

# Conventional superconductivity at 203 kelvin at high pressures in the sulfur hydride system

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A superconductor is a material that can conduct electricity without resistance below a superconducting transition temperature,  $T_c$ . The highest  $T_c$  that has been achieved to date is in the copper oxide system<sup>1</sup>: 133 kelvin at ambient pressure<sup>2</sup> and 164 kelvin at high pressures<sup>3</sup>. As the nature of superconductivity in these materials is still not fully understood (they are not conventional superconductors), the prospects for achieving still higher transition temperatures by this route are not clear. In contrast, the Bardeen–Cooper–Schrieffer theory of conventional superconductivity gives a guide for achieving high  $T_c$  with no theoretical upper bound—all that is needed is a favourable combination of high-frequency phonons, strong electron–phonon coupling, and a high density of states<sup>4</sup>. These conditions can in principle be fulfilled for metallic hydrogen and covalent compounds dominated by hydrogen<sup>5,6</sup>, as hydrogen atoms provide the necessary high-frequency phonon modes as well as the strong electron–phonon coupling. Numerous calculations support this idea and have predicted transition temperatures in the range 50–235 kelvin for many hydrides<sup>7</sup>, but only a moderate  $T_c$  of 17 kelvin has been observed experimentally<sup>8</sup>. Here we investigate sulfur hydride<sup>9</sup>, where a  $T_c$  of 80 kelvin has been predicted<sup>10</sup>. We find that this system transforms to a metal at a pressure of approximately 90 gigapascals. On cooling, we see signatures of superconductivity: a sharp drop of the resistivity to zero and a decrease of the transition temperature with magnetic field, with magnetic susceptibility measurements confirming a  $T_c$  of 203 kelvin. Moreover, a pronounced isotope shift of  $T_c$  in sulfur deuteride is suggestive of an electron–phonon mechanism of superconductivity that is consistent with the Bardeen–Cooper–Schrieffer scenario. We argue that the phase responsible for high- $T_c$  superconductivity in this system is likely to be  $H_3S$ , formed from  $H_2S$  by decomposition under pressure. These findings raise hope for the prospects for achieving room-temperature superconductivity in other hydrogen-based materials.

A search for high- (room)-temperature conventional superconductivity is likely to be fruitful, as the Bardeen–Cooper–Schrieffer (BCS) theory in the Eliashberg formulation puts no apparent limits on  $T_c$ . Materials with light elements are especially favourable as they provide high frequencies in the phonon spectrum. Indeed, many superconductive materials have been found in this way, but only a moderately high  $T_c = 39$  K has been found in this search (in  $MgB_2$ ; ref. 11).

Ashcroft<sup>5</sup> turned attention to hydrogen, which has very high vibrational frequencies due to the light hydrogen atom and provides a strong electron–phonon interaction. Further calculations showed that metallic hydrogen should be a superconductor with a very high  $T_c$  of about 100–240 K for molecular hydrogen, and of 300–350 K in the atomic phase at 500 GPa (ref. 12). However, superconductivity in pure hydrogen has not yet been found, even though a conductive and probably semimetallic state of hydrogen has been recently produced<sup>13</sup>. Hydrogen-dominated materials such as covalent hydrides  $SiH_4$ ,  $SnH_4$ , and so on might also be good candidates for showing high- $T_c$

superconductivity<sup>6</sup>. Similarly to pure hydrogen, they have high Debye temperatures. Moreover, heavier elements might be beneficial as they contribute to the low frequencies that enhance electron–phonon coupling. Importantly, lower pressures are required to metallize hydrides in comparison to pure hydrogen. Ashcroft's general idea was supported in numerous calculations<sup>7,10</sup> predicting high values of  $T_c$  for many hydrides. So far only a low  $T_c$  ( $\sim 17$  K) has been observed experimentally<sup>8</sup>.

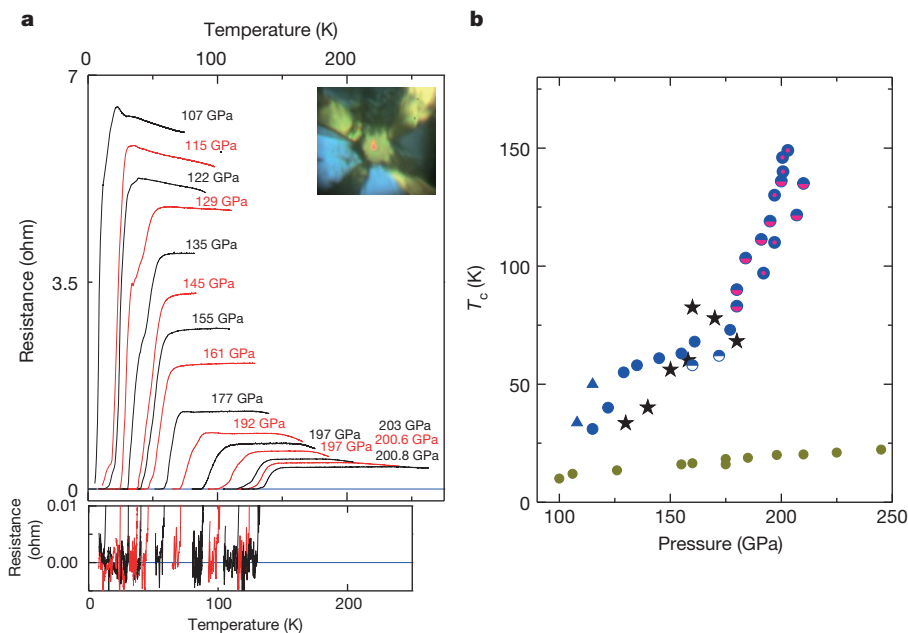
For the present study we selected  $H_2S$ , because it is relatively easy to handle and is predicted to transform to a metal and a superconductor at a low pressure  $P \approx 100$  GPa with a high  $T_c \approx 80$  K (ref. 10). Experimentally,  $H_2S$  is known as a typical molecular compound with a rich phase diagram<sup>14</sup>. At about 96 GPa, hydrogen sulphide transforms to a metal<sup>15</sup>. The transformation is complicated by the partial dissociation of  $H_2S$  and the appearance of elemental sulfur at  $P > 27$  GPa at room temperature, and at higher pressures at lower temperatures<sup>14</sup>. Therefore, the metallization of hydrogen sulphide can be explained by elemental sulfur, which is known to become metallic above 95 GPa (ref. 16). No experimental studies of hydrogen sulphide are known above 100 GPa.

In a typical experiment, we performed loading and the initial pressure increase at temperatures of  $\sim 200$  K; this is essential for obtaining a good sample (Methods). The Raman spectra of  $H_2S$  and  $D_2S$  were measured as the pressure was increased, and were in general agreement with the literature data<sup>17,18</sup> (Extended Data Fig. 1). The sample starts to conduct at  $P \approx 50$  GPa. At this pressure it is a semiconductor, as shown by the temperature dependence of the resistance and pronounced photoconductivity. At 90–100 GPa the resistance drops further, and the temperature dependence becomes metallic. No photoconductive response is observed in this state. It is a poor metal—its resistivity at  $\sim 100$  K is  $\rho \approx 3 \times 10^{-5}$  ohm m at 110 GPa and  $\rho \approx 3 \times 10^{-7}$  ohm m at  $\sim 200$  GPa.

