Raising the Curie Temperature of GaMnAs through Annealing by Arsenic Ion Beams within MBE Chamber

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Ferromagnetic semiconductors, such as (Ga,Mn)As, are ground breaking materials that have potential to become novel spintronic devices, like magnetic random access memory (MRAM) and spin-MOSFET, which can reduce energy needed to store memory in electronics devices. A current major obstacle that needs to be overcome is their low Curie temperature (T_c), the temperature at which the material loses its ferromagnetic properties. Because most semiconductors function at room temperature, the T_c has to be at least higher than room temperature. However, the current maximum T_c of (Ga,Mn)As is still 100 K short of room temperature. One way to increase T_c is by annealing of the sample, which is known to remove interstitial manganese ions that neutralize the magnetization of (Ga,Mn)As. In this study, we used a novel way to anneal the sample by exposing the sample to an arsenic ion beam inside the molecular beam epitaxy (MBE) machine during the annealing process. It is expected that the excess Mn bonds with As molecules and leaves the (Ga,Mn)As, improving the sample quality. In the experiment, four (Ga,Mn)As samples were prepared with MBE. After the growth of the samples, three samples were annealed in the MBE chamber with an As beam for 10, 30, and 60 minutes, respectively, while another sample was not annealed. Surface structures of all samples were monitored during the growth with RHEED, indicating MnAs formed on the (Ga,Mn)As surface. The T_c of all samples were determined through magnetization measurements with SQUID and through the analysis of the data with theory using the Brillouin function. We found that the T_c of the samples tends to increase by annealing with Arsenic beams, as expected. These facts indicate that the annealing of the sample improves the sample quality, most likely because the Mn interstitial was removed by forming magnetic precipitates on the surface. However, the resulting T_c seems independent of the annealing time. In summary, we suggested a novel way to increase the Curie temperature of the ferromagnetic semiconductor (Ga,Mn)As through the method of annealing the sample inside the MBE chamber with arsenic ion beams.

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General Background

Ferromagnetic semiconductors

Ferromagnetic semiconductors (FS) like (In,Mn)As and (Ga,Mn)As are materials, which behave as both semiconductor and ferromagnetism.

FS was invented in 1989 by H. Munekata and recently attracted large interest due to its potential for the novel devices like Magnetoresistive Random Access Memory (MRAM) and spin-MOSFET.

The most important challenge of the study in FS is its low Curie temperature, which is much lower than the room temperature (< 180 K). This fact prevents investigating commercial applications using FS.

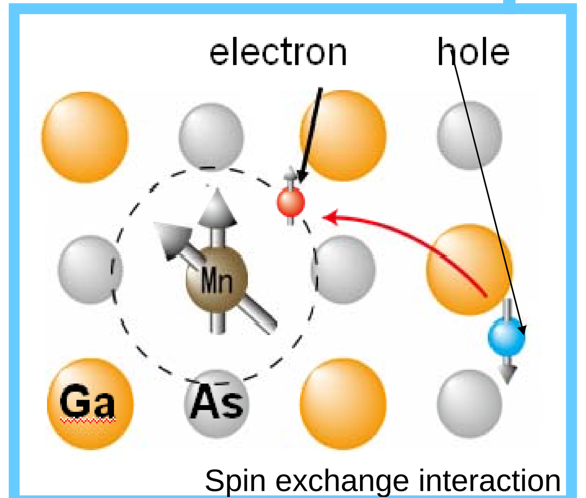
Properties of (Ga,Mn)As

What is Carrier Induced Ferromagnetism ?

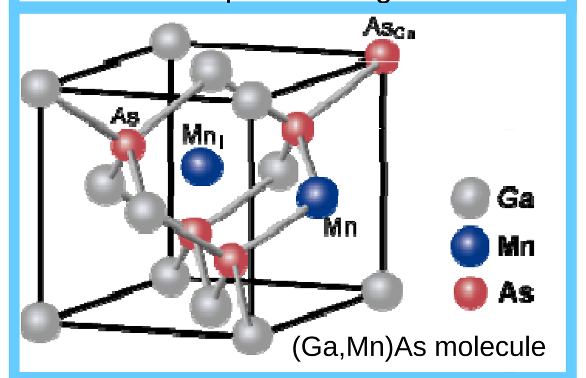
1. When a Ga site of GaAs is substituted by Mn ion, the Mn ion donates a hole to (Ga,Mn)As, in the spin exchange interaction diagram.
2. Spin-related interaction between a hole and a Mn ion orients their spins in anti-parallel configurations. This phenomenon is denoted as spin-exchange interaction
3. The result of spin-exchange interaction, causes ferromagnetic properties with Curie temperature is given by

$$T_c \propto xp^{1/3}$$

where *x* is the effective Mn ion density and *p* is the hole density. T. Dietl *et. al.* PRB **63**, 195205 (2001)



Spin exchange interaction



(Ga,Mn)As molecule

5. However, the interstitial Mn (Mn_i), Mn that resides in the middle of the molecule as shown in the (Ga, Mn)As diagram , acts as a double donor, which decreases *p* and thus decreases T_c. Also, when Mn_i is next to another Mn ion, the total spins of these Mn ions results to ± 0. The effective Mn concentration that contributes to the ferromagnetism decreases and T_c decreases.

