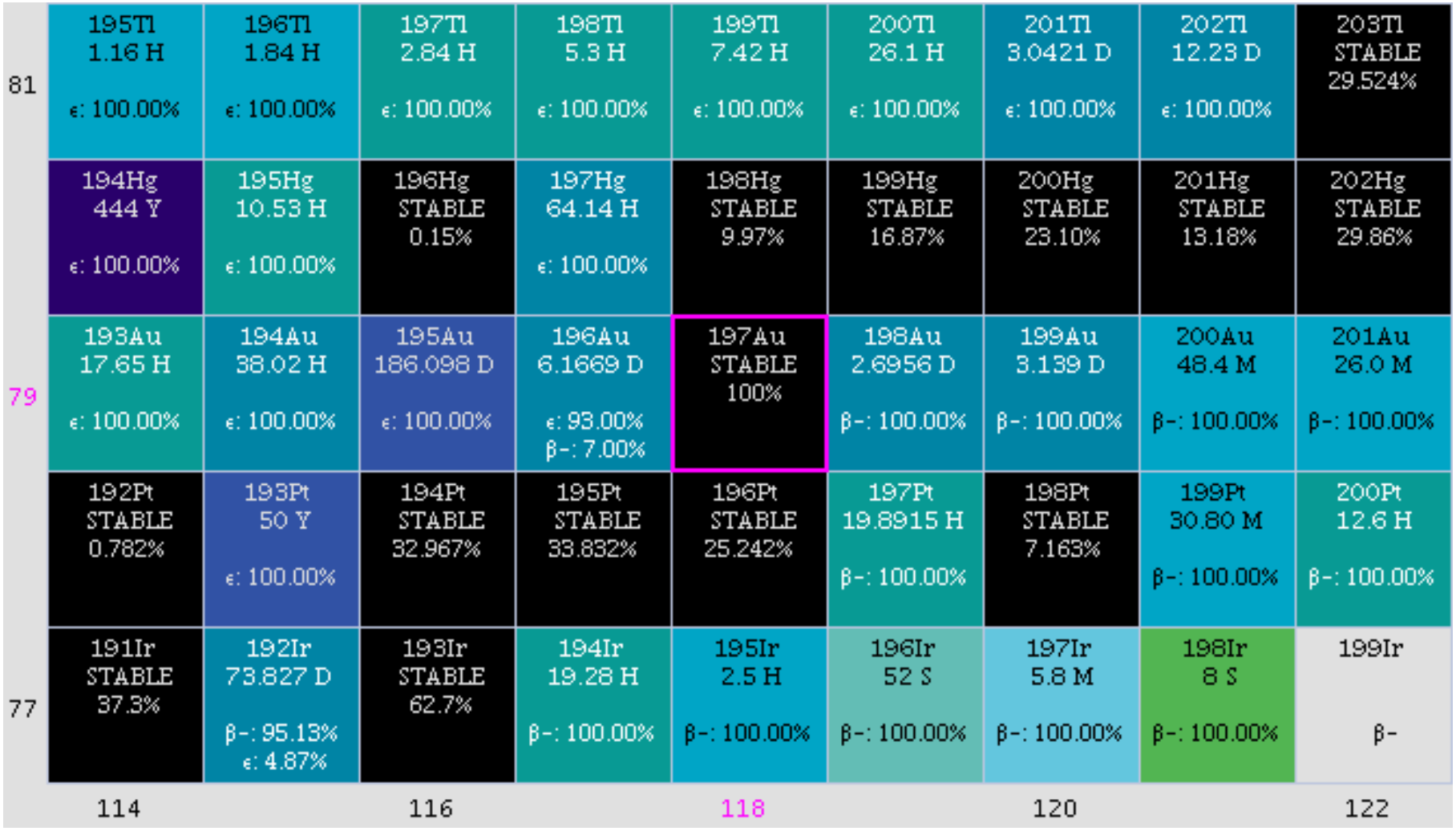


Growing evidence for the presence of multi-atom centres in Si involving Cu, Ag and Au, now revealed through "isotopic fingerprints"

