Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$: A ferromagnetic semiconductor with colloossal magnetoresistance

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Abstract – A bulk diluted magnetic semiconductor (Sr,K)/(Zn,Mn)$_2$As$_2$ was synthesized with degenerated charge and spin doping. It has a hexagonal CaAl$_2$Si$_2$-type structure with the (Zn,Mn)$_2$As$_2$ layer forming a honeycomb-like network. Magnetization measurements show that the sample undergoes a ferromagnetic transition with a Curie temperature of 12 K and magnetic moment reaches about 1.5 $\mu_B$/Mn under $\mu_0H = 5$ T and $T = 2$ K. Surprisingly, a colossal negative magnetoresistance, defined as $|\rho(H) - \rho(0)|/\rho(0)$, up to $-38\%$ under a low field of $\mu_0H = 0.1$ T and to $-99.8\%$ under $\mu_0H = 5$ T, was observed at $T = 2$ K. The colossal magnetoresistance can be explained based on the Anderson localization theory.

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Introduction. – Combining semiconductor and magnetism, diluted magnetic semiconductors (DMS) which possess both spin and charge degrees of freedom have attracted much attention because such DMS materials not only exhibit substantial novel phenomena such as quantum Hall effects, semiconductor lasers and single-electron charging, but also bring about numerous applications in sensors, memories as well as spintronics [1–5]. However, the III–V–based DMS materials, represented by (Ga,Mn)As, obtained by heterovalent substitution of Mn$^{2+}$ for Ga$^{3+}$, are only available as thin films due to the limited chemical solubility of manganese in bulk GaAs (< 1%), and lack independent control of local moment and carrier densities [6,7].

The I–II–V–based Li(Zn,Mn)As was theoretically proposed to be an n-type DMS and experimentally synthesized as a bulk p-type DMS material with Curie temperature ($T_C$) of 50 K [6,8]. In this bulk system charge and spin concentrations can be tuned separately via Li off-stoichiometry and the isovalent substitution of Mn$^{2+}$ for Zn$^{2+}$, and the solubility of Mn is significantly enhanced. Actually, only in a limited number of DMS systems the concentration of acceptor and Mn impurity (i.e., magnetic moment) can be tuned independently [9–11]. Inspired by the rapid development of iron-based superconductors [12–14], a series of DMS systems based on a similar layered structure were found [7,15–17].

The so-called colossal magnetoresistance (CMR) is usually observed in manganites, where the complex and intimate link among magnetic structure, crystallographic structure and electrical resistivity makes it a focus of research interest [18–20]. Materials exhibiting large MR can be exploited to enlarge the sensitivity of read/write heads of magnetic storage devices and thus to maximize the information density [19]. But magnetic fields of several teslas are typically required to obtain such a CMR effect, which limits the potential for applications [21,22]. The CMR effect under a low field is highly required from the viewpoint of practical use.

SrZn$_2$As$_2$ is a compound with hexagonal CaAl$_2$Si$_2$-type structure (shown in fig. 1(a) and (b)) [23], which belongs to the P-3m1 (No. 164) space group. The Zn$_2$As$_2$ layers form a honeycomb-like network. No detailed study on the physical properties of SrZn$_2$As$_2$ has been reported.

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yet. Attracting more and more attention, the honeycomb-like network is essential in recently extensively investigated topological insulators [24,25]. Recently, a first honeycomb-lattice bulk DMS (Ba,K)(Cd,Mn)$_2$As$_2$ with $T_C \sim 16$ K [26] has been reported. In this letter, we report successful synthesis of a honeycomb lattice bulk DMS (Sr,K)(Zn,Mn)$_2$As$_2$ with $T_C \sim 12$ K and magnetic moment of about 1.5 $\mu_B$/Mn under $\mu_0 H = 5$ T and $T = 2$ K. In this system, charge and spin degree of freedom can be controlled independently via K$^+$ for Sr$^{2+}$ and isovalent Mn$^{2+}$ for Zn$^{2+}$ substitution, respectively. Surprisingly, a large MR, defined as $|\rho(H) - \rho(0)|/\rho(0)$, up to $-38\%$ under a low field of $\mu_0 H = 0.1$ T and to $-99.8\%$ under $\mu_0 H = 5$ T was observed at $T = 2$ K. Only a limited number of systems (systems represented by (Ga,Mn)As and (La,Sr)MnO$_3$) exhibit such a remarkable CMR behavior [5,18,27]. The colossal magnetoresistance can be explained based on the Anderson localization theory. Although $T_C$ of this system may be too low for practical use, our work encourages to explore colossal magnetoresistance in the other recently discovered bulk DMS materials, such as (Ba,K)(Zn,Mn)$_2$As$_2$ [15] and (La,Sr)(Cu,Mn)SO [17] with much higher Curie temperatures, which makes them a truly appealing class of systems.