Mn_i decreases T_c of (Ga,Mn)As

Purpose of this study

We here suggest a novel way to decrease Mn_i in the ferromagnetic semiconductor (Ga,Mn)As; annealing the sample inside of Molecular Beam Epitaxy (MBE) chamber. The reason of fabricating this technique is the following.

When the (Ga.Mn)As sample touch air, surface of the sample is oxidized and thus the sample quality is disturbed. Therefore, in order to make (Ga,Mn)As sample with multi-layer structure like quantum wells, all of the processes including growth and annealing, must be done inside of the MBE chamber.

Hypothesis

Annealing procedure

The point of increasing T_c by annealing is to decrease Mn_i. Usually, the annealing has been performed in the atmosphere, while we here propose the following novel procedure.

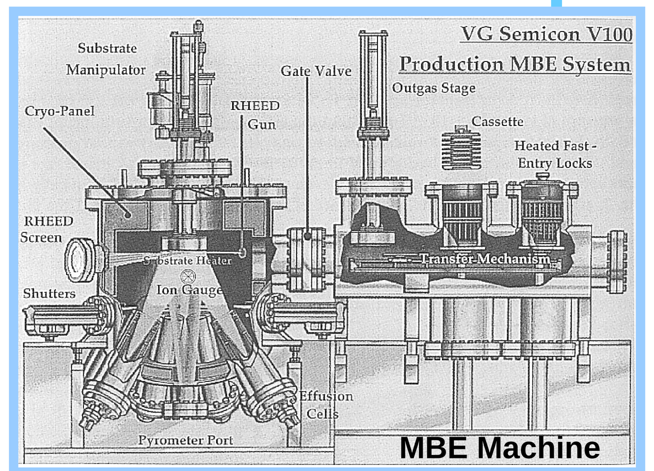
1. (Ga,Mn)As samples are grown in MBE chamber.
2. The sample inside of the MBE chamber is heated at 230 °C under Arsenic ion beam illumination.

In this condition, we expect that the phenomena shown in the following realized. By heating the sample, Mn_i moves about the (Ga,Mn)As layer. When the Mn_i is at the surface of the sample, the Mn_i interact with As, which is shined by As beam source, and forms precipitate like MnAs. These processes, which decrease Mn_i, is expected to increase T_c, as in the conventional way of annealing in the atmosphere.

