Pressure studies on the superconductor Mo$_3$Sb$_7$

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Abstract. The electrical resistivity ($\rho(T)$) and magnetization ($M(T)$) of the superconductor Mo$_3$Sb$_7$ ($T_c = 2.2$ K) has been measured under pressures up to 22 kbar and at temperatures from 0.4 to 80 K. Hydrostatic pressure is found to show up the spin-density wave state below $T_{SDW}$, which is revealed by a sharp jump in $\rho(T)$ and a maximum in $M(T)$ dependencies. With increasing pressure $T_{SDW}$ decreases while $T_c$ increases. The pressure dependencies of $T_c$ and $T_{SDW}$ can be understood on the basis of competition of the SDW and the superconducting states in this compound.

1. Introduction

Mo$_3$Sb$_7$ with the lattice parameter $a = 0.9591$ nm crystallizes in the cubic Ir$_3$Ge$_7$-type structure (space group Im-3m). This compound has been reported to enter into a superconducting state below $T_c = 2.2$ K [1, 2]. Moreover, specific heat and muon spin rotation experiments [2, 3] indicated that the effective mass of the charge carriers is enhanced ($\sim 16-18 m_0$), if compared to that of conventional BCS superconductors with a weak electron-phonon coupling. At present, Mo$_3$Sb$_7$ is concluded being nonmagnetic due to a spin dimerization below $T^* = 50$ K, which is reflected by anomalies in the heat capacity, magnetic susceptibility [4], muon spin rotation [5, 6, 7], and magnetic excitations in inelastic neutron scattering [5]. The crystal structure of this compound is certainly favorable for spin dimerization, because of a distinct difference between the Mo-Mo intradimer distance ($\sim 0.3$ nm) and Mo-Mo interdimer distance ($\sim 0.46$ nm). In general, the strength of magnetic interactions in a given compound can be modified by applying external hydrostatic pressure. Furthermore, taking into account the fact that the effective mass of superconducting carriers in Mo$_3$Sb$_7$ is sizable, approaching that of heavy-fermion superconductors, one would expect a similar response of its electronic state to applied pressure. In heavy-fermion superconductors, magnetic instabilities or magnetic fluctuations are responsible for enhancing the effective mass of the superconducting quasiparticles, and the pairing mechanism in unconventional superconductors is most probably associated with magnetism [8, 9, 10, 11]. In view of these arguments, a considerable pressure response on the magnetic state is expected in Mo$_3$Sb$_7$. Therefore, the electrical resistivity and magnetization of Mo$_3$Sb$_7$ was studied under pressure. In the following, we present pressure-dependent properties of Mo$_3$Sb$_7$. Results of our study revealed that a spin-density-wave exists in parallel with superconductivity in Mo$_3$Sb$_7$. 

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2. Experimental
Mo\textsubscript{3}Sb\textsubscript{7} was prepared from Mo and Sb (purity 99.95% from Alfa Aesar) by solid-state reaction. Synthesis, purity characterization and determination of crystal parameters of the sample have been carried out in a similar technique as given in Ref. [12]. Electrical resistivity under pressure up to 22 kbar was measured in the temperature range 0.3-80 K by means of a standard four-probe technique, using a \textsuperscript{3}He cryostat with a Cryogenics Ltd. 12 T magnet. Hydrostatic pressure was generated by a piston-cylinder cell using Daphne oil as the pressure-transmitting medium. The resistivity data were taken on cooling at a rate below 0.1 K/min. The magnetization was measured at a pressure of 7 kbar in several magnetic fields up to 0.07 T using a nonmagnetic pressure cell with a SQUID magnetometer (Quantum Design MPMS-5). High purity Pb was used as pressure indicator. The width of the superconducting transition of Pb did not exceed 10 mK, corresponding to an uncertainty of measured pressure ± 0.2 kbar.

