# Comment on "Retention of the Tetragonal to Orthorhombic Structural Transition in F-Substituted SmFeAsO: A New Phase Diagram for SmFeAs(O$_{1-x}$F$_x$)"

In their Letter, A. Martinelli *et al.* [1] draw a "new phase diagram" for SmFeAsO$_{1-x}$F$_x$ (Sm1111) from an analysis of their synchrotron powder diffraction data. This phase diagram is shown as well in their paper [2]. The authors mainly claim that, unlike the case of all other reported phase diagrams of all *RE*1111*s* (*RE* = La [3–6], Ce [7], Pr [8], Sm [9]), the tetragonal-to-orthorhombic structural transition extends almost unperturbed across the entire phase diagram. We show in the following that their conclusion is premature.

In the REFeAsO systems, the structural transition should be signaled in the x-ray powder diffraction spectra, as mentioned by the authors themselves, by the splitting of the 110 tetragonal peak into two clear distinct 020 + 200 orthorhombic peaks on cooling. Unfortunately, despite high resolution x-ray measurements, this is not seen in their data. Instead of a double peak (clearly visible with both x ray [8] and neutrons [7]), they observe just a broadening (absolute value unknown) of the tetragonal peak, even for the underdoped $x = 0.05$ composition sample that should clearly show a split into two distinct peaks at the structural transition. For unknown reasons, the authors chose low **q** scans, although the higher **q** 220 scan that their setup allows is known to give a larger separation between peaks. The immediate conclusion is poor quality samples, even if only the lack of a split into two peaks for the underdoped $x = 0.05$ and 0.1 is considered. That means that in the optimally doped sample as well, along with the majority phase, other underdoped minorities are present. This then explains the slight broadening of the tetragonal peak in the optimally doped sample. A range of fluorine doping instead of a single well-defined doping value will produce a broad Gaussian tetragonal peak in the two other underdoped samples. An indication of unwanted phases in their samples is the strong SmOF impurity peak from their x-ray powder diffraction spectra.

Furthermore, without a legend showing a quantitative scale of the intensities for the right graph of Fig. 1 the authors' statement "drastic suppression of intensities" is meaningless. In their left graph of Fig. 1, while for the $x = 0.05$ and 0.1 the full width at height maximum of the tetragonal 110 peak is almost doubled by changing in a step-like evolution from about 0.4 to 1 (in the normalized units used by the authors) in the [80, 275] K temperature interval, for the composition $x = 0.2$ the full width at height maximum steady increases slightly from 0.88 to 1 (data shown for a shorter temperature range, [100, 210] K). This is already a hint that the broadening in the $x = 0.2$, with a different temperature dependence, is of a different nature. As an effect, it is clear that if this slight broadening of the 110 peak for the optimally doped $x = 0.2$ sample is not due to structural phase transition, their phase diagram will look no different from what has been determined already [3–9]. While the authors claim that their samples were well characterized, there is no data in their articles showing quantitative determination of the fluorine doping and its homogeneity [1,10], which is necessary with such a claim. It has been shown, for instance, that in the very same system Sm1111, a $x = 0.25$ nominal fluorine doping resulted in an actual doping of 0.11 as determined from wavelength-dispersive x-ray spectroscopic microprobe measurements [6]. A considerably smaller actual fluorine doping than the nominal doping has been reported in Pr1111 as well [8]. This is in part because of a saturation of the doping but also because of resulting fluorine-based byproducts (which means less fluorine in the primary phase). As these are known, it is rather striking that the study under discussion is done on powders, although sizable crystals have been already available to the scientific community for more than two years [11].

To conclude, we presented briefly a few causes that contribute to the broadening of the tetragonal peak of the optimally doped SmFeAsO$_{0.8}$F$_{0.2}$ powder sample presented by A. Martinelli *et al.* [1], causes that mislead the authors in their claim of a different phase diagram. The accurate determination of the phase diagrams is of crucial importance for the understanding of the superconductivity phenomenon itself and this work must be further carried out on *RE*1111 high quality crystals, in order to settle these matters unambiguously.

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