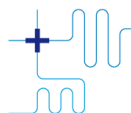


RESEARCH ARTICLE | APRIL 20 2016

**Superconductivity enhancement in the S-doped Weyl semimetal candidate  $\text{MoTe}_2$** F. C. Chen ; X. Luo; R. C. Xiao; W. J. Lu; B. Zhang; H. X. Yang; J. Q. Li; Q. L. Pei; D. F. Shao ; R. R. Zhang; L. S. Ling; C. Y. Xi; W. H. Song; Y. P. Sun*Appl. Phys. Lett.* 108, 162601 (2016)<https://doi.org/10.1063/1.4947433>**Articles You May Be Interested In**Origin of planar Hall effect in type-II Weyl semimetal  $\text{MoTe}_2$ *AIP Advances* (May 2019)Drastic enhancement of the superconducting temperature in type-II Weyl semimetal candidate  $\text{MoTe}_2$  via biaxial strain*APL Mater.* (February 2023)Anisotropic Shubnikov-de Haas effect in topological Weyl semimetal  $\text{MoTe}_2$ *Appl. Phys. Lett.* (December 2021)**Boost Your Lab's Performance  
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# Superconductivity enhancement in the S-doped Weyl semimetal candidate $\text{MoTe}_2$

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(Received 22 January 2016; accepted 11 April 2016; published online 20 April 2016)

Two-dimensional transition-metal dichalcogenide (TMDs)  $\text{MoTe}_2$  has attracted much attention due to its predicted Weyl semimetal state and a quantum spin Hall insulator in bulk and monolayer form, respectively. We find that the superconductivity in  $\text{MoTe}_2$  single crystal can be greatly enhanced by the partial substitution of the Te ions by the S ones. The maximum superconducting temperature  $T_C$  of  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal is about 1.3 K. Compared with the parent  $\text{MoTe}_2$  single crystal ( $T_C = 0.1$  K), nearly 13-fold in  $T_C$  is improved in the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  one. The superconductivity has been investigated through the resistivity and magnetization measurements.  $\text{MoTe}_{2-x}\text{S}_x$  single crystals belong to weak coupling superconductors and the improvement of the superconductivity may be related to the enhanced electron-phonon coupling induced by the S-ion substitution. A dome-shaped superconducting phase diagram is obtained in the S-doped  $\text{MoTe}_2$  single crystals.  $\text{MoTe}_{2-x}\text{S}_x$  materials may provide a new platform for our understanding of superconductivity phenomena and topological physics in TMDs. *Published by AIP Publishing.*  
[\[http://dx.doi.org/10.1063/1.4947433\]](http://dx.doi.org/10.1063/1.4947433)

The layered transition-metal dichalcogenides (TMDs) materials, named  $\text{MX}_2$ , where M is a transition metal (Ta, Mo, and W) and X is a chalcogen (S, Se, and Te), have attracted continued interest owing to their rich physical properties and promising potential applications.<sup>1–10</sup> For example, artificial van der Waals heterostructures with high on/off current ratio in TMDs are promising as an active channel for next-generation devices.<sup>2</sup> Topological field-effect transistors based on quantum spin Hall (QSH) insulators are another type of device.<sup>9</sup> At the same time, charge-density waves and superconductivity have also been observed in TMDs.<sup>10</sup> Particularly, a dome-shaped superconducting phase diagram is observed in a gate-tuned  $\text{MoS}_2$  device, and a pressure-driven superconducting state was also observed in non-magnetic  $\text{WTe}_2$  TMD.<sup>4–6</sup>

Predicted as a new type-II Weyl semimetal (WSM) candidate,  $\text{MoTe}_2$  has recently attracted much attention. WSMs were classified into two categories: type-I Weyl semimetals and type-II Weyl semimetals. For the type-I Weyl semimetals, such as the TaAs class, a point like Fermi surface at type-I Weyl point (WP) was found. As for the type-II Weyl semimetals, WPs occur in the boundaries between the electron pocket and hole pocket.<sup>11–13</sup> As we know,  $\text{MoTe}_2$  crystalizes into three different phases: 2H, 1T', and  $T_d$  ones. The 2H phase shows a semiconducting behavior. On the other hand, the 1T' and  $T_d$  phases present semi-metallic and

