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## Evidence for Superconductivity in $\text{Nb}_2\text{SC}_{0.90}$ Carbosulfide

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## Evidence for Superconductivity in Nb<sub>2</sub>SC<sub>0.90</sub> Carbosulfide

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Evidence for bulk superconductivity in Nb<sub>2</sub>SC<sub>0.90</sub> carbosulfide is presented. Diamagnetic volume fraction of Nb<sub>2</sub>SC<sub>0.9</sub> was precisely determined with low applied magnetic fields which were corrected by 6N lead rod as an internal standard. Based on magnetic field dependence of magnetic moment at several constant temperature's below T<sub>C</sub> = 5 K, Nb<sub>2</sub>SC<sub>0.9</sub> is found to be a type II superconductor with relatively high H<sub>C2</sub> and low H<sub>C1</sub>.

**Keywords:** carbosulfides; superconductivity; low-dimensional material

### INTRODUCTION

In the course of the studies toward a generation of new physical properties by chalcogenide materials, we recognized empirically that a presence of carbon is of importance. The carbon-containing sulfides are of particularly interest in that how carbon atom with the same electronegativity as sulfur may effect a change of structural dimensionality and physical properties of sulfide.

A little information has been reported for carbosulfide materials. In 1960 Kudielka and Rohde first synthesized the group 4 transition-metal (Tm = Ti, Zr) carbosulfides with the composition of Tm<sub>2</sub>SC and determined crystal structure which is now called H(hexagonal)-phase <sup>[1]</sup>. Subsequently the H-phase of the group 5 Nb carbosulfide, Nb<sub>2</sub>SC<sub>x</sub> (x = 1, 5/6, 2/3), was found by

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Beckmann *et al.* in 1968<sup>[2]</sup>. The H-phase has a layered structure with a layer stacking sequence of C-Tm-S-Tm-C-Tm-S-Tm-C. The Tm layer exists in every other layer and C atoms occupy octahedral holes or anti-prismatic sites between slabs of edge-sharing  $STm_6$  trigonal prisms. It is interesting to note that the structure of H-phase is analogous to that of transition-metal intercalated  $2H-MoS_2$ , but marked differences are the presence of  $STm_6$  trigonal prism and the absence of van der Waals gap. As we already reported,  $Nb_2SC_x$  ( $0.8 < x \leq 1.0$ ) exhibits superconductivity below 5.0 K<sup>[3,4]</sup>. In this paper, we describe a precise magnetic characterization for  $Nb_2SC_{0.9}$ .

### EXPERIMENTAL AND SAMPLE CHARACTERIZATION

The details of sample preparation were described in the previous reports<sup>[2-4]</sup>.  $Nb_2SC_{0.9}$  was characterized by powder XRD step scans (Philips, PW1800) following Rietveld analysis (RIETAN)<sup>[5]</sup>. Electron probe micro-analysis indicated that these compounds were exactly of the ternary system with an atomic ratio of Nb : S = 2 : 1. No Si and O were detected. It is convenient to define the amount of C in  $Nb_2SC_x$  determined by quantitative analyses in terms of "effective" against to "nominal" for initial composition. The effective C content of  $Nb_2SC_{0.90}$  was determined as 0.91 by C/S analyzer (Leco, CS-444LS) and as 0.89(2) by RIETAN, 0.90 on the average. The measured density of  $Nb_2SC_{0.9}$  was  $6.93 \text{ g}\cdot\text{cm}^{-3}$  with an accuracy of less than 1%.

The final conventional reliability factors obtained by Rietveld analysis for the single phase  $Nb_2SC_{0.9}$  was  $R_F = 1.01 \%$  and  $R_{wp} = 13.24 \%$ . The atomic coordinates for Nb, S, and C were  $4f$  ( $z = 0.0964$ ),  $2d$ , and  $2a$  in the space group  $P6_3/mmc$  (No. 194), respectively. The refined lattice parameters of  $Nb_2SC_{0.90}$  were  $a = 3.2940(1) \text{ \AA}$  and  $c = 11.5525(2) \text{ \AA}$ . The magnetic properties were measured by a Quantum Design SQUID (superconducting quantum interface device) with continuous low temperature control. Before low magnetic field ( $H$ ) measurements, in order to degauss the shield and to minimize the remnant field, the  $H$  oscillates from high field to 0 G. For an internal standard sample Pb (6N) rod from Rare Metallic Co. Ltd. was used.

### RESULTS AND DISCUSSION

Zero resistivity ( $\rho$ ) was confirmed for a series of  $Nb_2SC_x$ <sup>[3,4]</sup>. The  $\rho$  of  $Nb_2SC_{0.90}$  starts to decrease below 5.10 K and drops steeply at a middle

temperature of transition of 4.95 K with a 10-90% transition width of 0.06 K. The  $\rho$  becomes zero at 4.90 K. We define the superconducting transition temperature ( $T_C$ ) as the temperature showing half of the normal resistivity. The  $T_C$  shifted toward lower temperature with decreasing the effective C content in  $\text{Nb}_2\text{SC}_x$ .

