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Neutron diffuse-scattering intensities in niobium

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New neutron diffuse-scattering data on "pure" Nb and on Nb doped with N impurities are presented and compared to the results shown previously by Chang and Colella. The new data show that the effects previously observed in nominally pure Nb can be explained as arising from small concentrations of O or N impurities.

In a recent paper,[1] Chang and Colella (CC) compared x-ray and neutron diffuse scattering from nominally pure Nb, using neutron data obtained by Werner and Rowe.[2] As shown in CC, the earlier neutron data showed an intense diffuse-scattering peak centered near $\vec{Q}=(\frac{4}{3},\frac{4}{3},\frac{4}{3})2\pi/a$. Careful intensity calculations showed that this scattering should have been observable in the x-ray diffuse scattering. In addition, there were other unpublished neutron results indicating similar diffuse-neutron-scattering peaks, but the effect was not reproducible from sample to sample.

In an attempt to resolve the apparent discrepancy between the x-ray and neutron results, we have studied the diffuse neutron scattering from single crystals of "pure" Nb and niobium containing 0.15 atomic % nitrogen. The pure Nb sample was in the form of a cylinder with a [110] direction along the cylinder axis. The crystal was approximately 60 mm long by 12 mm in diameter and weighed 64 g. This sample was purified by outgassing in a vacuum of $<10^{-9}$ Torr for 30 hours at 2300 °C. On the basis of previous tests of this technique, we estimate that the total N and O concentration should be less than 5 ppm by weight. The crystal used for the doped sample was also a cylinder of approximate length 30 mm and diameter 12 mm and weighed 30 g. This crystal was first treated in a manner identical to that used for the "pure" Nb sample, then the temperature was lowered to 1800 °C and nitrogen was admitted to the vacuum system in a controlled flow at the equilibrium pressure for the desired concentration of 0.0015 nitrogen atoms per Nb atom. After 30 h, the temperature was lowered to 1100 °C (which is well above the precipitation temperature for this concentration), from which temperature it was rapidly quenched to room temperature by blowing He gas onto the sample. The nitrogen concentration was determined as 0.0015 ± 0.0002 by measuring the resistivity ratio of the sample between room temperature and 77 K and using the results of Schultz[3] and Meyerhof.[4] This corresponds to a concentration of 230 ppm by weight. Although we have not done extensive measurements to test for clustering in this sample, the results of Richter *et al.*[5] indicate that this should not be a problem. In addition, the close agreement between the concentration determined from gas pressure during loading and resistivity ratio is evidence that there is no significant clustering in the sample.

The neutron measurements were performed on a triple-axis spectrometer at the NBS reactor, using an incident energy of 41 MeV with a 4-cm-long pyrolytic graphite filter. Monochromator and analyzer crystals were pyrolytic graphite with a mosaic spread of 40 min of arc, and collimations of 40-20-20-40 min of arc were used before and after the monochromator and analyzer respectively.

The results of a scan along the (ζ,ζ,ζ) direction for $1.0\leq\zeta\leq 2.0$ are shown in Fig. 1. The data shown are the results of adding together counts from five adjacent measurements on a grid $5\times$ as fine. This addition is the dominant contribution to the resolution which is thus of the order of one step wide. The salient feature of Fig. 1 is the large peak which is present for the N-doped sample but is absent (or greatly weakened) for the "pure" Nb sample. This peak occurs at a wave vector (units of $2\pi/a$) of $(\frac{4}{3},\frac{4}{3},\frac{4}{3})$, or a reduced wave vector of $(\frac{2}{3},\frac{2}{3},\frac{2}{3})$, which is just the point at which the [111] LA phonon dispersion relation[6] has a deep local minimum.

However, as shown in Fig. 2, the diffuse scattering does not have the periodicity of the reciprocal lattice (open circles represent points where scans were made but no intensity was found; closed circles represent points where intensity was found). These results are similar to those found by Keating and LaPlaca[7] for the ω phase transition in Nb-Zr alloys, but are quite different from the results of Wakabayashi[8] for smaller concentrations of Zr in Nb (a substitutional alloy), where the diffuse scattering was

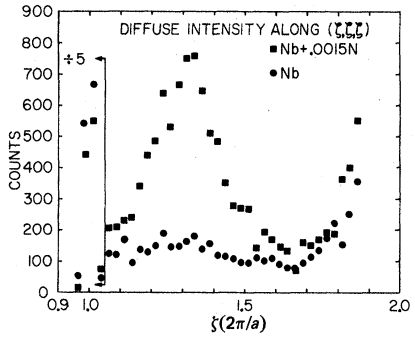

FIG. 1. Neutron diffuse scattering intensity for pure Nb and $NbN_{0.0015}$ observed along the [111] direction. Counts normalized to equal sample volumes.

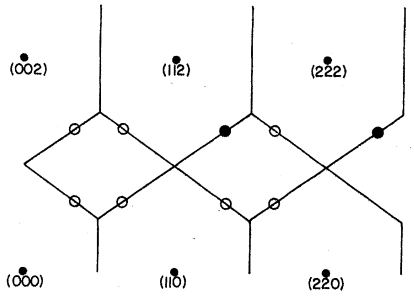

FIG. 2. Location of diffuse scattering observed for nominally pure Nb in Reference 2. Open circles represent points where scans were made but no peak in intensity observed; closed circles where intensity similar to that in Fig. 1 was observed. Size of circles is unrelated to intensity distributions.

periodic. In addition, Werner and Rowe[2] showed that the diffuse scattering was temperature independent. Similar results to those described here have been obtained for Nb-doped with oxygen impurities by Pynn and Shapiro.[9] We have also obtained similar results (of poorer quality) on a very small Nb sample which contained 0.015 atoms of O per Nb.

The results presented here, while shedding light on the earlier comparison of x-ray and neutron diffuse scattering data (which were obtained for different samples), raise many interesting questions. A more detailed study of the diffuse neutron intensity for the N-doped sample, and a comparison of x-ray and neutron data on the *same* sample are being undertaken to answer some of the questions. In particular, it is not at all clear why 0.0015 N atoms per Nb atom gives rise to a diffuse pattern similar to that of the ω phase rather than to a Huang scattering pattern such as was observed for substitutional defects.[8]

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