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Specific heat of $PbMo_6S_8$ in high magnetic field

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Abstract

In order to investigate the volume upper critical field of the Chevrel-phase superconductor $PbMo_6S_8$, specific-heat measurements have been performed in magnetic fields up to 24.5 T. The superconducting transition temperature T_c of 14.3 ($\pm$ 0.1) K is suppressed by the maximum magnetic field to 10.1 ($\pm$ 0.3) K, which extrapolates to an upper critical field of 56 T (confirming measurements at fields up to 14 T). For a mixed state in the magnetic field a linear term $\gamma_m T$, related to the core states, is present in the electronic contribution to the specific heat. At a magnetic field of 24.5 T, γ_m amounts to 50% of the normal state value γ of 6.5 mJ/K^2 g-at. By extrapolation, an upper critical field of 54 T can be derived.

1. Introduction

Of the class of Chevrel-phase superconductors, $PbMo_6S_8$ is a very interesting compound because of its very high upper critical field (of the order of 60 T), which makes it a promising material for realizing stationary magnetic fields above 20 T. In the past, the behaviour of the upper critical field B_{c2} has been investigated by critical current measurements and measurements of the specific heat up to 14 T [1]. In this work we will extend the investigation of Cors et al. to magnetic fields up to 24.5 T, and study the low temperature behaviour in the superconducting state. The calorimetric determination of B_{c2} has the advantage that a volume value is derived, whereas in inductive measurements the critical field is underestimated. This is seen, e.g. in experiments based on macroscopic screening currents, which indicate that the granular nature of the material may limit the transport current by possible weak links between grains [2,3].

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2. Experimental

The experiments have been performed in the High Field Magnet Laboratory of the University of Nijmegen, using a Bitter magnet with a maximum field of 20 T and a hybrid magnet with magnetic fields up to 24.5 T.

The homogeneous polycrystalline sample of $PbMo_6S_8$ has been prepared by a high temperature reaction of Mo, PbS and MoS_2 at 1600°C using a molybdenum crucible and applying pressure [4]. Using this method, a relatively large sample could be produced from which a cylinder of 1.457 g has been cut. This size has been chosen to be able to measure the heat capacity by a (semi-) adiabatic method. The advantage of this technique is that it is sufficiently rapid to measure in the high field installation, where the time available for measurement is limited. The

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measuring cell consisted of a sapphire plate sample holder of dimensions $10 \times 10 \times 0.5\,mm^3$ on one side of which, the sample was glued. On the other side the sample holder was equipped with a nickel–chromium film as heater and a thick-film RuO_2 resistance (ALPS 10 kΩ nominal resistance), used as a thermometer. The (small) magnetoresistance of this thermometer has been calibrated carefully in fields up to 24.5 T.

3. Measurements

The specific heat has been measured, below 20 K, in constant magnetic fields of 0, 5, 10, 12.5, 15, 17.5, 20, 22.5, and 24.5 T. In Fig. 1 a number of these measurements are presented in a plot of C/T versus T. The transition peak is still present at the highest field, showing that at 24.5 T there is still bulk superconductivity. Above T_c, the measured specific heat data coincide for all fields. The specific heat results confirm the previous measurements up to 14 T of Cors et al. [1] performed on a sample with an almost similar preparation, measured with a relaxation method.

The critical temperature T_c is obtained for each field by extrapolating the measured transition in C/T to an idealized transition, conserving the entropy. The superconducting transition temperature T_c of 14.3 ($\pm$ 0.1) K is suppressed by a magnetic field of 24.5 T to 10.1 ($\pm$ 0.3) K.

4. Discussion

From the suppression of the specific heat jump in field we can determine the initial slope dB_{c2}/dT for which we derive a value of -5.6 T/K (compared to -5.5 T/K reported in Ref. [1]). The upper critical field $B_{c2}(0)$ is given by

$$B_{c2}(0) = -AT_c \frac{dB_{c2}}{dT}\Big|_{T_c}, \quad (1)$$

where A varies between 0.693 (for the dirty limit) and 0.726 (for the clean limit) [4]. Taking the lower estimate, we arrive at $B_{c2}(0) = 56$ T (see Fig. 2(a)).