The magnetic susceptibility ( $\chi$  [emu/g]) for  $\text{Nb}_2\text{SC}_{0.90}$  changes drastically its sign from positive to negative at 5.0 K. But the field cooling (FC) diamagnetic volume fraction (VF) was smaller than an expected value. The FC diamagnetic VF decreased to 3% with an increase of  $H$  up to 200 G. On the contrary a decrease of  $H$  down to 0.1-0.05 G resulted in a remarkable increase of FC diamagnetic VF. It reached about 70% which was large enough to assign the sample to a bulk superconductor. To verify the above results, we used a 6N lead ( $T_C = 7.2$  K) rod as an internal standard to correct the low applied magnetic field. The diamagnetic VF of sample was also evaluated with respect to the ideal lead superconductor.

A mixture of Pb rod (6N, 98.62 mg) and  $\text{Nb}_2\text{SC}_{0.9}$  (132.13 mg) powder was used as a sample ( $\text{Pb/Nb}_2\text{SC}_{0.9}$ ). Figure 1(a) shows temperature dependence of

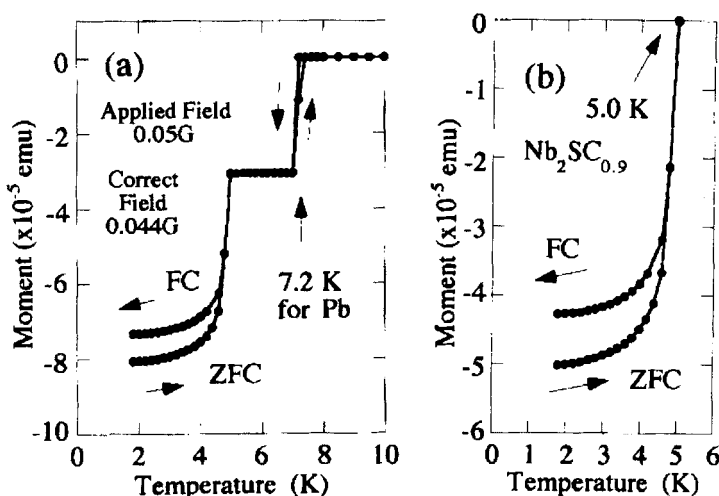


FIGURE 1 Temperature dependence of magnetic moment of a mixture of Pb rod (6N, 98.62 mg) and  $\text{Nb}_2\text{SC}_{0.9}$  (132.13 mg) powder (a). (b) is magnetic moment for  $\text{Nb}_2\text{SC}_{0.9}$  obtained by excluding the moment of Pb.

magnetic moment obtained for  $\text{Pb}/\text{Nb}_2\text{SC}_{0.9}$  under zero FC (ZFC) and FC. It is easily understandable from Fig. 1(a) that  $\text{Nb}_2\text{SC}_{0.9}$  is a bulk superconductor. Based on the density of Pb ( $11.34 \text{ g cm}^{-3}$ ), the applied field of  $0.05 \text{ G}$  was corrected to  $0.044 \text{ G}$ . Magnetic moment for  $\text{Nb}_2\text{SC}_{0.9}$  (Fig. 1(b)) was obtained by excluding that of Pb from Fig. 1(a). Figure 2 (a) and (b) show temperature dependence of magnetic susceptibility ( $\chi$  [emu/g]) for  $\text{Nb}_2\text{SC}_{0.90}$  under ZFC and FC, respectively. The diamagnetic VF for  $\text{Nb}_2\text{SC}_{0.90}$  was precisely determined by the correct applied magnetic field which was deduced from the internal standard of Pb. The diamagnetic VF for ZFC and FC at  $0.044 \text{ G}$  were determined as 75 % and 64 %, respectively. Similar temperature dependence of  $\chi$  was observed under the other low fields and the results are shown in Fig. 2(a) and (b) for ZFC and FC, respectively. The evaluated diamagnetic VF at each correct applied field is written within Fig. 2 (a) and (b). These results confirm the previous reports <sup>[3,4]</sup>. Even though the applied field is low,  $\text{Nb}_2\text{SC}_{0.9}$  exhibits a bulk superconductivity.

