Effect of Cd/As flux ratio and annealing process on the transport properties of Cd₃As₂ films grown by molecular beam epitaxy

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Abstract
To study how the Cd/As flux ratio affects the microstructure and transport properties for Cd₃As₂ films, we used molecular beam epitaxy (MBE) to grow Cd₃As₂ (224) thin films on CdTe (111)/GaAs (001) virtual substrates. The effects of Cd/As flux ratio, during the grown process, on the electrical properties and surface morphology of the sample was studied. The films grown at lower Cd/As flux ratio have higher electron mobility and longer effective dephasing length. With decreasing Cd/As flux ratio, the magnetoresistance (MR) of the film changes from negative to positive. These results show that a lower beam ratio is beneficial to improve the crystal quality. In order to optimize the electrical properties of the films, the effect of annealing on the electron mobility and MR have been studied. After annealing, the MR changes from negative to positive, the electron mobility increase by 8 times, and the MR increase from 15% to 360% at 9 T. These results indicate that annealing is an effective method to optimize the electrical properties of Cd₃As₂ epitaxial films.

1. Introduction

Three-dimensional (3D) Dirac semimetal materials have a linear dispersion relationship in the bulk state, and the surface state is protected by topology. Cd₃As₂ is a typical 3D Dirac semimetal material, with a pair of 3D Dirac cones along Γ-Z direction [1], and it has received significant research attention. Theoretical calculations show that the topological insulator and Weyl semimetal state can be obtained by breaking the symmetry or reducing the dimensions in Cd₃As₂ [1]. Angle-resolved photoemission spectroscopy indicate Cd₃As₂ materials have properties of 3D Dirac semimetal, and a pair of Dirac cones were observed in Brillouin zone [2, 3]. The two-dimensional (2D) topological surface state is an important feature of 3D Dirac semimetal, which was observed on the (112) and (001) planes, respectively [3, 4]. Transport experiments have shown that a bandgap opens in the bulk state, and the resistance is increase as temperature decrease to liquid helium temperature, thus the film shows semiconductor properties as Cd₃As₂ material thins to the quantum limit [5]. Besides, the quantum Hall effect can also be observed in thin films of Cd₃As₂ [6, 7], and the surface transport is virtually free of bulk conduction. Besides, the two-dimensional Dirac fermions appear in quantum-confined Cd₃As₂ thin films are probed by transport in the works of Luca Galletti [8]. In Cd₃As₂ nanolayers, a giant linear MR is observed when the Fermi level approaches the Dirac point [9]. Other phenomena in Cd₃As₂ that have received widespread attention include anisotropic longitudinal MR [10], the chiral anomaly [11], and topological phase transitions [12]. The rich physical properties of Cd₃As₂ have thus attracted extensive research attention [8, 13–16].

Investigating the material properties and possible applications of Cd₃As₂ films requires high-quality films. At present, Cd₃As₂ can be prepared by various methods, such as heating Cd₃(Cd₃As₄) in quartz ampoules [17, 18], chemical gas phase transport [9, 19], magnetron sputtering [20], pulsed laser deposition [21, 22], and molecular beam epitaxy (MBE). Among them, the MBE method has attracted significant attention because it allows for large lattice mismatch and can obtain large size films. To date, Cd₃As₂ thin films have been grown by MBE on substrates of mica [23], GaSb [12, 24], CdTe [7, 25] and Zn₁₋ₓCdₓTe [26]. Although these studies have
considered how temperature affects the microstructure and mobility of Cd$_3$As$_2$ films [24], the effect of the Cd/As flux ratio on the morphology and transport properties have not been studied.

It was reported that higher mobility of Cd$_3$As$_2$ thin films can be obtained at a higher growth temperature [24]. However, Cd$_3$As$_2$ has a high saturation vapor pressure, Cd$_3$As$_2$ film is usually grown at a lower temperature ($\leq 210 ^{\circ}$C) to reduce desorption and maintain the growth rate during film growth. However, the lower temperature can reduce the crystal quality through the formation of defects and increases disorder in the film, causing a higher carrier concentration and lower electron mobility. Due to the low-temperature growth conditions and the existence of a lattice mismatch, it is difficult to obtain high-quality Cd$_3$As$_2$ films. Thus, other methods should be considered to optimize the crystal quality and electrical properties of Cd$_3$As$_2$ epitaxial films.

