Higher Curie temperature in Fe_3GaTe_2 than in Fe_3GeTe_2: Role of hybridization exchange

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Fe_3GaTe_2, an itinerant van der Waals ferromagnet, has recently been reported to exhibit a Curie temperature (T_C) of 380 K [1], exceeding room temperature and higher than that of Fe_3GeTe_2 (T_C=230 K [2]). Fe_3GaTe_2 is isostructural with Fe_3GeTe_2 and has one less valence electron; however, its nearest-neighbor exchange coefficients (J_1 and J_2) are smaller, even though its T_C is higher. Here, we show that higher-order magnetic exchange interactions play a crucial role in the enhanced T_C of Fe_3GaTe_2, based on first-principles calculations of exchange coefficients. Due to a one-electron difference, higher-order exchange coefficients (J_3 and beyond) are positive in Fe_3GaTe_2 but negative in Fe_3GeTe_2, which can be understood in terms of hybridization exchange. In this, crystal-field-split ligand p orbitals play a key role for long-range ferromagnetism in Fe_3GaTe_2 and antiferromagnetism in Fe_3GeTe_2. Unlike super-exchange, hybridization exchange is nonperturbative and intrinsic to covalent bonding, providing a design principle for the search for materials with higher T_C. As a result, Fe_3GaTe_2 exhibits a larger total sum of all possible exchange coefficients. Using these J values, T_C of Fe_3GaTe_2 and Fe_3GeTe_2 is evaluated using both mean-field theory [3] and Monte Carlo simulations [4].

References:

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