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Electrical resistivity and magnetic susceptibility measurements of the itinerant ferromagnet CoS_2 at high pressures reveal that its magnetic ordering temperature is tuned to zero at a critical pressure 4.8 GPa, indicating the existence of a quantum-phase transition. The ambient pressure continuous magnetic phase transition in CoS_2 becomes first order on a very slight pressure increase, with a tricritical point probably located almost at ambient pressure. No deviations from Fermi liquid behavior were found in the vicinity of the quantum-phase transition in CoS_2, implying its strong first order nature.

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Cobalt disulphide, CoS_2, a metallic compound with the cubic pyrite-type crystal structure[1], experiences a phase transition to a ferromagnetic state at $T_c \sim 122$ K[2]. The magnetic moment per Co atom at low temperature is found to be equal to 0.84 μ_B. The magnetic susceptibility of CoS_2 cannot be fitted with a Curie-Weiss law at $T > T_c$ in a broad range of temperature; however, satisfactory fitting can be achieved at $T > 350$ K and yields an effective magnetic moment $\mu_{eff} = 1.76\ \mu_B/\mathrm{Co}$[3]. This situation indicates the itinerant nature of magnetism in CoS_2 that is supported by studies of the electrical and magnetic properties of CoS_2[2–6]. However CoS_2 occupies a special place among the others weak itinerant magnets like $ZrZn_2$, Ni_3Al, MnSi, (Fe,Co)Si, etc. The point is that CoS_2 becomes a nearly half metal at the ferromagnetic phase transition, when exchange splitting causes a significant decrease of the density of states at the Fermi level for minority spins, which influences its transport and thermodynamic properties [7,8]. That is why contrary to normal behavior, the electrical resistivity of CoS_2 increases upon transitioning to the spin-ordered ferromagnetic state, forming a hump below the phase-transition point, and the volume anomaly at the phase transition turned out to be so evident[9]. Some remarkable features of the magnetic phase transition in CoS_2 should be pointed out as well. The character of the phase transition in CoS_2 seemingly changes from continuous to first order on application of pressure or/and alloying CoS_2 with a small amount of Se[10,11]. The phase-transition temperature decreases with pressure and supposedly tends to zero at ~ 6 GPa, beyond which non-Fermi liquid behavior of resistivity is claimed in the paramagnetic phase[12].

The aim of this paper is twofold. The first goal is to resolve the controversial issue about the existence of a tricritical point on the phase transition line of CoS_2 by performing experiments in a hydrostatic helium-pressure environment for pressures less than 1 GPa. Second, we use a toroid-type quasi-hydrostatic cell to obtain additional information on the behavior of electrical resistivity and the phase-transition line in CoS_2 at low temperature and high pressure as it approaches a quantum regime. From these experiments, we find that a tricritical point does exist on the phase-transition line, probably close to ambient pressure and further that a strongly first-order quantum-phase transition from the ferromagnetic to the paramagnetic states in CoS_2 is located at 4.79 GPa. In contrast to an earlier report[12], there is no deviation from Fermi liquid behavior in CoS_2 at high pressures.

Single crystals of CoS_2 were grown from appropriate amounts of CoS_2 powder (99.5%) and ultra dry $CoBr_2$ -10 mesh beads (99.99%), according to the chemical vapor transport procedure used by Wang et al.[13]. The crystals that resulted following the two-week growth run had dimensions of 1 mm or less. The crystals have a cubic pyrite-type structure with a lattice parameter $a = 5.5362 \pm 0.0002$ Å. One measure of the quality of the crystals is their high residual resistivity ratio $RRR = \rho_{297K}/\rho_{2K} = 285$, which is the highest reported so far for CoS_2[12,13]. Additional characteristics of these CoS_2 crystals are given in Fig. 1, where the ambient pressure resistivity, dc magnetic susceptibility and thermal expansion of CoS_2 are plotted in the vicinity of its magnetic phase transition. Corresponding measurement techniques are described below. As seen in Fig. 1, the nature of the phase transition from these data is somewhat contrary. The temperature dependence of the resistivity agrees with the view of a continuous phase transition in CoS_2; whereas, the thermal expansion (and possibly susceptibility) data could be interpreted in favor of an impurity-smeared first-order phase transition. However, the intrinsic nature of the thermal expansion anomaly at T_c is obscured by a steep growth in thermal expansion below and above T_c. With this background thermal expansion subtracted, depicted in Fig. 1d, the expansion anomaly near T_c looks like the resistivity and probably confirms the continuous nature of the phase transition in CoS_2 at ambient pressure.

