

# Magnetism and superconductivity in expanded fullerenes

M. Menelaou,<sup>1,2</sup> Y. Takabayashi,<sup>1,3</sup> Peter J. Baker,<sup>4</sup> K. Prassides,<sup>1,5</sup>

<sup>1</sup> WPI-Advanced Institute for Materials Research, Tohoku University, Sendai, Japan

<sup>2</sup> Cyprus University of Technology, Limassol, Cyprus

<sup>3</sup> Nagoya Institute of Technology, Nagoya, Japan

<sup>4</sup> The ISIS Facility, Rutherford Appleton Laboratory, UK

<sup>5</sup> Osaka Prefecture University, Osaka, Japan

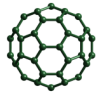
Second EUT+ Workshop on Nanomaterials and Nanotechnologies

Cartagena, Spain

1-2 February, 2022

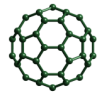
## Overview of the talk

### Fulleride Superconductivity - $A_3C_{60}$ superconductors



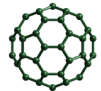
Structural and magnetic characterization of  $Rb_xCs_{3-x}C_{60}$

### Transverse-field muon spin rotation (TF- $\mu^+$ SR) technique

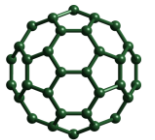


TF- $\mu^+$ SR and superconducting fullerides

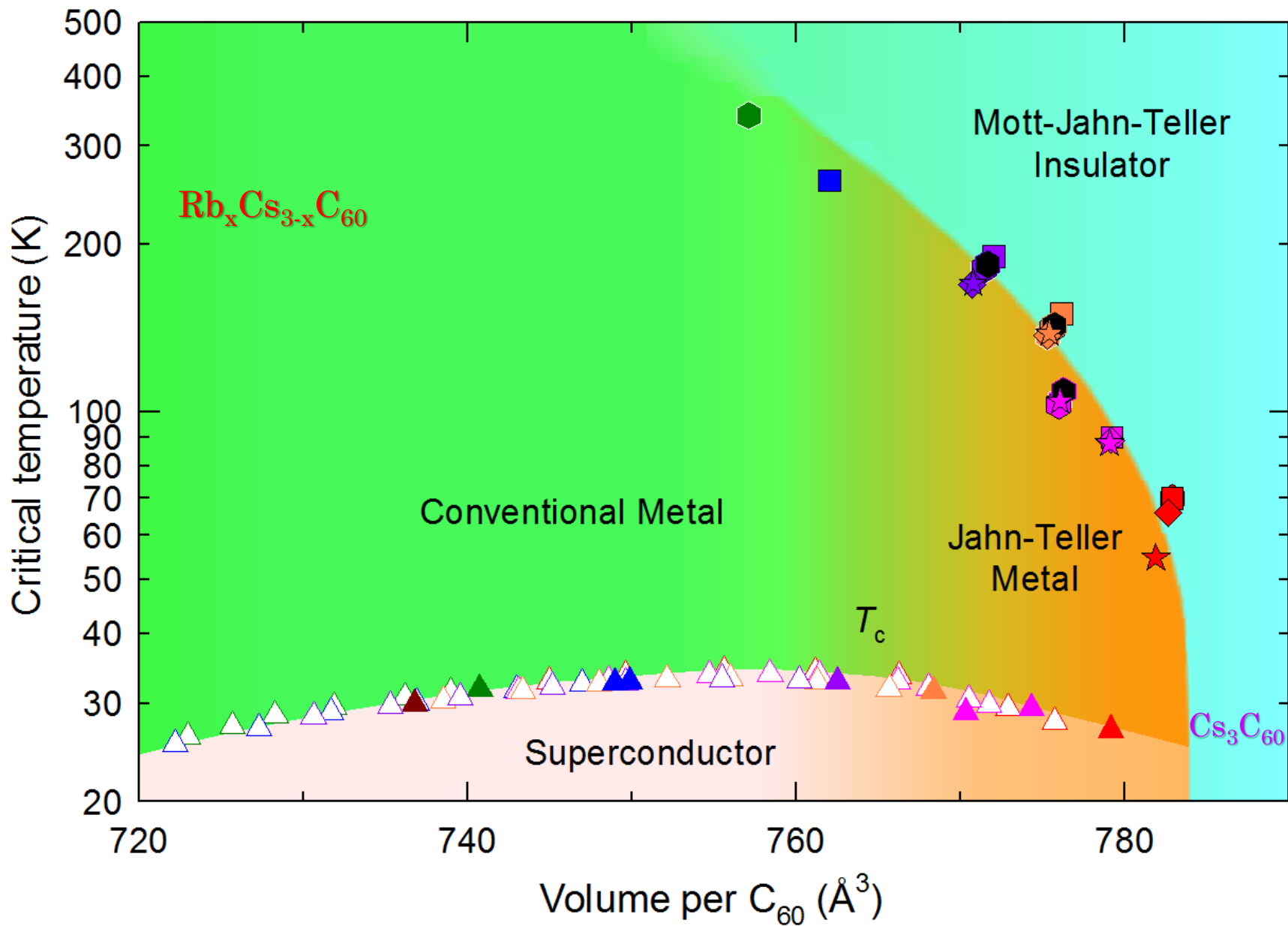
### Summary



Evolution of superconducting properties of  $Rb_xCs_{3-x}C_{60}$  compositions



# Global phase diagram of the $fcc A_3C_{60}$ fullerenes



# Synthesis of $fcc Rb_xCs_{3-x}C_{60}$ fullerides

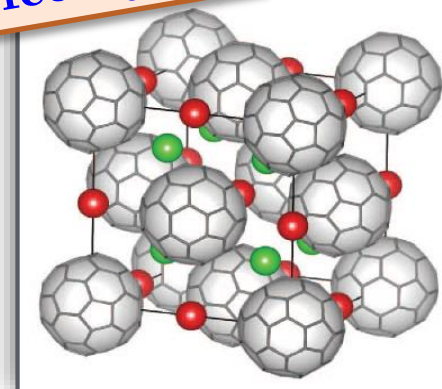



(x)