3. Results
The electrical resistivity of Mo\textsubscript{3}Sb\textsubscript{7} at temperatures from 3 to 80 K for various pressures up to 22 kbar is presented in Fig. 1 a. Above about 7 K, Mo\textsubscript{3}Sb\textsubscript{7} behaves metallic, without substantial changes upon a change of pressure. For temperatures around \(T^*\), a shoulder in \(\rho(T)\) becomes unveiled. We have analyzed the data around \(T^*\) by taking the temperature derivative of the resistivity \(d\rho(T)/dT\); results are plotted in the inset of Fig. 1 a. The maximum of \(d\rho(T)/dT\), associated with the spin dimerization transition, decreases with increasing pressure. The most important finding of this work, however, is the observation of a new phase transition as evidenced by a sharp anomaly of the 4.5 kbar resistivity at \(T_{SDW} = 6.6\) K, indicated by an arrow in Fig. 1 a, and it is shown in more detail in Fig. 1 b. Nevertheless, a very tiny anomaly around 6.7 K can be recognized for the data collected at 0.2 kbar (not shown here). It is reasonable to ascribe the upturn in the resistivity to the occurrence of a density wave phase, alike the spin-density wave in URu\textsubscript{2}Si\textsubscript{2} [13], and BaFe\textsubscript{2–x}Co\textsubscript{x}As\textsubscript{2} [14], or the charge-density wave (CDW) in Lu\textsubscript{5}Ir\textsubscript{4}Si\textsubscript{10} [15] and Lu\textsubscript{5}Rh\textsubscript{4}Si\textsubscript{10} [16]. We attribute the resistivity upturn in Mo\textsubscript{3}Sb\textsubscript{7} to the opening of an energy gap at some portions of the Fermi surface associated with an antiferromagnetic state.

To corroborate the magnetic origin of the transition at \(T_{SDW}\) the magnetization was measured in low magnetic fields up to 0.02 T and under pressure of 6 kbar (Fig. 2 a). While no anomalies occur at ambient pressure [4], the magnetization under applied pressure and at 0.01 T exhibits a transition at 6.8 K. In a manner typical for antiferromagnets, the application of higher magnetic fields shifts the transition to lower temperatures and in a field of 0.07 T a clear maximum suggesting an antiferromagnetic-type interaction appears at 2.8 K. According to the band structure calculations [4], the Fermi surface is nested. Such a property may trigger a SDW ordering.

Fig. 2b shows low-temperature resistivity \(\rho(T)\) data under pressure. The resistivity at 0.2 kbar is characterized by a superconducting transition at 2.15 K, in agreement with the previous report for ambient pressure.[12] With growing pressure, the critical temperature increases at an initial rate \(dT_c/dP \sim 0.02\) K/kbar, and at maximum applied pressure of 22 kbar \(T_c\) reaches a value of 2.37 K. Simultaneously, the width of the critical transition \(\Delta T_c\) decreases, from 0.2 K at 0.2 kbar to 0.1 K at 22 K, indicating that a more homogeneous superconducting state is realized under pressure. The observations of an increase of \(T_c\) and of a gradually sharpening of the phase transition with increasing pressure (see Fig. 2 b) imply that superconductivity is favored by pressure.

4. Summary
In summary, electrical resistivity and magnetization measurements were carried out under hydrostatic pressure for Mo\textsubscript{3}Sb\textsubscript{7}. The application of pressure increases the superconducting phase transition temperature \(T_c\). Also, pressure reveals a transition from a metallic to a SDW phase...
Figure 1. (color online) Electrical resistivity of Mo$_3$Sb$_7$ for different pressures. a) Electrical resistivity of Mo$_3$Sb$_7$ at temperatures up to 80 K. The inset shows temperature derivative of the resistivity vs. temperature around $T^\ast$. b) The resistivity data around $T_{SDW}$ are normalized at $T = 10$ K.

above 4.5 kbar. From the pressure response of $T_{SDW}$ and $T_c$, we suggest that superconducting Mo$_3$Sb$_7$ is an interesting example, where superconductivity competes with a SDW phase. The increasing $T_c$ in presence of antiferromagnetic interactions implies that superconductivity in Mo$_3$Sb$_7$ is most likely mediated by magnetic fluctuations. Studies of either NMR or neutron scattering under pressure should bring new insights to the spin-density-wave phase scenario or possibly "hidden order" in this binary compound.

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Figure 2. (color online) a) dc-magnetization of Mo$_3$Sb$_7$ at 7 kbar measured in the temperature range 2 - 8 K for several magnetic fields. b) Normalized electrical resistivity of Mo$_3$Sb$_7$ in the temperature range 1.6 - 2.6 K for different pressures.

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