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# Two Phase Spin Reversal Process in Co/Si/Co Trilayer Grown on GaAs(001)

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## Abstract

Three times larger coercivity of Co/GaAs(001) bilayer compared to that of Co/Si(001) led us to fabricate a multilayer of Co/Si/Co/GaAs(001). The magnetization process of the multilayer was studied as a function of the thickness of the Si spacer layer. The coercivity of top Co layer on Si was found to get decreased with the increase in the thickness of Si spacer layer. Perpendicular resistance of the multilayer was also increased with increasing Si spacer layer thickness. We suggest that localized electronics defect states in the gap of amorphous Si modulate the magnetic properties of the multilayer. Hysteresis loop was changed from two phase to single phase with decreasing temperature and Si spacer layer thickness. The nice correspondence between the magnetoresistance peak and the flat-field region found in the magnetization curves provided the direct evidence of the existence of antiparallel spin states in the two Co layers through the Si spacer in Co/Si/Co/GaAs(001) multilayer.

**Keywords**: Antiparallel spin state; Ferromagnet-semiconductor multilayer; Co/Si/Co trilayer; Spin reversal; Two-phase hysteresis loop

## 1. Introduction

In addition to mass and charge, electrons have another intrinsic quantity - angular momentum i.e. spins. Traditional electronic devices rely on the transport of charge carriers (electrons or holes) in semiconductors, such as silicon and germanium. For a long time, the spins of electrons have been ignored in the mainstream semiconductor electronics. The use of the spin degree of freedom of the electron in addition to its charge in spintronic devices promises additional functionalities, increased data processing speed and integration densities, as well as reduced power consumption [1-2]. The study of ferromagnetic thin film structures and their interfaces with semiconductor substrates has received much attention due to their potential technological applications as non-volatile magnetic memories, and in the field of spintronics [3-7]. The multilayers of *ferromagnet-semiconductor* hybrid structures have been the object of great recent interest. In such multilayer, a nonmagnetic semiconductor layer is sandwiched between two ferromagnetic layers. It is reported that the magnetic coupling between two ferromagnetic layers through amorphous Si spacer layer depends on the thickness of the Si spacer layer [8]. They found that the ferromagnetic layers are exchange coupled for Si spacer layer thicknesses equal to or less that 4 nm. For the thicknesses range of 1.4-2.2 nm they found antiferromagnetic coupling. Recent study has shown that the magnetic properties of Co/Ge/Co hybrid structure depend on temperature [7]. Their study revealed that in low magnetic field the magnetization was practically equal to zero up to some temperature ($T_c$). The magnetization increased sharply above the $T_c$ and began to saturate. The magnetic properties of *ferromagnet-semiconductor* multilayer are not only affected by the thickness of Si spacer layer but also the thickness of ferromagnetic layer itself. Patrin et al [9] showed that the thickness of ferromagnetic Fe films affects the magnitude of the interlayer exchange interaction through nonmagnetic Si spacer layer. Haque et al [10] investigated the magnetic properties of Ni/Si/Ni/GaAs multilayer and they reported the occurrence of spin reversal in two phases in the ferromagnetic layers through Si spacer layer. They observed antiparallel spin state below 250 K which became the strongest at the lowest (1.8 K) temperature. The main factor determining the read access time in a memory device is the signal-to-noise ratio. A new MRAM cell pseudo spin valve (PSV), capable of storing data at very high densities and giving improved signal levels, is suitable for either GMR memory or tunneling memory. The main advantage of PSV is that at the rest state of magnetizations, two ferromagnetic layers are antiparallel, giving small self-demagnetizing fields and small external stray fields on nearby memory cells, both of which could enhance cell stability. A PSV cell would have about 1/10 of the stray and self-demagnetizing fields of conventional spin valve cell where the magnetizations can be in the same direction within a cell when the memory is not being addressed. Therefore, the understanding of two phase hysteresis loop of PSV structure is very important in the field of magnetic memory applications. In this study the ferromagnetic Co are separated by semiconductor spacer (Si) layer instead of nonmagnetic metal. It is well known that the carrier density of semiconductor layer can be controlled precisely. The magnetic and magnetoresistive properties of *ferromagnet-semiconductor* multilayer can be modulated by the transport properties of semiconductor spacer layer [5-6, 11-12]. In this paper, we report two phase hysteresis loop found in Co/Si/Co/GaAs(001) multilayer and the effect of Si spacer layer thickness on magnetic properties of the multilayer. The correspondence between magnetization and magnetoresistance curves indicate the existence of an antiparallel spin state, which may have practical importance in connection to the giant magnetoresistance.

