**Method (theoretical part)**

**DMRG and Monte-Carlo simulations**

We employed the DMRG calculation (ref 6,7) with spin symmetry to investigate the ground state magnetic properties for the *J1-J2-J3-D* model on cylinders that have open boundary conditions (OBC) along the horizontal direction and periodic boundary conditions (PBC) along the vertical direction. We label the geometry of the cylinders as XCM or YCM. The labels X or Y specify if there is a nearest-neighbor bond parallel to the horizontal (X) or vertical (Y) direction. And the M labels the circumference of the cylinder. We pushed the kept state up to 15000 to obtain reliable results with truncation error for XC6-cylinder.

We adopted the Wolff cluster algorithm (2,3) combined with the Metropolis algorithm (1) in our classical Monte Carlo calculation. One Metropolis sweep of the entire spin lattice and 100 Wolff cluster updates were done for each measurement. samples were collected for calculating thermodynamic quantities at a given temperature. The first half samples were discarded for thermalization. The spin stiffness used to determine the transition temperature is defined by the second derivative of the free energy with respect to the twist angle about z-axis in spin space. In honeycomb lattice it can also be given by

where is the linear size of the torus, is the spin component at site in the direction, is the temperature, are vectors along interaction bonds, and is the direction of the twist in the lattice. The renormalization group calculation has predicted a universal jump of the stiffness at transition temperature , with a value of with a finite-size correction (4,5)

The blue dashed line in Fig. 4c depicts the relation . The transition temperatures for finite-size systems are determined by the intersection point of the blue line and the stiffness curves. Finally, the finite-size extrapolation between v.s. leads to the BKT transition temperature , as shown in the inset to Fig. 4c.

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