(3-x)

$$0.35 \leq x \leq 2$$

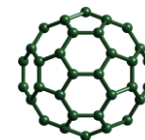
$fcc A_3C_{60}$



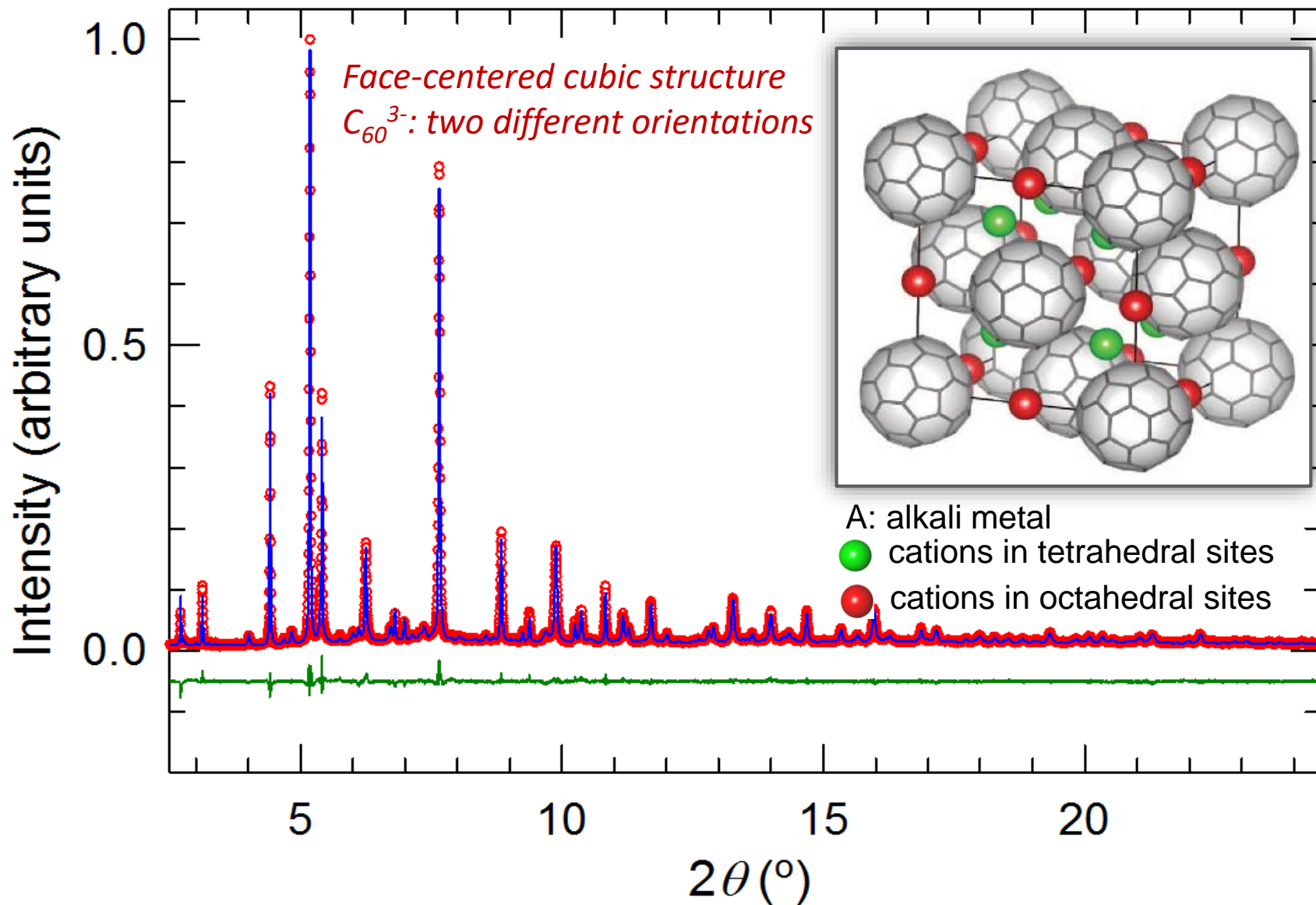
  $Rb_xCs_{3-x}C_{60}$  reproducibly synthesised using a solid state annealing route

 Phase pure samples (>1 g)

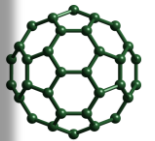
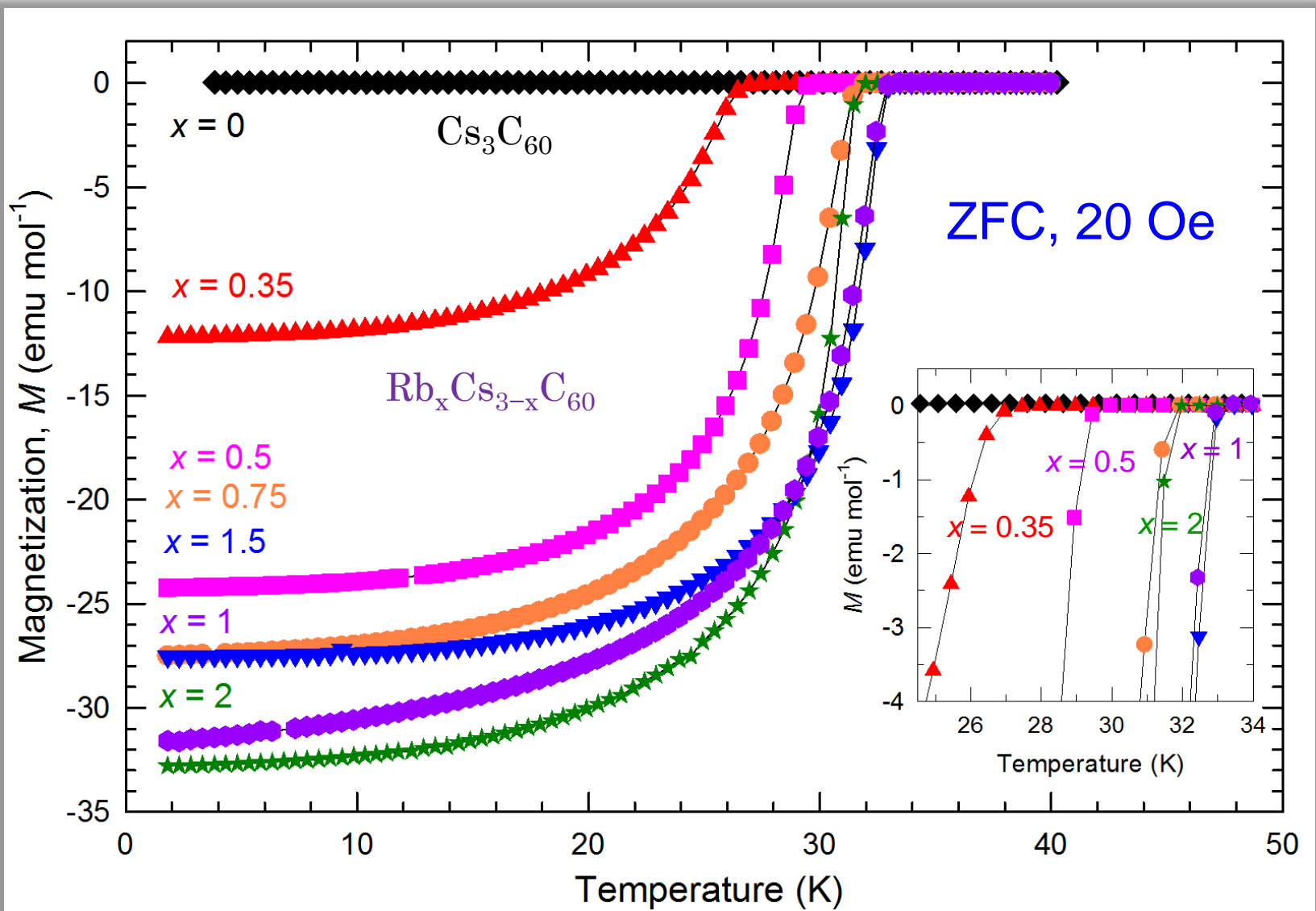
 Good stoichiometry control