In the work, we grow Cd$_3$As$_2$ films on CdTe (111)/GaAs (001) substrates with different Cd/As flux ratios, comparing the surface morphologies and transport properties among these samples. The results show that Cd$_3$As$_2$ can be grown in a large range of Cd/As flux ration (from 4.0 to 1.0). However, the Cd/As flux ratio has a great influence on the surface morphology and transport properties. At first, the Cd-rich atmosphere can prevent large island formation and increase the concentration of little island. Second the lower Cd/As flux ratio (about 1.0) can improve the electron mobility and change the MR from negative to positive. The annealing is an efficient methods to improve the crystal quality of semiconductor [27], which can reduce the defects such as point defect and disorder. In this work, the annealing is used to improve the crystal quality of Cd$_3$As$_2$. After annealing, the MR can be changed from negative to positive, and the electron mobility can be improved significantly.

2. Experiment

CdTe single crystals have a zinc blende crystal structure, and the atomic arrangement of its (111) plane is the same as that of the Cd$_3$As$_2$ (224) plane. Moreover, the lattice mismatch between CdTe (111) and Cd$_3$As$_2$ (224) is about 2.3%, so CdTe(111) film can be used as a substrate for epitaxial growth of Cd$_3$As$_2$ (224) film [7, 24]. In this experiment, CdTe compound was used as evaporation source, and the beam equivalent pressure (BEP) was about $1 \times 10^{-6}$ Torr. The CdTe films were nucleated at the low temperature of 280 $^{\circ}$C for 5 min on GaAs (001) substrate and then grown at 320 $^{\circ}$C, the thickness of CdTe(111) buffer layer is about 1 $\mu m$. The details of the growth of CdTe(111) can be found in our published article [28]. CdTe is a traditional semiconductor with the band gap about 1.47 eV, the transportation properties of Cd$_3$As$_2$/CdTe/GaAs(001) system is mainly contributed by the layer of Cd$_3$As$_2$. In the growth process of Cd$_3$As$_2$ films, the arsenic (As) flux and the growth temperature are fixed, and the Cd-source evaporation temperature was modified to obtain different Cd/As flux ratios. The growth temperature of Cd$_3$As$_2$ is about 170 $^{\circ}$C, the As flux is controlled by a valve-cracker effusion cell (As$_4$ mode), and the BEP was $1.8 \times 10^{-7}$ Torr. The Cd source evaporation temperature varied from 220 to 180 $^{\circ}$C to obtain different BEP of Cd, resulting in different Cd/As flux ratios.

The Al$_2$O$_3$, with a large gap about 3.4 eV, which is widely used as insulating layer in semiconductor device technology. To prevent the desorption and oxidation of Cd$_3$As$_2$, when the film is annealed at high temperature. A layer of Al$_2$O$_3$ film was grown on the Cd$_3$As$_2$ film at 150 $^{\circ}$C by atomic layer deposition, and the thickness is about 30 nm. The Cd$_3$As$_2$ annealing process was done in the air, by using an electric heating plate. The samples were annealed at 500 $^{\circ}$C for 10 min, and the samples are coded in the format of AS1, AS2, etc. The van der Pauw method was used for the magnetic transport test [29]. In the transport experiment, the sample size was 0.5 cm $\times$ 0.5 cm. The indium and silver wires served as electrodes to form ohmic contacts. The main magnetic transport data were acquired by using a 1.3 T magnetic-field transport system, the current is 1000 $\mu$A and the lowest temperature is 10 K. Besides, the magnetic transport data at 1.3 K were acquired by using a 9 T Oxford magnetic transport system, and the current is about 100 $\mu$A. In the experiment, the XRD and AFM were used to characterize the surface morphology and crystal properties of the sample.

3. Results and discussion

3.1. Effects of Cd/As flux ratios on MBE growth

The growth conditions of samples S1, S2, and S3 are listed in table 1. They are grown at the same substrate temperature for 60 min, keeping the As flux constant and changing the temperature of the Cd evaporation source. The Cd/As flux ratio of S1, S2, and S3 decreases continuously with decreasing temperature of the Cd evaporation source. These samples were used to study how the Cd/As flux ratios affect the surface morphology and electrical properties of Cd$_3$As$_2$ films.