exhibit pseudo-hexagonal layers with zig-zag metal chains. The  $T_d$  compound can be obtained by cooling the 1T' phase down to 245 K.<sup>8,14</sup> The  $T_d$  phase of  $\text{MoTe}_2$ , which is isostructural to  $\text{WTe}_2$ , is predicted to be a WSM candidate.<sup>7</sup> Very recently, the  $T_d$ - $\text{MoTe}_2$  is reported to be a superconductivity with  $T_C = 0.1$  K. The small applied pressure can dramatically enhance the  $T_C$ , and a dome-shaped superconducting phase diagram under the applied pressure is observed.<sup>8</sup> The  $\text{MoTe}_2$  thus opens a door to the study of the interaction of topological physics and superconductivity in the bulk materials. The superconductivity can be greatly enhanced by the applied pressure in the  $\text{MoTe}_2$  bulk crystal, indicating that the  $T_d$ - $\text{MoTe}_2$  may be also sensitive to the chemical pressure induced by the ion substitution. Therefore, in this paper, we did the partial substitution of Te ions by S ones in the  $\text{MoTe}_2$  single crystal. The enhanced superconducting temperature  $T_C$  is then observed, and the maximum of the  $T_C$  in  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals is about 1.3 K. A dome-shaped superconducting phase diagram is obtained in  $\text{MoTe}_{2-x}\text{S}_x$  single crystals, and the possible reasons of the enhanced superconductivity will be discussed. The  $\text{MoTe}_{2-x}\text{S}_x$  materials may open a new avenue to understand the superconductivity phenomena and topological physics in TMDs.

$\text{MoTe}_2$  and  $\text{MoTe}_{2-x}\text{S}_x$  ( $0 \leq x \leq 1$ ) single crystals were grown by the chemical vapor transport method using polycrystalline  $\text{MoTe}_2$  and  $\text{MoTe}_{2-x}\text{S}_x$  as raw materials and  $\text{I}_2$  as a transport agent. Some experimental and calculated details can be found in the supplementary materials.<sup>21</sup> Electron

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diffraction and scanning transmission electron microscopy (STEM) experiments were performed in the JEOL ARM200F equipped with double aberration correctors and cold field emission gun operated at 200 kV.

MoTe<sub>2</sub> crystallizes into three different phases, as shown in Figs. 1(a)–1(c). In 2H phase, the Mo atom has trigonal prismatic coordination with the Te atoms. The 1T' phase (space group  $P2_1/m$ ), which is also called the  $\beta$ -phase, is a monoclinic lattice and is stable at room temperature. The T<sub>d</sub> phase (space group  $Pmn2_1$ ) is an orthorhombic lattice, and the 1T' one is a slight sliding of the layer-stacking of the T<sub>d</sub> phase. We selected the MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal as a typical