*fcc*  $A_3C_{60}$




# Temperature dependence of the magnetization for $\text{Rb}_x\text{Cs}_{3-x}\text{C}_{60}$ compositions

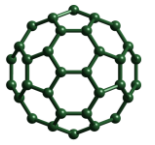
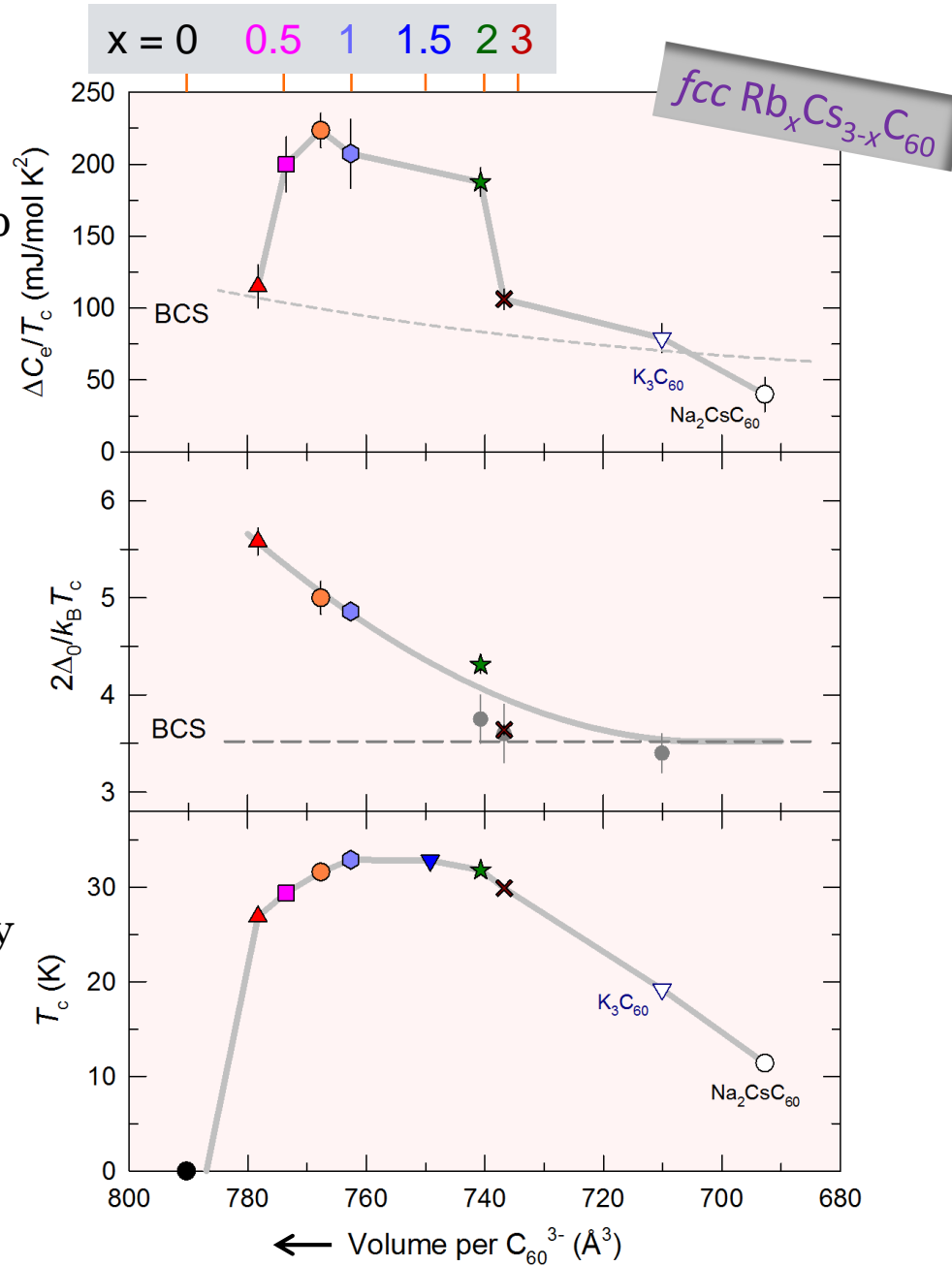