Figure 1(a) shows the XRD peak $(2\theta-\omega)$ of the Cd$_3$As$_2$ film and reveals the (224), (336), and (448) XRD peaks, which match with the peaks reported in the literatures [21, 23, 24]. According to literature reports, the spacing between Cd$_3$As$_2$ (112) planes is about 0.733 nm [30–32]. In our experiment, the interplane spacing for Cd$_3$As$_2$...
calculated by XRD results of figure 1(a) is about 0.728 nm. Figure 1(d) is the HRTEM image for the interface of CdTe(111) and Cd₃As₂(224), and the interplane spacing is about 0.73 nm. This result indicates that the Cd₃As₂ films with (224) crystal direction were grown on the CdTe(111)/GaAs(001) substrate. Figure 1(b) shows the high-resolution XRD pattern near the (224) diffraction peak, from which the thickness of Cd₃As₂ film can be obtained from the Laue fringes. As shown in figure 1(b), the film thickness gradually increases with increasing the Cd/As flux ratio. This is attributed to the high saturation vapor pressure of Cd₃As₂ and the Cd-rich atmosphere can effectively suppress the desorption. The mismatch between Cd₃As₂(224) and CdTe(111) is about 2.3%, and the substrate produces a compressive strain in the Cd₃As₂ film [12]. In figure 1(b), the Cd₃As₂(224) diffraction peak gradually shifts to the right as the film thickness increases, which is indicative of strain relaxation in the film, meaning that the initial compressive strain in the Cd₃As₂ films is not fully released. Figure 1(c) shows the RHEED pattern of Cd₃As₂ film. During growth, the RHEED lines in the pattern remain clear and sharp, and the reconstituted stripes appear. When the sample is rotated 60°, the RHEED pattern remains completely equivalent, as expected from the six-fold rotational symmetry of the Cd₃As₂(224) surface [24]. The RHEED pattern remains essentially constant in the larger Cd/As flux ratio range (from 4.0 to 1.0), but when the Cd/As flux ratio decreases below unity, the RHEED image gradually becomes point-like and the polycrystalline rings appear. It was observed in experiments that a higher growth temperature required a larger

| Sample | Growth temperature (°C) | As flux × 10⁻⁷ (Torr) | Cd flux × 10⁻⁷ (Torr) | Cd/As flux ratio | Cd₃As₂ thickness (nm) | Mobility (cm² V⁻¹ s⁻¹) | Carrier density × 10¹⁸ (1/cm³) |
|--------|------------------------|------------------------|------------------------|-------------------|-----------------------|------------------------|-----------------------------|
| S1     | 170                    | 1.71                   | 7.05                   | 4.12              | 110                   | 986                    | 4.3                         |
| S2     | 170                    | 1.69                   | 4.75                   | 2.81              | 93                    | 1469                   | 2.0                         |
| S3     | 170                    | 1.86                   | 1.96                   | 1.05              | 78                    | 1700                   | 2.8                         |

Table 1. Growth conditions and properties of Cd₃As₂ samples S1, S2 and S3, at the temperature of 300 K.

Figure 1. (a, b) Results of x-ray diffraction (XRD) measurement of samples S1, S2 and S3 from table 1. (a) The XRD peak (2θ-ω) of the Cd₃As₂ films. (b) XRD results near the Cd₃As₂(224) diffraction peak. (c) Reflection high energy electron diffraction (RHEED) pattern during the growth of Cd₃As₂ sample. (d) The High-resolution transmission electron microscopy (HRTEM) image of Cd₃As₂ films.
Cd/As flux ratio to reduce the desorption. A higher growth temperature is beneficial to improve the electron mobility of the Cd₃As₂ film, which is consistent with results published [33]. Besides, different Cd/As flux ratios can strongly affect the electron mobility. As shown in table 1, the Hall mobility of S1, S2, and S3 is 986, 1469, and 1700 cm² V⁻¹ s⁻¹, respectively. It is shown that a smaller flux ratio is beneficial to improve the sample mobility.

