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Electron-phonon and magnetoelastic interactions in ferromagnetic $Co[N(CN)_2]_2^1$ TATIANA BRINZARI, University of Tennessee, JASON HARALDSEN, Los Alamos National Laboratory, PENG CHEN, QI SUN, University of Tennessee, YOUNGHEE KIM, LI-CHUN TUNG², National High Magnetic Field Laboratory, ALEXANDER LITVINCHUK, University of Houston, JOHN SCHLUETER, Argonne National Laboratory, DMITRY SMIRNOV, National High Magnetic Field Laboratory, JAMIE MANSON, Eastern Washington University, JOHN SINGLETON, Los Alamos National Laboratory, JANICE MUSFELDT, University of Tennessee — Many of the most attractive properties of multifunctional materials can be traced to the competition between charge, structure, and magnetism. The discovery that these interactions can be tuned with various physical stimuli has accelerated investigation of their behavior under extreme conditions. In this work, we combined Raman and infrared vibrational spectroscopies with complementary lattice dynamics calculations and magnetization measurements to highlight the signatures of two different coupling processes in the molecule-based magnet $Co[N(CN)_2]_2$. In addition to a large anisotropy, our work reveals electron-phonon coupling as a field-driven avoided crossings of the low-lying Co^{2+} electronic excitation with the ligand phonons and a magnetoelastic effect that signals a flexible local CoN₆ environment. These findings broaden our understanding of charge-lattice-spin interactions under extreme conditions and demonstrate rich new aspects of multifunctionality in tunable molecular materials.

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