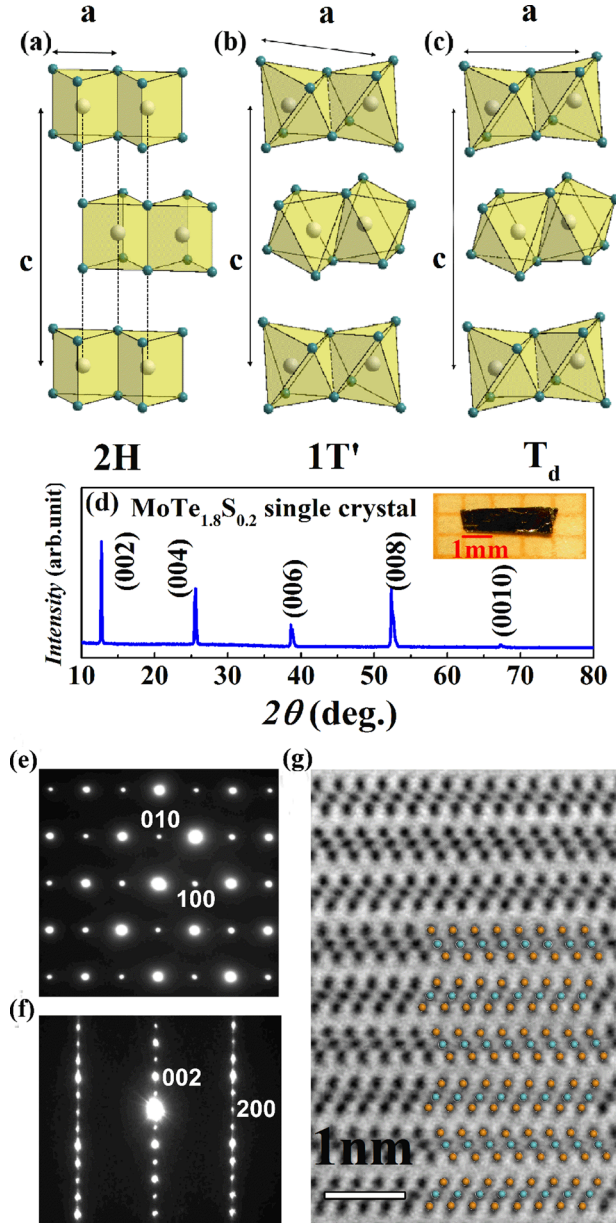


FIG. 1. (a)–(c) The crystalline structure of 2H, 1T', and T<sub>d</sub> structures of MoTe<sub>2</sub>; (d) XRD patterns of the MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal measured on the (001) surface. The inset presents the picture of the single crystal used for this study. The size is approximately  $3 \times 1 \times 0.5$ ; (e) and (f) the electron diffraction patterns of MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal taken along the [001] and [010] zone axis directions, respectively; (g) the ABF-STEM image of the MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal taken along the [010] zone direction, with the orange and cyan spheres representing the Te/S and Mo ions, respectively. The scale bar is 1 nm.

sample to perform further experiments. Figure 1(d) shows the X-ray patterns of MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystals along [001] direction. We performed the powder X-ray studies of the crushed MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystals. However, we found that the X-ray data can be well fitted based on the orthorhombic or monoclinic symmetry, as shown in Fig. S1.<sup>21</sup> It is hard for us to distinguish the structure of the MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal at the room temperature. We also carried out TEM characterization of the MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal. As shown in Fig. 1(e), the diffraction spots on the [001] selected area electron diffraction (SAED) pattern can be well indexed by the orthorhombic or monoclinic symmetry. However, the SAED pattern (Fig. 1(f)) and annular bright field (ABF) STEM image (Fig. 1(g)) taken along the [010] zone axis direction demonstrate evident structure difference compared with the previously reported orthorhombic or monoclinic structure. It is recognized that the MoTe(S)X<sub>6</sub> octahedral are much less distorted.

In order to compare the lattice parameters of MoTe<sub>2</sub> and MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystals, we also performed the XRD analysis of the crushed MoTe<sub>2</sub> single crystals. Figure S2 shows the result of the calculated XRD patterns. The calculated lattice parameters of the crushed MoTe<sub>2</sub> and MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystals and the reported MoTe<sub>2</sub> ones are summarized in Table SI of the supplementary material.<sup>21</sup> In addition, in order to get more structural information on the MoTe<sub>2-x</sub>S<sub>x</sub> single crystal, we also performed the Raman experiments. As shown in Fig. S3, except for the MoTeS single crystal (changing into 2H phase), all another crystals show similar Raman signals at room temperature. The temperature dependence on Raman spectra of the MoTe<sub>2</sub> single crystal is shown in Fig. S4. It seems that it is even more difficult to explain the phase transition from present Raman experiments. Thus, we still cannot give the information on the structure of the S-doped MoTe<sub>2</sub> crystals, and further experiments are needed in the future.<sup>21</sup>

Figure 2(a) shows the evolution of the resistance as a function of temperature  $\rho(T)$  in a MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal down to 0.35 K. For the MoTe<sub>2</sub> single crystal, we performed the measurement down to 2 K under the cooling and warming modes. As shown in Fig. 2(a), an anomaly with a hysteresis in the  $\rho(T)$  of MoTe<sub>2</sub> single crystals is observed, which is associated with the first-order structural phase transition from the 1T' phase to T<sub>d</sub> phase around 240 K and is consistent with the previous findings.<sup>8,14</sup> We applied the Fermi-liquid model  $\rho(T) = \rho_0 + A \times T^2$ , in which  $\rho_0$  and  $A$  are the residual resistivity and a constant, respectively, to the curve below 50 K, as shown in the left inset of Fig. 2(a). The model analysis yielded the residual resistivity  $\rho_0$  of  $1.36 \times 10^{-4} \Omega \text{ cm}$  and the coefficient  $A$  of  $0.00238 \mu\Omega \text{ cm}$ , indicating Fermi-liquid-like behavior for MoTe<sub>2</sub> single crystal. For the MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal, as shown in the right inset of Fig. 2(a), a clear zero-resistivity behavior is observed around 1.3 K. We measured the  $\rho(T)$  under the applied magnetic field  $H = 0.5 \text{ T}$ , and the zero-resistivity temperature moves to 0.7 K, indicating that superconductivity may occur in the MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal. On the other hand, we also find that there exists the minimum of the resistivity around  $T_{\min} = 8 \text{ K}$  before the superconducting transition in MoTe<sub>1.8</sub>S<sub>0.2</sub> single crystal, which is similar to the transport