Although the RHEED patterns of the films do not change significantly for different Cd/As flux ratios, the flux ratio strongly affects the surface morphology and Mr. As shown in figure 2(a), when the Cd/As flux ratio is relatively large (≥4.12), many 10–20 nm island-like clusters appear on the surface. These clusters can be observed in figure 2(d), and they are evenly distributed on the sample surface. Figure 2(b) shows layered steps that appear on the film surface, indicating that the material exhibits a two-dimensional (2D) layer-island mutual growth mode. As can be seen in figure 2(e), reducing the Cd/As flux ratio in S2, the clusters disappear in S1, indicating that the generation of these clusters is related to the flux of Cd. In figure 2(c), as the Cd/As flux ratio is close to unity, the layered steps disappear on the surface. Compared with S1 and S2, the sample of S3 has a larger 3D island structure (200–300 nm). The main reason is that the Cd-rich atmosphere can prevent large island formation and increase the concentration of little island [34]. During the growth of sample S1, Cd-rich atmosphere increases the film deposition rate. The atoms on the surface cannot fully migrate, which increases the nucleation density on the surface, resulting in a large number of micro-island (the clusters mentioned above) in the atomic force microscopy image. During the island coalescence, grain boundaries and defects will be introduced into the film [22]. These defects can reduce the electron mobility and affect MR properties [35]. Sample S1 has a higher nucleation density and a smaller island, which means more defects in the film. Therefore, S1 has low mobility and exhibits a negative Mr. Besides, the Cd-rich growth conditions tend to cause the As vacancies. The extra Cd atoms contribute electrons to the film, resulting in a higher carrier concentration in the sample.

3.2. Effects of Cd/As flux ratios on electron mobility and magnetoresistance

We now compare the transport properties of films grown under different Cd/As flux ratios. Figure 3 shows the MR and weak antilocalization effects of samples S1, S2, and S3. Figures 3(a)–(c) show the longitudinal MR, which is defined as 

\[ \text{MR} \left( \% \right) = \left[ \frac{R_{xx}(B) - R_{xx}(0)}{R_{xx}(0)} \right] \times 100\% \]

Figures 3(d) and (e) show the conductance as a function of the magnetic field at different temperatures. The conductance is defined as \( \sigma = 1/R_{xx} \), and the normalized conductance is defined as

\[ \Delta \sigma = \sigma - \sigma(0) \]

Figure 2. AFM images characterize the surface morphology of Cd₃As₂ films. The Cd/As flux ratio during sample growth was (a) 4.12, (b) 2.81, and (c) 1.05. The image of (d)–(f) are the microstructures in images (a)–(c).
Figures 3(a)–(c) show that the MR changes from negative to positive as the Cd/As flux ratio decreases. In figure 3(a), when the magnetic field is higher than 0.1 T, the resistance decreases as the magnetic field increases, giving negative Mr. However, the positive and negative MR coexist in figure 3(b), and the negative MR is observed in the magnetic field range of 0.1 to 0.5 T. As the Cd/As flux ratio continues to decrease, the negative MR disappears completely in sample S3 (see figure 3(c)). The negative MR of Cd$_3$As$_2$ materials with parallel magnetic fields has been extensively studied [33, 36–38]. According to these literatures, both the chiral anomaly, current-jetting effect, and conductivity fluctuation can cause negative Mr. However, the negative MR has not been discussed in detail when the magnetic field is perpendicular to the film surface. The origin of the negative MR will be discussed at the end of this paper.
Another obvious feature in figures 3(a)–(c) is the WAL near the zero magnetic field. In the magnetic field range of ±0.1 T, the resistance increases rapidly as the increase of the magnetic field, due to the strong spin-orbit coupling (SOC) in Cd₃As₂. Spin-orbit scattering destroys the interference between the electron paths with time-inversion symmetry, which can reduce the resistance at the zero magnetic field [39]. The applied magnetic field will destroy the time-inversion symmetry, resulting in a rapid increase of resistance. The WAL phenomenon exists widely in materials with strong spin-orbit coupling, such as topological insulators [40–42] and graphene [43]. In addition, the WAL phenomenon also occurs in Cd₃As₂ [5, 6]. According to the literatures, the WAL phenomenon in 2D and quasi-2D systems can be described by the Hikami-Larkin-Nagaoka (HLN) mode [44]. The HLN model is also used to describe the weak local phenomenon in Cd₃As₂ films, studying the mechanism of the dephasing and electron scattering [5, 15]. The HLN model is expressed as follows:

$$\sigma(B) - \sigma(0) = -\frac{e^2}{\pi \hbar} \left[ \psi\left(\frac{1}{2} + \frac{B_{\text{c}}}{B}\right) - \ln\left(\frac{B_{\text{c}}}{B}\right) \right] + \beta B^2$$

where \(\alpha\) is a fitting parameter that is related to the number of independent conductive channels in the sample. For systems with WAL and weak localization (WL), \(\alpha\) varies from 0.5 to 1. \(\psi(x)\) is the digamma function, \(L_{\text{c}}\) is the effective dephasing length, and \(B_{\text{c}} = \hbar/(4eL_{\text{c}}^2)\). The parameter \(\beta\) is a correction factor related to the parabolic conductivity background. In figures 3(d)–(f), the phenomenon of WAL mentioned above is fitted by HLN mode, and the results are shown in figures 4(a) and (b).

Figure 4(a) is the WAL fitting results of \(L_{\text{c}}\) for samples S1, S2 and S3 shown in figure 3. The effective dephasing lengths \(L_{\text{c}}\) of samples S1, S2, and S3 are all greater than 200 nm in the temperature range 12–30 K, which is greater than the film thickness (<110 nm). Therefore, samples S1, S2, and S3 can be considered as quasi-2D systems. In figure 4(b), the parameter \(\alpha\) for samples S1, S2, and S3 are close to 0.33 at 12 K. According to the literature, topological materials such as Bi₃Te₂Se and Bi₃Se₂ with WAL usually have \(\alpha = 0.5\) [45], which is much greater than our results. Figure 4(a) shows that the effective dephasing lengths of S1, S2, and S3 are 320, 420, and 480 nm, respectively, which increase with decreasing Cd/As flux ratio. The longer \(L_{\text{c}}\) means that the electrons are subject to weaker inelastic scattering. The samples grown with a lower Cd/As flux ratio have a longer \(L_{\text{c}}\), indicating the better electrical properties. For samples S1, S2, and S3, we obtain \(L_{\text{c}} \propto T^{-0.328}\), \(L_{\text{c}} \propto T^{-0.345}\), and \(L_{\text{c}} \propto T^{-0.364}\), respectively. According to the literature, when \(L_{\text{c}} \propto T^{-1/2}\), the dephasing mechanism is dominated by the electron-phonon interactions [15, 46, 47]. However, if \(L_{\text{c}} \propto T^{-3/2}\), the dephasing mechanism is dominated by electron-photon scattering [39, 48]. In some topological materials, there is also the relationship of \(L_{\text{c}} \propto T^{-1/3}\) [39, 49]. Based on the fitting results, the dephasing mechanism in S1, S2, and S3 is mainly dominated by the scattering between the electrons.

### 3.3. The effect of annealing on electron mobility and magnetoresistance

Table 2 compares the carrier concentration and electron mobility of a series of Cd₃As₂ films before and after annealing. These samples were grown with different substrate temperatures and Cd/As flux ratios. During annealing, the samples were maintained at 500 °C for 10 min and then cooled naturally to room temperature. After annealing, the carrier concentration in the samples decreases significantly, and the mobility increases by two to eight times. The possible reason is that the annealing reduces the point defects and disorder in the material. This shows that annealing improves the electron mobility of Cd₃As₂ epitaxial film and reduces the carrier concentration. It can be seen from the table that the annealing is more effective for samples with lower...
electron mobility ($\leq 1000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$). These samples (S4 and S6) are mainly grown under Cd-rich conditions, which have more point defects, grain boundaries in the samples (similar to the sample of S1 discussed in figure 2). These defects can act as potential barriers to carrier transport. When Cd$_3$As$_2$ is annealed at high temperature, it may undergo a recrystallization process. This process can enlarge the grain size and reduce the number of grain boundaries in the film [50]. In addition, the disorder and impurities at the grain boundary can also be decreased. Therefore, the mobility in the Cd$_3$As$_2$ film increases significantly after annealing.