During the cooling of the metal at pressures of about 100 GPa (Fig. 1a) the resistance abruptly drops by three to four orders of magnitude, indicating a transition to the superconducting state. At the next increase of pressure at low temperatures of  $T < 100$  K,  $T_c$  steadily increases with pressure. However, at pressures of  $> 160$  GPa,  $T_c$  increases sharply (Fig. 1b). As higher temperatures of 150–250 K were involved in this pressure range, we supposed that the increase of  $T_c$  and the decrease of sample resistance during warming (Fig. 1a) could indicate a possible kinetic-controlled phase transformation. Therefore in further experiments, after loading and after the initial pressure increase at 200 K, we annealed all samples by heating them to room temperature (or above) at pressures of  $> \sim 150$  GPa (Fig. 2a, see also Extended Data Fig. 2). This allowed us to obtain stable results, to compare different isotopes, to obtain the dependence of  $T_c$  on pressure and magnetic field, and to prove the existence of superconductivity in our samples as follows. (We note that additional information on experimental conditions are given in the appropriate figure legends.)

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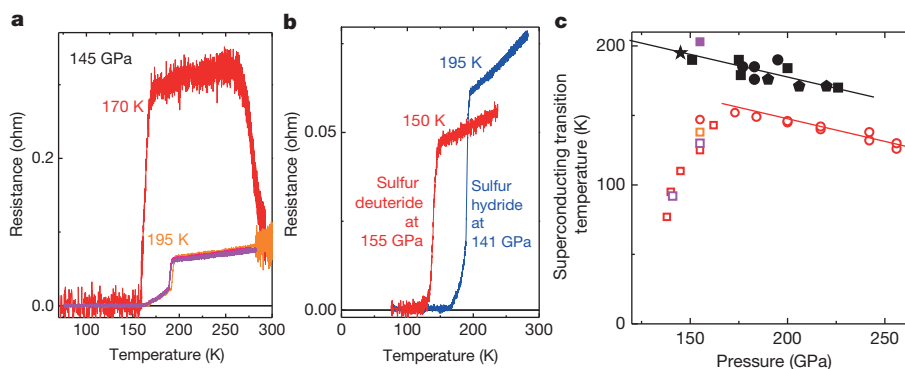


**Figure 1 | Temperature dependence of the resistance of sulfur hydride measured at different pressures, and the pressure dependence of  $T_c$ .** **a**, Main panel, temperature dependence of the resistance ( $R$ ) of sulfur hydride at different pressures. The pressure values are indicated near the corresponding plots. At first, the sample was loaded at  $T \approx 200$  K and the pressure was increased to  $\sim 100$  GPa; the sample was then cooled down to 4 K. After warming to  $\sim 100$  K, pressure was further increased. Plots at pressures  $< 135$  GPa have been scaled (reduced) as follows—105 GPa, by 10 times; 115 GPa and 122 GPa, by 5 times; and 129 GPa by 2 times—for easier comparison with the higher pressure steps. The resistance was measured with a current of 10  $\mu$ A. Bottom panel, the resistance plots near zero. The resistance was measured with four electrodes deposited on a diamond anvil that touched the sample (top panel inset). The diameters of the samples were  $\sim 25$   $\mu$ m and the thickness was

$\sim 1$   $\mu$ m. **b**, Blue round points represent values of  $T_c$  determined from **a**. Other blue points (triangles and half circles) were obtained in similar runs. Measurements at  $P > \sim 160$  GPa revealed a sharp increase of  $T_c$ . In this pressure range the  $R(T)$  measurements were performed over a larger temperature range up to 260 K, the corresponding experimental points for two samples are indicated by adding a pink colour to half circles and a centred dot to filled circles. These points probably reflect a transient state for these particular  $P/T$  conditions. Further annealing of the sample at room temperature would require stabilizing the sample (Fig. 2a). Black stars are calculations from ref. 10. Dark yellow points are  $T_c$  values of pure sulfur obtained with the same four-probe electrical measurement method. They are consistent with literature data<sup>30</sup> (susceptibility measurements) but have higher values at  $P > 200$  GPa.

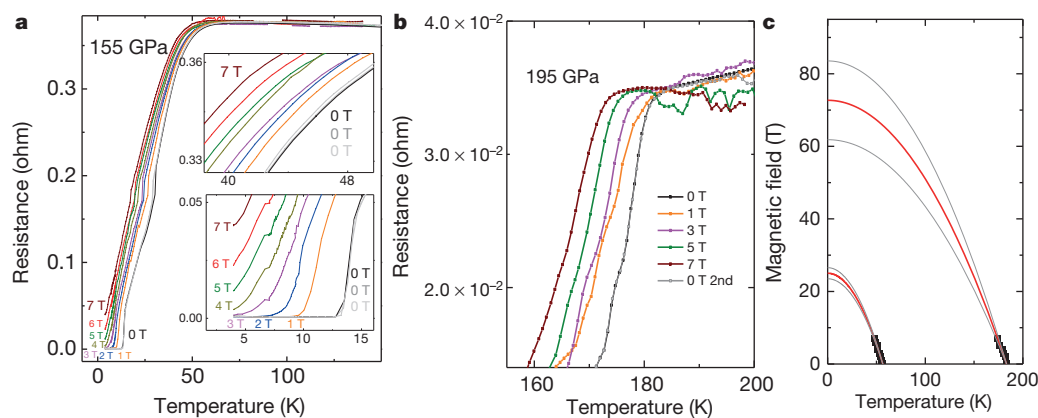
(1) There is a sharp drop in resistivity with cooling, indicating a phase transformation. The measured minimum resistance is at least as low,  $\sim 10^{-11}$  ohm m—about two orders of magnitude less than for pure copper (Fig. 1, Extended Data Fig. 3e) measured at the same temperature<sup>19</sup>. (2) A strong isotope effect is observed:  $T_c$  shifts to lower

temperatures for sulfur deuteride, indicating phonon-assisted superconductivity (Fig. 2b, c). The BCS theory gives the dependence of  $T_c$  on atomic mass  $m$  as  $T_c \propto m^{-\alpha}$ , where  $\alpha \approx 0.5$ . Comparison of  $T_c$  values in the pressure range  $P > 170$  GPa (Fig. 2c) gives  $\alpha \approx 0.3$ . (3)  $T_c$  shifts to lower temperatures with available magnetic field ( $B$ ) up to 7 T



**Figure 2 | Pressure and temperature effects on  $T_c$  of sulfur hydride and sulfur deuteride.** **a**, Changes of resistance and  $T_c$  of sulfur hydride with temperature at constant pressure—the annealing process. The sample was pressurized to 145 GPa at 220 K and then cooled to 100 K. It was then slowly warmed at  $\sim 1$  K  $\text{min}^{-1}$ ;  $T_c = 170$  K was determined. At temperatures above  $\sim 250$  K the resistance dropped sharply, and during the next temperature run  $T_c$  increased to  $\sim 195$  K. This  $T_c$  remained nearly the same for the next two runs. (We note that the only point for sulfur deuteride presented in ref. 9 was determined without sample annealing, and  $T_c$  would increase after annealing at room temperature.) **b**, Typical superconductive steps for sulfur hydride

(blue trace) and sulfur deuteride (red trace). The data were acquired during slow warming over a time of several hours.  $T_c$  is defined here as the sharp kink in the transition to normal metallic behaviour. These curves were obtained after annealing at room temperature as shown in **a**. **c**, Dependence of  $T_c$  on pressure; data on annealed samples are presented. Open coloured points refer to sulfur deuteride, and filled points to sulfur hydride. Data shown as the magenta point were obtained in magnetic susceptibility measurements (Fig. 4a). The lines indicate that the plots are parallel at pressures above  $\sim 170$  GPa (the isotope shift is constant) but strongly deviate at lower pressures.



