Longitudinal Magnetic Excitation in KCuCl₃ Studied by Raman Scattering under Hydrostatic Pressures

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The pressure-induced quantum phase transition in a three-dimensional spin-dimer system KCuCl₃ has been studied by using Raman scattering at low temperatures under hydrostatic pressures P with He gas as a pressure medium. Around 3 K, we observe a one-magnon Raman peak above a quantum critical point at $P_c = 0.82$ GPa. The origin of the one-magnon Raman peak above P_c is related to the change of the magnetic ground state. Below P_c , the ground state is a spin-singlet state together with a spin gap. Above P_c , the ground state is a mixed singlet-triplet one, resulting in emergence of two massless transverse modes and a massive longitudinal one in magnetic excitations. Because the longitudinal magnon mode is Raman active, the observed one-magnon Raman peak is assigned to it. The peak energy, the Raman intensity, and the halfwidth increase as a function of $\sqrt{P-P_c}$ above P_c . These P dependences are discussed based on the bond-operator mean-field theory taking decay channels from one longitudinal mode to two transverse ones into consideration.