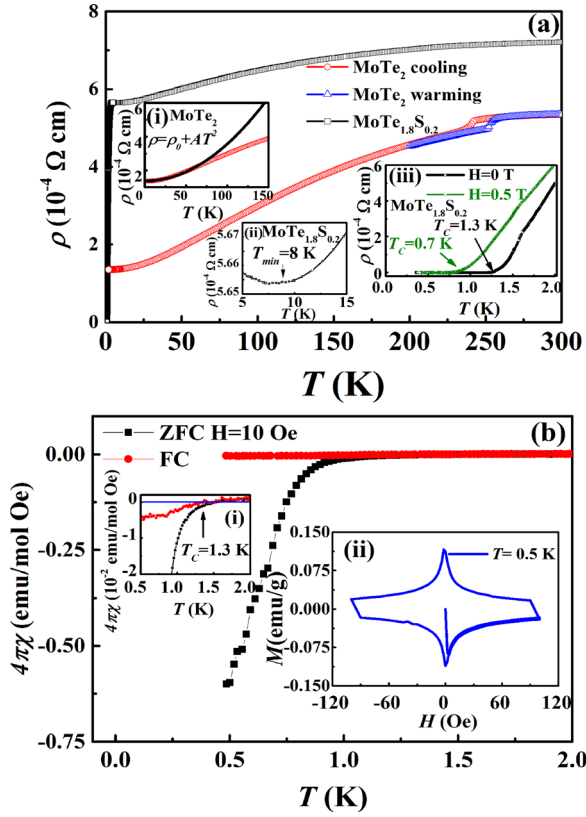


FIG. 2. (a) The temperature dependence of resistivity of the  $\text{MoTe}_2$  (warming and cooling modes) and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals. The left inset presents the fitting result according to the Fermi liquid theory. The middle inset shows the minimum of the resistivity  $\rho_{\min}$  with a larger scale. The right inset presents the temperature dependence of resistivity of the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal under  $H = 0 \text{ T}$  and  $0.5 \text{ T}$  at the low temperature. (b) The temperature dependence of magnetization of the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal with ZFC and FC modes under  $H = 10 \text{ Oe}$ . The left inset presents the temperature dependence of magnetization under a large scale around the  $T_C$ . The right inset shows the magnetic field dependence of magnetization at  $T = 0.5 \text{ K}$ .

properties of the  $\text{WTe}_2$  single crystal under the applied high pressure.<sup>5</sup> The presence of the minimum of resistivity may be related to the structural abnormalities induced by the substitution of Te ions by S ones in the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal. Figure 2(b) presents the temperature dependence of magnetization  $M(T)$  with the zero-field and field cooling (ZFC and FC) modes under an applied magnetic field of 10 Oe. The diamagnetic signal, in other words, the superconducting transition, can be obviously seen at  $T_C = 1.3 \text{ K}$  in the left inset of Fig. 2(b), which is in good agreement with the resistance measurements. The calculated shielding superconducting volume fraction at 0.5 K is about 60%, indicating the bulk superconducting behavior of the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal. The magnetization as a function of magnetic fields at 0.5 K is shown in the right inset of Fig. 2(b). The clear hysteresis is observed, which indicates a typical type-II superconducting behavior of  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals. Compared with the parent  $\text{MoTe}_2$  ( $T_C = 0.1 \text{ K}$ ), nearly 13-fold of the superconducting temperature is observed in the S doped  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal.

The temperature dependence of specific heat  $C_P$  also provides more information about the normal state properties. Figure 3 shows the temperature dependence of the heat capacity  $C_P$  of the  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals at

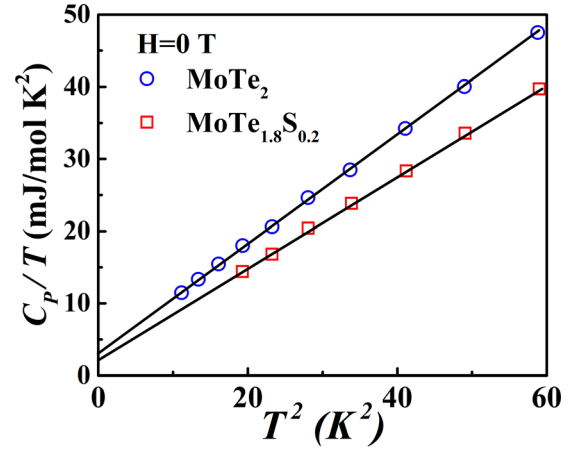


FIG. 3.  $T^2$  dependence of  $C_P/T$  of the  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  crystals under zero field. The solid lines show the heat capacity data fitting with the equation  $C_P/T = \gamma_N + \beta T^2$ .

