New Plasma Source With Low Electron Temperature for Fabrication of an Insulating Barrier in Ferromagnetic Tunnel Junctions

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***Abstract*—A new plasma source, characterized as low electron temperature of 1 eV and high density of 10^{12} cm^{-3}, is introduced to the Al oxidation process in the magnetic tunnel junction (MTJ) fabrication. The MTJ fabricated with this new plasma source shows a high magnetoresistance ratio of about 50%. As a peculiar feature, the monotonous decrease of resistance area (RA) product is observed with increasing the postannealing temperature of MTJ. The decrease of the RA product is due to the decrease of the effective barrier width, which is a favorable feature to realize a low-resistance MTJ.**

***Index Terms*—Annealing-temperature dependence, magnetic tunnel junction (MTJ), plasma source, tunnel resistance.**

I. INTRODUCTION

SINCE THE discovery of a large tunnel magnetoresistance (TMR) over 10% at room temperature [1], [2], magnetic tunnel junction (MTJ) has been a strong candidate for several applications. Up to the present, MTJs with TMR in excess of 40% have been demonstrated by several groups in a Co–Fe–Al–O–Co–Fe system [3]–[5]. These MTJs with large TMR are fabricated with a plasma oxidation process after the deposition of thin metallic Al layer. However, in general, the plasma oxidation process may introduce some defects in the insulating layer, due to the bombardment of ions in plasma [6]. When there exist some defect states in the barrier of the MTJs, electrons can tunnel the barrier via defect states, and such a two-step tunneling process [7] lowers the magnetoresistance (MR) ratio and the bias voltage V_h at which the MR ratio decreases to its half value [8].

Since the ion-bombarding energy strongly depends on the space potential of plasma, which is mainly determined by the electron temperature, one can expect to provide the Al–O layer without introducing any defects by using an oxidizing plasma with a low electron temperature.

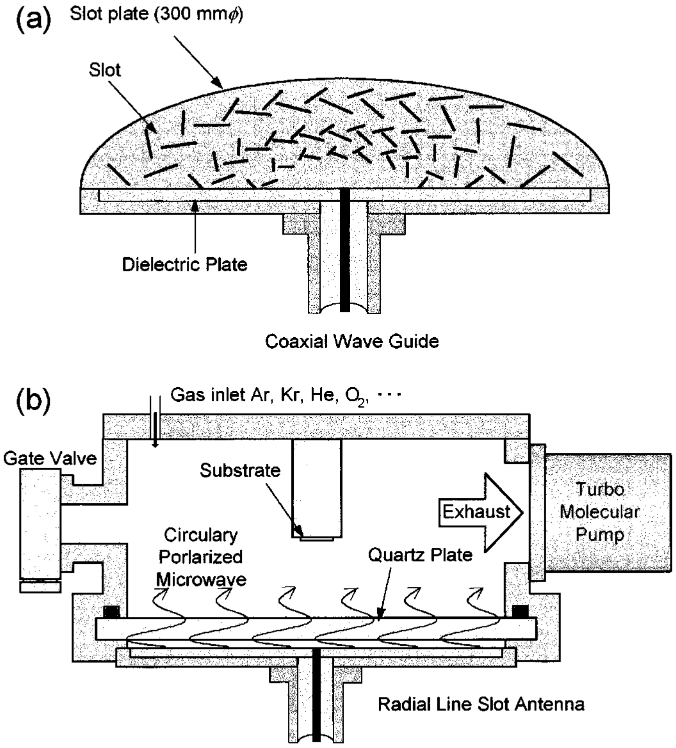

Fig. 1. Schematic illustration of (a) an RLSA and (b) newly introduced microwave-excitation plasma process equipment using an RLSA.

In the present study, we introduce a plasma source, which can produce a plasma, having low electron temperature, for the oxidation process of Al layer in the MTJs, and we investigate the magnetotransport properties of the MTJs as a function of the postannealing temperature.

II. EXPERIMENTAL PROCEDURE

Fig. 1(a) illustrates a radial line slot antenna (RLSA), and (b) shows a newly introduced microwave-excitation plasma process equipment using an RLSA [9], [10]. The RLSA, which is a waveguide planar array, consists of two metal disks separated by a dielectric plate, and the top one has many slot pairs that are the unit radiators of microwave. A 2.45-GHz microwave is introduced through a coaxial waveguide, and the microwave power is radiated from the slots into the vacuum chamber though the quartz window. A low electron temperature of 1 eV and high-density plasma of 10^{12} cm^{-3} is obtained at the substrate position, 50 mm distant from the quartz window [11].

Manuscript received February 13, 2002; revised May 22, 2002.

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Digital Object Identifier 10.1109/TMAG.2002.803167.

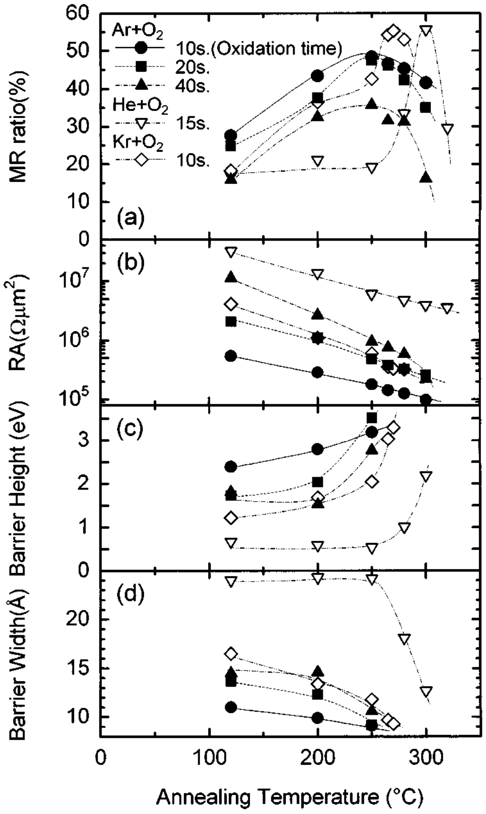

Fig. 2. Annealing-temperature dependence of (a) MR ratio, (b) RA product, (c) barrier height, and (d) barrier width for the MTJs fabricated under the different oxidation conditions.