Figure 5 is used to illustrate the effect of annealing on the MR of samples S1, S2, S4, and S5. These films are annealing at 500 °C for 10 min. The growth conditions of S1 and S2 are shown in table 1, while the S4 and S5 are shown in table 2. As shown in figure 5(a), the MR of the AS1 sample after annealing is positive, while the original S1 sample in figure 3(a) has a negative MR. Different form S1, the sample of S2 has a positive MR before and after annealing. Comparing figures 3(b) and 5(d), the MR of S2 has increased from 0.8% to 14% under the magnetic of 1.3 T. In order to further verify the effect of annealing on MR, the MR of S4 and S5 before and after annealing is compared in figure 5(c). The thickness for S4 and S5 is about 30 nm and 90 nm. As shown in the insert of figure 5(c), S4 has a negative MR in the range of 0–9 T magnetic field. However, in figure 5(c), both AS4 and AS5 have positive MR after annealing. This result is similar with that of AS1 and AS2 mentioned above. Indicating that the annealing can change the MR from negative to positive. Besides, the MR in the sample increases significantly after annealing, in figure 5(c). Before annealing, the MR of S4 and S5 is $-7\%$ and $15\%$, whereas the MR after annealing is $170\%$ and $360\%$, respectively.

According to the work of L N Wei and C Z Li et al Applying a gate voltage to the sample, the MR increases significantly when the Fermi level is close to the Dirac point [51, 52]. Figures 5(e) and (f) shows that the annealing can decrease the carrier concentration, and which can approve the explanation for the increases of MR, mentioned above. Figure 5(f) shows that the $R_{xy}$ maintains a linear shape, before and after annealing, and a single carrier type dominates the transport properties. For the S4 sample shown in figure 5(e), the relationship between transverse $R_{xy}$ and the magnetic field strength is no longer linear after annealing. The reason is that there is a bandgap in the bulk state of S4 (thickness is 30 nm) [5], and the Fermi level moved into the bandgap after annealing.

### 3.4. Discussion on the origin of negative magnetoresistance

When the electric field is parallel to the surface of a topological semimetal, negative longitudinal MR has been observed. Which can be related to chiral anomaly [16]. In the present experiment, the magnetic field is perpendicular to the electric field and to the sample surface, which differs from the negative MR phenomenon mentioned above. There are several possible explanations for the negative longitudinal MR when the magnetic field is perpendicular to the sample surface.

1. The twinning is difficult to eliminate when Cd$_3$As$_2$ (224) films grown on CdTe(111) substrate. The existence of twins can reduce electron mobility [53], which may also lead to negative Mr. The phi ($\Phi$) scans of XRD for Cd$_3$As$_2$ (4416) peaks is used to characterize the twinning in previous reports [7, 24, 26]. In the (224) plane of Cd$_3$As$_2$ film, the (4416) diffraction peak has triple rotational symmetry. However, the six-fold rotational symmetry is observed in figure 6(a), indicate the micro twin exists in the film. After annealing, the micro twins still exist in the film, but the negative MR of S4 and S5 is disappeared (As shown in figure 5). This indicates that the micro twin defects may not be the main origin of negative MR in the manuscript. In figure 6(b) the 2$\theta$–$\omega$ scan for Cd$_3$As$_2$ film does not change significantly before and after annealing. This result proves that annealing process does not change the crystal structure of the film.

2. The magnetic polaron model is another possible explanation for negative MR, and the magnetic doping can form magnetic polarons [14, 54]. Increasing the magnetic field, the scattering related to magnetic polarons is suppressed, resulting in the negative Mr. In the manuscript, there are no magnetic impurities in the films, and the origin of negative MR can’t be explained by the magnetic polaron model.

