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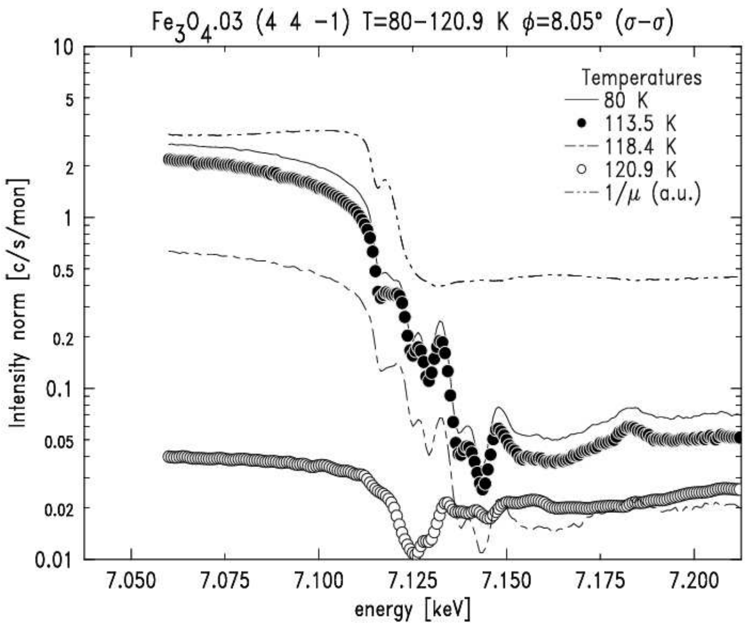
Report:

The determination of the charge ordering pattern below the Verwey transition in Fe_3O_4 constitutes one of the toughest problems in crystallography which, despite numerous efforts, still remains unsolved. We report here our first attempt to solve the problem by means of resonant X-ray diffraction , and a preliminary analysis of the data collected in this experiment. The complete data analysis is currently under way.

In this experiment we have used a Fe_3O_4 sample grown by us and where susceptibility experiments and our diffraction data have yielded a transition temperature T_V=120.5 K. The phase transition in the studied sample is very sharp and the intensity of the Bragg spots originating below T_V (see figure) drops by a good factor of 20 between T_V and T_V-1 K. The diffraction was performed in reflexion, off a {110} surface and a magnetic field of 0.3T was applied along the (001) direction (the hard-magnetization direction) in order to favor one ferromagnetic domain. The number of structural domains allowed as a consequence of the phase transition is thus divided by a factor of three. The effect of the magnetic field was optimal as Bragg peaks indexed as Q+$(0, \frac{1}{2}, 0)$ and Q+$(\frac{1}{2}, 0, 0)$ (Q being a Bragg peak of the high temperature cubic phase) had null intensity. A total of 20 different reflections originating at the phase transition were collected as a function of the incident photon energy around the Fe $K-$edge and at T=30K. These reflections, being orders of magnitude weaker than that of the main cubic structure, are often found polluted by multiple scattering.

Therefore these reflections were systematically measured at several azimuthal angles in order to remove any uncertainty in their energy spectra. All these measured reflections require a careful analysis, and adequate absorption corrections are mandatory. Some exhibit the characteristic features of the Templeton scattering, as it has been observed in previous experiments in peaks such as (002) and (006). In others, as shown below, the absorption edge contains a sharp structure, a distinct feature that is free from multiple scattering pollution. And in most of Bragg peaks the spectra are characterized by interplay of f' and f''. More intriguing as to the charge order problem itself is the temperature dependence shown in the figure below.



Intensity of the (4 4 -1) reflection in the neighborhood of the Fe $K-$edge as a function of temperature. No absorption correction has been applied to the spectra. The entire spectra scales as a function of temperature, and the spectra right above T_V shows a radically different anomalous lineshape. For a comparison, the absorption ($1/\mu$ in arbitrary units) has been included.

The energy region around the edge of the (4 4 -1) Bragg reflection, where we expect the strongest contribution from a *hypothetical* charge order, is manifestly different above and below T_V. We believe that this feature is a signature of the disappearance of a static charge order, which therefore should be present below T_V. Above T_V each Fe of the unit cell experiences *the same* average charge state and the fine structure around the edge vanishes. We interpret the anomalous scattering left remaining in the energy scan above T_V as originating from long-range lattice fluctuations which persists up to high temperatures. Other signatures of the charge order (not shown here) are evident in the energy scans performed at low temperatures, where most of the experiment have been carried out.

The data shown in the figure can be considered as preliminary evidences of the presence of an *actual* charge order below T_V that requires further analysis and obviously more experiments. These will help to stablish the charge order *pattern*, that is our final goal.