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Exotic Ground States: A Study in the Structural Effects of Frustration and Dimensionality

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Structural disorder and dimensionality play central roles in the characterization of structure and properties of crystalline materials. Structural disorder can be a desired property when searching for new materials with exotic properties and can be used as a tunable parameter by considering atomic sizes, coordination preferences, and electronegativity differences when substituting elements in a given structure. Dimensionality can also be viewed as an adjustable parameter when searching for new materials presenting unique challenges when characterizing new materials. Tuning of disorder and dimensionality often involves tuning at the edge of structural stability. Understanding the effects of dimensionality and structural disorder on magnetic properties could lead to better understanding of emergent phenomena. Here we present the growth and characterization of single crystals of 3-*d* structurally disordered intermetallic phases $Ln_2Ag_{1-x}Ga_{10-y}$ (*Ln* = La, Ce) and β-LnNiGa$_4$ (*Ln* = Tb-Ho), 2-*d* frustrated spin glasses MAl_2S_4 (*M* = Mn, Fe, Co, Ni), and 1-*d* quantum antiferromagnets $A_4Cu(MoO_4)_3$ (*A* = K, Rb) to highlight the effects of structural disorder and dimensionality on magnetic ground states.

I. Disordered Intermetallics. Single crystals of $Ln_2Ag_{1-x}Ga_{10-y}$ (*Ln* = La, Ce) and β-$LnNi_{1-x}Ga_4$ (*Ln* = Tb-Ho) were grown by the self-flux method and structurally characterized by single crystal X-ray diffraction. Magnetic and transport properties on crystal aggregates illustrate the spin glassiness in $Ce_2Ag_{1-x}Ga_{10-y}$ and the magnetic frustration in β-$LnNi_{1-x}Ga_4$ (*Ln* = Tb-Ho) attributable to strong site disorder in these phases. The disorder in $Ln_2Ag_{1-x}Ga_{10-y}$ (*Ln* = La, Ce) and β-$LnNi_{1-x}Ga_4$ (*Ln* = Tb-Ho) stems from the tendency of Ga and Ni, respectively, to achieve the optimum coordination and is consistent with the disorder found in similar distorted gallium networks.

II. Frustrated Spin Glasses. Single crystals of MAl_2S_4 (*M* = Mn, Fe, Co, Ni) were grown by the chemical vapor transport method and structurally characterized by single crystal X-ray diffraction at the Advanced Light Source Synchrotron facility in Berkeley California at the Small-Crystal Crystallography Beamline 11.3.1 (Lawrence Berkeley National Laboratory). Magnetic property measurements exhibit spin freezing at 1.8 K, 10.5 K, 5 K and 4 K for MAl_2S_4 (*M* = Mn, Fe, Co, Ni), respectively with large frustration parameters, $|\theta_W|/T^*$, which can be attributed primarily to strong site disorder and 2-dimensionality with geometrical frustration also playing a role.

III. One-dimensional Magnets. Single crystals of $K_4Cu(MoO_4)_3$ were grown by the flux growth method and structurally characterized by single crystal X-ray diffraction. Magnetic measurements illustrate spin liquid behavior down to 2 K with no indication of long-range or short-range magnetic ordering. Structural comparison of $A_4Cu(MoO_4)_3$ (*A* = K, Rb) allows for the isolation of the dimensionality and structural distortion parameters in these quasi-1-*d* quantum antiferromagnets.

Keywords: disorder, frustrated, dimensional