**Experimental.** – The polycrystalline samples of (Sr,K)(Zn,Mn)$_2$As$_2$ were synthesized by the solid state reaction method. All the starting materials, Sr granules, K lumps, and the powders of Zn, Mn and As are of high purity (≥ 99.9%). As a first step, SrAs was presynthesized by reacting stoichiometric Sr granules and As powder at 1123 K for 48 h, and KAs was presynthesized by heating stoichiometric K lumps and As powder with a ramping rate of 0.25 K/min to 773 K and kept at that temperature for 10 h. Then the resultant SrAs and KAs, and the powders of Zn, As and Mn were weighted according to their stoichiometric ratio and then fully ground in an agate mortar. The mixture was then pressed into pellets, heated in evacuated quartz tubes at 1223 K for 33 h, and finally furnace-cooled to room temperature. The process was repeated once again in order to get the pure phase.

Powder X-ray diffraction (XRD) was performed at room temperature using a PANalytical X-ray diffractometer (Model EMPIREAN) with a monochromatic CuK$_{\alpha 1}$ radiation. The electrical resistivity was measured by the four-terminal method. The dc magnetization was measured on a Quantum Design magnetic property measurement system (MPMS-5). The magnetoresistance and Hall coefficient was measured using a Quantum-Design physical property measurement system (PPMS).

**Results and discussion.** – Figure 1(c) shows the X-ray diffraction patterns of (Sr,K)(Zn,Mn)$_2$As$_2$. The crystal structure is also sketched in fig. 1(a) and (b). The diffraction peaks of both SrZn$_2$As$_2$ and Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ can be well indexed based on the P-3m1 (No. 164) space group with a hexagonal CaAl$_2$Si$_2$O$_6$ type structure. The XRD patterns indicate that the samples are essentially single phase. The lattice parameters of the two samples were obtained by least-squares fit of more than 20 reflection peaks with the correction of zero shift, using space group of P-3m1 (No. 164). The resulting room temperature lattice constants $a$ and $c$ are 4.223 Å and 7.270 Å, respectively for the SrZn$_2$As$_2$ sample, compound, consistent with the previously reported values in [15].

In fig. 1(d), the Rietveld refinement of the Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ sample based on the CaAl$_2$Si$_2$O$_6$ type structure shows that the calculated profile well matches the experimental data. The weighted reliable factor $R_w$ and the goodness of fit $S$ are 12.8% and 1.27, respectively.

In fig. 2(a), the temperature dependence of dc magnetization of Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ is shown for both zero-field-cooling (ZFC) and field-cooling (FC) procedures.
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Fig. 2: (Color online) (a) Temperature dependence of dc magnetic susceptibility measured under $H = 1$ kOe and 10 Oe for Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ with solid symbols standing for FC and open ones for ZFC. No significant difference can be found between ZFC and FC data when $H = 1$ kOe. The inset displays the plot of $M^2$ vs. $H/M$ (Arrott plot) at various temperatures. (b) Field dependence of magnetization measured at $H = 1$ kOe and 10 Oe under a small field of 100 Oe, with solid symbols standing for FC and open ones for ZFC. No significant difference can be found about 12 K. Under an applied field of 100 Oe, a bifurcation between ZFC and FC curves can be observed below the temperature $T_C$ of Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ is about 12 K. Under an applied field of 100 Oe, a bifurcation between ZFC and FC curves can be observed below the temperature $T_f = 6$ K, where $T_f$ stands for the freezing temperature of individual spins or domain wall motion. As discussed in ref. [16,28], the bifurcation of ZFC and FC curves and the hysteresis loops can be found not only in regular ferromagnets but also in spin glasses. In typical spin glass systems, the moment size is usually small, i.e., \( \sim 0.01 \mu_B/\text{Mn} \) for the II-VI (Zn,Mn)Se and other typical diluted alloy spin glasses [29-31]. Considering that the magnetic moment of our system is 0.22 \( \mu_B/\text{Mn} \) under a small field of $H = 10$ Oe and can reach 0.80 \( \mu_B/\text{Mn} \) under $H = 1000$ Oe, we tentatively assign it to a ferromagnetic ordering rather than a spin glass. The small magnetization anomaly at $T \sim 320$ K should be due to the traceable MnAs impurity phase, [32,33] whose content is too small to be detected by X-ray diffraction. The large increase of magnetization and magnetoresistance (MR) below temperature of about 12 K cannot be attributed to the traceable MnAs impurity phase. In the inset (i) of fig. 2(b), we plot the $H/M$ vs. $T$ curve, which goes to zero at $T_C$ (around 12 K), consistent with the Arrott plot.

