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Magnetic and transport properties of III–V diluted magnetic semiconductor $\text{Ga}_{1-x}\text{Cr}_x\text{As}$

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Magnetic and transport properties of Cr-based III–V diluted magnetic semiconductor $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ Cr concentrations up to $x=0.10$ have been investigated. For all the films, no long-range magnetic order was observed down to 2 K. A sign of paramagnetic Curie temperature is positive, indicating that the dominant magnetic interaction between Cr atoms is ferromagnetic. The effective number of Bohr magneton is estimated to be 5.1 ± 0.4 , which is close to that of Cr^{2+} ion. The magnetization curve shows superparamagnetic behavior at low temperatures. The radius of the local spin order in $x=0.034$ is estimated to be 1.5–2.0 nm at $T=5$ K and it decreases with increasing temperature. From Hall effect measurements at room temperature, the magnitude of mobility is estimated to be less than $0.5 \text{ cm}^2/\text{V s}$. This low mobility strongly suggests that the hopping conductivity is dominant in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ films. © 2001 American Institute of Physics. [DOI: 10.1063/1.1359475]

I. INTRODUCTION

Recent progress in crystal growth techniques allow one to synthesize new III–V diluted magnetic semiconductors (DMS) such as $(\text{In}_{1-x}\text{Mn}_x)\text{As}$ (Ref. 1) and $(\text{Ga}_{1-x}\text{Mn}_x)\text{As}$.² Their unique magnetic properties and excellent matching with III–V optoelectronics and electronics materials make them very important for the development of a new class of spin-electronic devices. Contrary to II–VI DMSs, up to now III–V DMSs have been limited only to the Mn-based DMSs. Therefore it is quite interesting to synthesize new III–V DMSs with other magnetic transition metals.

Recently, we have succeeded in synthesizing a Cr-based III–V DMS $\text{Ga}_{1-x}\text{Cr}_x\text{As}$.³ Since the physical properties of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ have not been clarified, we report the magnetic and transport properties of a new Cr-based III–V DMS $\text{Ga}_{1-x}\text{Cr}_x\text{As}$.

II. EXPERIMENT

$\text{Ga}_{1-x}\text{Cr}_x\text{As}$ films were grown by a low-temperature molecular beam epitaxy (LTMBE) method. A GaAs buffer layer was first grown on (001) GaAs substrates at a substrate temperature $T_S \sim 600^\circ\text{C}$. Then a 400-nm-thick $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ layer was grown at $T_S \sim 250^\circ\text{C}$ under the As to Ga beam flux ratio of ~ 30 . Cr concentration x was determined by an electron probe micro-analysis. A streaky reflection high energy electron diffraction (RHEED) patterns was observed for the films with $x < 0.10$. With increasing x , the RHEED patterns became spotty, indicating a roughening of grown films. Also, x-ray diffraction showed that the films with low Cr concentration ($x < 0.05$) are twins-free, but with increasing x the density of twins sharply increases. Magnetic circular dichroism (MCD) spectra were measured in a reflection mode under the magnetic field applied perpendicularly to the film.⁴

Magnetization measurements were carried out using a superconducting quantum interface device magnetometer (SQUID). To derive the magnetization of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ films, the magnetization of GaAs substrate was subtracted from the total magnetization.

III. RESULTS AND DISCUSSION

Figure 1 shows the MCD spectrum of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x=0.034$) in a magnetic field $H=10$ kOe at temperature $T=5$ K, together with that of $x=0.0$. The MCD signals for $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ are strongly enhanced near the energies corresponding to the Γ (~ 1.5 eV) and Λ (~ 3.0 eV) critical points (CP) of $x=0.0$. The MCD spectral shape of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ film is very similar to that of $\text{Ga}_{1-x}\text{Mn}_x\text{As}$.⁴ For Cr concentration $x > 0.10$, no enhancement MCD signals at the CP was observed. Therefore it can be concluded that $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x < 0.10$) is a DMS with a zinc-blende type band structure and the strong sp - d mixing occurs at each CP.

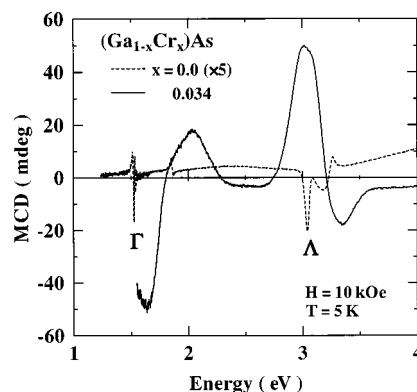


FIG. 1. MCD spectrum of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x=0.034$) in $H=10$ kOe at $T=5$ K (solid line) together with that of $x=0.0$ (broken line) for comparison.

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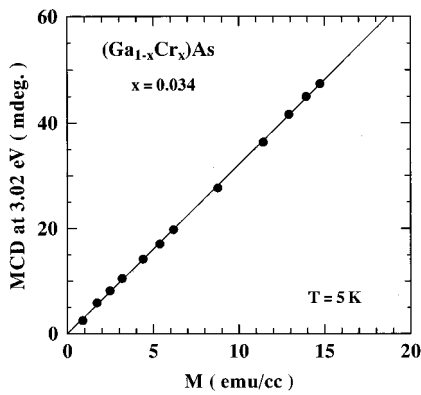


FIG. 2. MCD amplitude of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x=0.034$) at the Λ critical point (3.02 eV) as a function of the magnetization of the sample measured by SQUID.