## 2. Experimental Details

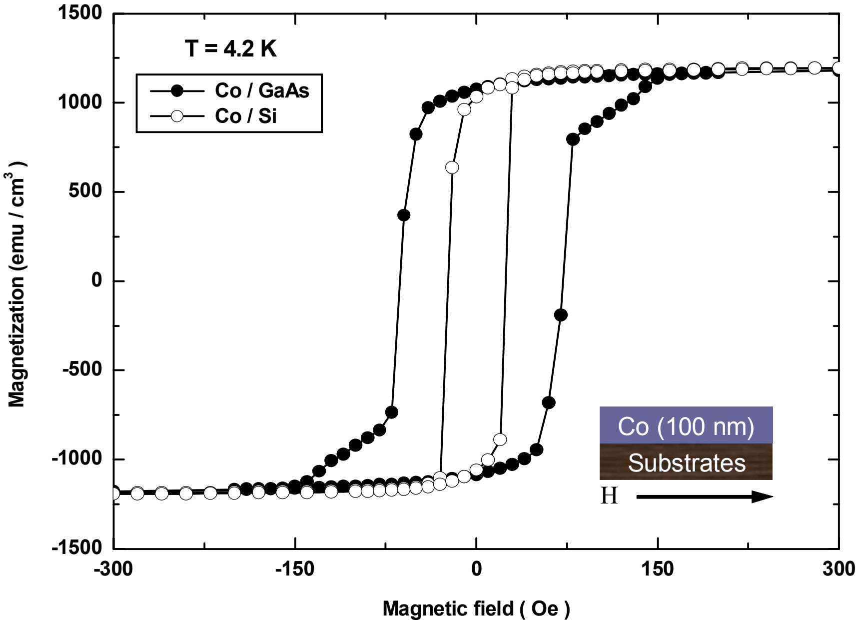
Before depositing Co (99.9% pure: Cojundo Chemical Lab. Co., Japan) thin films, the substrates were chemically etched by semiconductor grade organic compound (Semico-23-clean, Japan) and rinsed with deionized water. The GaAs(001) substrate (GIRMET Ltd., Russia) was Si-doped, with an electron carrier concentration of $1\times10^{24}$ $m^{-3}$. The Co films were deposited at a rate of 1.5 $Ås^{-1}$ onto the GaAs(001) substrate by e-beam (OSAKA VACUUM: VCB2-200E, Japan) evaporation method at a base pressure of $5\times10^{-6}$ Torr. On the other hand, the deposition rate of Si (99.9999% pure, NewMet, UK) was only 0.3 $Ås^{-1}$. The substrate was kept at room temperature during the film deposition. In this experiment, a multilayer was prepared by depositing 125 nm Co film on GaAs(001) and a subsequent deposit of 50 and 50 nm Si and Co films, respectively. The structure of the multilayer finally became Co(50 nm)/Si(50 nm)/Co(125 nm)/GaAs(001). To investigate the effect of Si spacer layer thickness on magnetic properties of the trilayer, the thickness of Si was varied in the range of 5-70 nm. The film thickness was measured by a quartz crystal oscillator (ULVAC: CRTM-7000, Japan), which was calibrated by an atomic force microscope (JSPM-5200: JEOL Scanning Probe Microscope). In X-ray diffraction measurement (MAC Science, Japan), the Co films grown on n-GaAs(001) substrate exhibited polycrystalline *hcp* structure. The measurements were performed using an X-Ray diffractometer with Cu $K\alpha_1$ radiation. A superconducting quantum interference device (SQUID-Quantum Design) with a moment resolution of the order of $10^{-9}$ emu was used to measure the magnetization after cooling the sample to the desired temperature in absence of any magnetic field. In order to investigate the effect of field cooling on magnetic properties, a DC magnetic field of 100 mT was applied during the cooling process but no significant difference was observed. The magnetic field was applied parallel to the film plane, which was [100] crystallographic direction of the substrate. The magnetoresistance was measured using a Physical Property Measurement System (PPMS: Quantum-Design, Model-6000).