The magnetic field dependence of magnetization, i.e. $M(H)$ curves, of Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ at $T = 2$ K and 30 K is shown in fig. 2(b). At $T = 2$ K, the magnetization reaches 1.5 \( \mu_B/\text{Mn} \) under an applied field of $H = 50$ kOe, which is comparable with the result in Li(Mn,Zn)As [8] and (Ba,K)(Zn,Mn)$_2$As$_2$ [15]. At $T = 30$ K, the $M(H)$ curve indicates that the sample is mainly in a paramagnetic state, but the traceable MnAs impurity with FM order can be observed. As shown in the inset (ii) of fig. 2(b), the coercive field of Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ is less than 10 Oe at $T = 2$ K. Such a coercive field is even smaller than that of Li(Mn,Zn)As (30–100 Oe) [8], thus the material should be appealing in spin manipulation.

As shown in fig. 3, for the SrZn$_2$As$_2$ parent compound, the resistivity clearly exhibits a thermally activated behavior with decreasing temperature, and it increases beyond our measurement limitation below 70 K. The thermal activation energy ($E_a$) obtained by fitting with the thermal activation formula

$$\rho(T) = \rho_0 \exp(E_a/k_B T)$$

in the temperature range from 250 to 300 K is 0.16 eV, as shown in the inset of fig. 3. With 10% K-for-Sr substitution, $p$-type carriers can be introduced, and the resistivity of Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ decreases rapidly compared with the SrZn$_2$As$_2$ parent compound, but it still increases with decreasing $T$, which should be due to the localization effect. The Hall effect of Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ was also measured at $T = 300$ K and the charge carrier density is calculated to be $7.6 \times 10^{12}/\text{cm}^2$ by using the one-band model formula $R_H = 1/\rho e$. The positive Hall coefficient demonstrates that the hole-type charge carrier is indeed dominant in the system, consistent with that 10% K-for-Sr
substitution. However, the Hall coefficient is hard to measure at low temperatures, due to the extremely large resistivity.

We then measured the magnetoresistance (MR), defined in the standard way as \([\rho(H) - \rho(0)]/\rho(0)\), (the magnitude of MR could be much larger if it is defined in the other way as \([\rho(H) - \rho(0)]/\rho(H)\)), for Sr0.9K0.1Zn1.8Mn0.2As2, as shown in fig. 4. Without magnetic field, the resistivity is \(9.0 \Omega \text{ m at 2 K}\), which decreases rapidly with applied magnetic field. It reaches \(5.84 \Omega \text{ m under a low field of } \mu_0 H = 0.1 \text{T (MR = -38%)},\) then to \(0.19 \Omega \text{ m under } \mu_0 H = 1 \text{T (MR = -98%)},\) and even to \(0.018 \Omega \text{ m under } \mu_0 H = 5 \text{T (MR = -99.8%)}.\) Such a colossal magnetoresistance has only been observed in a limited number of systems (represented by (Ga,Mn)As and (La,Sr)MnO3) [5,18,27]. These materials have attracted much attention in the field of condensed matter physics.

In a typical manganite CMR system, i.e. (La,Sr)MnO3, the scattering by fluctuating local moments plays an important role in the charge transport [18], and thus MR is large around \(T_C\), but it usually becomes smaller far below \(T_C\) [34]. However, CMR in Sr0.9K0.1Zn1.8Mn0.2As2 keeps increasing below \(T_C\), which implies that the mechanism of CMR in this system could essentially be different. CMR in this system has two interesting features: 1) low coercivity field, 2) maximum in MR at temperatures far below \(T_C\).

A similar negative magnetoresistance was observed in p-(Zn,Mn)Te, which also occurred below \(T_C\) and was thought to be related with the localization effect [35]. On crossing the metal-insulator transition (MIT), the extended states become localized. However, according to the scaling theory of the MIT, their localization radius \(\xi\) decreases rather gradually from infinity at the MIT toward the Bohr radius deep in the insulator phase, so that on a length scale smaller than \(\xi\), the wave function retains an extended character [36]. Previous studies have demonstrated that ferromagnetic interactions can be mediated by the weakly localized holes showing an extended character [36,37].

