#### Yasutomo J. Uemura, list of significant research contributions:

(reference numbers correspond to those in the list of 10 most important papers)

## I. High-Tc cuprates and other unconventional superconductors (SCs):

(a) Scaling of Tc on the superfluid density and the effective Fermi energy: "Uemura Plot" By using MuSR, Uemura and co-workers discovered a nearly linear scaling of the superfluid density  $n_s/m^* \propto 1/\lambda^2$  with the transition temperature  $T_c$  of underdoped cuprates [1] in 1989. They also showed a similar scaling followed by  $A_3C_{60}$  (Uemura et al, Nature 1991), overdoped cuprates [4] in 1993, organic BEDT SCs (Le et al., PRL 1992), FeAs SCs (Aczel et al., PRB 2008; Carlo et al., PRL 2009), Fe(Se,S) (Matsuura et al, PNAS 2022), and in NbSe<sub>2</sub> and other transition metal di-chalcogenides (TMDC) (von Rohr et al., Science Adv. 2019). In 1991 [2], Uemura and co-workers initiated an energy-scale considerations by deriving an effective Fermi energy  $E_F$  from  $n_s/m^*$  and plotting vs  $T_c$ . Both the  $n_s/m^*$  vs  $T_c$  and the  $E_F$  vs  $T_c$  plots demonstrated that these unconventional SCs exhibit behaviors quite different from those expected in simple BCS systems, in terms of the scaling and the magnitudes of  $T_c/E_F$ , while suggesting possible relevance to the concept of Bose Einstein Condensation (BEC). These plots have been commonly referred to as "Uemura plot" among researchers of superconductivity, and used for discussions of condensation mechanisms in unconventional SCs.

In the energy-scale considerations using Uemura plot, in 2019 Uemura [10] also included the transient superfluid density and the transient  $T_c$  of several unconventional SC's observed in optical conductivity measurements after ultrafast laser excitations by Andrea Cavalleri and coworkers. The results are consistent with a picture that the laser excitations suppress competing antiferromagnetic order and promotes transient superconductivity with the  $T_c$  values close to those expected for BEC of preformed superconducting pairs. Uemura plot was also employed by researchers of superconductivity of magic angle twisted bilayer graphene and other 2-dimensional materials, including Pablo Jarillo Herrero (Nature 2018), in comparing 2-d SCs with bulk unconventional SCs. Thus, Uemura and his plot have made significant impacts both in measurements of the superfluid density and subsequent discussions of a wide range of unconventional SCs.

#### (b) Magnetic phase diagrams of various unconventional SCs

In tuning with carrier doping and/or pressure, many unconventional SCs are found to be adjacent to magnetically ordered phase, and MuSR has made significant contributions in discovery of magnetic order and characterization of magnetic phase diagrams. Using TRIUMF and BNL muon facilities, Uemura and co-workers made pioneering contributions in this study: (i) by discovering magnetic order of La<sub>2</sub>CuO<sub>4</sub> (Uemura et al., PRL 1987), electron-doped cuprates (Luke et al., Nature 1989), CeCu<sub>2</sub>Si<sub>2</sub> (Uemura et al., PRB 1989), and in TMDC MoTe<sub>2</sub> (at PSI) (Guguchia et al., Science Adv. 2018); (ii) by elucidating phase diagrams of YBCO (Brewer et al., PRL 1988), and (Sr,Ca)<sub>2</sub>RuO<sub>4</sub> (Carlo et al., Nature Materials 2012); (iii) by observing coexistence of magnetically ordered and superconducting phases in CeCu<sub>2</sub>Si<sub>2</sub> (Luke et al., PRL 1994), oxygen overdoped and Eu doped LSCO cuprates (Savici et al., PRB 2002; Kojima et al, Physica B 2003), and FeAs systems (Cheung et al., PRB 2018, Tam et al., PRB 2017); and (iv) by detecting incommensurate / stripe spin modulations in LBCO with the 1/8 doping (Luke et al., Physica B 1991) and TMTSF<sub>2</sub>-PF<sub>6</sub> (Le et al., PRB 1993). Together with independent contributions from other MuSR researchers using PSI, ISIS, KEK and JPARC, these MuSR studies made very significant impacts on elucidating magnetic phase diagrams of "parent systems" of unconventional SCs.

