Structure of a Phase Ordering CuZn Alloy

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The order-disorder transition occurring at T_C=737 K in a 1:1 alloy of Cu and Zn (β-brass) is continuous and thus of second order [1]. It is a model realization of the theoretical Ising model for critical phenomena which was honored by the 1982 Noble Prize in Physics to Ken Wilson for his renormalization theory. The neutron diffraction data obtained in 1967 [2] is still the best experimental basis for the three dimensional Ising model, but the data could be dramatically improved using the high resolution provided by synchrotron X-rays if β-brass crystals with sufficiently small mosaic spread were available

The Ising model was originally conceived as a model of ferro-magnetism with atomic spins up or down arranged in a crystal lattice. The magnetic susceptibility χ diverges as the temperature approaches the Curie temperature like $\chi \propto [(T-T_c)/T_c]^{-\gamma}$. Conceptually, one could imagine that the magnetic field varies sinusoidally in space with a corresponding wave vector dependent susceptibility $\chi(q)$. The same model can be used for the order-disorder transition in β-brass with spin up or down corresponding to Cu or Zn occupation of a lattice site. The critical scattering can by the fluctuation-dissipation theorem be shown to be proportional to $\chi(q)$ which approximately is of Lorentzian shape, $\chi(q) \propto 1/[1+(\xi q)^2]$ with a diverging correlation length ξ as the temperature approaches T_C from above. Figure 1 summarizes the results obtained by neutron scattering, and the results potentially feasible with synchrotron X-ray radiation, assuming that the crystal mosaicity gives a negligible contribution to the *q*-resolution, or in other words is less than about one milli-radian.

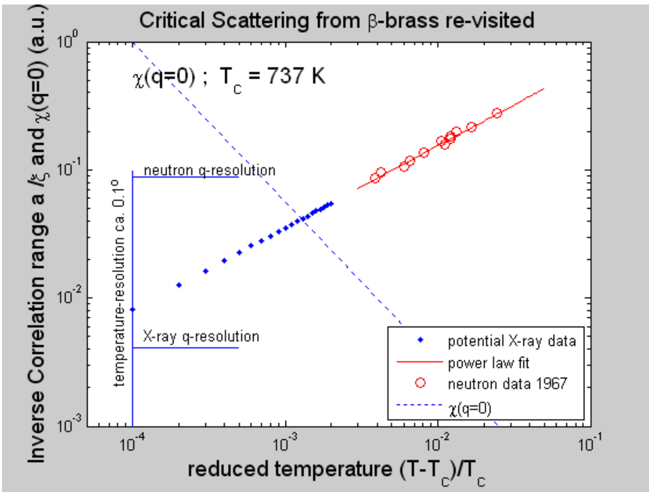

Figure 1: 1967 Neutron data of the correlation length in CuZn compared to what is potentially possible to obtain with high resolution X-ray scattering at a synchrotron source.

A thin β-brass crystal with <100> surface orientation initially showed a mosaic spread of around 1 degree. Polished and cut into 4 pieces of 3 x 3 mm^2, each about 0.2 mm thick, these 4 pieces were put into indentations with a bottom thickness of about 20 micrometer made in Si single crystal wafer and subsequently covered by bonding of a 0.3 mm thick Pyrex glass. This encapsulation is necessary to prevent Zn evaporation near T_C. Zn evaporation has been the major difficulty in previous studies of β-brass with X-rays but our container provide an efficient encapsulation which at the same time is transparent to X-rays through the 20 micron thick Si window. After encapsulation one of the crystals was examined at ESRF in Bragg reflection geometry at 9 keV. The mosaic spread was drastically reduced by almost 3 orders of magnitude, i.e. from about one degree to a few milli-degrees. One could observe the long range order disappearing at the canonical critical temperature T_C, but surprisingly the transition was not accompanied by the usual critical scattering in a range around T_C, the hallmark of a 2^{nd} order transition [1]. We assumed that both of these surprising findings could be due to surface effects, since the X-ray penetration depth was only a few micrometers in reflection geometry.

In September 2012 we had access to the P08 beamline at PETRAIII with an X-ray energy of 25 keV, and one could then illuminate the entire crystal Laue transmission geometry, again with the surprising result that the mosaic spread was reduced from about 1 degree to only a few milli-degrees, which in fact equals the Darwin width of a perfect crystal. This measurement was on the very same crystal that we had previously used at ESRF and which during that beamtime had been held at temperatures near T_C for several days. Similarly to the results from ESRF we could not detect any critical scattering in Laue geometry with a sample thickness of 200 micrometer and a similar size beam height, thus illuminating almost 2 million reflecting atomic planes. A second crystal which had not been exposed to any heat treatment (other than during the encapsulation process explained above) showed similar behaviour.

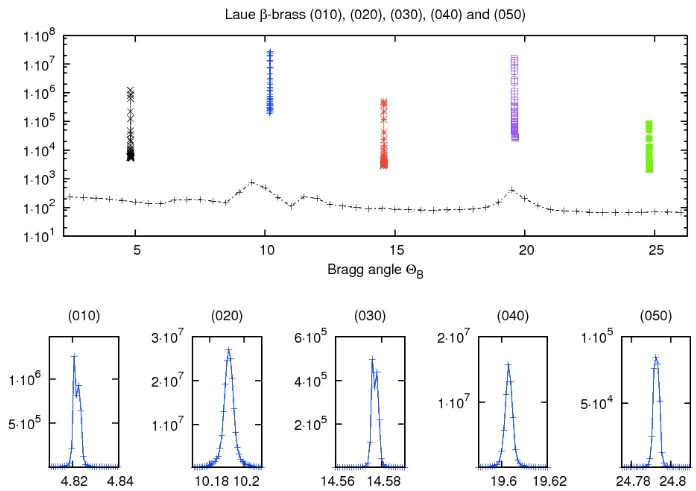

Figure 2. Data on CuZn in the ordered state (P08, September 2012). The X-ray energy is 25.06 keV, the sample the same as used at ESRF, but here in Laue geometry illuminating a thickness of about 200 micrometer and more than one million reflecting atomic planes as determined by the beam height. The intensity ratios between the fundamental reflections (020) and (040) and the superlattice reflections (010), (030) and (050) are consistent with dynamical diffraction theory from a perfect crystal. Surprisingly, no critical scattering is observed upon approaching Tc.

We need to understand how the mosaic crystals become perfect crystals, and why the critical scattering in the encapsulated perfect crystals is totally suppressed without any apparent change in Tc. To solve this mystery we need a suitable, highly stable X-ray transparent furnace and more encapsulated crystals. These will be the major ingredients of the investigations that will continue at P08 in an experiment scheduled for June 2013.

References

[1] B. E. Warren, X-ray Diffraction, Addison-Wesley (1969)
[2] J. Als-Nielsen and O. W. Dietrich, Phys. Rev. 153, 717 (1967); O. W. Dietrich and J. Als-Nielsen, Phys. Rev. 153, 711 (1967)