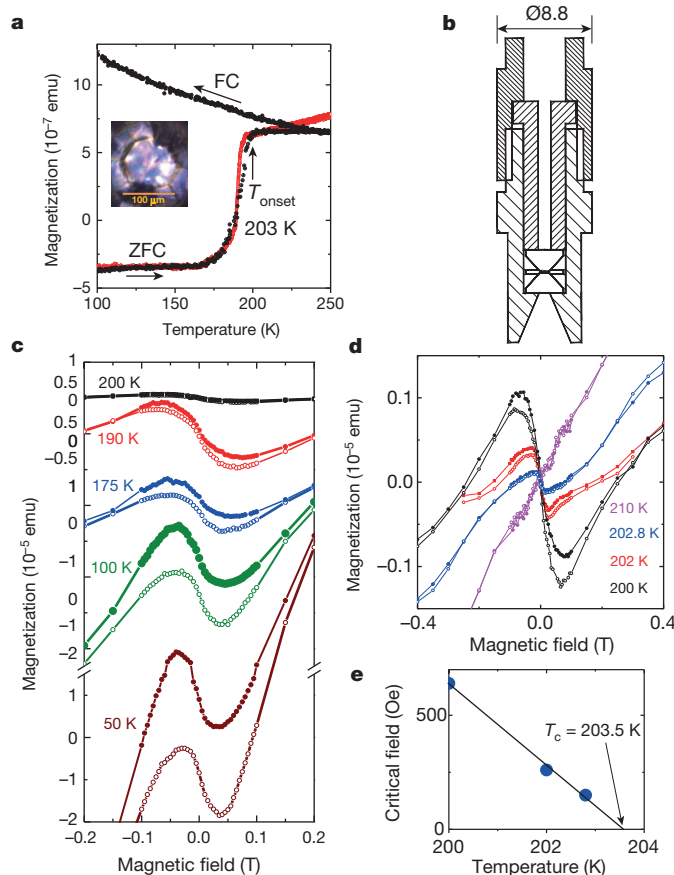
**Figure 3 | Temperature dependence of the resistance of sulfur hydride in different magnetic fields.** **a**, The shift of the  $\sim 60$  K superconducting transition in magnetic fields of 0–7 T (colour coded). The upper and lower parts of the transition are shown enlarged in the insets (axes as in main panel). The temperature dependence of the resistance without an applied magnetic field was measured three times: before applying the field, after applying 1, 3, 5, 7 T and finally after applying 2, 4, 6 T (black, grey and dark grey colours). **b**, The

same measurements but for the 185 K superconducting transition. **c**, The temperature dependence of the critical magnetic field strengths of sulfur hydride.  $T_c$  (black points deduced from **a**, **b**) are plotted for the corresponding magnetic fields. To estimate the critical magnetic field  $H_c$ , the plots were extrapolated to high magnetic fields using the formula  $H_c(T) = H_{c0}(1 - (T/T_c)^2)$ . The extrapolation has been done with 95% confidence (band shown as grey lines).

(Fig. 3). Much higher fields are required to destroy the superconductivity: extrapolation of  $T_c(B)$  gives an estimate of a critical magnetic field as high as 70 T (Fig. 3). (4) Finally, in magnetic susceptibility measurements (Fig. 4) a sharp transition from the diamagnetic to the paramagnetic state (Fig. 4a) was observed for zero-field-cooled (ZFC) material. The onset temperature of the superconducting state  $T_{\text{onset}} = 203(1)$  K, and the width of the superconducting transition is nearly the same as in electrical measurements (Fig. 4a). Magnetization measurements  $M(H)$ , where  $H$  is magnetic field, at different

temperatures (Fig. 4c) revealed a pronounced hysteresis indicating type II superconductivity with the first critical field  $H_{c1} \approx 30$  mT. The magnetization decreases sharply at temperatures above 200 K showing the onset of superconductivity at 203.5 K, in agreement with the susceptibility measurements (Fig. 4a). A list of key properties of the new superconductor is given in Methods.

We have presented purely experimental evidence of superconductivity in sulfur hydride. However the particular compound responsible for the high  $T_c$  is not obvious. The superconductivity measured in the



**Figure 4 | Magnetization measurements.** **a**, Temperature dependence of the magnetization of sulfur hydride at a pressure of 155 GPa in zero-field cooled (ZFC) and 20 Oe field cooled (FC) modes (black circles). The onset temperature is  $T_{\text{onset}} = 203(1)$  K. For comparison, the superconducting step obtained for sulfur hydride from electrical measurements at 145 GPa is shown by red circles. Resistivity data ( $T_{\text{onset}} = 195$  K) were scaled and moved vertically to compare with the magnetization data. Inset, optical micrograph of a sulfur hydride sample at 155 GPa in a  $\text{CaSO}_4$  gasket (scale bar 100  $\mu\text{m}$ ). The high  $T_{\text{onset}} = 203$  K measured from the susceptibility can be explained by a significant input to the signal from the periphery of the sample which expanded beyond the culet where pressure is smaller than in the culet centre ( $T_c$  increases with decreasing pressure (Fig. 2b)). **b**, Non-magnetic diamond anvil cell (DAC) of diameter 8.8 mm. **c**, Magnetization measurements  $M(H)$  of sulfur hydride at a pressure of 155 GPa at different temperatures (given as curve labels). The magnetization curves show hysteresis, indicating a type II superconductor. The magnetization curves are however distorted by obvious paramagnetic input (which is also observed in other superconductors<sup>31</sup>). In our case, the paramagnetic signal is probably from the DAC, but further study of the origin of this input is required. The paramagnetic background increases when temperature is decreased. The minima of the magnetization curves ( $\sim 35$  mT) are the result of the diamagnetic input from superconductivity and the paramagnetic background. The first critical field  $H_{c1} \approx 30$  mT can be roughly estimated as the point where magnetization deviates from linear behaviour. At higher fields, magnetization increases due to the penetration of magnetic vortices. As the sign of the field change reverses, the magnetic flux in the Shubnikov phase remains trapped and therefore the back run (that is, with decreasing field) is irreversible—the returning branch of the magnetic cycle (shown by filled points) runs above the direct one. Hysteretic behaviour of the magnetization becomes more clearly visible as the temperature decreases. **d**, At high temperatures  $T > 200$  K, the magnetization decreases sharply. **e**, Extrapolation of the pronounced minima at the magnetization curves to higher temperatures gives the onset of superconductivity at  $T = 203.5$  K.