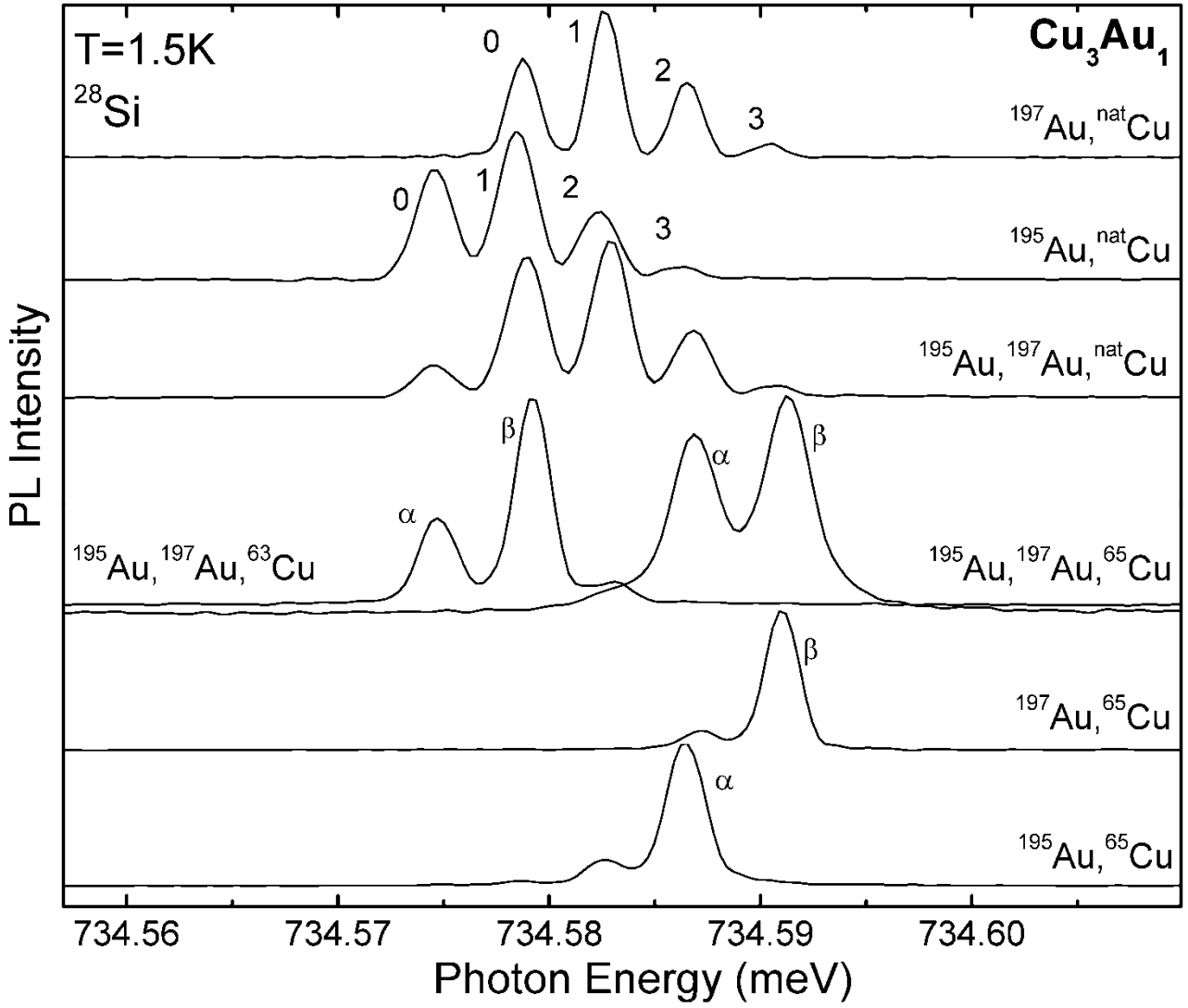
Label	E(meV)	Model
'Cu-pair center'	1014 meV	Cu_4
'perturbed Cu-pair center'	944 meV	Ag_1Cu_3
'Ag_s center'	778 meV	Ag_4
'Fe center'	735 meV	$AuCu_3$
'Fe-B center'	1066 meV	$AuCu_4$

How many Au?

