Magnetic field-temperature phase diagram of the high-temperature magnetoelectric multiferroic CuO

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ABSTRACT: Encouraged by recent experiments on multiferroic systems using high-resolution ultrasonic measurements [1], we measured the temperature and field dependence of the velocity of acoustic modes in order to determine the magnetic phase diagram of the monoclinic multiferroic cupric oxide (CuO). A new transition at TN3 = 230 K, corresponding to an intermediate state between the antiferromagnetic incommensurate non-collinear spiral phase (AF2) observed below TN2 = 229.3 K and a paramagnetic phase (PM), is revealed. Anomalies associated with a rst order transition to a commensurate collinear phase (AF1) are also observed at TN1 = 213 K. Our dielectric constant measurements con rm that only the spiral phase supports a spontaneous electric polarization. In addition, we report on a spin-flop transition between 11 T - 13 T in the low temperature AF1 collinear phase with B // b.

Based on rigorous symmetry arguments, a non-local Landau-type free energy is developed for CuO and similar monoclinic multiferroics. In contrast with previous results from several studies, but in support of a recent proposal, our analysis clearly reveals the necessity for an incommensurate collinear phase (AF3) between the PM and the spiral AF2 states. Such a phase has been shown, both theoretically and experimentally, to occur in other geometrically frustrated antiferromagnets where symmetry allows for uniaxial anisotropy at second order [2,3] and in multiferroic compounds similar to CuO. Landau models have also been successful in explaining the magnetic phase diagrams of a number of multiferroic systems [4,5] based on a mean- field treatment of a Heisenberg-type Hamiltonian that includes explicit temperature and magnetic field (B) dependences. We compare the model predictions to the B-T phase diagram of CuO obtained using ultrasonic velocity data. The same sequence of magnetic phase transitions is observed as in other multiferroic systems with spiral spin-driven ferro-electric order such as MnWO4, AMSi2O6, some of the orthorhombic systems RMnO3, the Kagom_e-related compound Ni3V2O8 and LiCuVO4.

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[5] G. Quirion et al., Phys. Rev. B 84, 014408 (2011).

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