## 3. Result and discussion

### 3.1. Co/GaAs(001) and Co/Si(001) Bilayers

The magnetization process of Co thin films on both the GaAs(001) and Si(001) substrates are shown in Fig. 1. The magnetization process of the Co film deposited onto GaAs substrate was found to be quite anomalous at low temperature. The sample (100 nm Co film) exhibited high coercive field, $H_c$ (85 Oe at 4.2 K) and high saturation field, $H_s$ (larger than 140 Oe at liquid helium temperature). The squareness, $M_r/M_s$ of the sample was found to be as high as 0.94. The corresponding magnetic properties of the same thickness Co film on Si were comparatively lower ($H_c = 30$ Oe, $H_s = 30$ Oe and $M_r/M_s = 0.85$ observed at 4.2 K). The value of coercivity found in Co/Si(001) was very close to the reported value of coercivity of Co films grown onto glass as well as onto Si(111) substrates [13-14]. The coercivity of Co films onto Si(001) substrate remained constant within the thickness range of 20-100 nm. On the other hand, the coercivity increased with increasing film thickness in the case of Co films deposited on GaAs substrate. It was also noticed from the figure that the hysteresis loop of Co/GaAs is quite different than the hysteresis loop observed in Co/Si. Further details of the anomalous magnetization behavior observed in Co/GaAs(001) and its possible reasons could be found elsewhere [15].

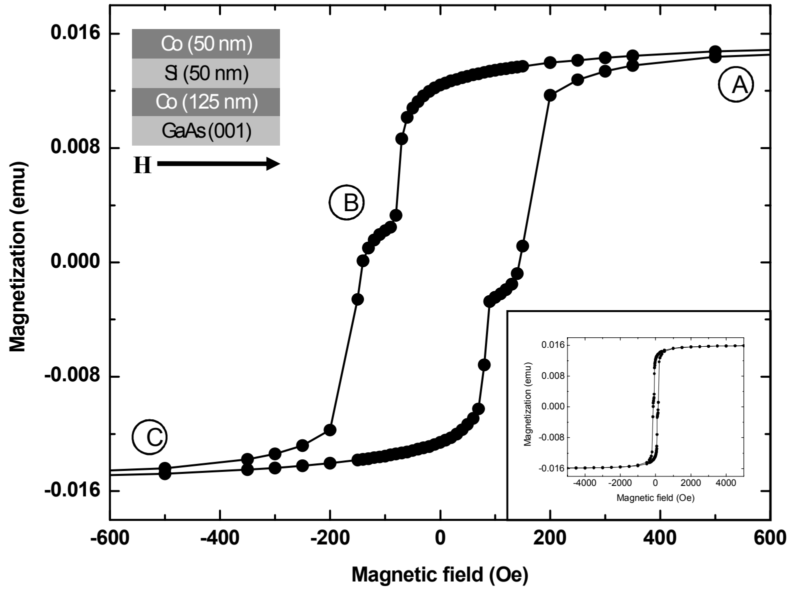
**Figure 1:** Magnetization process of 100 nm Co film on both the GaAs(001) and Si(001) substrates observed at 4.2 K. The direction of the applied magnetic field was parallel to the film plane as shown in the figure.