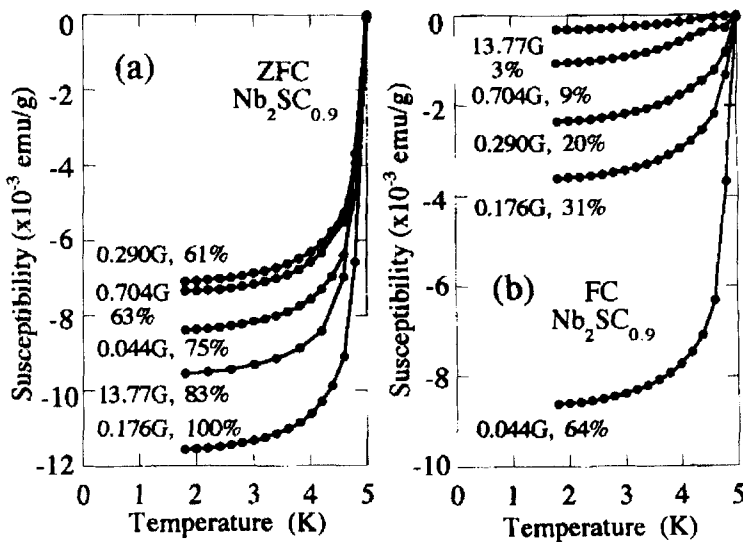


FIGURE 2 Temperature dependence of magnetic susceptibility of  $\text{Nb}_2\text{SC}_{0.90}$  for ZFC (a) and FC (b). The lines drawn are guides to the eye.

To examine a type of superconductivity of  $\text{Nb}_2\text{SC}_{0.9}$ , field dependence of magnetic moment was measured at 2.00 K, 3.25 K, and 4.50 K as shown in Fig. 3(a) and (b) with SQUID for high field measurement. Figure 3(c) shows field dependence of magnetic moment in the low magnetic field region, which was measured precisely after zero-field correction. It is easily understandable from Fig. 3(a)-(c) that  $\text{Nb}_2\text{SC}_{0.9}$  belongs to a type II superconductor. The upper critical field  $H_{C2}$  is determined as 1.6 T, 0.9 T, and 0.3 T for 2.00 K, 3.25 K, and 4.50 K, respectively. As shown in Fig. 3(c) the lower critical field  $H_{C1}$  is determined as 66 G at 2.00 K, 38 G at 3.25 K, and 8 G at 4.50 K.

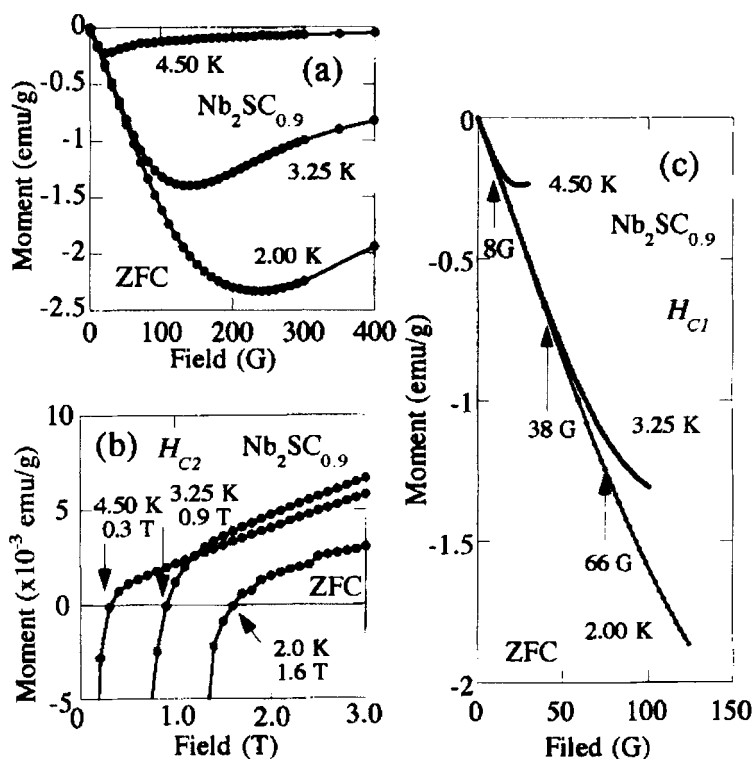


FIGURE 3 Field dependence of magnetic moment of  $\text{Nb}_2\text{SC}_{0.9}$  at 2.00 K, 3.25 K, and 4.50 K in low field (a) and in high field (b). (c) is a precise measurement for low field.

## CONCLUSIONS

By using 6N lead rod as the internal standard, diamagnetic volume fraction for  $\text{Nb}_2\text{SCo}_{0.9}$  was definitively determined under low applied magnetic fields. The field cooling diamagnetic volume fraction reached about 64%, which was large enough to assign the sample to a bulk superconductor. Based on magnetic field dependence of magnetic moment at several constant temperature's below  $T_C$ ,  $\text{Nb}_2\text{SCo}_{0.9}$  is found to be a type II superconductor with relatively high  $H_{C2}$  and low  $H_{C1}$ . As we demonstrated in this paper, the  $T_C$  is substantially low. But further investigation of this class of compounds may lead to find a higher  $T_C$  superconductor.

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