A molecular Mott system with a quasi triangular lattice, β'-EtMe₃Sb[Pd(dmit)₂]₂, has been found to exhibit a quantum spin liquid (QSL) state [1, 2]. In the crystal with space group C2/c, Pd(dmit)₂ units are strongly dimerized with an eclipsed overlapping mode to form a dimer [Pd(dmit)₂]²⁻. A unit cell contains two crystallographically equivalent anion layers with different stacking directions of the dimer units. They are separated from each other by the insulating cation (EtMe₃Sb⁺) layer. Because the cations are located on the two-fold axis, the EtMe₃Sb cation without the two-fold symmetry shows two possible orientations with an occupancy of 50% for each one. Within each anion layer, the dimer units form a quasi triangular lattice where the spin frustration operates. The anisotropy of the triangular lattice (deviation from the regular-triangular lattice) and the band width can be tuned by the choice of the cation. In the salts with larger anisotropy, a transition toward antiferromagnetic long-range order (AFLO) occurs to remove the spin frustration. The salts with a nearly regular-triangular lattice and strong frustration exhibit various ground states instead of AFLO. For example, Me₄Sb, EtMe₃Sb, and Et₂Me₂Sb salts show AFLO, QSL, and charge order (2[dimer]⁻ → [dimer]⁰ + [dimer]²⁻; CO), respectively.

In the β'- Pd(dmit)₂ system, QSL is situated between AFLO and CO [1]. We have prepared mixed crystals (alloys) in an isostructural series (for example, (Me₄Sb),,(EtMe₃Sb)₁₋ₓ[Pd(dmit)₂]₂, where 0≤x≤1). This provides a powerful experimental tuning knob of the triangular lattice. Structural and physical properties of these mixed crystals will be demonstrated.

This work has been done in collaboration with T. Fukunaga (Tokyo Univ. of Science), H. Cui, S. Yamashita, Y. Ishii, H. M. Yamamoto and A. Tajima, (RIKEN).