In an Anderson localized system, the Fermi level is located on the localized side of the mobility edge. The application of a magnetic field introduces Zeeman shifts of each eigenstate dependent on spin directions and the repopulation among Anderson localized states. For one of the spin subbands, the mobility edge moves toward the Fermi level. This results in a negative magnetoresistance because of the exponential dependence of the wave function (\(\psi(r) \propto e^{-r/\xi}\)) overlap on the localization length \(\xi \propto (E_C - E)^{\nu}\), where \(E_C\) is the mobility edge and \(E\) is the energy of the localized state, \(\nu\) is an exponent on the order of unity [27,38]. We plot in fig. 5 the magnetic field dependence of conductivity for Sr0.9K0.1Zn1.8Mn0.2As2 under \(\mu_0 H = 3 \text{T in } \rho \text{ vs. } \log T\) plot. 

![Fig. 4: (Color online) Temperature dependence of resistivity (left axis) under \(\mu_0 H = 0, 1, 3, 5 \text{T and magnetoresistance (MR) (right axis), defined as } [\rho(H) - \rho(0)]/\rho(0)\), under \(\mu_0 H = 5 \text{T for Sr0.9K0.1Zn1.8Mn0.2As2}.\) The inset displays the field dependence of MR at \(T = 2 \text{K}\).](67007-p4)

![Fig. 5: (Color online) Magnetic field dependence of conductivity of Sr0.9K0.1Zn1.8Mn0.2As2 at \(T = 2 \text{K}\). Inset (a): resistivity of Sr0.9K0.1Zn1.8Mn0.2As2 under zero magnetic field in ln \(\rho\) vs. \(T^{-1/3}\) (2D-VRH) plot; (b) resistivity of Sr0.9K0.1Zn1.8Mn0.2As2 under \(\mu_0 H = 3 \text{T in } \rho \text{ vs. } \log T\) plot.](67007-p4)
and reduce the disorder of the system, and then resistivity exhibits the $\rho \propto -\log T$ behavior when the applied field $\mu_0 H \geq 3$ T (shown in the inset (b) of fig. 5), which might be attributed to quantum correlations to the conductivity in the weakly localized regime [37,42,45]. All the results indicate that our studied system can be well described by the localization theories, and the negative CMR could be understood based on the Anderson localization.

Since most applications of the MR effect require operating magnetic fields of less than several hundreds of Oe, to reduce the required magnetic field for CMR in manganeseites has been a major goal [21,22]. In the studied Sr$_{0.9}$K$_{0.1}$Zn$_{1.8}$Mn$_{0.2}$As$_2$ system, MR reaches $\sim 38\%$ only under an applied field of 1000 Oe. The low-field MR may stem from the spin-dependent scattering at domain walls or grain boundaries in our polycrystalline sample [21,22]. An external field can align the magnetic domains, and thus the spin-dependent scattering will be reduced, resulting in a negative MR. Moreover, under a high magnetic field, based on the Anderson localization theory, a delocalization effect occurs, and thus a negative colossal magnetoresistance is induced.

It should also be mentioned that the CMR effect is often accompanied by a metal-insulator phase transition induced by applying the magnetic field near magnetic order temperatures as observed in III-V–based (Ga,Mn)As induced by applying the magnetic field near magnetic or grain boundaries in our polycrystalline sample [21,22]. An external field can align the magnetic domains, and thus the spin-dependent scattering will be reduced, resulting in a negative MR. Moreover, under a high magnetic field, based on the Anderson localization theory, a delocalization effect occurs, and thus a negative colossal magnetoresistance is induced.

Concluding remarks. – In summary, we have successfully prepared a bulk hexagonal DNS, (Sr,K)(Zn,Mn)$_2$As$_2$, with $T_c$ of 12 K, and magnetic moment reaches about $1.5 \mu_B$/Mn under $\mu_0 H = 5$ T and $T = 2$ K. It is a soft magnet with a relatively small coercive field of less than 10 Oe. A low-field CMR, defined as $[\rho(H) - \rho(0)]/\rho(0)$, reaches $\sim 38\%$ under $\mu_0 H = 0.1$ T and up to $\sim 99.8\%$ under $\mu_0 H = 5$ T. Further studies suggest that the Anderson delocalization due to applying magnetic field may account for the observed CMR effect. With decoupled charge and spin doping, the hexagonal (Sr,K)(Zn,Mn)$_2$As$_2$ combines semiconductor, ferromagnetism and colossal magnetoresistance, which makes it an appealing system.

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