$H = 0$  above  $T_C$ .  $C_P/T$  varies almost linearly with  $T^2$ . The Sommerfeld constant  $\gamma_N$  is obtained from the fit  $C_P/T = \gamma_N + \beta T^2$  ( $\gamma_N$  and  $\beta$  are  $T$ -independent coefficients), where  $\gamma_N$  is the normal-state electronic contribution and  $\beta$  is the lattice contribution to the specific heat. The fitting results yield  $\gamma_N = 3.06 \text{ mJ mol}^{-1} \text{ K}^{-2}$  and  $\beta = 0.758 \text{ mJ mol}^{-1} \text{ K}^{-4}$  for the  $\text{MoTe}_2$  single crystal and  $\gamma_N = 2.07 \text{ mJ mol}^{-1} \text{ K}^{-2}$  and  $\beta = 0.635 \text{ mJ mol}^{-1} \text{ K}^{-4}$  for the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  one, respectively. The resulting DOS at Fermi surface  $N(E_F)$  are 1.33 states/eV and 0.88 states/eV for the  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals, respectively. The Debye temperature  $\Theta_D$  can be determined from the coefficient of the  $T^2$  term  $\beta = N(12/5)\pi^4 R \Theta_D^{-3}$ , where  $R = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}$  and  $N = 3$  for the  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals. The Debye temperatures  $\Theta_D$  are 135 K and 143 K for the  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals, respectively. All the fitting results are summarized in Table I. An estimation of the strength of the electron-phonon coupling (EPC) can be derived from the McMillan formula<sup>15,16</sup>

$$\lambda_{ep} = \frac{\mu^* \ln \left( \frac{1.45 T_C}{\Theta_D} \right) - 1.04}{1.04 + \ln \left( \frac{1.45 T_C}{\Theta_D} \right) (1 - 0.62 \mu^*)}. \quad (1)$$

By assuming the Coulomb pseudopotential  $\mu^* = 0.1$ , the EPC constant  $\lambda_{ep}$  is estimated to be 0.32 and 0.49 for  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals, respectively. The calculated EPC show that both of  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals are weak coupling superconductors.<sup>15</sup> Because the  $N(E_F)$

TABLE I. The physical properties of  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystals.

| Parameters              | Units                               | $\text{MoTe}_2$ | $\text{MoTe}_{1.8}\text{S}_{0.2}$ |
|-------------------------|-------------------------------------|-----------------|-----------------------------------|
| $T_C$                   | K                                   | 0.1             | 1.3                               |
| $\gamma$                | $\text{mJ mol}^{-1} \text{ K}^{-2}$ | 3.06(1)         | 2.07(1)                           |
| $\beta$                 | $\text{mJ mol}^{-1} \text{ K}^{-4}$ | 0.758(1)        | 0.635(1)                          |
| $N(E_F) \text{ (exp.)}$ | States/eV                           | 1.3(1)          | 0.88(1)                           |
| $\Theta_D$              | K                                   | 135(1)          | 143(1)                            |
| $\lambda_{ep}$          |                                     | 0.32(1)         | 0.49(1)                           |

of the  $\text{MoTe}_2$  single crystal is larger than that of the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  one, the improvement of the superconductivity in the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal cannot be explained according to the variation of DOS of Fermi surface  $N(E_F)$  and it may be related to the enhancement of the EPC by the S-ion substitution.

In order to study the superconducting evolution with S-doping level, we also grew the  $\text{MoTe}_{2-x}\text{S}_x$  ( $0 \leq x \leq 1$ ) single crystals and studied its electrical transport properties. All the zero-resistivity temperature  $T_C$ , the minimum temperature of the resistivity  $T_{\min}$ , and the structural phase transition temperature  $T_S$  vs the doped S contents  $x$  are summarized in the  $T$ - $x$  phase diagram shown in Fig. 4. The structural phase transition temperature  $T_S$  is defined from the  $\rho(T)$  data and similar to that of the  $\text{MoTe}_2$  single crystal,<sup>14</sup> as shown in the inset of Fig. S5(a).<sup>21</sup> Note that the  $T_S$  anomaly slowly increases with the little S-doped content  $x$ , then decreases rapidly as  $x$  increases. And the  $T_S$  disappears at  $x = 0.18$ . For the superconducting temperature  $T_C$ , it increases nearly linearly up to a maximum  $T_C = 1.3$  at  $x = 0.2$ , as shown in Fig. S6.<sup>21</sup> From this maximum temperature, the  $T_C$  begins to decrease as  $x$  increase, with the transition eventually disappearing at  $x = 0.37$ . Thus, a dome-shaped superconducting phase diagram is obtained for the  $\text{MoTe}_{2-x}\text{S}_x$  single crystal. For the S doped  $\text{MoTe}_{2-x}\text{S}_x$  crystals, the lower S-doping at Te sites can sharply increase in  $T_C$ , which is concomitant with the enhancement of structural transition. Then, the  $T_C$  still increases slowly; however, the  $T_S$  decreases and disappears eventually as  $x$  increases, which is similar to the results from the applied pressure in the  $\text{MoTe}_2$  single crystal.<sup>8</sup> On the other hand, when the content  $x$  is larger than 0.18, which is in accord with the content where the structural phase disappears, the minimum of resistivity  $T_{\min}$  emerges and increases slowly with the  $x$ .  $T_{\min}$  then tends to a constant value when the  $x$  is larger or equal to 0.37. For  $x$  equals 1, the crystal presents 2H phase and shows a semiconducting behavior, as shown in Fig. S3 and S5.<sup>21</sup> However, we did not get the single crystals when  $x$  is larger than 1, which means the solution limit of S ions may be  $x = 1$  in the  $\text{MoTe}_{2-x}\text{S}_x$  single crystals, the XRD patterns of the  $\text{MoTe}_{2-x}\text{S}_x$  crystals except for  $x = 0$  and 0.1 are shown in Fig. S8.<sup>21</sup>