Since the MCD signal at the Λ CP comes from $\text{Ga}_{1-x}\text{Cr}_x\text{As}$, its amplitude should be proportional to the magnetization M of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$. To determine whether the magnetic impurities or precipitation exist in our samples, we compare the field dependence of MCD and M of the samples measured by SQUID. Figure 2 shows the MCD amplitude at 3.02 eV of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x=0.034$) at $T=5$ K as a function of M . The data are on a single straight line, indicating that the measured magnetization comes from $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ and is free from the possible magnetic precipitations.

The temperature dependence of susceptibility χ and inverse susceptibility χ^{-1} for $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x=0.034$) in $H=10$ kOe are given in Fig. 3. The shape of the curve for χ shows a Curie–Weiss type paramagnetism and no long-range magnetic order down to 2 K. Fitting χ^{-1} versus T curve using a Curie–Weiss law, the effective number of Bohr magneton P_{eff} and paramagnetic Curie temperature θ_p are obtained to be 5.1 ± 0.4 and $15 \text{ K} \pm 5 \text{ K}$, respectively. The value of P_{eff} is close to that of a Cr^{2+} ion (4.9), suggesting that Cr ions act as acceptors in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$. It should be remarked that a sign of θ_p is positive, indicating that the dominant magnetic interaction between Cr ions is ferromagnetic. Moreover, the χ^{-1} versus T curve deviates from a Curie–

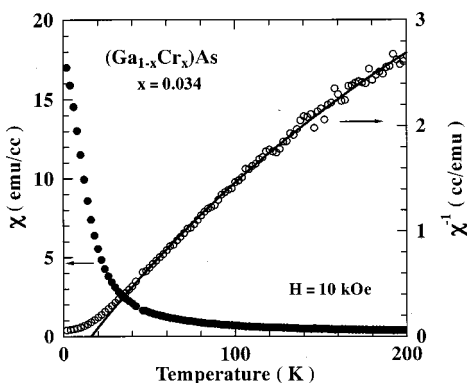


FIG. 3. Temperature dependence of susceptibility χ (●) and inverse susceptibility χ^{-1} (○) for $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x=0.034$) in $H=10$ kOe. The solid line shows a fitting result by a Curie–Weiss law.

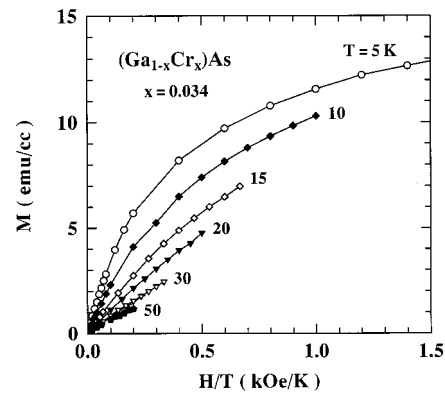


FIG. 4. Magnetization vs H/T curves for $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x=0.034$).

Weiss law below about $T=30$ K, implying that the effect of the ferromagnetic interaction becomes pronounced at low T .

To clarify the magnetic ground state of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$, we measured magnetization curves as a function of T . Figure 4 shows M versus H/T curves of $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ ($x=0.034$). Fitting the curve by the Brillouin function at $T=5$ K, the value of effective spin S_{eff} is obtained to be 20–50, showing superparamagnetism in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$. Using a spin of Cr^{2+} ion $S_{\text{Cr}}=2$ and the lattice constant $a=0.565$ nm, the radius of the local spin order is estimated to be 1.5–2.0 nm at $T=5$ K for $x=0.034$. If S_{eff} does not change with T and H , the dependence of M versus H/T should follow a single Brillouin function irrespective of T . However, from Fig. 4, M versus H/T curves clearly change with T . Also, M decreases with increasing T . This indicates that the size of the local spin ordering in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ decreases with increasing T .

Hall voltages for $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ at room temperature is too small to determine the conduction type. The magnitude of mobility is roughly estimated to be less than $0.5 \text{ cm}^2/\text{V s}$. Such a low mobility strongly suggests that the hopping conductivity is dominant. On the other hand, the p -type conduction was clearly observed in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$, even the valence of most of the Mn ions is the same as Cr ions, that is, Mn^{2+} .⁵ This difference in type of conductivity is qualitatively explained by the difference of the isolated acceptor level of the magnetic ion, that is, an acceptor level of Cr in GaAs [0.89 eV (Ref. 6)] is several times as deep as that of Mn [0.11 eV (Ref. 7)].

There are two possible origins for the observed ferromagnetic interaction between Cr in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$. One is the carrier-induced interaction.⁸ A band calculation⁹ predicts the carrier-induced ferromagnetic interaction between localized spins in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ as the same as $\text{In}_{1-x}\text{Mn}_x\text{As}$ and $\text{Ga}_{1-x}\text{Mn}_x\text{As}$. The other one is the superexchange interaction.⁸ At present, it is not clear which mechanism is responsible in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$. More detailed studies are needed to elucidate the origin of magnetic interaction in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$.

IV. CONCLUSIONS

In summary, we have investigated magnetic and transport properties of Cr-based III–V DMS $\text{Ga}_{1-x}\text{Cr}_x\text{As}$ having Cr concentrations up to $x=0.10$. No magnetic order was ob-

served down to 2 K in all the films. A positive sign of the paramagnetic Curie temperature indicates that the dominant interaction between Cr ions is ferromagnetic. The effective number of a Bohr magneton is close to that of a Cr^{2+} ion, suggesting that Cr ions act as acceptors. The magnetization curve shows the superparamagnetic behavior with a large effective spin at low temperature T . The radius of the local spin order in $x=0.034$ at $T=5$ K is calculated to be 1.5–2.0 nm and it decreases with increasing T . Low mobility at room temperature estimated from Hall measurement suggests the hopping conductivity is dominant in $\text{Ga}_{1-x}\text{Cr}_x\text{As}$.

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