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High pressure sequence of $Ba_3NiSb_2O_9$ structural phases: new S= 1 quantum spin-liquids based on Ni^{2+}[1]

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A quantum spin-liquid (QSL) is a ground-state where strong quantum- mechanical ?uctuations prevents a phase-transition towards conventional magnetic order and makes the spin ensemble to remain in a liquid-like state. Most QSL candidates studied to date are two-dimensional frustrated magnets with either a triangular or a kagome lattice composed of $S = 1/2$ spins. Here, we report the use of a high pressure, high temperature technique to transform the antiferromagnetically ordered ($T_N = 13.5$ K) 6H-A phase of $Ba_3NiSb_2O_9$ into two new QSL candidates with larger $S = 1$ (Ni^{2+}) moments: the 6H-B phase of $Ba_3NiSb_2O_9$ which crystallizes in a triangular lattice and the 3C-phase of $Ba_3NiSb_2O_9$ which forms a three-dimensional edge-shared tetrahedral lattice. Both compounds show no evidence for magnetic order down to $T = 0.35$ K despite Curie-Weiss temperatures θ_{CW} of -75.5 K (6H-B) and -182.5 K (3C), respectively. Below $\sim$25 K the magnetic susceptibility of the 6H-B phase is found to saturate at a constant value $\chi = 0.013$ emu/mol which is followed below 7 K, by a linear in temperature dependence for the magnetic contribution to the specificheat (C_M) which displays a giant coefficient $\gamma = 168$ mJ/mol-K^2 comparable to values observed in heavy-fermion metallic systems. Taken together, both observations indicate the development of a Fermi-liquid like ground-state characterized by a Wilson ratio of 5.6 in this otherwise insulating material It also points to the formation at finite temperatures of a well defined Fermi surface of $S = 1$ spin-excitations which behave as charged quasiparticles. For the 3C phase one observes $C_M \propto T^2$ indicating a unique $S = 1$ three-dimensional QSL ground-state as previously reported for $Na_3Ir_4O_8$ although this later compound is composed of Ir^{4+}ions having $S = 1/2$.

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