Standard four-terminal dc and ac techniques, with

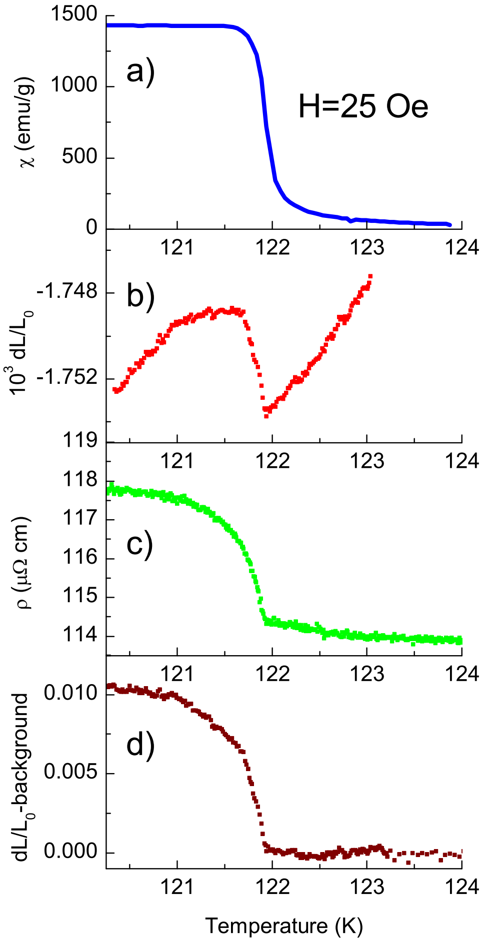

FIG. 1: (Color online) Various properties of CoS_2 at ambient pressure: (a) dc magnetic susceptibility, (b) thermal expansion and (c) resistivity of CoS_2 in the vicinity of the magnetic phase transition. (d) The thermal expansion of CoS_2 with background contribution subtracted. The background was taken as a linear approximation of the thermal expansion in the paramagnetic phase.

welded Pt-wire contacts, were used to measure the resistivity. The dc magnetic susceptibility at ambient pressure was measured with a Quantum Design SQUID magnetometer. Measurements of the ac susceptibility were carried out with a three- and two-coil set up making use of standard modulation techniques. The thermal expansion experiment at ambient pressure was performed with a home-made capacitance dilatometer in which temperature was measured by a calibrated Cernox sensor. Accuracy of the Cernox sensor in the temperature range under study was about 0.02 K.

High-pressure measurements were performed with two kinds of high pressure apparati. A helium-gas apparatus was used in the pressure range 0-1 GPa and, therefore, provided ideal hydrostatic compression of the sample. The helium pressure was measured with a calibrated manganin gauge. Pressures up to 6 GPa were generated in a miniature clamped toroid-type anvil pressure cell.

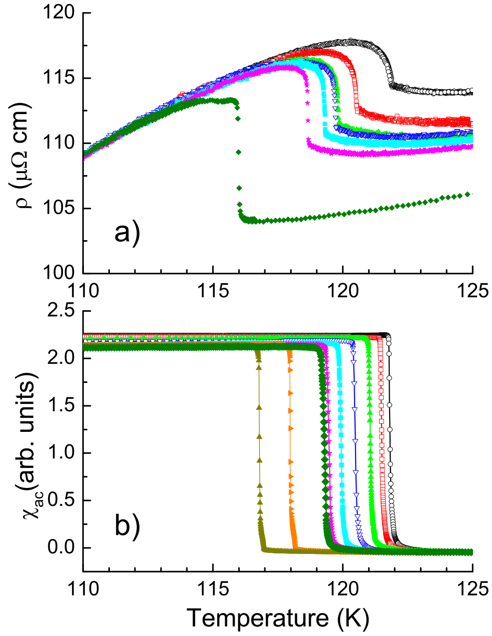

FIG. 2: (Color online) Temperature dependences of resistivity (a) and ac magnetic susceptibility (b) of CoS_2 measured in a helium-gas pressure cell. (a) Pressure from highest to lowest paramagnetic resistivity: 0, 0.19, 0.31, 0.34, 0.42, 0.53 and 0.99 GPa. (b) Pressure from right to left: 0, 0.06, 0.11, 0.2, 0.3, 0.37, 0.45, 0.67 and 0.86 GPa.

Pressure was estimated from a variation of the superconducting transition temperature of Pb[14]. A glycerol/water (3:2 by volume) mixture was used as a pressure medium. This mixture was found to be liquid at pressures up to 5.3 GPa at room temperature and passed through a glass transition near 180 K at ambient pressure. Both pressure systems have been described briefly in Refs.[15,16].