Only one stable isotope of Au → solution implant with ^{195}Au at ISOLDE


81
79
77
195Tl 1.16 H ε: 100.00%
196Tl 1.84 H ε: 100.00%
197Tl 2.84 H ε: 100.00%
198Tl 5.3 H ε: 100.00%
199Tl 7.42 H ε: 100.00%
200Tl 26.1 H ε: 100.00%
201Tl 3.0421 D ε: 100.00%
202Tl 12.23 D ε: 100.00%
203Tl STABLE 29.524%
194Hg 444 Y ε: 100.00%
195Hg 10.53 H ε: 100.00%
196Hg STABLE 0.15%
197Hg 64.14 H ε: 100.00%
198Hg STABLE 9.97%
199Hg STABLE 16.87%
200Hg STABLE 23.10%
201Hg STABLE 13.18%
202Hg STABLE 29.86%
193Au 17.65 H ε: 100.00%
194Au 38.02 H ε: 100.00%
195Au 186.098 D ε: 100.00%
196Au 6.1669 D ε: 93.00% β-: 7.00%
197Au STABLE 100%
198Au 2.6956 D β-: 100.00%
199Au 3.139 D β-: 100.00%
200Au 48.4 M β-: 100.00%
201Au 26.0 M β-: 100.00%
192Pt STABLE 0.782%
193Pt 50 Y ε: 100.00%
194Pt STABLE 32.967%
195Pt STABLE 33.832%
196Pt STABLE 25.242%
197Pt 19.8915 H β-: 100.00%
198Pt STABLE 7.163%
199Pt 30.80 M β-: 100.00%
200Pt 12.6 H β-: 100.00%
191Ir STABLE 37.3%
192Ir 73.827 D β-: 95.13% ε: 4.87%
193Ir STABLE 62.7%
194Ir 19.28 H β-: 100.00%
195Ir 2.5 H β-: 100.00%
196Ir 52 S β-: 100.00%
197Ir 5.8 M β-: 100.00%
198Ir 8 S β-: 100.00%
199Ir β-
114
116
118
120
122