In order to study the superconducting behaviour, we obtain the electronic part of the specific heat (C_e) subtracting the lattice contribution (C_L) from the specific heat. Using a method developed by Junod [5] a simple spectral analysis can be made, in which the lattice specific heat is described by a summation of three Debye components, representing a simple approximation to the phonon spectrum:

$$C_L(T) = \sum_{i=1}^{3} D_i C_D\left(\frac{T}{T_i}\right). \quad (2)$$

Here $C_D(T/T_i)$ is the Debye specific heat function, with Debye temperature T_i. This description has been applied in the past to the normal state specific heat [4,6], fitting the expression $C_N(T) = \gamma T + C_L(T)$, using specific heat data above T_c and low temperature specific heat data measured on a Fe-doped sample (where the superconductivity is suppressed). For the normal linear coefficient γ, a value of $6.5 \pm 0.2\,mJ/K^2$g-at. is derived. The normal state specific heat calculated with these parameters gives

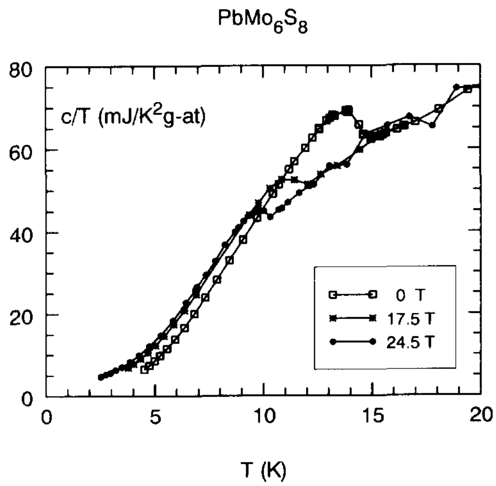

Fig. 1. Specific heat of $PbMo_6S_8$ in magnetic field.

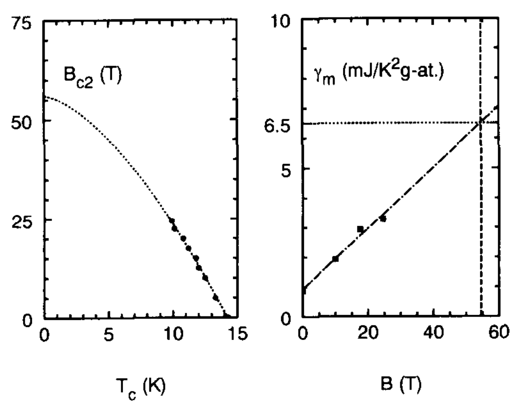

Fig. 2. (a) Applied field versus critical temperature, the dashed line is drawn as a guide to the eye; (b) The linear coefficient in the mixed state γ_m.

a good description of our data above T_c in all fields. We have used this phonon contribution to obtain the electronic part of the specific heat in the superconducting state, also for the measurements in field, assuming that the phonon contribution is field independent below T_c.

The electronic specific heat of a strong coupling superconductor can be described satisfactorily by the α-model of Padamsee [7], where the superconducting gap is scaled relative to the BCS-gap by a factor α. As a good approximation to this description, the simple BCS-like relation: $C_e = A_1 \exp(-bT_c/T)$ can be used, in the low temperature region ($T/T_c < 0.5$).

An acceptable fit to the zero field data is only obtained including a linear term in the above expression, corresponding to a residual γ^* of 0.9 mJ/K^{2}g-at. The occurrence of a residual γ^* is excluded however, in the study of Cors[4] on samples prepared in a similar way. The most probable cause for γ^* is an inaccuracy in the low temperature part of the phonon term C_L, as the sample preparation technique plays a role in the phonon contribution [6].

The data of C_e/T clearly indicate the presence of a linear contribution γ_m in the specific heat in magnetic field. Using the relation $C_e = A_1 \exp(-bT_c/T) + \gamma_m(B)T$ to fit these values, at low temperatures, we can derive for several fields values of $\gamma_m(B)$; γ_m(24.5 T) amounts to about 50% of the normal state value γ_0. In Fig. 2(b), $\gamma_m(B)$ is presented for several fields. The non-zero linear contribution $\gamma_m(B)T$ has its origin in the normal cores in the mixed state of the superconducting material. A linear increase of γ_m with the applied magnetic field is expected, until the value of the normal state coefficient γ at the critical field B_{c2} is reached (see Fig. 2(b) where $B_{c2} = 54 \pm 5$ T is derived).

5. Conclusions

The critical field B_{c2} of $PbMo_6S_8$ is confirmed to be as high as 56 T, by specific heat measurements up to 24.5 T. In the mixed state, a linear term in the specific heat is present associated with the normal cores.

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