### 3.2. Two Phase Hysteresis Loop of Co/Si/Co Trilayer

It became clear that the Co films had dissimilar coercivities on GaAs and Si substrates. These dissimilar coercivities lead to the fabrication of a multilayer of Co/Si/Co/GaAs in particular to achieve an antiparallel spin state in it. The magnetization process of the multilayer was measured at liquid helium temperature in an applied magnetic field parallel to the film plane and is presented in Fig. 2. The magnetization curve at high magnetic field value is shown in the inset, which shows quite typical ferromagnetic characteristic. However, the hysteresis loop was interesting since it reflected the resultant of the two dissimilar characteristics of Co/GaAs(001) and Co/Si(001). The spins in both hard (125 nm Co) and soft Co (50 nm Co) layers were unidirectional at positive remanent point (H=0) on the M-H curve and around point A. The soft Co (top Co) layer altered its spin direction at a negative field value of 75 Oe. This corresponded to the sharp vertical fall of magnetic moment on the M-H curve. The negative applied magnetic field at this point was still trying to alter the magnetization direction of the hard Co layer (125 nm Co) on the GaAs substrate. The antiparallel spin state existed in the multilayer (the magnetic field region around the point B) until the magnetization reversal took place in the hard Co layer. After the first magnetization reversal, there was a short region of magnetic field having a tendency to be flat. The antiparallel spin state existed in this field-region. The moment was positive in this field- region. This was due to the fact that the magnetic moment data from the SQUID was a resultant of both the Co layers. The magnetization reversal of the hard Co layer (bottom Co) occurred at a magnetic field value of 140 Oe and the spins in both the Co layers became unidirectional at around C. The coercivity of soft Co layer on Si in the multilayer was enhanced than that of Co/Si bilayer. The coercivity of 50 nm Co on Si(001) was found to be 30 Oe. But in the multilayer it was increased to 75 Oe. This was due to the fact that the Si spacer layer in the multilayer was amorphous. But the Si(001) substrate in the bilayer was single crystal. The coercivity of 125 nm Co film on GaAs(001) was 120 Oe. On the other hand, the coercivity of the same thickness Co layer on GaAs(001) in the multilayer was increased to 140 Oe. The hard Co layer had two contacts namely Co/GaAs(001) as well as Co/Si. Thus, the hard Co layer might be under the influence of both semiconductors. To understand the origin of the enhancement of coercivity of hard Co layer in the multilayer, a trilayer Si(10 nm)/Co(125 nm)/GaAs(001) was prepared and its magnetization was measured. It was found from the hysteresis curve that the coercivity of 125 nm Co film in the trilayer increased to 140 Oe. Although the thickness of Si spacer layer in the multilayer was 50 nm, other study showed that the coercivity of hard Co layer in the multilayer was independent of Si layer thickness.

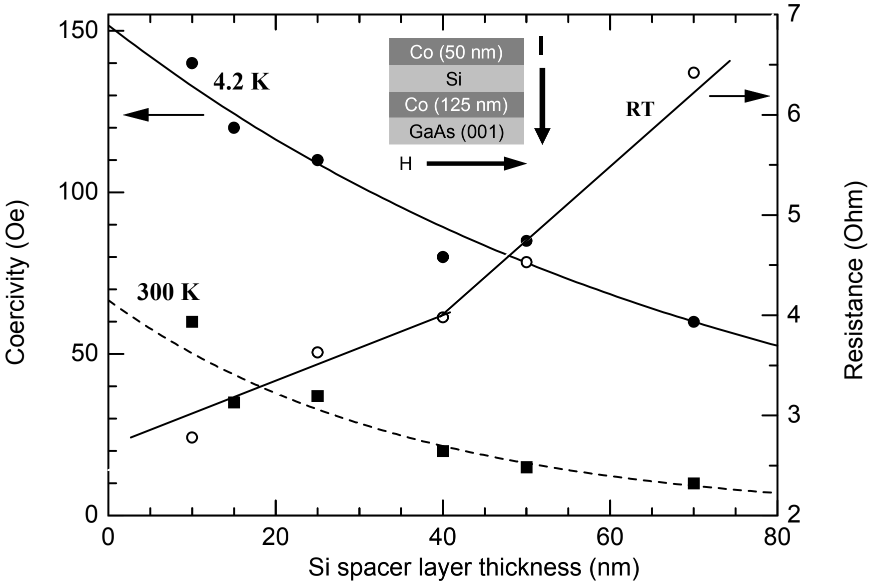
**Figure 2**: Magnetization process of Co/Si/Co/GaAs(001) multilayer measured at 4.2 K. Thicknesses of hard (125 nm) and soft (50 nm) ferromagnetic layers and the semiconductor interlayer (50 nm) and magnetic field direction are shown in the figure.