Process

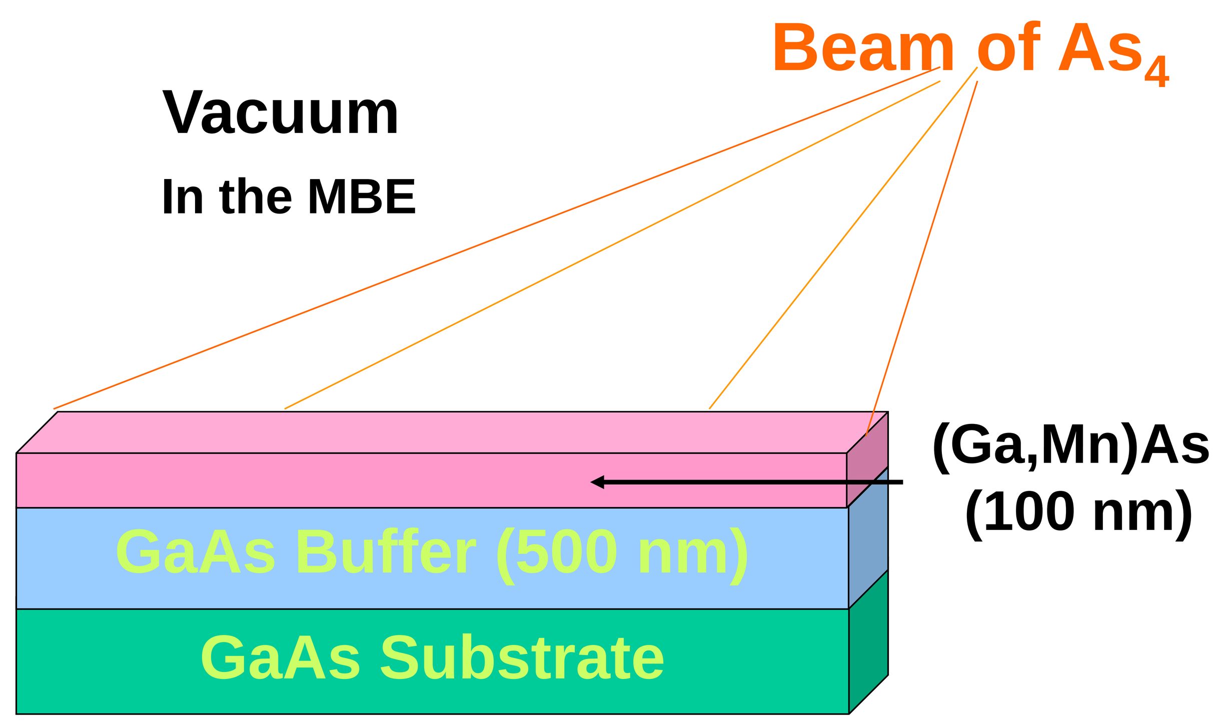
Sample Growth

1. Each layer was grown on the GaAs substrate layer by MBE shown on the right.
2. The substrate in which the sample is grown on is inserted in the right chamber on the picture, passing through many vacuum gates.
3. GaAs buffer layer was grown to smoothen the surface of the substrate.
4. (Ga,Mn)As layer was then grown at the substrate temperature of 230 °C and the Mn content ~ 5 %.



MBE Machine

As Annealing and RHEED



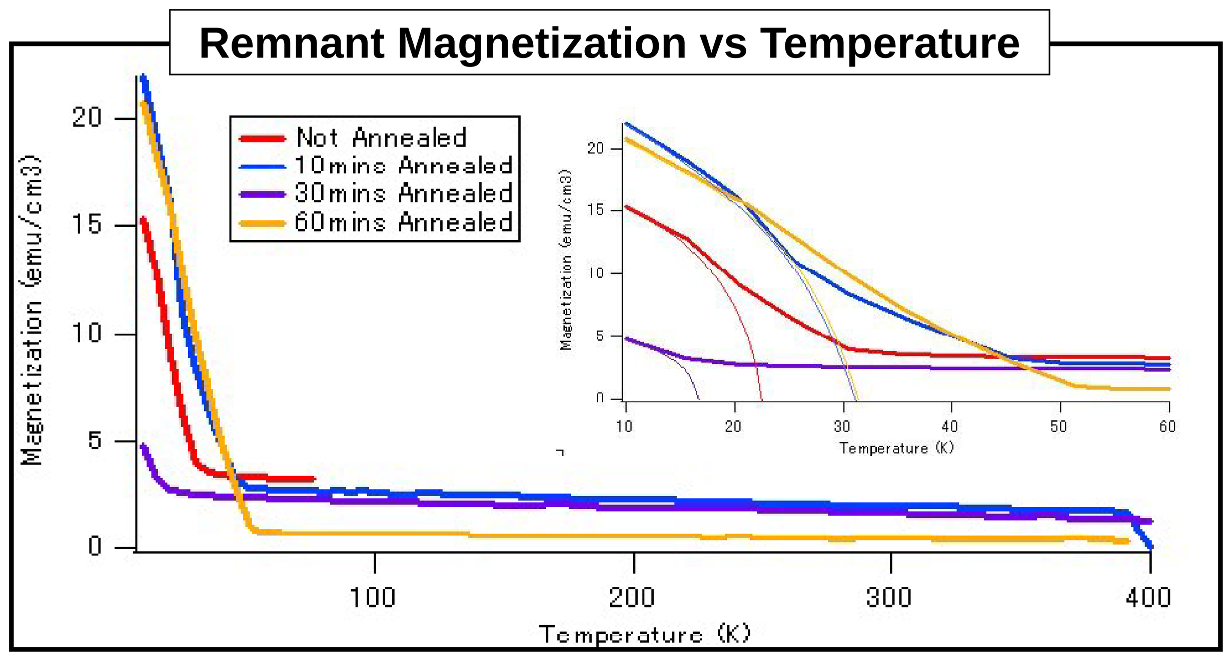
1. The sample is annealed at 230 °C under shining As_4 beam.
2. Three samples grown in the same condition were annealed in the different annealing time of 10 mins, 30 mins, and 1 hour.
3. The annealing process was monitored with reflection high-energy electron diffraction (RHEED).

Analysis/ Results

Determination of Curie temperature

Temperature dependence of magnetization is observed with Superconducting Quantum Interference Device (SQUID).

The Curie temperature of the samples were extrapolated with two procedure of visual guess and fitting of the data with a model calculation using Brillouin function.



Annealing Time	Visual Guessing	Fitting
0 min	32K	22K
10 mins	50K	32K
**30 mins	16K	16K
1 hour	51K	32K

**During the growth of this sample, it was shown that there was no change on this sample. Because of this, we choose to ignore this data point.

RHEED Analysis

We monitored the growth surface with RHEED. The graphs showed a change in surface materials for all samples as the material is annealed, indicating that something, most probably MnAs, is formed on (Ga,Mn)As during annealing.

These two results may indicate that the annealing of the sample successfully eliminates Mn_i as expected, while further confirmation is strongly needed!

Summary

A novel procedure for annealing sample, which is applied to (Ga,Mn)As sample in MBE chamber has been suggested.

Systematic investigations of the annealing effects with SQUID and RHEED indicate that Mn_i in (Ga,Mn)As samples were successfully eliminated by the annealing, and results in an increase in T_c, while further confirmation is strongly needed.

RICE
SUMMER NANOTECHNOLOGY STUDY PROGRAM IN JAPAN

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