# Superconducting properties as functions of packing density

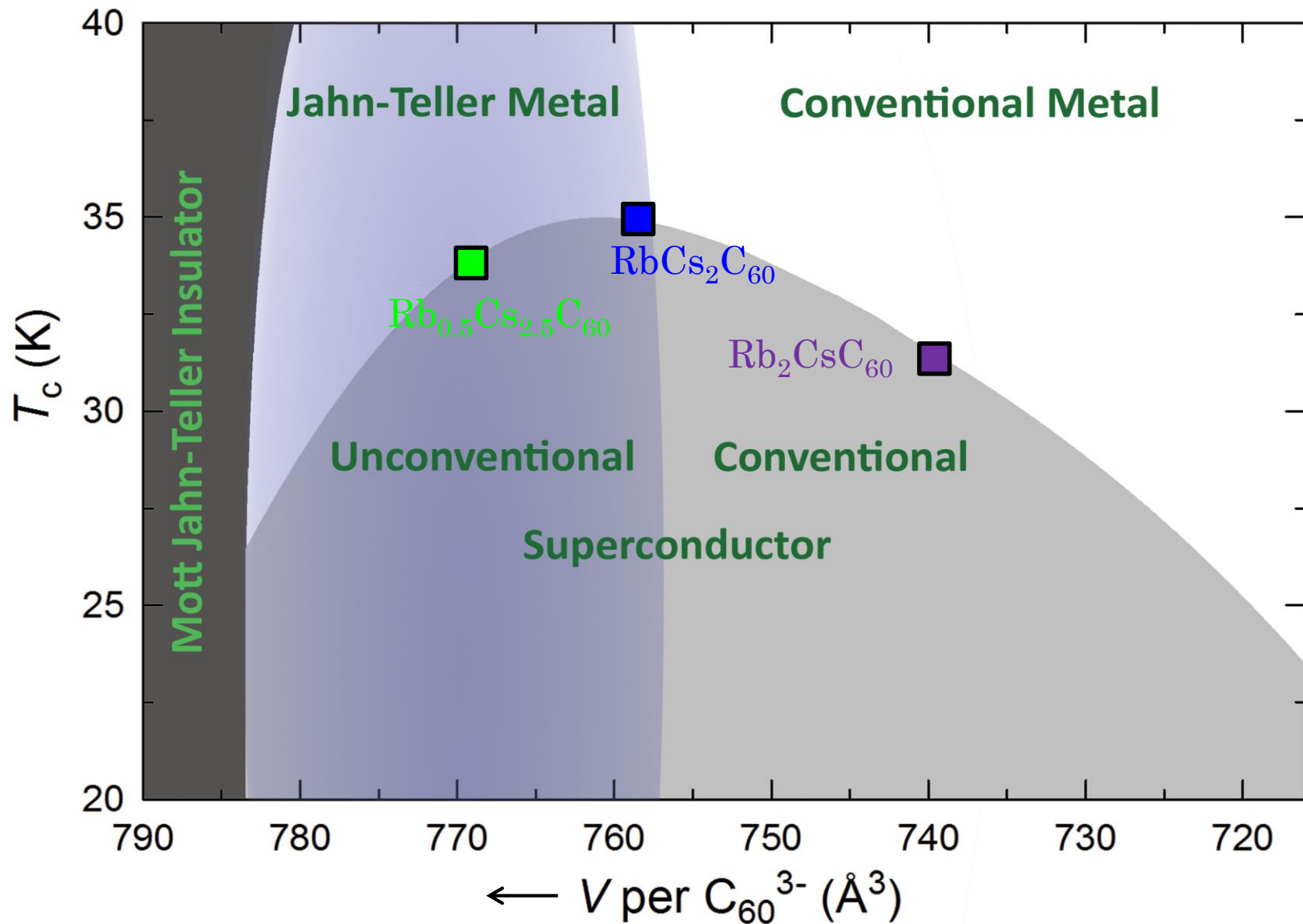
 Specific heat jump at  $T_c$

 Superconducting gap from NMR

 Superconductivity dome



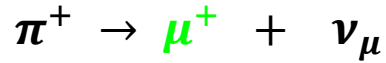
# Electronic phase diagram of the *fcc* $A_3C_{60}$ fullerenes



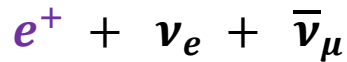


# Transverse-field muon spin rotation (TF- $\mu^+$ SR) technique

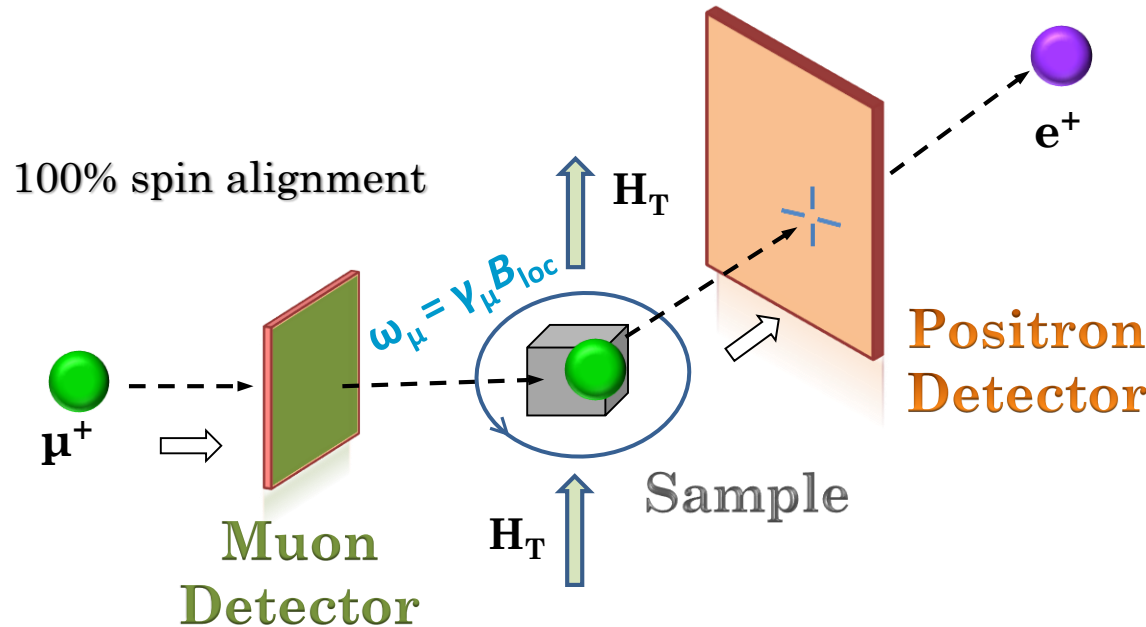
## Pion Decay



## Muon Decay



## Transverse-field geometry



## Selected properties of the muon

Mass:  $207 m_e = 0.1126 m_p$

Spin:  $\frac{1}{2}$

Charge:  $+e$

Lifetime:  $2.2 \mu s$

Magnetic moment:  $4.84 \times 10^{-3} \mu_B$

Gyromagnetic ratio:  $(2\pi) \times 13.554 \text{ kHz G}^{-1}$

$\mu^+$

MuSR@ISIS, UK



## Why TF- $\mu^+$ SR?

- The muon acts as a microscopic probe of the local magnetic field distribution in the vortex state and provides a direct way to measure the London penetration depth,  $\lambda$

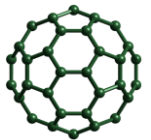
$$\frac{1}{\lambda^2} = \frac{4\pi n_s e^2}{m^* c^2} \times \frac{1}{1 + \xi/\ell}$$