To understand the enhanced superconductivity in the  $\text{MoTe}_{2-x}\text{S}_x$  single crystals, we did the first principle

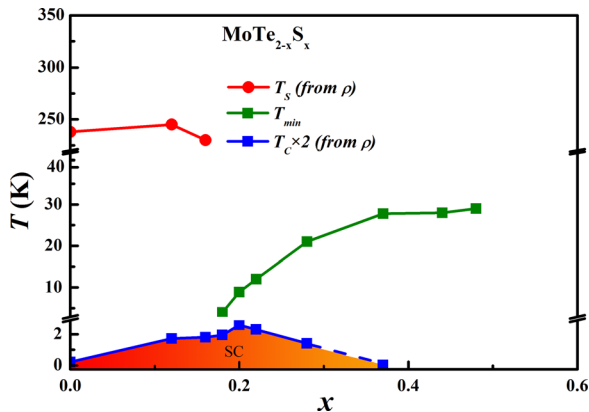


FIG. 4. The superconducting phase diagram of the  $\text{MoTe}_{2-x}\text{S}_x$  single crystals. SC presents the superconductivity.

calculations. In order to simulate the S-doping effect, we substitute one Te atom in  $\text{MoTe}_2$  unit cell by one S atom, i.e., the doped  $\text{MoTe}_{7/4}\text{S}_{1/4}$  was theoretically considered. The results show that the general gradient approximation (GGA) slightly overestimates the lattice parameters of  $\text{MoTe}_2$  with  $1T'$  and  $T_d$  structures compared with those obtained from experiments (see Table SI in the supplementary material; the difference is less than 2% (Ref. 21)). For the  $\text{MoTe}_{7/4}\text{S}_{1/4}$  sample, we test the cases that the S atom locates at four inequivalent Te-sites in the unit cell, respectively. By comparing the total energies, we found both in  $1T'$  and  $T_d$  structures, the S atom tends to replace the Te atom for forming the shortest Mo-Te bond. As expected, the S substitution makes the structures shrinking. Figure S7 shows the calculated DOS for  $\text{MoTe}_2$  and  $\text{MoTe}_{7/4}\text{S}_{1/4}$  with  $1T'$  and  $T_d$  structures. For the  $\text{MoTe}_2$ , our calculation is well consistent with previous calculations.<sup>17,21</sup> One can note that in both structures, S doping rarely changes the overall structure of DOS. S doping slightly increases the bandwidths, which should be due to the enhanced  $p$ - $d$  hybridization in the shrunked structures. In both structures, the DOS at  $E_F$  ( $N(E_F)$ ) decreases upon S doping, as shown in Fig. S5(c). It is in good agreement with the results from the  $C_P$  measurements. That is, the value of Sommerfeld constant  $\gamma_N$  of  $\text{MoTe}_{1.8}\text{S}_{0.2}$  crystals is lower than that of  $\text{MoTe}_2$ . Therefore, the decreased  $N(E_F)$  does not support the S-doping induced superconductivity enhancement in the  $1T'$  and  $T_d$  structures, which is consistent with the experimental results. The real structure of our experimentally prepared  $\text{MoTe}_{2-x}\text{S}_x$  crystals might be different from  $1T'$  and  $T_d$ , which needs to be further investigated both via experimental methods and theory calculations.