Table 2. Effect of annealing on electron mobility and carrier concentration for Hall measurements at 300 K.

| Sample     | S4       | S5       | S6       | S7       | S8       |
|------------|----------|----------|----------|----------|----------|
| Before annealing | $\mu$ (cm$^2$ V$^{-1}$ s$^{-1}$) | 758 | 1779 | 602 | 2420 | 2045 |
|            | $n_0$ (10$^{17}$/cm$^3$) | 15 | 39 | 60 | 35 | 24 |
| After annealing | $\mu$ (cm$^2$ V$^{-1}$ s$^{-1}$) | 3334 | 5342 | 5281 | 5909 | 3988 |
|            | $n_0$ (10$^{17}$/cm$^3$) | 20 | 10 | 0.58 | 0.59 |
| Cd/As flux ratio |          | 1.6 | 1.31 | 4.26 | 2.4 | 1.08 |
| Thickness (nm) |          | 30 | 110 | 118 | 82 | 65 |
It is well known that the weak localization effect (WL) in disordered systems can lead to negative MR [40]. Increasing temperature can strengthen the electron-phonon interaction, and WL effect with increasing temperature. In general, the negative MR caused by WL is saturated when the magnetic field is up to 1 T. When the temperature is above 120 K, the negative MR disappears completely. Before annealing, the negative MR in samples S2 (figure 2(b)) disappears at room temperature and only occurs at a magnetic field of about 0.5 T. Thus, the origin of negative MR in S2 is related to WL effects. However, this explanation is not suitable for S1 and S4. Because the negative MR still exists at room temperature, and the MR is not saturated in the magnetic field of 1 T.

Figure 5. Influence of annealing at 500 °C for 10 min on the transport properties of Cd₃As₂ films under different magnetic fields. (a) and (b) is the MR of ample S1 and S2, after annealing. Image (c) and (d) show the MR of S4 and S5 before and after annealing, the temperature is 1.5 K and measured with a 9 T hall system. (e) and (f) shows the Transverse resistance (Rₓᵧ) of S4 and S5 before and after annealing.
(4) In highly disordered materials, diffuse scattering at the microcrystalline boundary may lead to negative Mr. The scattering of electrons at the crystallite boundary can affect the mean-free path of carriers, which leads to an increase in resistance \([55, 56]\). Applying an external magnetic field, the carriers are constrained by the magnetic field, and the electron path becomes arc-shaped (the radius of curvature is \(r = m^* \mu / eB\)). The stronger the magnetic field, the smaller is the radius of curvature, and scattering at the boundary is suppressed. Therefore, the resistance decreases with increasing magnetic field, and the film exhibits negative Mr. As the Cd/As flux ratio decreases, the MR changes from negative to positive for samples S1, S2 and S3. Combine with the results of AFM analysis, the possible reason is that the Cd-rich atmosphere can increase the nucleation density of S1 and reduce the size of 3D island. Therefore, there are more grain boundaries and defects in the S1 sample and the film exhibits negative Mr. Before annealing, both S1 and S4 showed negative MR properties, and the negative MR still exists when the temperature rises to 300 K. In addition, S4 maintains negative MR in the 9 T magnetic field range, which is similar to the phenomena observed in graphene material related to grain boundary \([57]\). Annealing at high-temperature can reduce the grain boundaries and point defects \([22]\) and reduce the disorder in the film. Thus, the sample of S4 and S5 shows positive MR (as shown in figure 5) after annealing.

4. Conclusions

We grew Cd\(_3\)As\(_2\) (211) films on CdTe (111)/GaAs (001) virtual substrates. We found that Cd\(_3\)As\(_2\) film grown under a lower Cd/As flux ratio is beneficial to improve the electrical properties. Decreasing Cd/As flux ratio (Cd/As = 4.12, 2.81, 1.05), the MR changes from negative to positive. At the same time, the film grown under a lower Cd/As flux ratio (1.05) have higher electron mobility and longer effective dephasing length. In this work, the effect of annealing on the electrical properties of Cd\(_3\)As\(_2\) films was studied. The experimental results show that the annealing process can suppress the negative MR, improves the electron mobility, and reduces the carrier concentration. This indicates that annealing is an effective method to optimize the electrical properties of Cd\(_3\)As\(_2\) epitaxial films. The origin of negative MR is discussed in the manuscript, and the micro twins are not the main contribution for negative Mr. The diffuse scattering and WL effect are considered to explain the phenomenon of negative Mr.

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