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1991 J. Phys.: Condens. Matter 3 5227

(http://iopscience.iop.org/0953-8984/3/27/018)

J. Phys.: Condens. Matter 3 (1991) 5227–5229. Printed in the UK

LETTER TO THE EDITOR

Arrhenius behaviour of the degradation of the superconducting transition temperature in the A15 compound Nb_3Au

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Received 5 April 1991

Abstract. Nb_3Au was synthesized by arc melting appropriate amounts of Nb and Au together. The compound was quenched from different high temperatures. The degradation of the superconducting transition temperature as a function of the quenching temperature turns out to follow an Arrhenius behaviour. This behaviour is similar to V_3Ga, $Ca_3Rh_4Sn_{13}$ and $Yb_3Rh_4Sn_{13}$ which were measured previously. The degradation is ascribed to anti-site disorder in the A15 structure.

Intermetallic compounds are not necessarily completely ordered up to the melting temperature. The A15 and related ternary superconducting compounds which we have studied so far (Van Winkel *et al* 1984, Westerveld and Bakker 1986 and Westerveld *et al* 1987) all exhibit atomic (chemical) disorder at higher temperatures, i.e. the one atomic species is substituted on the other sublattice and vice versa. Thus when the compound is denoted by A_3B, A atoms substitute on the B sublattice and B atoms on the A sublattice. This type of disorder is called anti-site disorder. It results in a degradation of the superconducting transition temperature, when the disorder is frozen by fast quenching.

In previous papers (Westerveld and Bakker 1986, Westerveld *et al* 1987) it was demonstrated than an Arrhenius relation holds between the quenching temperature T_Q and the degradation of superconducting transition temperature $\Delta T_c = T_{cmax} - T_c$, where T_{cmax} corresponds to T_c for perfect order. Moreover it was derived that from the slope of the Arrhenius plot the disordering enthalpy, i.e. enthalpy corresponding to the exchange of an A and B atom, can be obtained. The Arrhenius relation was shown to be:

$$\frac{\Delta T_c}{T_{cmax}} = A \exp\left(-\frac{W}{k_B T_Q}\right) \tag{1}$$

where A is a constant containing the entropy, W is half the disordering enthalpy and k_B is Boltzmann's constant.

In order to extend these investigations to more compounds and in order to obtain a better insight into values of disordering energies in superconducting A15 compounds we studied the result of quenching the A15 compound Nb_3Au.

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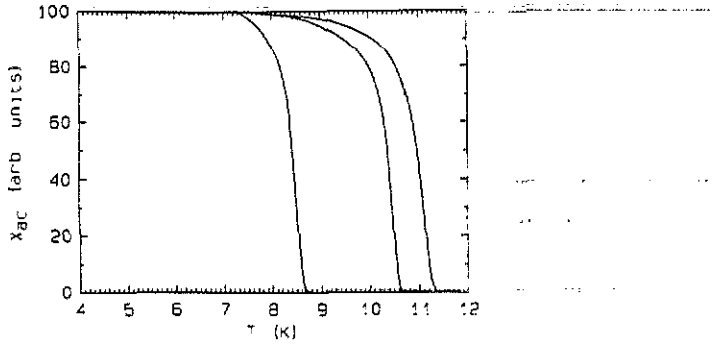

Figure 1. AC susceptibility of Nb_3Au versus temperature for three different quench temperatures: from left to right 1152.9, 862.6 and 819.8 °C.

The Nb_3Au compound was prepared by arc melting Nb and Au (both with a purity better than 99.5%) together in the 3 to 1 composition. X-ray diffraction showed a pure A15 diffraction pattern with a lattice parameter of 5.2037 Å. After annealing the sample for 20 hours at 770 °C the superconducting transition temperature became 11.39 K. A special quenching device, called HIPOQ (Riemersma *et al* 1991), was used to quench samples from high temperatures into water. The quenching rate is estimated to be 5×10^4 K s^{-1}. To ensure fast quenching powdered samples with powder diameters between 0.21 mm and 0.48 mm were used. After annealing long enough in the HIPOQ to obtain thermodynamic equilibrium, the samples were quenched into water from temperatures between 820 and 1150 °C. The superconducting transition temperature was determined by AC susceptibility measurements. The superconducting transition temperature was taken as the temperature where the susceptibility becomes 'zero', the so-called onset T_c. It was checked that there was negligible dependence of T_c on the small measuring field (4.6, 23, 46, 92 A m^{-1}).

The lattice parameter of our starting material of 5.2037 Å corresponds to 5.2024 Å from the literature (Waterstrat 1968). Quenching turned out to degrade T_c, but an anneal at relatively low temperature restored T_c to its original value, showing that there were no irreversible effects. Figure 1 gives magnetic susceptibility measurements for three samples quenched from different temperatures, showing different transition temperatures.

By using a least-mean-square fit to equation (1) we obtained the three parameters for Nb_3Au:

$T_{cmax} = 11.75(2)$ K $\quad 2W = 1.72(6)$ eV (the disordering enthalpy) $\quad \ln A = 5.7(3)$.

This T_{cmax} is comparable to literature values of 11.5 K (Vonsovsky *et al* 1982). In figure 2 the Arrhenius curve is presented, i.e. a plot of $\ln(\Delta T_c/T_{cmax})$ versus $1/T_Q$. Following equation (1) this should yield a straight line and in fact the measuring points follow the predicted relation well.

Let us compare the results of $2W$ and $\ln A$ with our previous results on V_3Ga. On the one hand it is imaginable that the disordering enthalpies could scale with the melting temperatures. V_3Ga does not melt, but transforms to a disordered BCC phase at high temperatures. The average of the solidus and liquidus temperatures of this phase is 1910 K (Massalski 1986) and the disordering enthalpy is 1.23(6) eV (Westerveld and

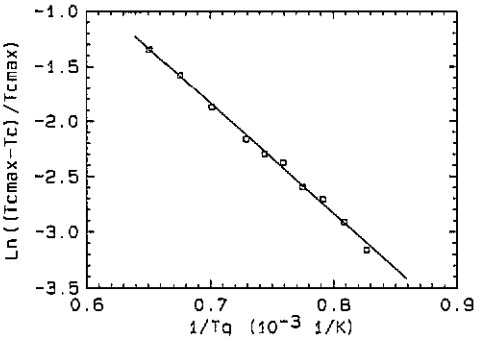

Figure 2. The logarithm of the degradation of the superconducting transition temperature $T_{c\max} - T_c/T_{c\max}$ versus the reciprocal of the quenching temperature $1/T_q$ for Nb_3Au.

Bakker 1986). Nb_3Au melts at about 2400 K (Massalski 1986). For the ratio of these temperatures we obtain 0.80 and for the corresponding ratio of disordering enthalpies 0.72(4), so that within experimental error this relation seems approximately to hold. More experimental results on other compounds are needed to explore such a relation. On the other hand, we could speculate that the disordering enthalpies could scale with the logarithm of the pre-exponential factors, such as is approximately valid for analogue quantities determining atomic diffusion coefficients. In fact this means a scaling of enthalpies and entropies. For V_3Ga this ratio is 0.33(3) eV (in Westerveld and Bakker (1986) the quantity 'A' should be read as 'ln A'), Nb_3Au this ratio is 0.30(2) eV. Within experimental error the relation is fulfilled, but experimental data for other compounds are also necessary.

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