### 3.3. Effect of Si layer thickness on magnetization process

The two phase hysteresis loop depended not only on the temperature but also on the Si spacer layer thickness. At thinner Si spacer layer, two Co layers were coupled strongly to each other and the resultant hysteresis loop became single phase through all the temperature ranges studied. But the coupling strength between two Co layers decreased as the Si spacer layer thickness increased and an antiparallel spin state exist in the multilayer having thicker Si spacer layer in the temperature range of 4.2-300 K. Although the mechanism of antiferromagnetic coupling in metal/metal multilayer is well-established, it is still not clear how the mechanism can be modified for coupling across semiconducting spacer layer. The interlayer coupling strength between ferromagnetic layers exponentially decays with Si spacer layer thickness [6]. There is also report that with increasing Si spacer layer thickness the coupling oscillates from ferro- to antiferromagnetic and back to ferromagnetic [16]. In this study, the coupling strength between Co layers was found to decrease with increasing Si spacer layer thickness. Spacer (Si) layer thickness dependency of two phase hysteresis loop found in the multilayer could be better quantified by measuring Si layer thickness dependent coercivity at various temperatures. The coercivity of soft Co layer in the multilayer varied with Si layer thickness. Fig. 3 shows Si layer thickness-dependent coercivity of soft Co layer and perpendicular resistance of the multilayer at various temperatures. The coercivity of soft Co (top Co) layer decreased from 140 to 60 Oe as the Si spacer layer thickness increased from 10 to 70 nm observed at 4.2 K. It is worthy of mention that the coercivity of hard Co layer (bottom Co) was thickness independent (Si layer). Whereas it is decreased from 60 to 10 Oe as the thickness of the Si spacer increased from 10 to 70 nm measured at room temperature. On the other hand, the perpendicular resistance of the multilayer increased with increasing Si layer thickness but it showed two different slopes in the curve. The first slope lied in the thickness range of 10-40 nm, whereas the second slope lied in the thickness range of 40-70 nm.

**Figure 3**: Spacer layer (Si) thickness-dependent coercivity of soft Co layer (50 nm Co/Si) and perpendicular resistance of the multilayer. The current was perpendicular to the film plane but the magnetic field was parallel to the film plane.