The experimental results are displayed in Figs. 2, and 3. Resistivity data in the range 0-6 GPa and 1.5-200 K are shown in Fig. 2a and Fig. 3a. As is seen, resistivity isobars of CoS_2 reveal a distinct anomaly at the phase transition, with the amplitude and sharpness of the anomaly increasing with pressure. Figures 2b and 3b illustrate the temperature and pressure dependences of the ac magnetic susceptibility of CoS_2 in the vicinity of its phase transition. Sharp discontinuities of ac magnetic susceptibility identify the phase-transition points. Note that the amplitude of the discontinuity only slightly depends on pressure.

On the basis of these resistivity and susceptibility measurements we are able to draw the phase-transition line shown in Fig. 4 and compare it with available literature data. Our low pressure data agree nicely with early results[9,17,18], but the striking disagreement with high pressure data of Barakat et al.[12] probably can be understood as a result of pressure inhomogeneity generated by the steatite pressure medium used for those measurements and difficulties with a precise determination of the transition point, as mentioned by these authors.

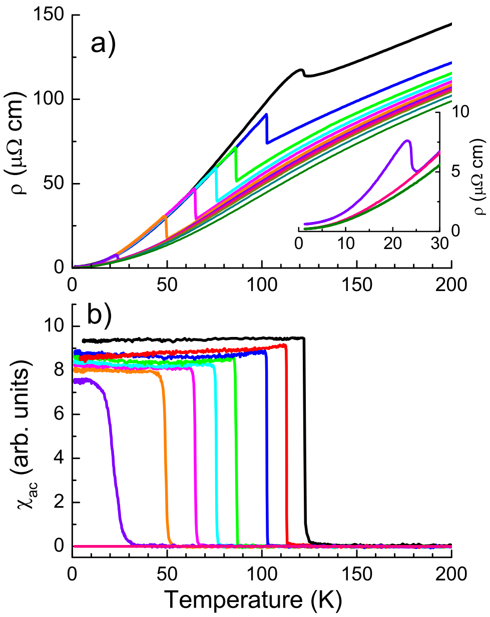

FIG. 3: (Color online) Temperature dependences of (a) resistivity and (b) ac magnetic susceptibility of CoS_2 measured in a toroid-type pressure cell. (a) Pressures from highest to lowest paramagnetic resistivity: 0, 2.34, 3.39, 3.88, 4.24, 4.54, 4.78, 4.92, 5.11, 5.49 and 5.85 GPa. (b) Pressures from right to left: 0, 1.22, 2.34, 3.39, 3.88, 4.24, 4.54, 4.78 and 4.92 GPa. The insert of (a) shows the low temperature resistivity at 4.78, 4.92 and 5.85 GPa. The susceptibility curve at 4.78 GPa appears spread out due to the special trajectory of the measurement that goes along the almost vertical phase boundary.

It is clearly seen in Fig. 4 that the transition line crosses the pressure axis at ~ 4.8 GPa, therefore indicating the existence of quantum phase transition in the ferromagnet CoS_2. However, one must distinguish between quantum and quantum-critical transitions. In the latter case, a phase transition is continuous (second order) and quantum fluctuations of an order parameter are expected to produce a vast area of abnormal behavior of thermodynamic and transport properties. In particular, a non-Fermi liquid temperature variation in resistivity is expected in the quantum-critical regime, which is claimed in Ref.[12]. Fitting the temperature dependence of resistivity $\rho(T)$ at different pressures in the range 1.3-6 K by the function $\rho(T) = \rho_0 + AT^n$ yields pressure dependences of the parameters ρ_0, A, and n that are depicted in Fig. 5. The residual resistivity ρ_0 jumps discontinuously from 0.6 $\mu\Omega$ cm just below the critical pressure $P_c = 4.79$ GPa to 0.2 $\mu\Omega$ cm just above P_c, where the residual resistivity ratio RRR reaches a value of 700, pointing to the very good crystal quality and to the high degree of pressure homogeneity. The parameter A exhibits no essential anomaly at 4.79 GPa, which implies the absence of a divergence in the effective mass of charge carriers at the quantum-phase transition in CoS_2. The temperature exponent n does not show any notable deviation

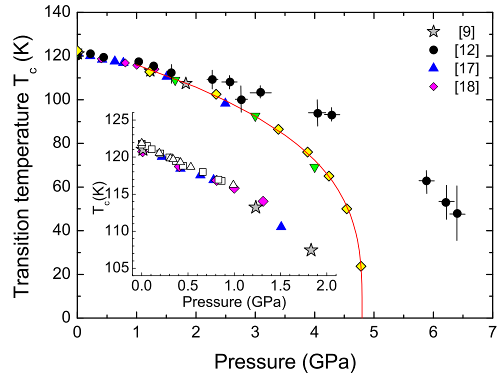

FIG. 4: (Color online) The magnetic P-T diagram of CoS_2. Data obtained with the toroid-type anvil pressure cell are labeled as inverted green triangles and yellow diamonds. Data obtained with the helium gas apparatus are labeled as open squares (susceptibility) and open triangles (resistivity). Other symbols correspond to results published by others (Refs. 9, 12, 17, 18).