Now, we turn to the EPC strength  $\lambda_{ep}$ . As we know, the EPC strength  $\lambda_{ep}$  for a material can be qualitatively expressed as

$$\lambda_{ep} = \sum_{\alpha} \frac{\langle I_{\alpha}^2 \rangle N_{\alpha}(E_F)}{M_{\alpha} \langle \omega_{\alpha}^2 \rangle}, \quad (2)$$

where the  $\langle I_{\alpha}^2 \rangle$ ,  $M_{\alpha}$ , and  $\langle \omega_{\alpha}^2 \rangle$  are the mean square EPC matrix element averaged over Fermi surface, atomic mass, and averaged squared phonon frequency of the  $\alpha$  th atom in the unit cell, respectively.<sup>18–20</sup> We can simply compare the variations of  $\langle I_{\alpha}^2 \rangle$ ,  $M_{\alpha}$ ,  $\langle \omega_{\alpha}^2 \rangle$  and  $N(E_F)$  of  $\text{MoTe}_2$  and those of  $\text{MoTe}_{1.8}\text{S}_{0.2}$  crystals. As we know, the S ions are lighter than the Te ones, so the  $M_{\alpha}$  of  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal (332.03 g/mol) is smaller than that of the  $\text{MoTe}_2$  one (351.14 g/mol). Meanwhile, the Debye temperature  $\Theta_D$  is in proportion to the phonon frequency  $\langle \omega_{\alpha}^2 \rangle$  and that of the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  crystal ( $\Theta_D = 143$  K) is little larger than that of  $\text{MoTe}_2$  one ( $\Theta_D = 135$  K). So, the denominators  $M_{\alpha} \langle \omega_{\alpha}^2 \rangle$  of  $\text{MoTe}_2$  and  $\text{MoTe}_{1.8}\text{S}_{0.2}$  crystals may be close. Let us pay attention to the numerator, the  $N(F)$  of the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal ( $N(F) = 0.88$  states/eV) is just 67% of that of the  $\text{MoTe}_2$  one ( $N(F) = 1.33$  states/eV), indicating that the mean square EPC matrix element averaged over Fermi surface  $\langle I_{\alpha}^2 \rangle$  of the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal should be much larger than that of  $\text{MoTe}_2$  one. The EPC strength  $\lambda_{ep}$  of  $\text{MoTe}_{1.8}\text{S}_{0.2}$  ( $\lambda_{ep} = 0.49$ ) is over one and half times than that of  $\text{MoTe}_2$  ( $\lambda_{ep} = 0.32$ ). Thus, we propose that the improvement of superconductivity in  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal may be related to the dramatic

change or reconstruction of the Fermi surface in the new structure induced by the substitution of Te ions by the S one. Nevertheless, the further exploration of superconductivity in  $\text{MoTe}_{2-x}\text{S}_x$  from both experimental and theoretical perspectives is needed in the future.

We find that the superconductivity in the  $\text{MoTe}_2$  single crystal can be greatly enhanced by the partial substitution of the Te ions by the S ones. The maximum of the superconducting temperature  $T_C$  of the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  single crystal is about 1.3 K. Compared with the parent  $\text{MoTe}_2$  single crystal ( $T_C = 0.1$  K), nearly 13-fold in  $T_C$  is improved in the  $\text{MoTe}_{1.8}\text{S}_{0.2}$  one. The superconductivity has been investigated via the resistivity and magnetization measurements.  $\text{MoTe}_{2-x}\text{S}_x$  single crystals belong to weak coupling superconductors, and the improvement of the superconductivity may be due to the enhanced EPC induced by the S-ion substitution. A dome-shape superconducting phase diagram is obtained in the S-doped  $\text{MoTe}_2$  single crystals. The  $\text{MoTe}_{2-x}\text{S}_x$  materials may provide a new platform to understand the interplay between the superconductivity and topological physics in TMDs.

This work was supported by the Joint Funds of the National Natural Science Foundation of China and the Chinese Academy of Sciences' Large-Scale Scientific Facility under Contracts (U1432139, U1232139), the National Nature Science Foundation of China under Contracts (51171177, 11404342), the National Key Basic Research under Contract No. 2011CBA00111, and the Nature Science Foundation of Anhui Province under Contract Nos. 1508085ME103 and 1408085MA11. The authors thank Dr. Chen Sun from the University of Wisconsin-Madison for her assistance in editing the revised manuscript.

<sup>1</sup>D. H. Keum, S. Cho, J. H. Kim, D.-H. Choe, H.-J. Sung, M. Kan, H. Kang, J.-Y. Hwang, S. W. Kim, H. Yang, K. J. Chang, and Y. H. Lee, *Nat. Phys.* **11**, 482 (2015).