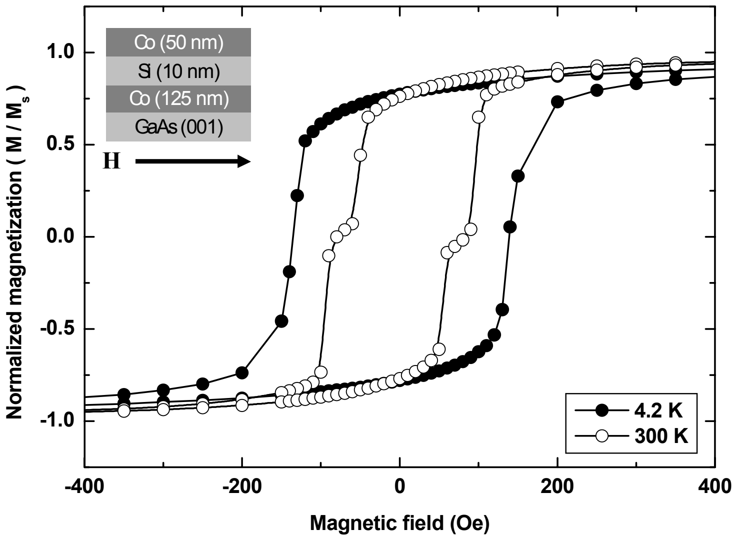


It is known that cobalt silicide phase formed at the interface of Co and Si [4,6]. The electrical conductivity of cobalt silicide is much higher than that of Si semiconductor. Therefore, the two slopes found in the curve can be explained with the help of silicide formation at the interfaces. The effective resistance of the multilayer was low at thinner Si spacer layer because of the cobalt silicide formation at both sides of Si layer. Thus, the total resistance is low in the Si thickness region of 10-40 nm. However, the interface width was limited to a certain extent in the Si layer. The effect of cobalt silicide on the total resistance of the multilayer became less dominant as the Si layer thickness increased. Thus, the slope of resistance curve in the thicker Si layer (40-70 nm) was increased. It appeared from the above results that certain electronic properties like carrier density in the amorphous Si modulate the magnetic properties of the multilayer. We suggest that localized electronic defect states in the energy gap of amorphous Si modulate the magnetic properties of soft Co layer in the multilayer. One could ask the question why the coercivity of hard Co layer was not influenced by the thickness of Si spacer layer. This could not be well answered from Fig. 3. But it could be well answered from the magnetization of Si/Co/GaAs(001) trilayer. The coercivity of Co layer changed a little bit due to the influence of Si spacer layer. In the trilayer, Co film exhibited the threshold phenomenon and it also kept the nature of the magnetization process unchanged. No sign of the two steps magnetization reversal like the Co/Si/Co/GaAs(001) was observed. This observation of the trilayer confirmed that the hard Co layer of the multilayer was almost under the influence of the GaAs(001), but the soft Co layer was indeed under the influence of Si layer. It could be worth of studying a trilayer by altering the deposition order, GaAs/Co/Si instead of Si/Co/GaAs. But, deposition of GaAs on Co was quite uncertain in the present deposition method. However, the study of magnetization process of Co/GaAs(001) at various thicknesses showed that GaAs(001) had much influence on the coercivity of Co films [15].

### 3.4. Effect of temperature on magnetization process

It was found that at and below a critical thickness of 5 nm (Si), the hysteresis loop appeared in single phase in the temperature range of 4.2-300 K. It was found from Fig. 3 that the coercivity of top Co layer became comparable with the coercivity of bottom Co layer in the multilayer having thinner Si spacer. Therefore, it showed single phase hysteresis loop in the temperature range studied. When the thickness of Si spacer layer increased to 10 nm, the two phase hysteresis loop became single phase in the temperature range of 4.2-100 K. The same sample showed two phase hysteresis loop in the temperature range of 200-300 K (Fig. 4).

**Figure 4**: Magnetization process of Co/Si/Co/GaAs(001) multilayer measured at 4.2 K. Thicknesses of hard (125 nm) and soft (50 nm) ferromagnetic layers and the semiconductor interlayer (10 nm) and magnetic field direction are shown in the figure.