low-temperature runs (Fig. 1) possibly relates to  $\text{H}_2\text{S}$ , as it is generally consistent with calculations<sup>10</sup> for  $\text{H}_2\text{S}$ : both the value of  $T_c \approx 80$  K and its pressure behaviour. However superconductivity with  $T_c \approx 200$  K (Fig. 2) does not follow from these calculations. We suppose that it relates to the decomposition of  $\text{H}_2\text{S}$ , as high temperatures are required to reach the high  $T_c$  (Fig. 2b). Precipitation of elemental sulfur on decomposition could be expected (which is well known at low pressures of  $P < 100$  GPa; ref. 14); however the superconducting transition in elemental sulfur occurs at significantly lower temperatures (Fig. 1b). Another expected product of decomposition of  $\text{H}_2\text{S}$  is hydrogen. However, the strong characteristic vibrational stretching mode from the  $\text{H}_2$  molecule was never observed in our Raman spectra (nor was it observed in ref. 14). Therefore we suppose that the dissociation of  $\text{H}_2\text{S}$  is different and involves the creation of higher hydrides, such as  $3\text{H}_2\text{S} \rightarrow \text{H}_6\text{S} + 2\text{S}$  or  $2\text{H}_2\text{S} \rightarrow \text{H}_4\text{S} + \text{S}$ . It is natural to expect these reactions, as sulfur can be not only divalent, but also exhibits higher valencies. In fact, calculations<sup>10</sup> indirectly support this hypothesis, as the dissociation  $\text{H}_2\text{S} \rightarrow \text{H}_2 + \text{S}$  was shown to be energetically very unfavourable. We found further theoretical support in ref. 20. In that work, the van der Waals compound<sup>21</sup>  $(\text{H}_2\text{S})_2\text{H}_2$  was considered, and it was shown that at pressures above 180 GPa it forms an  $Im\text{-}3m$  structure with  $\text{H}_3\text{S}$  stoichiometry. The predicted  $T_c \approx 190$  K and its pressure dependences are close to our experimental values (Fig. 2c). Our hypothesis of the transformation of  $\text{H}_2\text{S}$  to higher hydrides (in the  $\text{H}_3\text{S}$  stoichiometry each S atom is surrounded by 6 hydrogen atoms) is strongly supported by further calculations<sup>22,23</sup>. All the numerous works based on the  $Im\text{-}3m$  structure<sup>23–27</sup> are consistent in their prediction of  $T_c > 200$  K, which decreases with pressure. The hydrogen sublattice gives the main contribution to superconductivity<sup>20,25,26</sup>. Inclusion of zero point vibrations and anharmonicity in the calculations<sup>24</sup> corrected the calculated  $T_c$  to  $\sim 190$  K, and the isotope coefficient from  $\alpha = 0.5$  to  $\alpha = 0.35$ —both in agreement with the present work.

The highest  $T_c$  of 203 K that we report here has been achieved most probably in  $\text{H}_3\text{S}$  having the  $Im\text{-}3m$  structure. It is a good metal; interestingly, there is also strong covalent bonding between H and S atoms in this compound<sup>20</sup>. This is in agreement with the general assumption (see for instance ref. 28) that a metal with high  $T_c$  should have strong covalent bonding (as is realized in  $\text{MgB}_2$ ; ref. 29) together with high-frequency modes in the phonon spectrum. This particular combination of bonding type and phonon spectrum would probably provide a good criterion when searching for the materials with high  $T_c$  at ambient pressure that are required for applications. There are many hydrogen-containing materials with strong covalent bonding (such as organics) but typically they are insulators. In principle, they could be tuned to a metallic state by doping or gating. Modern methods of structure prediction could facilitate exploration for the desired materials.

**Online Content** Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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**Author Contributions** A.P.D. performed the most of the experiments and contributed to the data interpretation and writing the manuscript. M.I.E. designed the study, wrote the major part of the manuscript, developed the DAC for SQUID measurements, and participated in the experiments. I.A.T. participated in experiments. V.K. and S.I.S. performed the magnetic susceptibility measurements and contributed to writing the manuscript. M.I.E. and A.P.D. contributed equally to this paper.

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## METHODS

**Experimental procedure.** For electrical measurements we used diamond anvils (DACs) with anvils of the following shape: tip diameter of 200–300  $\mu\text{m}$  bevelled at  $7\text{--}8^\circ$  to a culet of 40–80  $\mu\text{m}$ . An insulating gasket is required to separate the metallic gasket from the electrodes. It was prepared in the following way (Extended Data Fig. 3). First a metallic gasket of T301 stainless steel (or Re) 250  $\mu\text{m}$  thick was indented with about 17–20 GPa pressure. Then the bottom of the imprint of diameter  $\sim 200\text{ }\mu\text{m}$  was drilled out, and a powder insulating material was put in the imprint and pressed between the anvils to form a layer. The insulating layer was made of either Teflon, NaCl or  $\text{CaSO}_4$  as these materials do not react with  $\text{H}_2\text{S}$ . The layer was pressed to obtain a thickness in the centre of  $\sim 3\text{--}5\text{ }\mu\text{m}$  to provide stable clamping. A larger thickness leads to instability in the sample—it shifts or escapes under pressure—while with a thinner gasket it is difficult to reach high pressures. A hole of diameter  $\sim 10\text{--}30\text{ }\mu\text{m}$  was then drilled in the insulating layer. Four Ti electrodes were sputtered on the diamond anvil. The electrodes were capped with Au to prevent oxidation of the Ti. (To check a possible contribution of the diamond surface to the conductivity, we prepared a different configuration of electrodes for a once-only experiment: two electrodes were sputtered on one anvil and another two on another anvil, similar to ref. 13). After preparation of the electrodes the gasket was put back on the anvil and the DAC was assembled so that the separation between the anvils was about 20–100  $\mu\text{m}$  (measured by interference fringes). The DAC was placed into a cryostat and cooled down to  $\sim 200\text{ K}$  (within the temperature range of liquid  $\text{H}_2\text{S}$ ) and then  $\text{H}_2\text{S}$  gas was put through a capillary into a rim around the diamond anvil where it liquefied (Extended Data Fig. 4).  $\text{H}_2\text{S}$  of 99.5% and  $\text{D}_2\text{S}$  of 97% purity were been used. The filling was monitored visually (Extended Data Figs 4, 5) and the sample was identified by measuring Raman spectra. Then liquid  $\text{H}_2\text{S}$  was clamped in the gasket hole by pushing the piston of the DAC with the aid of screws outside the cryostat. The thickness of the sample can be estimated to be few micrometres, as measured from interference spectra through the clamped transparent sample. The thickness might be  $\sim 1\text{ }\mu\text{m}$  if the sample expanded over the culet (Fig. 4). After the clamping, the DAC was heated to  $\sim 220\text{ K}$  to evaporate the rest of the  $\text{H}_2\text{S}$ , and then the pressure was further increased at this temperature. The pressure remained stable during the cooling within  $\pm 5\text{ GPa}$ . The pressure was determined by a diamond edge scale at room temperature and low temperatures<sup>32</sup>. For optical measurements a Raman spectrometer was equipped with a nitrogen-cooled CCD and notch filters. The 632.8 nm line of a He–Ne laser was used to excite the Raman spectra and to determine pressure.