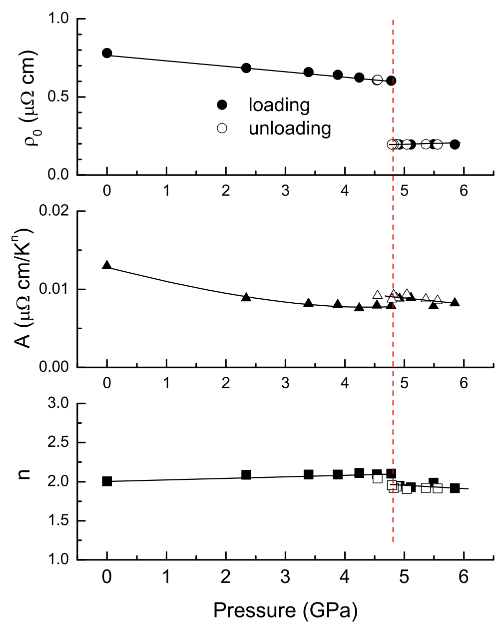

FIG. 5: (Color online) Pressure dependences of the parameters ρ_0, A, and n obtained from fits of resistivity ($1 < T < 6$ K) to the form $\rho(T) = \rho_0 + AT^n$.

from the Fermi-liquid value $n = 2$ in the entire pressure range 0-6 GPa, including the critical pressure $P_c = 4.79$ GPa. These data are again different from that of Ref.[12], probably for the reasons indicated above.

Our results obviously are a signature for the strongly first-order nature of the quantum-phase transition in CoS_2. If the phase transition in CoS_2 at ambient pressure is identified as second order, a tricritical point should exist somewhere on the phase-transition line as claimed in the literature[17,19,20]. However, exact coordinates of the tricritical point in CoS_2 have not been established with

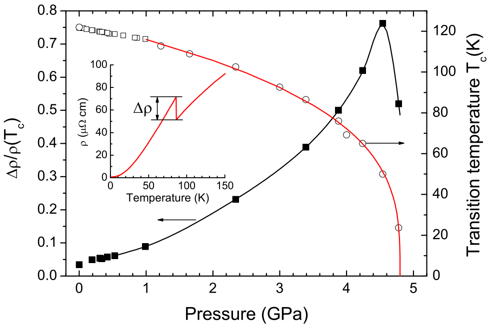

FIG. 6: (Color online) Pressure dependence of the relative change of resistivity at the phase transition $\Delta\rho/\rho(T_c)$ (see inset). The maximum in $\Delta\rho/\rho$ at ~ 4.5 GPa should be ascribed to "freezing" of classical fluctuations, leading to the general resistivity drop. Transition temperatures measured in both the helium gas pressure cell (open squares) and in the toroid-type cell (open circles) are shown. The red curve through $T_c(P)$ data is a guide for the eyes.

precision. Some information can be gained from Fig. 6, where the dependence of the relative resistivity change at the phase transition is shown as a function of pressure. We see that the ratio $\Delta\rho/\rho(T_c)$ grows continuously and almost exponentially with pressure at least to 4.5 GPa, but there is no indication for a tricritical behavior at high pressure. From inspection of Figs. 3, 4 and 6, we conclude that the ambient pressure continuous (second order) magnetic transition in CoS_2 becomes first order with a very slight increase of pressure and then remains first order at pressures to P_c. We note again that the resistivity anomaly at the phase transition becomes progressively sharper with pressure increasing. These observations probably locate the tricritical point almost at ambient pressure, consistent with conclusions in Ref.[20].

In conclusion, measurements of the electrical resistivity and magnetic susceptibility of the itinerant ferromagnet CoS_2 at pressure up to 6 GPa indicate an almost immediate evolution of the magnetic phase transition in CoS_2 from continuous to first order with a slight pressure increase, and, hence, with a tricritical point located close to ambient pressure. The strongly first order quantum-phase transition from ferromagnetic to the paramagnetic states occurs at 4.79 GPa. As a consequence of the first-order nature of the phase transition, no deviations from Fermi-liquid behavior were detected. Both the strong sensitivity of the character of the phase transition in CoS_2 to pressure and the peculiar behavior of resistivity of CoS_2 at the phase transition should find explanations in spin-dependent band structure effects.

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