when:  $\xi \ll \ell$  clean limit  
 $\xi \gg \ell$  dirty limit

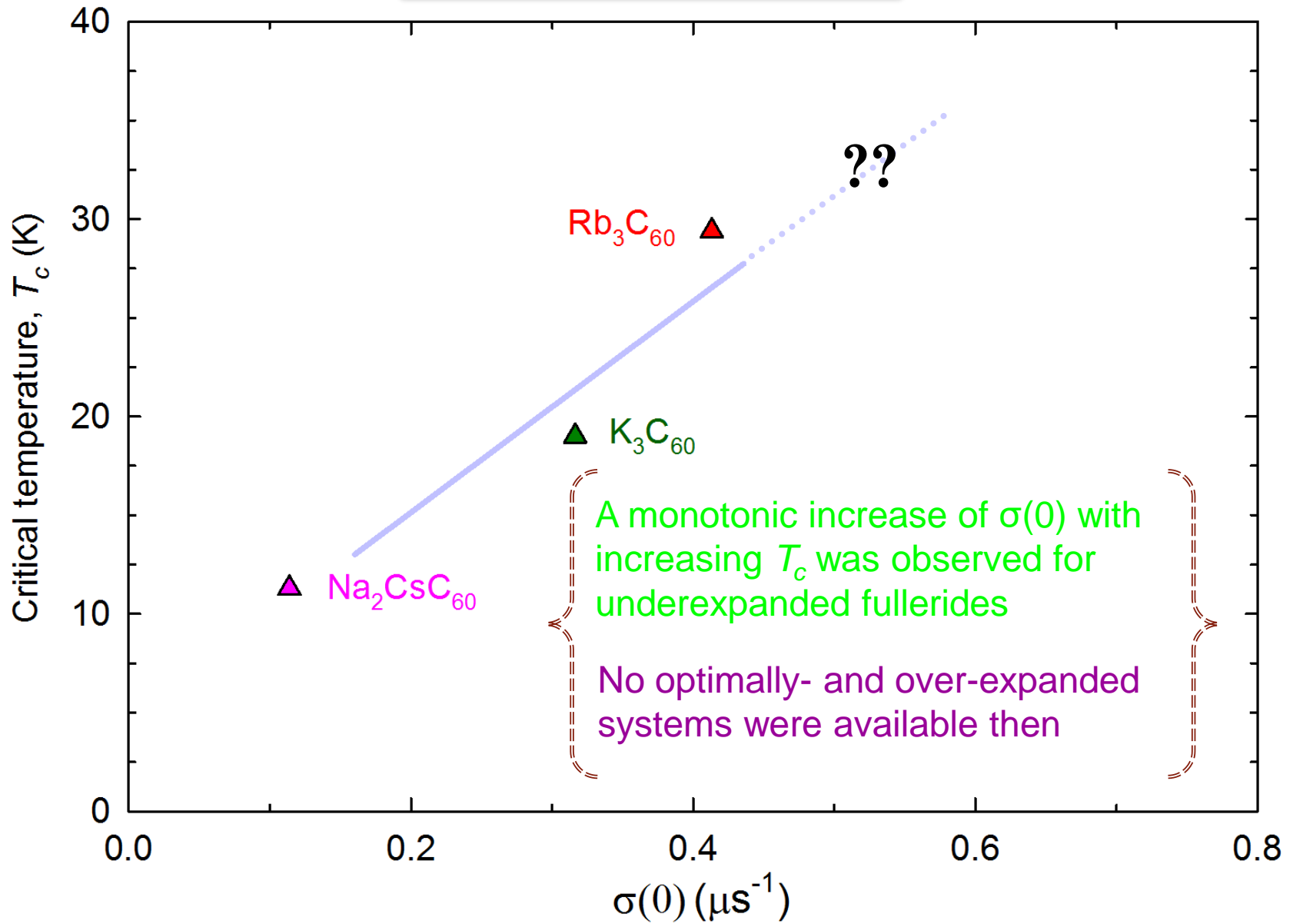
$$\sigma(0) \propto \frac{1}{\lambda^2} \propto \frac{n_s}{m^*} \left( \frac{1}{1 + \xi/\ell} \right)$$

$\sigma$  : relaxation rate  
 $n_s$  : superfluid density  
 $m^*$  : effective mass  
 $\xi$  : coherence length  
 $\ell$  : mean free path

- The temperature dependence of the relaxation rate,  $\sigma$  contains information on the pairing mechanism (s- or d-wave, weak/strong coupling, clean/dirty limit)



Back in 1994



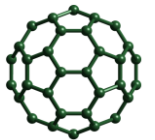
## GOAL

Investigate the superconducting properties  
of  $\text{Rb}_x\text{Cs}_{3-x}\text{C}_{60}$  compositions  
across the superconductivity dome:

underexpanded ( $x = 2$ )

optimally expanded ( $x = 1$ )

overexpanded ( $x = 0.5$ )

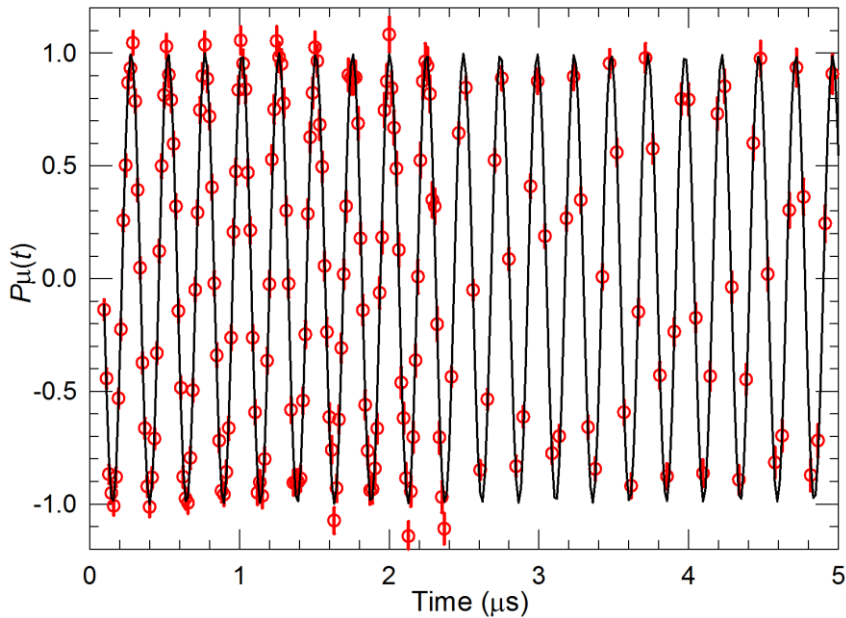


# TF- $\mu^+$ SR spectra

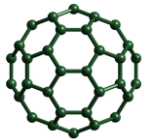
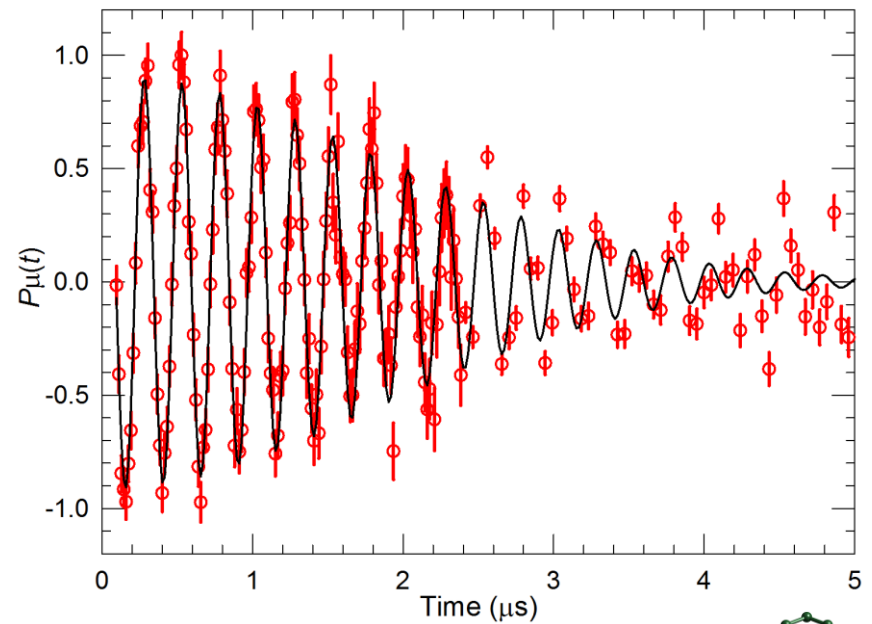