The low temperature loading seems to be required to prepare samples with high  $T_c$ . If  $\text{H}_2\text{S}$  was loaded at room temperature in the gas loader, for example, only sulfur was detected in Raman and X-ray scattering. Apparently in this route the sample decomposes before reaching the required high-pressure phase of  $\text{H}_3\text{S}$ . We did not explore all ( $P, T$ ) paths to reach the state with high  $T_c$ . We found however that superconductivity is not observed in sample loaded at  $\sim 200\text{ K}$  but heated to room temperature at low pressure  $< \sim 100\text{ GPa}$ .

The resistance and Raman spectra were measured during the pressurizing using the four-probe van der Pauw method (Extended Data Fig. 3) with a current of 10–10,000  $\mu\text{A}$ . The temperature was reliably determined by using a slow warming rate ( $\sim 1\text{ K min}^{-1}$ ) and allowing the DAC to equilibrate with attached thermometer. The determined  $T_c$  was well reproduced in measurements with the PPMS6000 (Physical Property Measurement System from Quantum Design)

and other set-ups.  $T_c$  was determined as the point of steepest change of resistance from the normal state (Fig. 2b).

The influence of the magnetic field on superconducting transitions has been measured with a non-magnetic DAC (diameter 25 mm) in a PPMS6000 in a 4–300 K temperature range and fields up to 7 T.

Magnetic susceptibility measurements were performed in an MPMS (Magnetic Property Measurement System) from Quantum Design. For these measurements a miniature non-magnetic cell made of Cu:Ti alloy working up to 200 GPa was designed (Fig. 4b). Samples of diameter  $\sim 50\text{--}100\text{ }\mu\text{m}$  and a thickness of a few micrometres were prepared to provide a sufficient signal. Magnetic susceptibility measurements using a high-pressure cell were performed using a background subtraction feature of the MPMS software of the SQUID magnetometer (Extended Data Fig. 6).

**Results.** We present here some important key features of our new high- $T_c$  sulfur hydride superconductor:

(1) The new superconductor is of type II. This fact is clearly supported by (i) a difference in temperature-dependent ZFC and FC magnetization (Fig. 4a), which is due to the Meissner effect (ZFC) and magnetic flux capture when the sample is cooled down from its normal state (FC); and (ii) the magnetic hysteresis curves (Fig. 4c, d). The magnetic hysteresis curves also have all the features of typical type II superconductors with a mixed state between  $H_{c1}$  and  $H_{c2}$ .

(2) A typical value of the coherence length  $\xi_{\text{GL}}$  in the framework of the Ginzburg–Landau theory can be estimated on the basis of the measured upper critical fields from conductivity measurements (Fig. 3c). Using the experimental estimation  $60\text{ T} < H_{c2} < 80\text{ T}$  and the relation

$$\xi_{\text{GL}} = \frac{1}{2} \sqrt{\frac{\hbar}{\pi e H_{c2}}}$$

we find limits for the coherence length:  $2.3\text{ nm} > \xi_{\text{GL}} > 2.0\text{ nm}$ . We note that this relatively short coherence length is of the same order as, for instance, the values for superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (1.3 nm) and  $\text{Nb}_3\text{Sn}$  (3.5 nm).

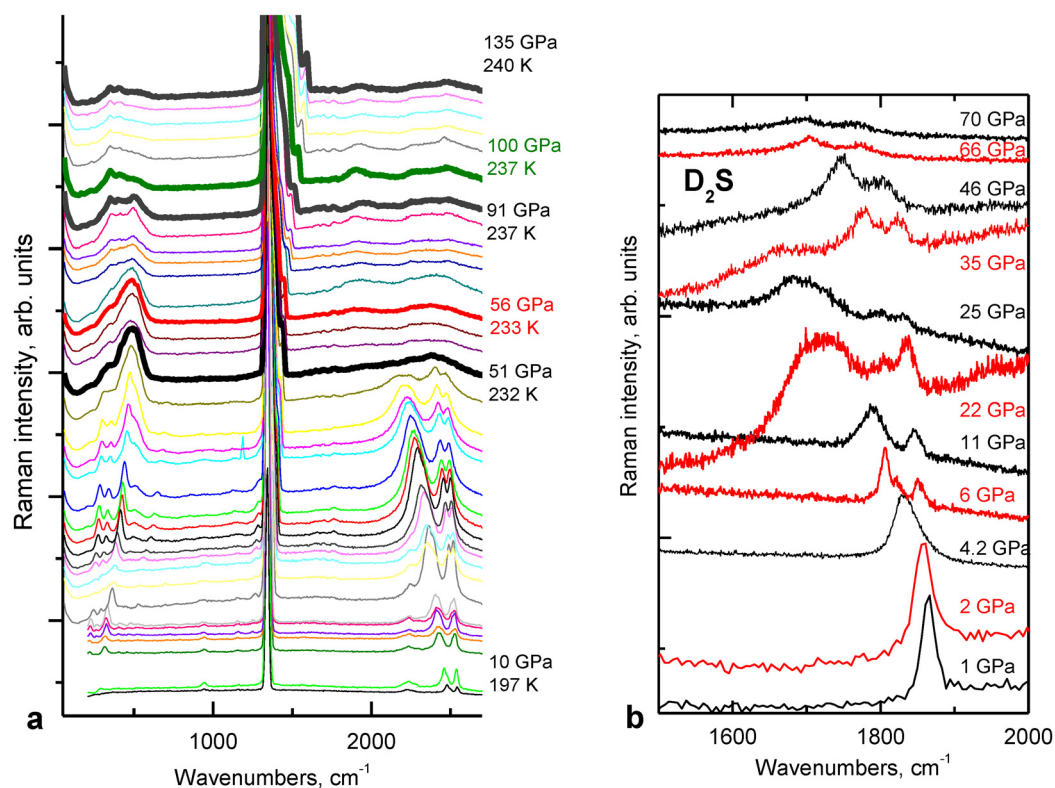
(3) The London penetration depth  $\lambda_L$  can be estimated from the known relation of the lower critical field  $H_{c1}$  to the upper critical field  $H_{c2}$  for a type II superconductor

$$\frac{H_{c1}}{H_{c2}} \approx \frac{\ln \kappa}{2\sqrt{2}\kappa^2}$$

in the limit  $\kappa \gg 1$  of the Ginzburg–Landau parameter  $\kappa = \frac{\lambda_L}{\xi_{\text{GL}}}$ . Considering the experimental value of the first critical field of  $3 \times 10^{-2}\text{ T}$  (Fig. 4c) and the above-mentioned relation  $60\text{ T} < H_{c2} < 80\text{ T}$ , we can obtain the following estimate for the London penetration depth:  $\lambda_L \approx 125\text{ nm}$ .

