Electron-phonon and magnetoelastic interactions in ferromagnetic $Co[N(CN)_2]_2$

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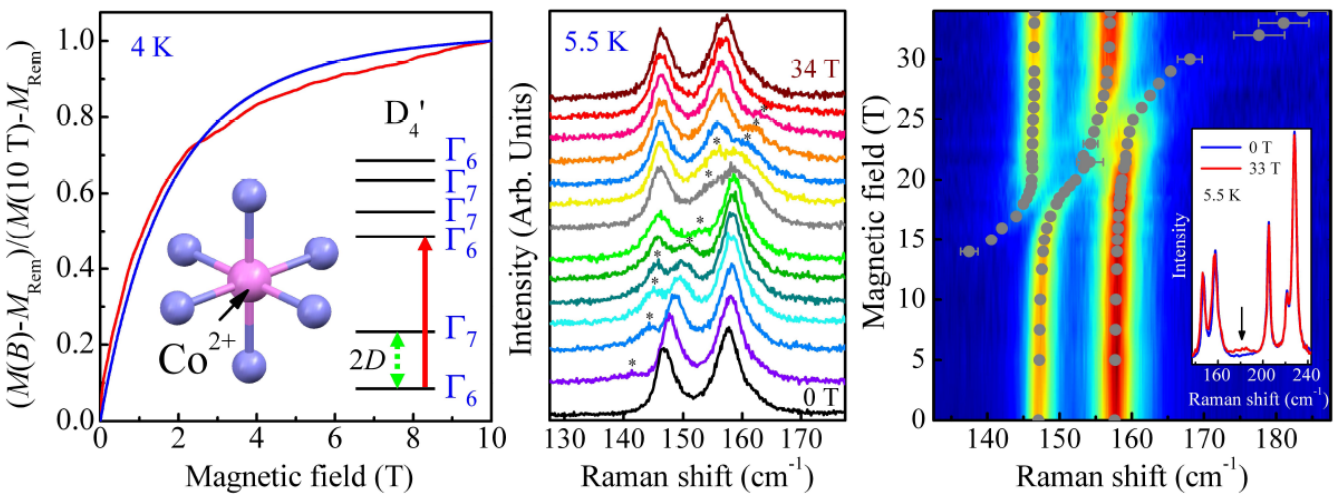
Introduction

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. These interactions take place over multiple energy scales and impact a variety of physical properties. For instance, the competition between charge, lattice, and spin degrees of freedom is influential in determining static properties like *dc* conductivity and crystal structure. These interactions affect the dynamical properties as well; electron-phonon and magnetoelastic coupling are flagship examples. Here, we combined vibrational spectroscopy with complementary lattice dynamics calculations and magnetization measurements to highlight the signatures of these coupling processes in $Co[N(CN)_2]$, findings that adds to the rich character of $M[N(CN)_2]$ molecule-based magnets.



Magnetization, magneto-Raman response, and B – frequency diagram of $Co[N(CN_2)]$ at low temperature. We extract an anisotropy of ≈12 cm^{-1} and discovey a field-driven avoided crossing of an on-site Co^{2+} electronic excitation with two wagging phonons from which we extract electron-phonon coupling constants of 2 - 3 cm^{-1}.

Results and Discussion

In this work, we measured the vibrational response of $Co[N(CN)_2]$ under two different external stimuli, magnetic field and temperature, and compare our findings to complementary dynamics calculations and magnetization measurements. Our work reveals a large anisotropy and rich charge-lattice-spin coupling including (i) an avoided crossing between a field-dependent Co^{2+} crystal field excitation and low frequency wagging phonons that we discuss in terms of electron-phonon interactions and (ii) magnetoelastic effects in which the local Co^{2+} environment distorts and the C≡N environment relaxes in response to applied field. These findings broaden our understanding of charge-lattice-spin interactions under extreme conditions and demonstrate rich new aspects of multifunctionality in tunable molecular materials. Similar processes appear in oxides where again the exotic properties also derive from charge-spin-lattice interaction rather than a separation of the different degrees of freedom.

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