$$(H_{c1} = 10 \text{ mT} < H_{\text{ext}} = 30 \text{ mT} < H_{c2} = 68 \text{ T})$$

$$T = 50 \text{ K} > T_c = 31.8 \text{ K}$$
$$\sigma = 0.03 \mu\text{s}^{-1}$$



$$T = 3 \text{ K} < T_c = 31.8 \text{ K}$$
$$\sigma = 0.54 \mu\text{s}^{-1}$$



## Temperature dependence of the muon spin relaxation rate

Fitting is under way

Clean Limit

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = 1 + 2 \int_{\Delta(T)}^{\infty} \left( \frac{\partial f}{\partial E} \right) \frac{E}{\sqrt{E^2 - \Delta(T)^2}} dE$$

Dirty Limit

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \frac{\Delta(T)}{\Delta(0)} \tanh \left[ \frac{\Delta(T)}{2k_B T} \right]$$

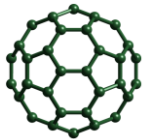
Fermi function:  $f = [1 + \exp(E/k_B T)]^{-1}$

$\lambda^{-2}(0)$ : zero-temperature value of the magnetic penetration depth

s-wave symmetry

Temperature dependence of the gap:

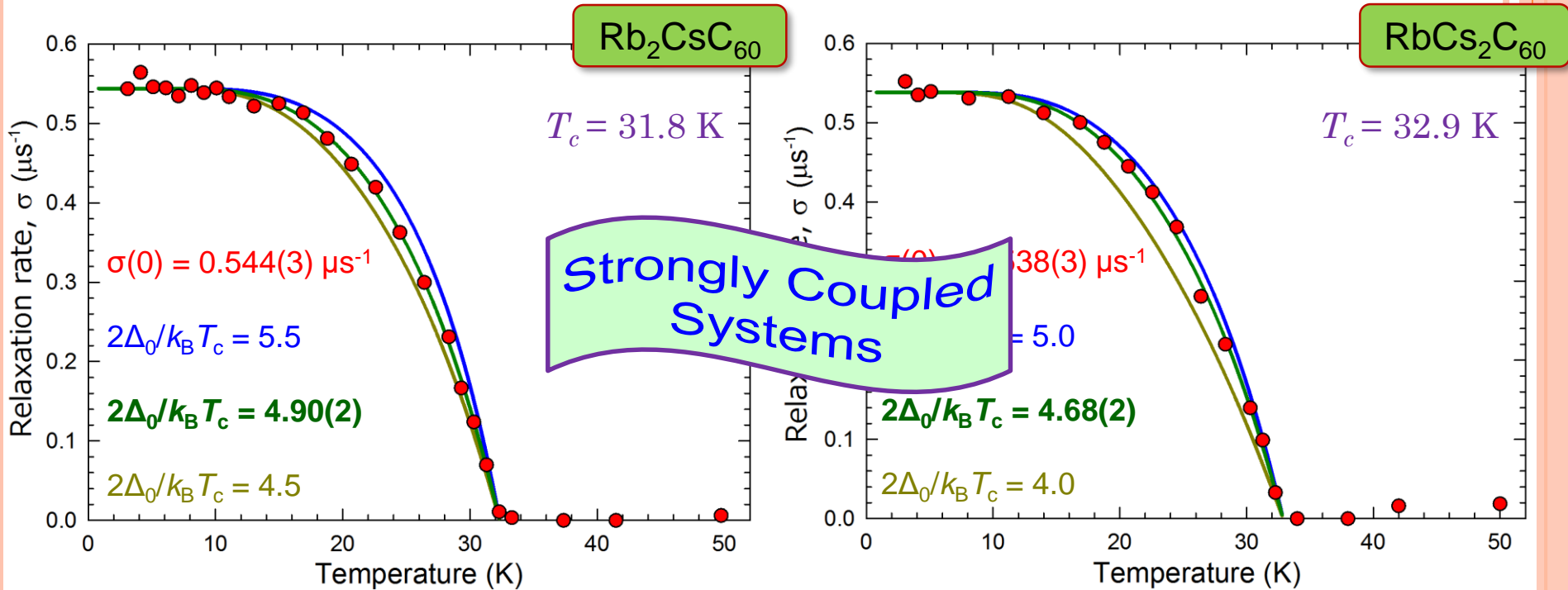
$$\Delta(T) = \Delta(0) \tanh\{1.82 [1.018(T_c/T - 1)]^{0.51}\}$$



# Temperature dependence of the muon spin relaxation rate

## s-wave isotropic superconductors

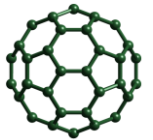
Clean Limit



Dirty Limit

$$2\Delta_0/k_B T_c = 5.52(2)$$

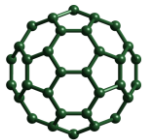
$$2\Delta_0/k_B T_c = 5.31(2)$$



Rb<sub>0.5</sub>Cs<sub>2.5</sub>C<sub>60</sub>: different behavior, data analysis in progress

If we assume  
“clean limit”

Compound	$\sigma(0)$ ( $\mu\text{s}^{-1}$ )	$\lambda(0)$ ( $\text{\AA}$ )	$n_s/(m^*/m_e)$ ( $\text{cm}^{-3}$ )	$T_F$ (K)
* $\text{Na}_2\text{CsC}_{60}$	0.11	8000	$0.4 \times 10^{20}$	315
* $\text{K}_3\text{C}_{60}$	0.32	4800	$1.2 \times 10^{20}$	470
* $\text{Rb}_3\text{C}_{60}$	0.42	4200	$1.6 \times 10^{20}$	650
$\text{Rb}_2\text{CsC}_{60}$	0.55	3660	$2.1 \times 10^{20}$	790
$\text{RbCs}_2\text{C}_{60}$	0.54	3680	$2.1 \times 10^{20}$	830
$\text{Rb}_{0.5}\text{Cs}_{2.5}\text{C}_{60}$	0.63	3400	$2.5 \times 10^{20}$	890