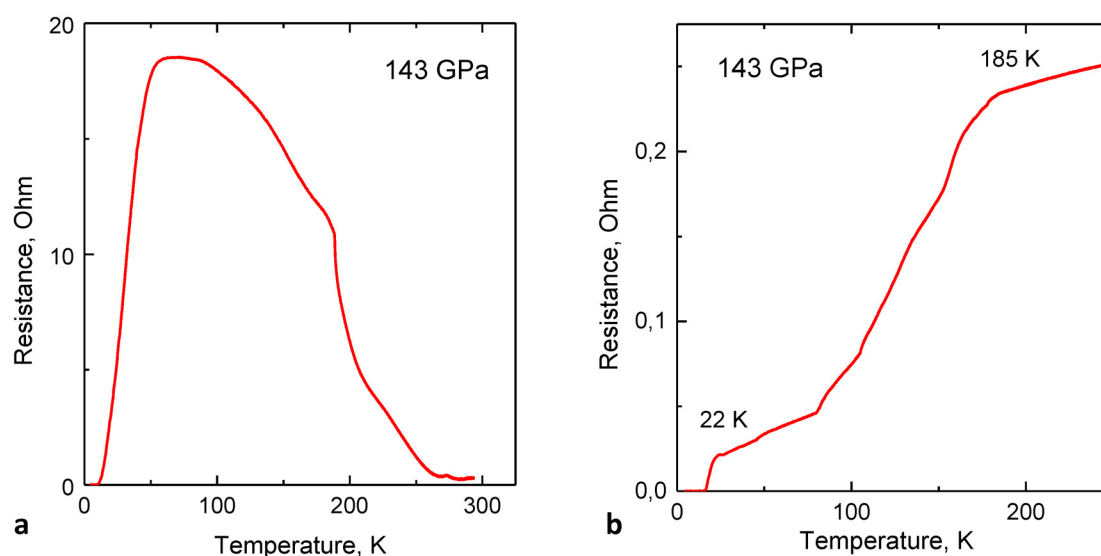
(4) According to Bean's model, the magnetic critical current density of the superconductor can be estimated from the distance between the direct and the returning branches of the magnetic hysteresis loop at a given magnetic field (Fig. 4c). Provided grain radii are about 0.1  $\mu\text{m}$ , the intra-grain critical current  $J_c$  is about  $10^7\text{ A cm}^{-2}$ .

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33. Landau, L. D. & Lifshitz, E. M. *Electrodynamics of Continuous Media* Vol. 8, 1st edn, 173 (Pergamon, 1960).



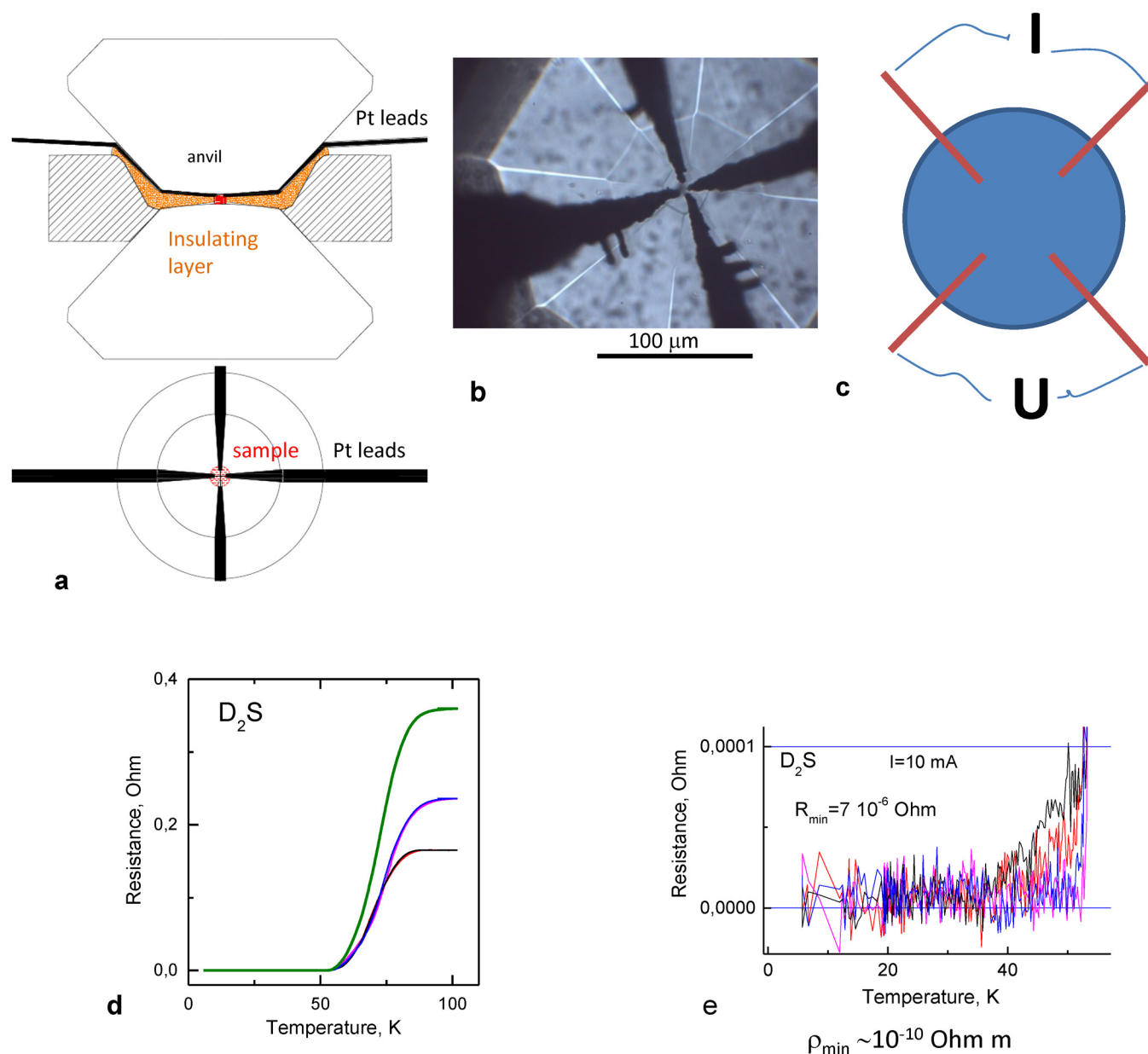
**Extended Data Figure 1 | Raman spectra of sulfur hydride at different pressures.** **a**, Spectra of sulfur hydride at increasing pressure at  $\sim 230$  K. The spectra are shifted relative to each other. At 51 GPa there is a phase transformation, as follows from disappearance of the characteristic vibron peaks in the  $2,100\text{--}2,500\text{ cm}^{-1}$  range. The corresponding spectrum is

highlighted as a bold curve. Bold curves at higher pressure (and the temperature of the measurement) are shown to follow qualitatively the changes of the spectra. The pressure corresponding to the unassigned plots can be determined from the Raman spectra of the stressed diamond anvil<sup>32</sup>. **b**, Raman spectra of sulfur deuteride measured at  $T \approx 170$  K and over the pressure range 1–70 GPa.



