INFLUENCE OF NITROGEN INTERSTITIELS ON THE THERMAL CONDUCTIVITY OF H ON D DOPED SUPERCONDUCTING Nb

M. Locatelli, K. Neumaier, H. Wiph

To cite this version:

HAL Id: jpa-00217918
https://hal.archives-ouvertes.fr/jpa-00217918
Submitted on 1 Jan 1978

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.
INFLUENCE OF NITROGEN INTERSTITIELS ON THE THERMAL CONDUCTIVITY OF H ON D DOPED SUPERCONDUCTING Nb

M. Locatelli t, K. Neumaier tt and H. Wipf ttt

t Centre d’Etudes Nucléaires de Grenoble, Service des Basses Températures
85X-38041 Grenoble Cedex (France)

tt Zentralinstitut für Physikalische Forschung der Bayerischen Akademie der Wissenschaften
D 8046 Garching (Germany)

ttt Department of Metallurgy and Mining Engineering and Materials Research Laboratory,
University of Illinois, Urbana, Ill. 61801 (U.S.A.)

Résumé.— La conductivité thermique de cristaux de niobium contenant H, D, N a été mesurée entre 0,07 K et 1,5 K. Les résultats mettent en évidence la présence de niveaux tunnel de l’hydrogène ou de deutérium piégés sur l’azote en position d’interstitiel.

Abstract.— The thermal conductivity of H, D, N doped Nb crystals has been measured between 0.07 K and 1.5 K. The results show the evidence of tunneling of hydrogen or deuterium trapped by interstitial nitrogen in niobium.

The addition of hydrogen to superconducting Nb produces an isotope dependent specific heat anomaly and resonant phonon scattering at temperatures below 2 K. The latter was interpreted as tunneling of H and D in the Nb lattice /1,2/. Electrical resistivity measurements have shown that the behaviour of hydrogen in Nb is changed considerably by additional interstitials such as N or O providing traps. Since traps are energetically favorable compared to ordinary sites the H and D atoms are attracted to traps at low temperature and do not precipitate as in the case of pure Nb /3/.

The aim of the present measurements of thermal conductivity is to study the influence of N interstitials in H doped Nb. Our thermal conductivity data (temperature range 0.07-1.4 K) were obtained with the standard two heater method using a dilution refrigerator. Wire samples (1.25 mm diameter and 5 cm long) rather than bulk samples were used since they are easy to purify via ohmic heating (2200°C, 10^10 torr) and can be quenched rapidly enough to prevent nitrogen clustering after doping at 1800°C. The interstitial impurities content (N, O, C) can be reliably controlled by electrical resistivity measurements /3/. The samples were doped electrolytically with hydrogen isotopes at room temperature. Since we are only interested in thermal conduction by phonons we have made measurements only below 1.4 K. Below this temperature the contribution of electrons to the heat transport is negligible, due to the high superconducting transition temperature of Nb. To show clearly the phonon scattering, thermal conductivity data are plotted in figures 1, 2, 3 on a reduced scale K/T^3 which is proportional to the phonon mean free path. Its variation therefore can be directly observed.

Fig. 1: K/T^3 versus T experimental points

The dashed line in figure 1 was calculated assuming a boundary limited I equal to the sample diameter (1 = 0.12 cm) with K = 0.71 1 T^3 (W cm^-1 K^-1) /2/. Data of the N doped sample below 0.1 K are a factor 2 larger than theoretically expected (figure 1). This is not unreasonable if one supposes specular reflections of phonons, since the sample was not abraded. At higher temperatures one observes the
influence of N. Due to rapid quenching after doping nitrogen occupies isolated interstitial sites and scatters phonons like a point defect.

The most striking feature shown in figure 1 is the large isotope effect in samples doped with both N and H or D. Therefore some anomalies below 1 K must be due to H or D trapped on interstitials N in our case. This is particularly clear in the case of D doped samples shown again in figure 3. The high temperature scattering due to precipitates or dislocations, in the only D doped sample disappears nearly completely in the D.N. doped sample.

A quantitative analysis of the thermal conductivity data of doubled doped samples, using the Callaway model, indicates that at least two resonances are necessary to describe the results: one at high frequency (above 1 K) and the other showing the isotope dependence (0.3 K for D and about 0.9 for H). In the case of H, it is somewhat difficult to define the low resonance temperature because the two resonances are not well separated.

The low frequency resonance displaying isotope effect can be explained by a tunneling process of the N-H and H-D pairs. The concentrations in our samples are such that all the H and D are trapped in principle on the N interstitials, so that there are no more precipitates and dislocations /2/ and we can suppose that the resonance at high frequency is also due to some tunneling energy.

So far we are not aware of a detailed model for the levels of these pairs. The ratio of about 3 between the H and D lowest frequencies cannot be simply explained; such interstitial pairs are certainly strongly coupled to the Nb lattice and it is therefore not permitted to separate the motion of the N-H pair from that of the surrounding lattice.

Furthermore the concentration of N-H pairs (~10²⁰ pairs/cm³) in our samples is more than an order of magnitude too high to neglect interactions which could impede or prevent tunneling. We note finally that the small reduction observed at the lowest temperature for the only D doped sample can be attributed to spurious contamination with N, O, C which introduce some pairs.

The presence of residual impurities can also explain the results of Seller et al. who have observed the same kind of thermal conductivity cur-
ves as ours. This hypothesis is supported by the fact that the residual resistivity ratio of their samples was only $100^{2}$. In conclusion we have shown evidence of strong coupling of $H$ or $D - N$ pairs with the Nb lattice. Detailed interpretation requires additional measurements on samples with lower $N, H$ concentration, together with a theoretical model for the tunnelling level system.

References