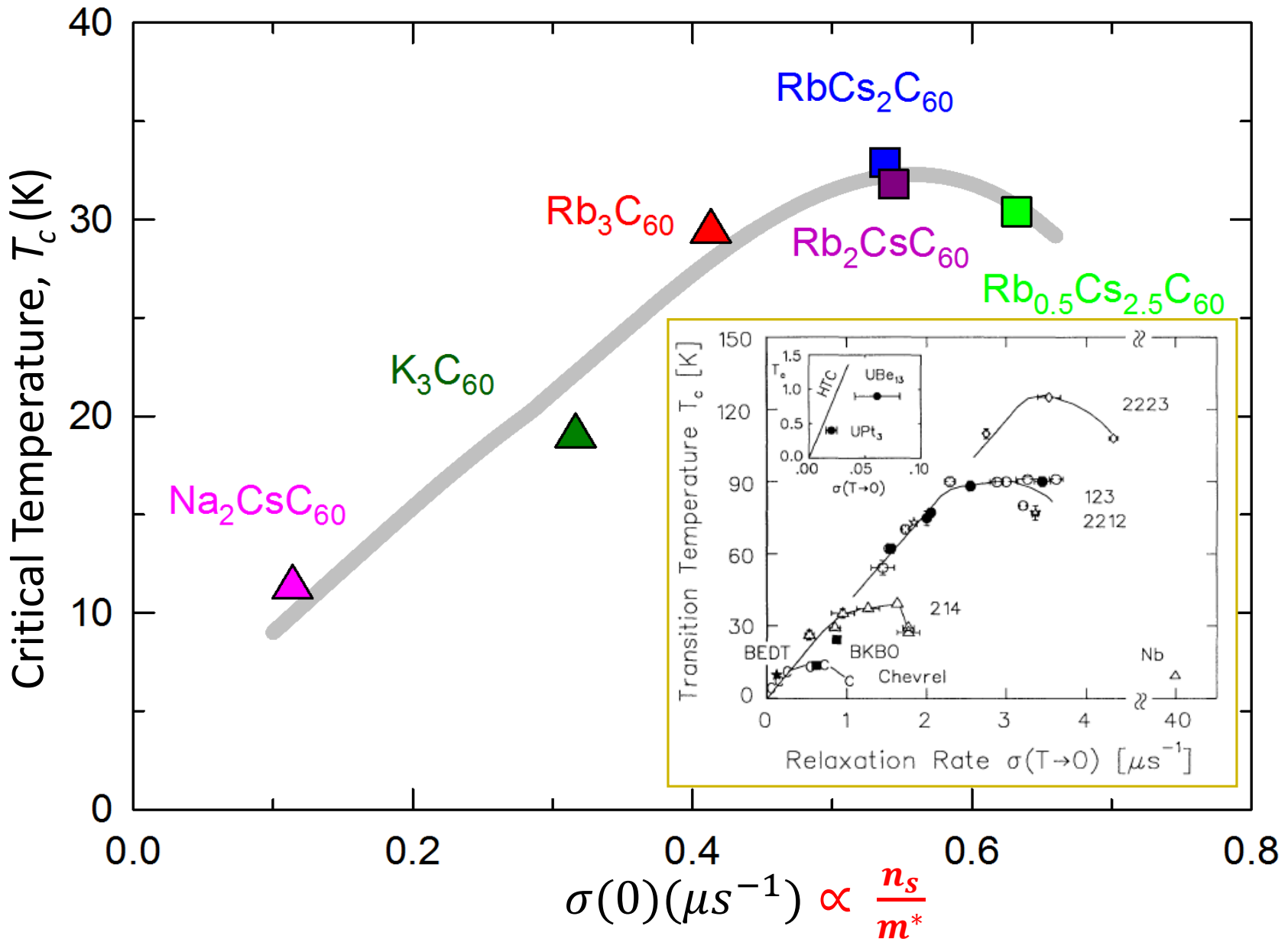


\* Y. J. Uemura *et al.*, Physica C **235-240**, 2501 (1994)

Y. J. Uemura *et al.*, Nature **352**, 605 (1991)





# T<sub>c</sub> vs σ(0) for fcc fullerides

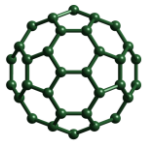
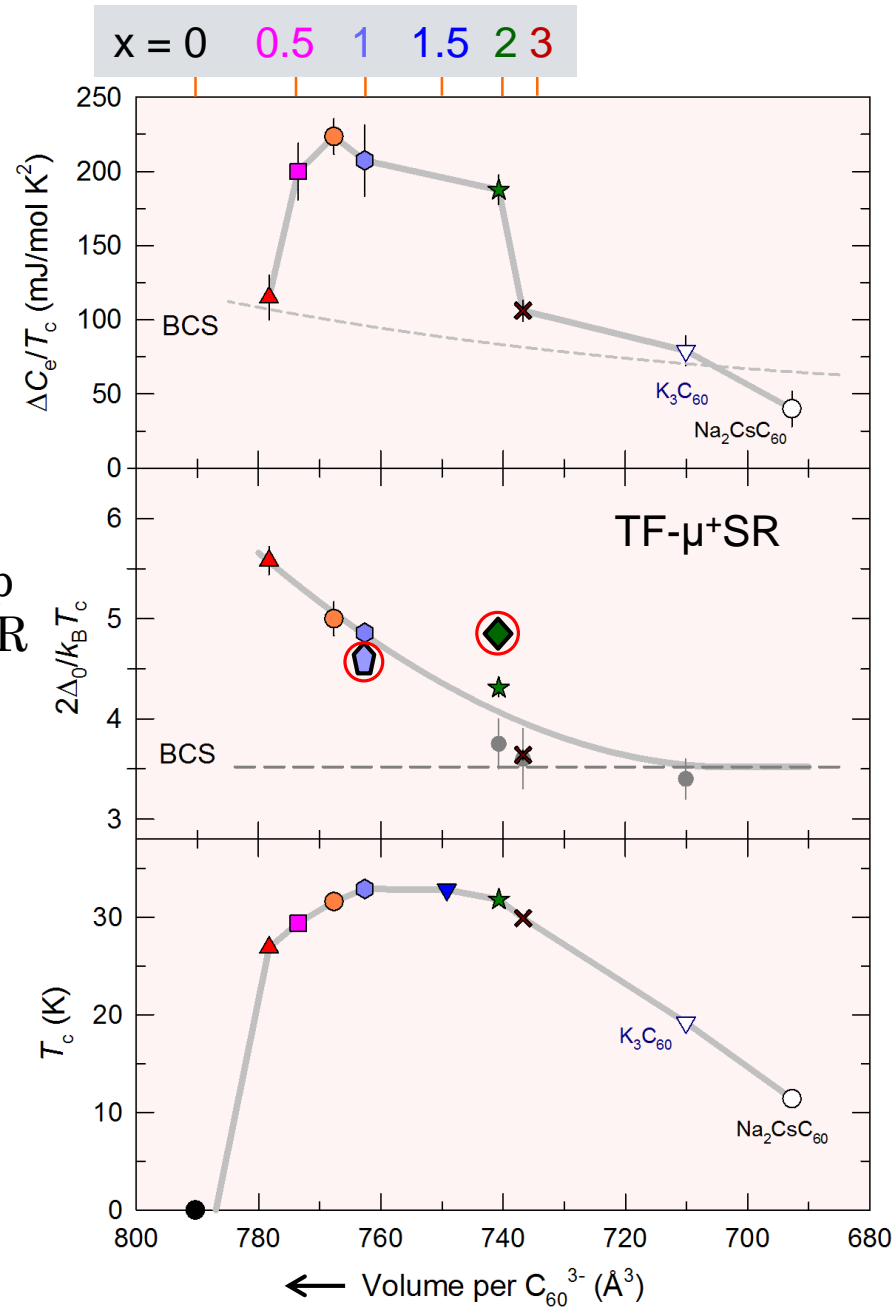