## (c) Time reversal symmetry breaking (TRSB) in exotic SCs

By using its extremely high sensitivity to small static magnetic fields, MuSR has made unique contributions in detecting static magnetic field originating from superconductivity due to unconventional symmetry of superconducting order parameter and/or a loop orbital current. Such phenomena are referred to as time reversal symmetry breaking (TRSB) of exotic SCs. Detection of TRSB was pioneered by

Heffner et al. in (U,Th)Be<sub>13</sub> (PRL 1990) and by the Uemura group in UPt<sub>3</sub> (Luke et al. PRL 1993) and in Sr<sub>2</sub>RuO<sub>4</sub> [4] in 1998. More recent MuSR observations of TRSB include a study by Uemura and coworkers on Fe(Se,S) (Matsuura et al., PNAS 2023) and independent works at PSI in uniaxial pressure on Sr<sub>2</sub>RuO<sub>4</sub> by Grinnenko et al. (Nature Physics 2021), in ambient / hydrostatic pressure on Kagome SC's AV<sub>3</sub>Sb<sub>5</sub> by Mielke III et al., (Nature 2022), and in several other SCs. In all these cases, the magnitude of the observed TRSB field at the muon site is typically less than 1 Gauss. The TRSB phenomena have also been detected by optical Carr rotation, which subsequently confirmed the MuSR results in UPt<sub>3</sub> and Sr<sub>2</sub>RuO<sub>4</sub>. In the underdoped region of YBCO and other cuprates, the TRSB phenomena have been reported by neutron scattering, but conflicting negative results have been obtained by MuSR and NMR. Although major tasks remain to explain the extremely small magnitudes and the origin of the TRSB fields and to resolve the controversy with neutrons in cuprates, MuSR has initiated the research field of TRSB in unconventional SCs, to which Uemura and co-workers have made major impactful contributions.

II. Spin Fluctuations in Spin Glasses and Spin Liquids: spin freezing vs persistent fluctuations In his PhD thesis (1982) and subsequent paper [5] in 1985, Uemura reported MuSR studies of dilute alloy spin glasses AuFe and CuMn, which demonstrated that MuSR can detect critical slowing down and freezing of spin fluctuations with a unique time window interpolating those of neutron spin echo and ac susceptibility. In 1994, Uemura and co-worders initiated MuSR studies in Kagome lattice SrCrGaO (SCGO) [6] and other Kagome and pyrochlore spin systems with geometrical frustrations, which suggested persistence of dynamic spin fluctuations at T  $\rightarrow$ 0 without static spin freezing. The persistent dynamic fluctuations have been subsequently reported in many spin liquid candidate systems with geometrical frustrations by various MuSR researchers, while absence of both dynamic and static spin responses have been reported in some more perfect Kagome systems including Herbertsmithite by Philip Mendels and co-workers. Above-mentioned pioneering studies by Uemura initiated the activity of highly informative MuSR studies on classical and quantum spin liquids.

# III. Phase separation and first-order magnetic transitions in itinerant ferromagnets and Mott systems

In addition to the size of ordered magnetic moments, MuSR has a unique capability of detecting the volume fraction of magnetically ordered regions. Using this advantage, Uemura and co-workers have demonstrated strongly first-order magnetic phase transitions associated with phase separation in itinerant magnets MnSi tuned with hydrostatic pressure and  $(Sr,Ca)RuO_3$  with chemical pressure [7] and in Mott transition systems *RE*NiO<sub>3</sub> with Rare-Earth (*RE*) element substitutions and V<sub>2</sub>O<sub>3</sub> with pressure tuning [8]. They also showed (Goko et al, npj Quant. Mat. 2017) that second order transition is restored in more disordered (Mn,Fe)Si with pressure tuning, consistent with theories of Belitz and Kirkpatrick. The abovementioned studies highlighted importance of first order behaviors in quantum magnetic phase transitions.

## IV. Development and characterization of new diluted ferromagnetic semiconductors

(Ga,Mn)As is a representative diluted magnetic semiconductor (DMS) based on III-V compounds. The Uemura group performed MuSR studies on thin films of (Ga,Mn)As (Dunsiger et al., Nature Materials 2010), and demonstrated a sharp phase transition prevailing in the entire volume fraction. The III-V DMS systems, however, have strongly limited chemical solubility due to  $Mn^{2+}$  / Ga<sup>3+</sup> substitutions, which prevents formation of bulk specimens. Following a proposal by Jungwirth, Uemura and co-workers [9] synthesized Li(Zn,Mn)As based on the I-II-V semiconductor LiZnAs, and confirmed ferromagnetic order using MuSR in a bulk specimen obtained thanks to iso-valent  $Zn^{2+}/Mn^{2+}$  doping. They subsequently synthesized bulk specimens of (Ba,K)(Zn,Mn)<sub>2</sub>As<sub>2</sub> with ferromagnetic T<sub>c</sub> up to 230 K (Zhao et al., Nature Commun. 2013), which has the same crystal structure with the 122 FeAs SC. These efforts opened door to synthesis of more than 30 new DMS compounds. Common crystal structures may hopefully allow formation of heterostructure spintronics devices consisting of ferromagnetic DMS, FeAs SC, and antiferromagnetic BaMn<sub>2</sub>As<sub>2</sub> in junctions with epitaxial films and/or exfoliated single crystals.