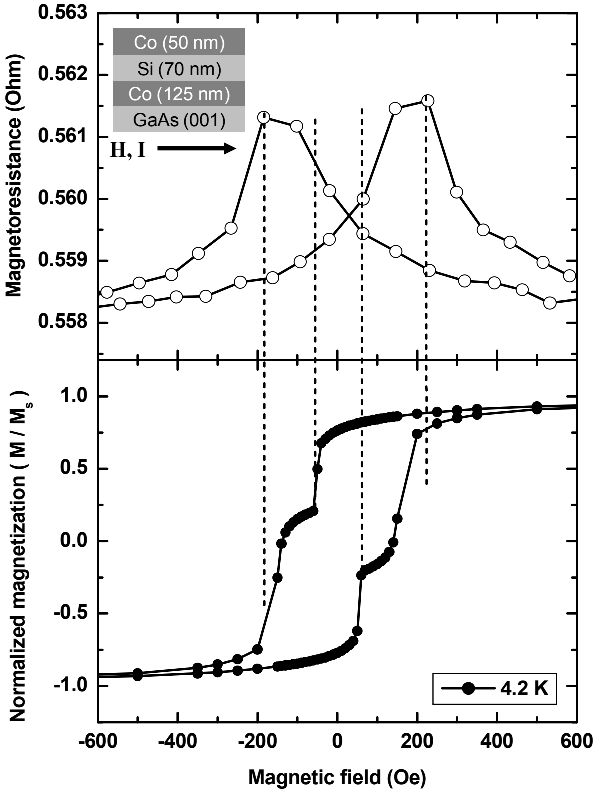


The coercivity of the top Co layer in the multilayer (with 10 nm Si spacer) was 140 Oe, which was exactly equal to the coercivity (140 Oe) of the bottom Co layer at 4.2 K. Thus, the multilayer showed single phase hysteresis loop at 4.2 K. When the temperature increased to 300 K, the coercivity of the top Co layer became 60 Oe, whereas the coercivity of the bottom Co layer decreased to 85 Oe. Due to the dissimilar coercivities of two Co layers, the multilayer showed two phase hysteresis loop. When the thickness of the Si spacer layer increased to 70 nm, two phase spin reversal occurred in all the temperature range (4.2-300 K) studied. This result can also be explained with the aid of Fig. 3. At 4.2 K, the coercivities of top and bottom Co layers were 60 and 140 Oe, respectively. The coercivities of top and bottom Co layers decreased to 10 and 85 Oe, respectively, at 300 K. Therefore, the multilayer having thickness of 70 nm Si spacer showed two phase hysteresis loop in the temperature range of 4.2-300 K. It became clear that certain electronic properties (properties like carrier density) of Si spacer layer modulate the magnetic properties of the multilayer. It was also found that temperature dependency of coercivity of top Co layer was much more than that of bottom Co layer. For example, in the multilayer having thickness of 70 nm Si spacer, the change in coercivities of top and bottom Co layers were 500% and 65%, respectively, as the temperature increased from 4.2 to 300 K. This was due to the fact that the coercivity of bottom Co layer was under the influence of GaAs(001) at all temperatures. On the other hand, it was found that the magnetic properties of top Co layer depended on the transport properties of the amorphous Si spacer.

### 3.5. Magnetoresistance Effect

The two phase spin reversal observed in Co/Si/Co/GaAs(001) multilayer was confirmed by the magnetoresistance measurement. The two phase hysteresis loop and its corresponding magnetoresistance curves are shown in Fig. 5. It was seen that the magnetoresistance became maximum when the moments in the two Co layers were antiparallel. On the other hand, the magnetoresistance became minimum when the magnetic moments in the two Co layers were parallel to each other. It was also noticed from the magnetoresistance curve that there appeared a short region of field having a tendency to be flat and was corresponding to the flat-field region in the magnetization curve. We have studied multilayer having different Si layer thicknesses and found that same results. The above results were due to the current parallel to the film plane (CIP) configuration. We have also measured the magnetoresistance in the perpendicular current (CPP) configuration and found the same results. Therefore, the nice correspondence between the magnetoresistance peak and the flat-field region found in the magnetization curves provided the direct evidence of the existence of antiparallel spin states in the Co/Si/Co/GaAs(001) multilayer. Therefore, the two phase hysteresis loop (antiparallel spin states) found in Co/Si/Co/GaAs(001) multilayer might be used in pseudo spin valve (PSV) based MRAM application.

**Figure 5:** Magnetic field dependent resistance and its corresponding magnetization curve of Co/Si/Co/GaAs(001) multilayer at 4.2 K. Both the magnetic field and electric current were parallel to the film plane.



## 4. Conclusion

We have presented two phase hysteresis loop found in Co/Si/Co trilayer deposited on GaAs(001) substrate. The two steps magnetization process in the multilayer confirmed that the spins in the two Co layers were aligned antiferromagnetically in the multilayer. The coercivity of the soft Co layer was found to decrease with the increase of Si spacer layer thickness. Perpendicular resistance of the multilayer increased with increasing Si spacer layer thickness. This result indicates that certain electronic properties of Si layer might modulate the magnetic properties of the multilayer. For 10 nm Si spacer layer thickness, the two phase hysteresis loop in the multilayer became single phase in the temperature range of 4.2-100 K. The same sample showed two phase hysteresis loop in the temperature range of 200-300 K. Spacer layer (Si) thickness-dependent coercivity of Co layer in the multilayer was able to give explanation of such phase transformation. Magnetoresistance became maximum when the two ferromagnetic Co layers aligned antiferromagnetically and it showed minimum value when the two Co layers aligned ferromagenetically. The nice correspondence between the magnetoresistance peak and the flat-field region found in the magnetization curves provoded the direct evidence of the existence of antiparallel spin states in the Co/Si/Co/GaAs(001) multilayer.

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