Adjusting chemical disorder stabilizes multiferroic magnetic spirals at higher temperatures

A research team at the Paul Scherrer Institute in Switzerland has demonstrated that the spiral magnetic order phase-transition temperature in $YBaCuFeO_5$, a magnetoelectric (ME) multiferroic, can be systematically tuned by changing the annealing rate. ME multiferroics possess coupled magnetic and electrical properties. "This is always interesting because if you act on one of them, then you can change the other one for free," says Marisa Medarde, group leader and corresponding author of their work, which was published in a recent issue of *Nature Communications* (doi: 10.1038/ncomms13758).

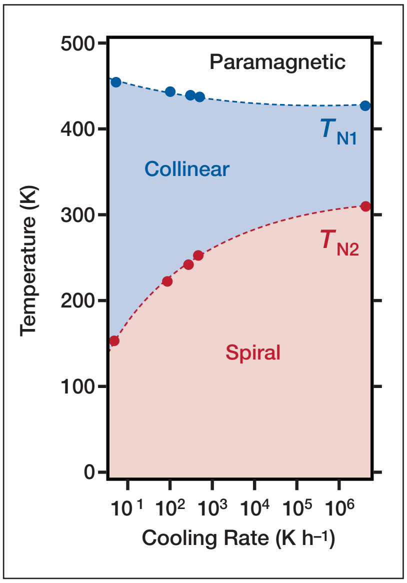
Multiferroic materials have garnered attention as potential next-generation data storage devices. It is fundamentally challenging, however, to find a suitable multiferroic that exhibits coupled behavior at room temperature. Strong multiferroic coupling relies on a specific ordering, or structure, of the magnetic spins in the material that is typically only stable at low temperatures. Magnetic spiral order, an example of one of these structures, leads to a twisting of the magnetic spins into a helix along a particular direction in a material.

The key to these exotic ordered phases lies in the frustration of magnetic interactions between the spins. A novel device could exploit phase transitions into the magnetic spiral phase because they are often accompanied by spontaneous electrical polarization of the material. These devices are still many years away from reality, as few materials, to date, have demonstrated a spiral magnetic order close to room temperature.

One such material is $YBaCuFeO_5$, a multiferroic perovskite, studied by Medarde and her colleagues. $YBaCuFeO_5$ is structurally related to the $YBa_2Cu_3O_{6+x}$ family of high-temperature superconductors. The multiferroic material was initially created by doping the superconductor with iron. According to Medarde, the "magnetism of iron kills superconductivity," so the result was not superconducting; however, researchers were soon attracted to the "interesting magnetism" observed in $YBaCuFeO_5$.

Her research team demonstrated that the spiral phase-transition temperature, T_{N2}, can be methodically shifted from 150 K to 310 K by changing the cooling rate over several orders of magnitude. Other fundamental properties, such as the periodicity and inclination of the rotation plane of the spiral, can be tuned by changing the annealing rate. The ability to tune these characteristics of the spiral phase is exciting to materials scientists, including John Mitchell, group leader of the Emerging Materials Group at Argonne National Laboratory, who sees this behavior as an indication "that there's some underlying hidden variable that's controlling this, and that turns out to be local disorder."

Previous neutron and x-ray powder diffraction experiments and density functional theory (DFT) calculations done by Medarde's group with collaborators at ETH Zürich, the Bragg Institute in Australia, and Institut Laue Langevin in France, have connected the observation of incommensurate magnetic ordering to the chemical disorder in the occupation of the copper and iron B-sites in the perovskite crystal lattice (see *Physical Review B*, doi:10.1103/PhysRevB.91.064408). In their most recent work, the researchers noticed



Stability range of the collinear and spiral phase of the $YBaCuFeO_5$ with cooling rate. Dashed lines are included as a guide to the eye. Courtesy of *Nature Communications.*

an annealing-dependent distortion in the bipyramidal bow-tie units that contain the copper and iron cations. The quenched samples had the highest average Cu/Fe occupation disorder and, surprisingly, exhibited the highest spiral phase-transition temperature.

Recent theoretical models have also confirmed the importance of chemical disorder in creating the competing magnetic interactions necessary in the spiral order phase of $YBaCuFeO_5$ (see arXiv:1610.00783v3). This could establish what Medarde calls "design rules," where researchers introduce chemical disorder in a precise way to synthesize multiferroic materials with desired properties. Being able to tune the properties of multiferroics will drive their utilization in future devices and applications.

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