- <sup>2</sup>S. Cho, S. Kim, J. H. Kim, J. Zhao, J. Seok, D. H. Keum, J. Baik, D.-H. Choe, K. J. Chang, K. Seunaga, S. W. Kim, Y. H. Lee, and H. Yang, *Science* **349**, 625 (2015).
- <sup>3</sup>M. N. Ali, J. Xiong, S. Flynn, J. Tao, Q. D. Gibson, L. M. Schoop, T. Liang, N. Haldolaarachchige, M. Hirschberger, N. P. Ong, and R. J. Cava, *Nature (London)* **514**, 205 (2014).
- <sup>4</sup>J. T. Ye, Y. J. Zhang, R. Akashi, M. S. Bahramy, R. Arita, and Y. Iwasa, *Science* **338**, 1193 (2012).
- <sup>5</sup>D. Kang, Y. Zhou, W. Yi, C. Yang, J. Guo, Y. Shi, S. Zhang, Z. Wang, C. Zhang, S. Jiang, A. Li, K. Yang, Q. Wu, G. Zhang, L. Sun, and Z. Zhao, *Nat. Commun.* **6**, 7804 (2015).
- <sup>6</sup>X.-C. Pan, X. Chen, H. Liu, Y. Feng, Z. Wei, Y. Zhou, Z. Chi, L. Pi, F. Yen, F. Song, X. Wan, Z. Yang, B. Wang, G. Wang, and Y. Zhang, *Nat. Commun.* **6**, 7805 (2015).
- <sup>7</sup>Y. Sun, S.-C. Wu, M. N. Ali, C. Felser, and B. Yan, *Phys. Rev. B* **92**, 161107 (2015).
- <sup>8</sup>Y. Qi, P. G. Naumov, M. N. Ali, C. R. Rajamathi, O. Barkalov, Y. Sun, C. Shekhar, S.-C. Wu, V. Süß, M. Schmidt, E. Pippel, P. Werner, R. Hillebrand, T. Förster, E. Kampert, W. Schnelle, S. Parkin, R. J. Cava, C. Felser, B. Yan, and S. A. Medvedev, *Nat. Commun.* **7**, 11038 (2016).
- <sup>9</sup>X. Qian, J. Liu, L. Fu, and J. Li, *Science* **346**, 1344 (2014).
- <sup>10</sup>D. E. Moncton, J. D. Axe, and F. J. DiSalvo, *Phys. Rev. B* **16**, 801 (1977).
- <sup>11</sup>A. A. Soluyanov, D. Gresch, Z. Wang, Q. Wu, M. Troyer, X. Dai, and B. Andrei Bernevig, *Nature* **527**, 495 (2015).
- <sup>12</sup>Z. Wang, D. Gresch, A. A. Soluyanov, W. Xie, S. Kushwaha, X. Dai, M. Troyer, R. J. Cava, and B. Andrei Bernevig, e-print [arXiv:1511.07440v1](https://arxiv.org/abs/1511.07440v1).
- <sup>13</sup>S.-Y. Xu, I. Belopolski, N. Alidoust, M. Neupane, G. Bian, C. Zhang, R. Sankar, G. Chang, Z. Yuan, C.-C. Lee, S.-M. Huang, H. Zheng, J. Ma, D. S. Sanchez, B. Wang, A. Bansil, F. Chou, P. P. Shibayev, H. Lin, S. Jia, and M. Zahid Hasan, *Science* **349**, 613 (2015); S.-M. Huang, S.-Y. Xu, I. Belopolski, C.-C. Lee, G. Chang, B. Wang, N. Alidoust, G. Bian, M. Neupane, C. Zhang, S. Jia, A. Bansil, H. Lin, and M. Zahid Hasan, *Nat. Commun.* **6**, 7373 (2015).
- <sup>14</sup>H. P. Hughes and R. H. Friend, *J. Phys. C: Solid State Phys.* **11**, L103 (1978); T. Zandt, H. Dwelk, C. Janowitz, and R. Manzke, *J. Alloys Compd.* **442**, 216 (2007).
- <sup>15</sup>H. Padamsee, J. E. Neighbor, and C. A. Shiffman, *J. Low Temp. Phys.* **12**, 387 (1973).
- <sup>16</sup>W. L. McMillan, *Phys. Rev.* **167**, 331 (1968).
- <sup>17</sup>M. Ríflková, R. Martoňák, and E. Tosatti, *Phys. Rev. B* **90**, 035108 (2014).
- <sup>18</sup>L. N. Cooper, *Phys. Rev.* **104**, 1189 (1956).
- <sup>19</sup>J. Bardeen, L. N. Cooper, and J. R. Schrieffer, *Phys. Rev.* **106**, 162 (1957).
- <sup>20</sup>J. Bardeen, L. N. Cooper, and J. R. Schrieffer, *Phys. Rev.* **108**, 1175 (1957).
- <sup>21</sup>See supplementary material at <http://dx.doi.org/10.1063/1.4947433> for experiment details, structural information, sample characterization and calculated details.