**Extended Data Figure 2 | Temperature dependence of the resistance of sulfur hydride at 143 GPa.** In this run the sample was clamped in the DAC at  $T \approx 200$  K, and the pressure then increased to 103 GPa at this temperature; the further increase of pressure to 143 GPa was at  $\sim 100$  K. **a**, After next cooling to  $\sim 15$  K and subsequent warming, a superconducting transition with  $T_c \approx 60$  K was observed, then the resistance strongly decreased with increasing temperature. After successive cooling and warming (**b**; only the warming curve

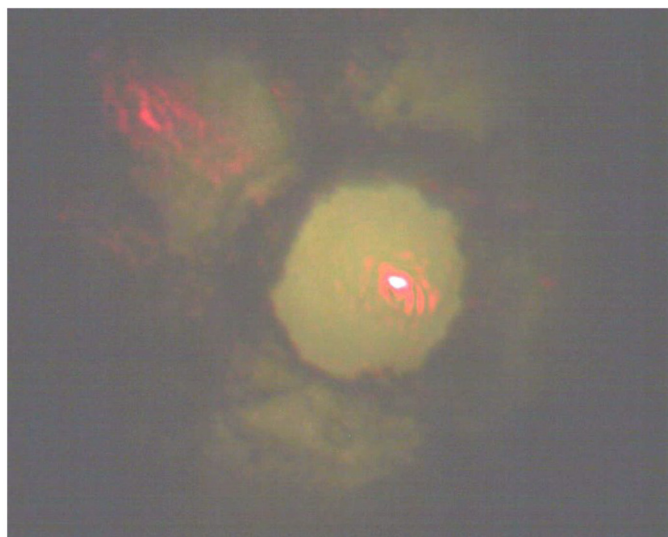
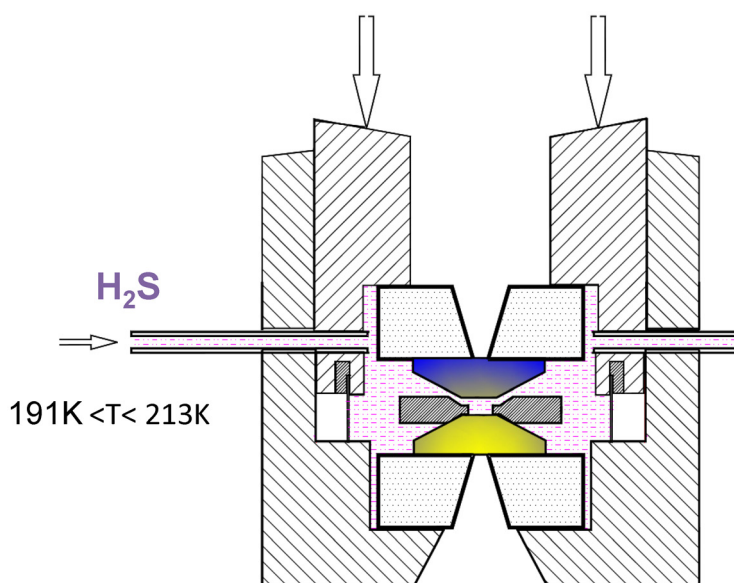
is shown) a kink at 185 K appeared, indicating the onset of superconductivity. The superconducting transition is very broad: resistance dropped to zero only at  $\sim 22$  K. There are apparent 'oscillations' on the slope. Their origin is not clear, though they probably reflect inhomogeneity of the sample in the transient state before complete annealing. Similar 'oscillations' have also been observed for other samples (see, for example, figure 3 in the Supplementary Information of ref. 9).



**Extended Data Figure 3 | Electrical measurements.** **a**, Schematic drawing of diamond anvils with electrical leads separated from the metallic gasket by an insulating layer (shown orange). **b**, Ti electrodes sputtered on a diamond anvil shown in transmitted light. **c**, Scheme of the van der Pauw measurements: current leads are indicated by  $I$ , and voltage leads as  $U$ . **d**, Typical superconducting step measured in four channels (for different combinations of

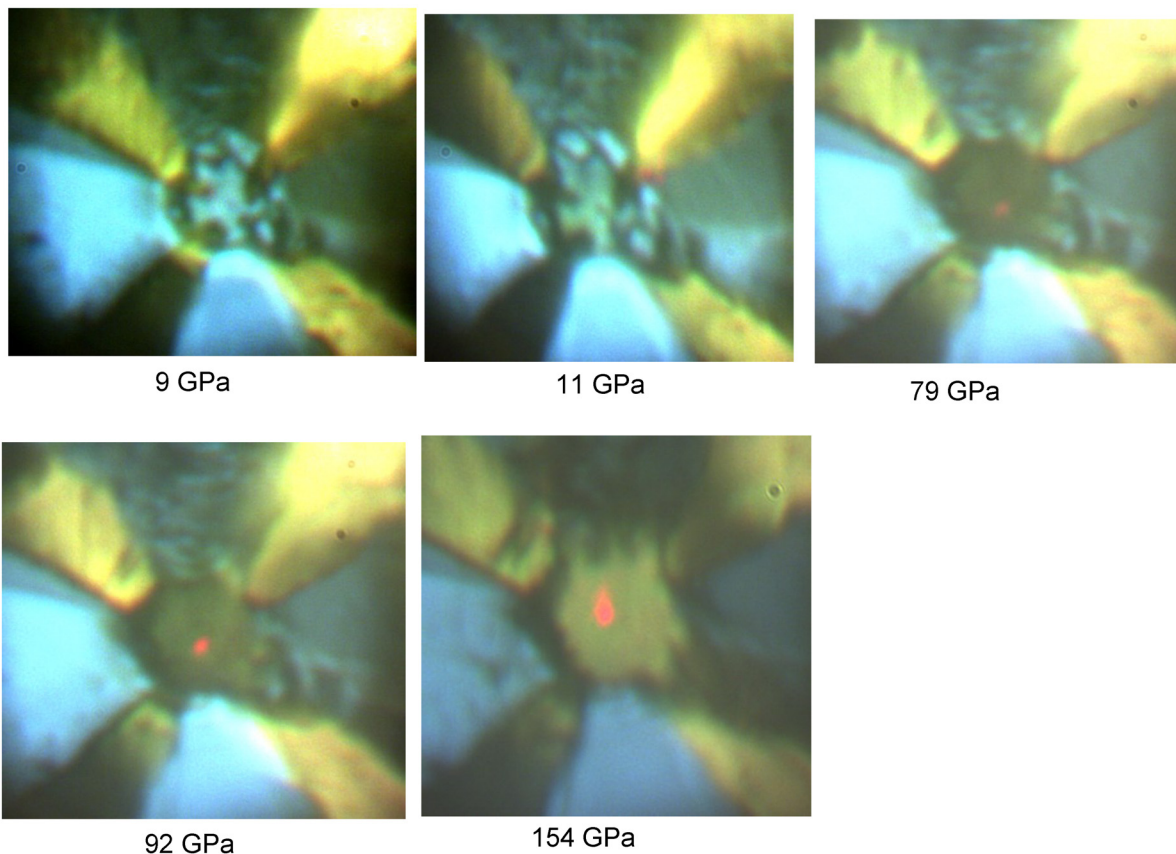
current and voltage leads shown in **c**). A sum resistance obtained from the van der Pauw formula is shown by the green line. Note here that the superconducting transition was measured with the un-annealed sample<sup>9</sup>. After warming to room temperature and successive cooling,  $T_c$  should increase. **e**, Residual resistance measured below the superconducting transition (**d**).  $R_{\min}$  and  $\rho_{\min}$  are averaged over four channels shown by different colours.