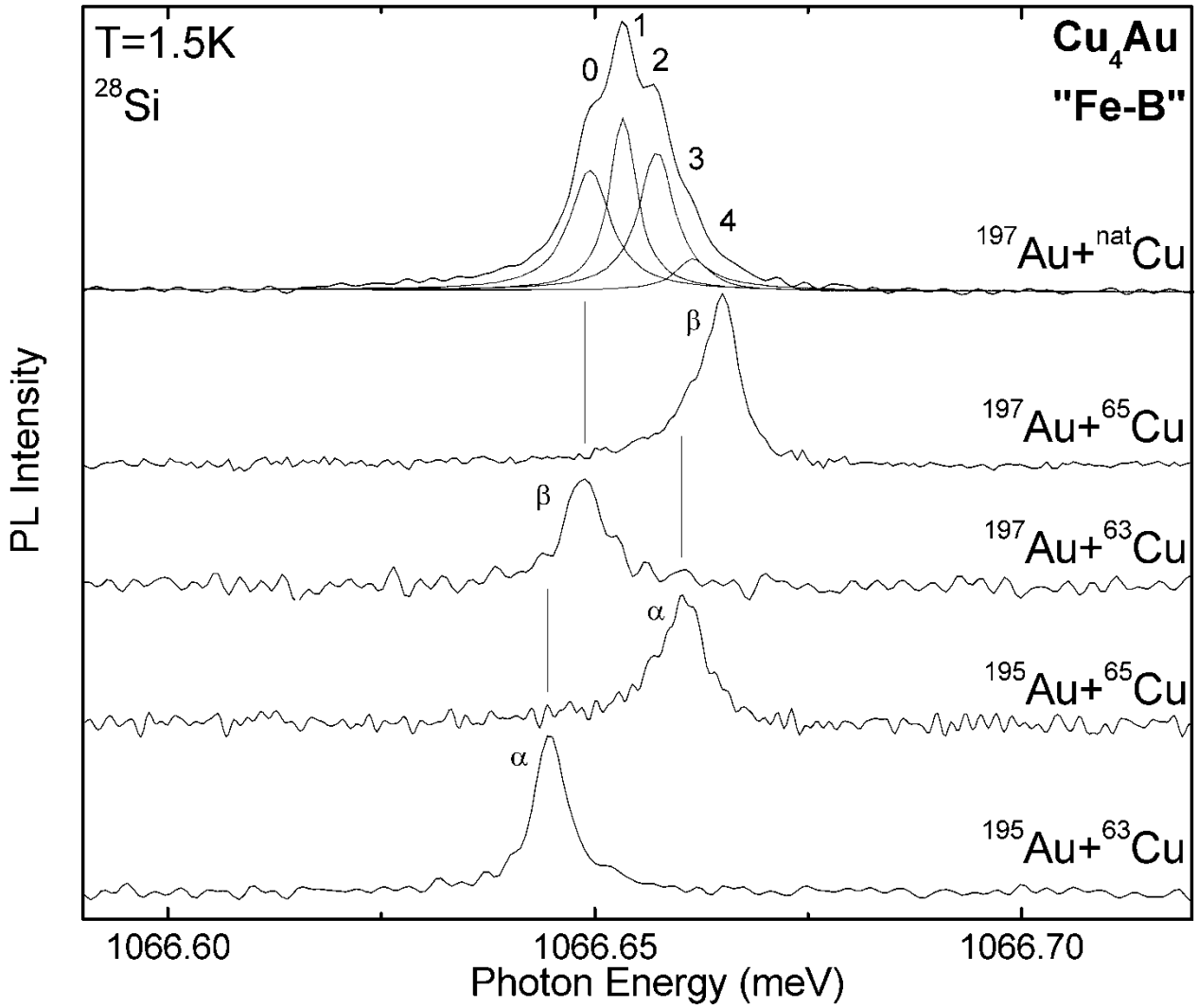
~735 meV Cu_3Au center, previously thought to be related to Fe:



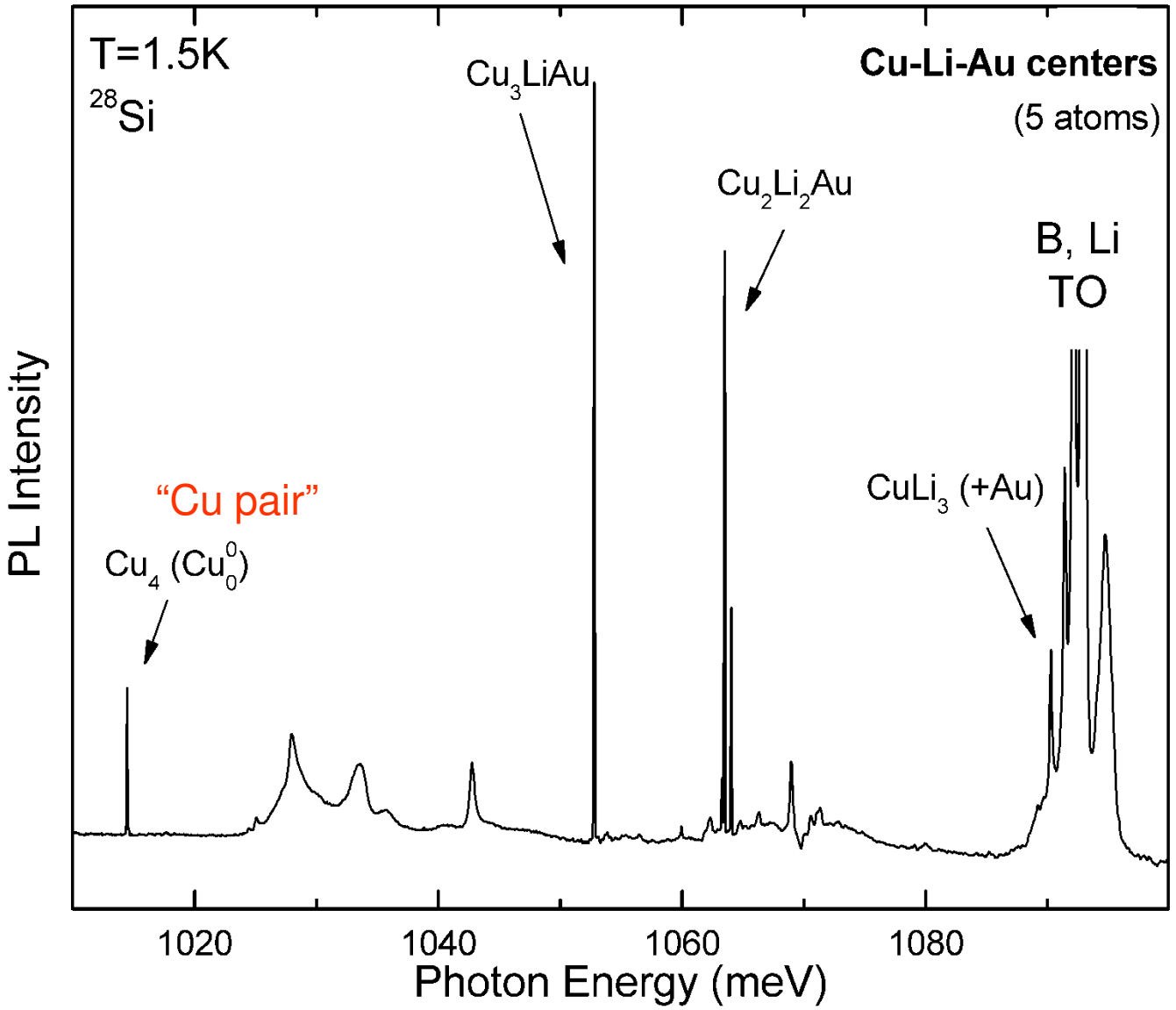
Proof that the Cu_3Au center contains **one** gold atom (α, β label # ^{197}Au)

So-called “Fe-B” center:

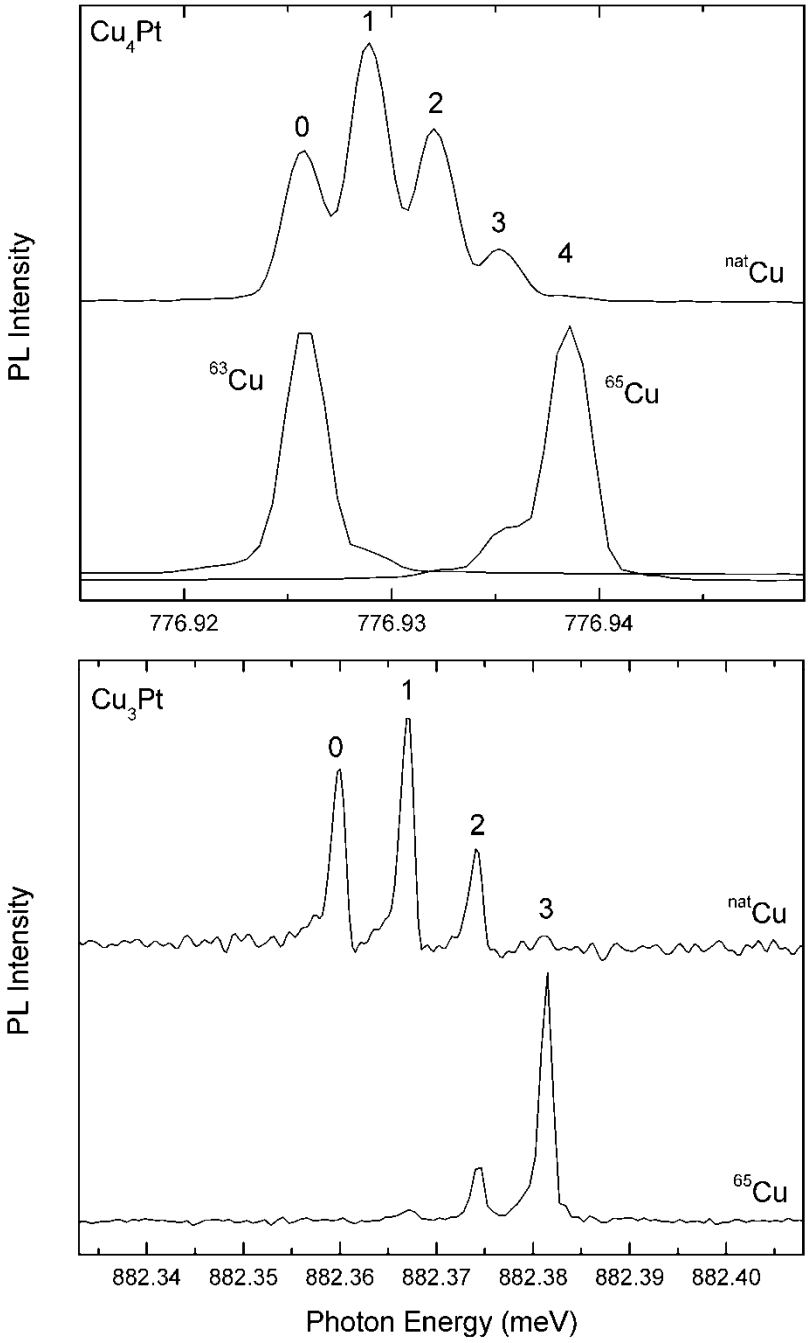
previously shown to contain 4 Cu, also contains Au



CuLi centers – we previously did not realize that Au was involved!



NOT 4!



Bonus!

^{195}Au decays to ^{195}Pt, so we also see previously studied Pt PL centers at ~777 meV and ~882 meV

The isotopic fingerprint versus Cu shows that these centers contain four and three Cu, respectively

So, these Pt centers are again examples of four and five atom centers (the Pt isotope shifts are currently under study)

meV	IBE Complex	old labels
652.9	Ag_2Au_2	
712.0	Ag_3Au	
735.0	Cu_3Au	“Fe” or “Fe-Au”
735.3	Cu_2AuLi	
746.8	$CuAuLi_2$	
765.2	Li_3Au	
778.0	Ag_4	“single Ag”
867.4	Cu_2Ag_2 (**Cu)	
944.0	Cu_3Ag (*Cu)	“Cu-pair”
1014.5	Cu_4 (Cu^0_0)	“Cu-pair”
1052.8	Cu_3Li	
1063.5	Cu_2Li_2	
*1066.6	4 Cu + Au?	“Fe-B”
1090.2	$CuLi_3$	

CONCLUSIONS

Multi-atom impurity centres are revealed in single isotope silicon, overturning previous data on “well-known” impurities.

Isotopic fingerprints can be obtained even for Au, which has only one stable isotope.

These four and five atom luminescence centers based on (Cu, Ag, Au, Pt, Li) are ubiquitous in quenched Si containing these impurities

~ 20 such centers have now been identified

Time for some theory!

How do they form after thermal quenching?

Why are they apparently so stable?

Can their properties (energies, LVM replicas, isotope shifts) be understood?