# Superconducting properties in addition to TF- $\mu^+$ SR

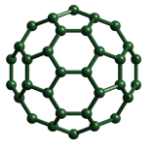
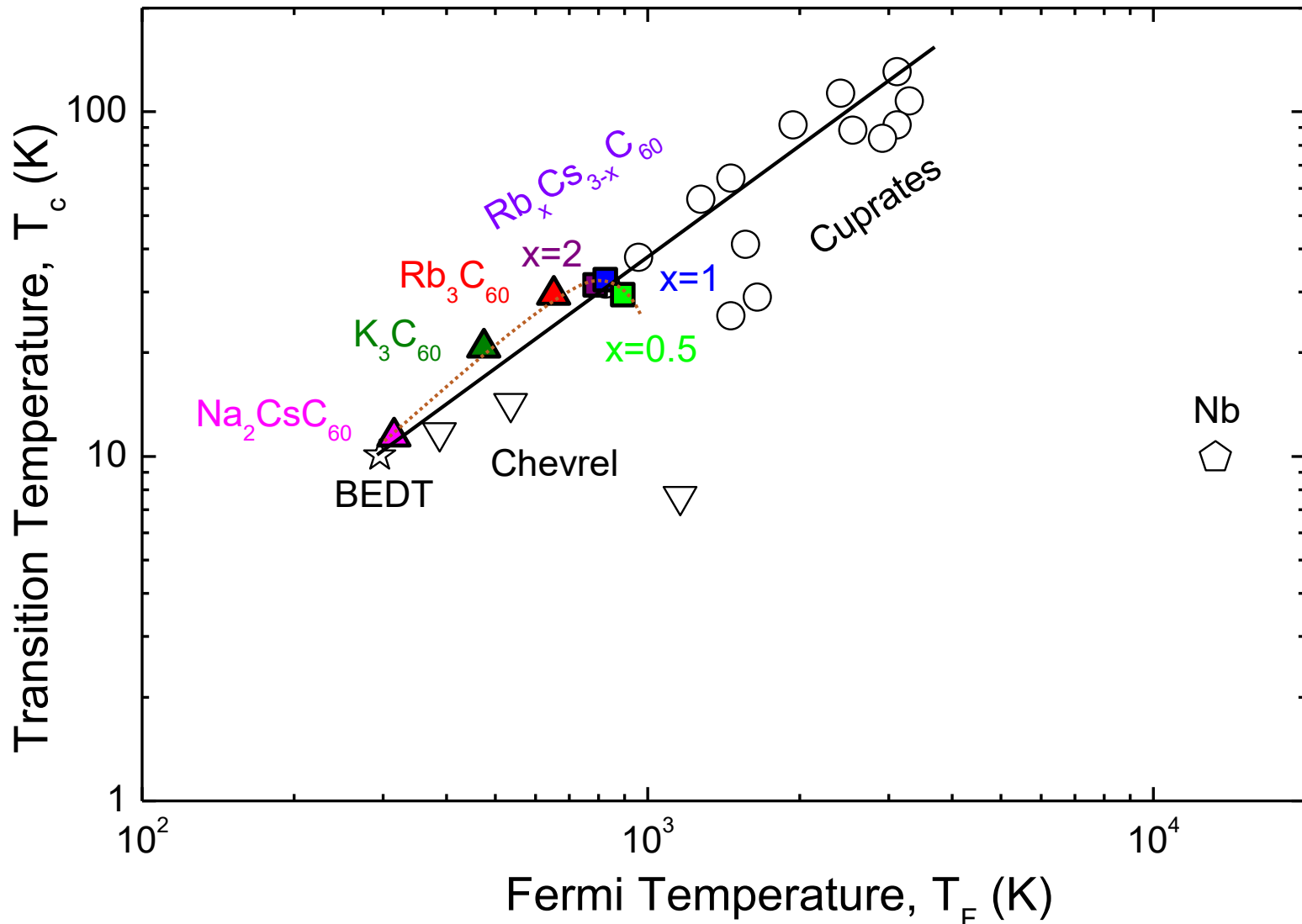
 Specific heat jump at  $T_c$

 Superconducting gap from NMR + TF- $\mu^+$ SR

 Superconductivity dome

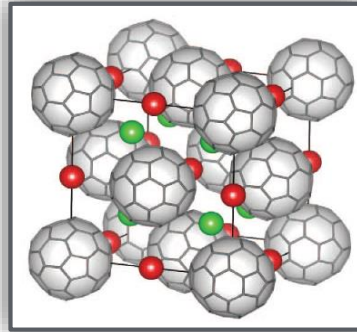


$T_c$  vs effective Fermi temperature,  $T_F$   
(Uemura plot)



## Summary

*fcc*  $\text{Rb}_x\text{Cs}_{3-x}\text{C}_{60}$  superconductors



TF- $\mu^+$ SR

$x = 0.5$

$x = 1$

$x = 2$

- ⊗ s-wave superconductors
- ⊗ strongly coupled systems
- ⊗ large enhancement of the gap in  $\text{Rb}_2\text{CsC}_{60}$
- ⊗ similar trend as high- $T_c$  cuprates and other exotic superconductors

## Acknowledgements

### **Prassides group (AIMR, Tohoku University)**

Y. Takabayashi

T. Nakagawa

### **Prassides group (Durham University)**

R. H. Zadik

### **P. J. Baker (ISIS, RAL)**