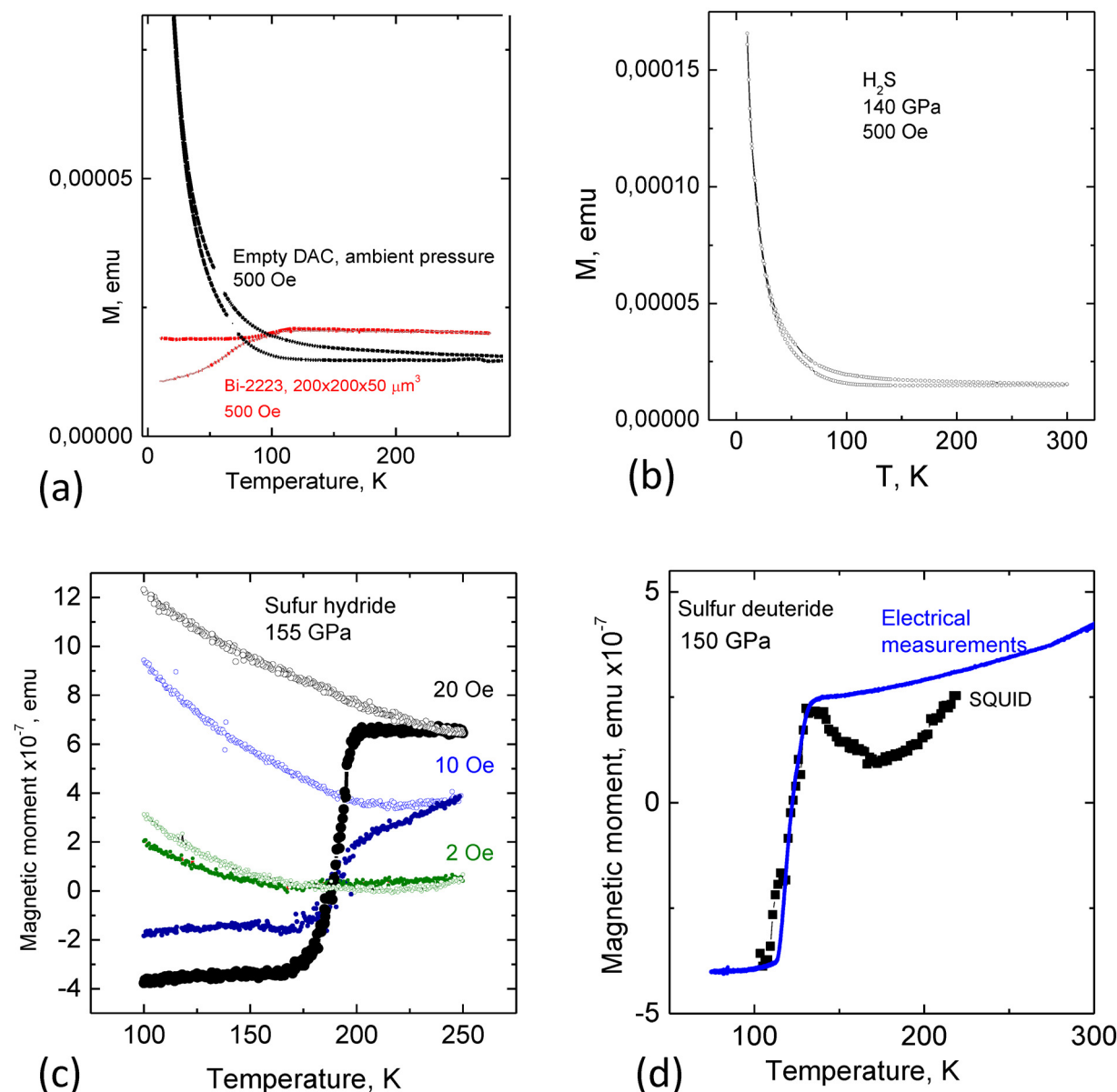
**Extended Data Figure 4 | Loading of H<sub>2</sub>S.** Gaseous H<sub>2</sub>S is passed through the capillary into a rim around the diamond anvils (upper panel). When the sample liquefies, in the temperature range 191 K < *T* < 213 K, it is clamped. The process of loading is shown on a video (<https://vimeo.com/131914556>) and a still is shown here (lower panel). On the video, the camera is looking through a hole in the transparent gasket (CaSO<sub>4</sub>), and shows a view through the

diamond anvil. At *T* ≈ 200 K, the line to the H<sub>2</sub>S gas cylinder was opened and the gas condensed. At this moment, the picture changes due to the different refractive index of H<sub>2</sub>S. The second anvil with the sputtered electrodes was then pushed forward, and the hole was clamped. The sample changed colour during the next application of pressure. The red point is from the focused HeNe laser beam.



**Extended Data Figure 5 | View of  $D_2S$  sample with electrical leads and transparent gasket ( $CaSO_4$ ) at different pressures.** The  $D_2S$  is in the centre of these photographs, which were taken in a cryostat at 220 K with mixed illumination, both transmitted and reflected. Under this illumination, the

insulating transparent gasket shows blue, and the electrodes yellow. The red spot is the focused HeNe laser beam. The sample, which is initially transparent, becomes opaque and then reflective as pressure is increased.



**Extended Data Figure 6 | Magnetic susceptibility measurements with a SQUID.** A typical sample (Fig. 4) has a disk shape (diameter 50–100  $\mu\text{m}$  and thickness of few micrometres). In the superconductive state the magnetic moment for this disk is estimated as  $M(\text{disk}) \approx 0.2r^3H$  (ref. 33). For a disk of radius  $r = 40 \mu\text{m}$  (a sample size typical for DACs in the megabar range) and  $H = 2 \text{ mT}$  the expected diamagnetic signal,  $M(\text{disk})$  is estimated as  $2.6 \times 10^{-7} \text{ emu}$ . This value is well above the sensitivity of the SQUID which is  $\sim 10^{-8} \text{ emu}$  and, therefore, the signal can be detected. A high-pressure DAC made of Cu:Ti alloy has its own magnetic background signal (a) which increases sharply at low temperatures due to residual paramagnetic impurities. Signal from a large superconducting sample (for example, a Bi-2223 superconductor)

could still be detected without magnetic background subtraction. However, the sulfur hydride sample is not seen (b) unless background has been subtracted (c, d). The background signal acquired in the normal state immediately above  $T_{\text{onset}}$  has been used for subtraction over all the temperature range taking into account that the magnetic moment of the DAC is fairly temperature independent above 100 K. c, Magnetic measurements for the sample of sulfur hydride at different magnetic fields (labels on curves). The data on sulfur deuteride (d) are compared with the superconducting transition in resistivity measurements (blue curve) which has been scaled to fit the susceptibility data (black points).