The MTJs of substrate/Ta 50 Å/Cu 200 Å/Ta 200 Å/Ni-Fe 20 Å/Cu 50 Å/$Mn_{75}Ir_{25}$ 100 Å/$Co_{70}Fe_{30}$ 25 Å/Al-O/$Co_{70}Fe_{30}$ 25 Å/Ni–Fe 100 Å/Ta 350 Å/Cu 4000 Å/Ta 50 Å were prepared. All the metallic films were deposited by the dc magnetron sputtering method. The barrier formation was performed by depositing a 15-Å-thick Al film and subsequently oxidizing it in the oxidation chamber (described above). The applied microwave power density was 1.1 W/cm^2. A photolithographic process and ion milling were used to pattern the tunnel junction in a normal area of 36 μm^2–3600 μm^2. The MR measurements were performed with a four-point probe method at the bias voltage of 5 mV. The scaling of the resistance inversely with the area of the junction and the constant TMR regardless of the size of the junction exclude the possibility of geometrical enhancement of the TMR. The barrier height and the barrier width were obtained by fitting the current–voltage (I–V) curves with Simmons' formula [12]. The thermal treatment consisted of consecutive 60-min vacuum annealing at each temperature, followed by furnace field cooling (1 kOe).

III. RESULT AND DISCUSSION

Fig. 2 shows the annealing-temperature dependence of a) MR ratio, b) resistance area (RA) product, c) barrier height, and d) barrier width for the MTJs fabricated with several oxidation conditions. The detail oxidation conditions are shown in Table I.

For all MTJs, the MR ratio increases by thermal annealing and reaches a peak around the annealing temperature T_a of 250 °C to 300 °C. The MR ratio of 48.4% is obtained at $T_a =$ 250 °C in the case of the MTJ fabricated with Ar + O_2 gas and 10-s oxidation time. The V_h of 420 mV, separately measured, for the same MTJ indicates that a good quality barrier is formed with using the RLSA oxidation plasma process. When the mixed inert gas is changed from Ar to He or Kr in the plasma oxidation process, the achievable MR ratio during thermal annealing is enhanced to over 50%. The details of this enhancement have been reported in [13]. The monotonous decrease of the RA product in coincidence with the decrease of the barrier width with an increasing annealing temperature is the peculiar feature for the MTJs fabricated with the RLSA plasma oxidation technique. In the case of the MTJ fabricated by using 10-s oxidation of the Al layer with Ar + O_2 plasma, RA decreases from $5.4 \times 10^5\ \Omega \cdot \mu\text{m}^2$ to $1.8 \times 10^5\ \Omega \cdot \mu\text{m}^2$, and the barrier width decreases from 11–9 Å with increasing T_a from 120 °C to 250 °C. Even though the oxidation time and mixed inert gas are changed, the RA and the barrier width of MTJs show similar behaviors against T_a.

TABLE I
OXIDATION CONDITIONS OF MTJs

Gas	O_2 mixing ratio	Gas pressure	Oxidation time
Ar+O_2	3%	1 Torr	10, 20, 40 seconds
He+O_2	3%	1 Torr	15 seconds
Kr+O_2	3%	0.8 Torr	10 seconds

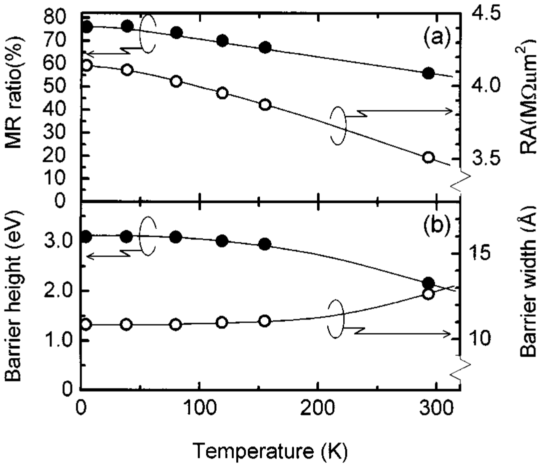

Fig. 3. Temperature dependence of (a) MR ratio and RA product and (b) barrier height and barrier width about the MTJ oxidized He + O_2 plasma for 15 s.

In order to clarify the mechanism of the RA decrease with increasing T_a, we investigated the temperature dependence of the magnetotransport properties for the MTJ after thermal annealing. Fig. 3 shows the changes of (a) MR ratio and RA and (b) barrier height and barrier width, as a function of the measuring temperature, for the MTJ oxidized by He + O_2 plasma and annealed at $T_a = 300$ °C. The MR ratio increases up to 76% with decreasing the temperature down to 4.2 K. The RA and the barrier height increase, and the barrier width decreases for decreasing temperature. Taking into account the criteria for tunnel conduction [14], one says that the conduction of this annealed MTJ is dominated by tunneling and not by pinhole. Namely, the decrease of RA during thermal annealing is due to the decrease

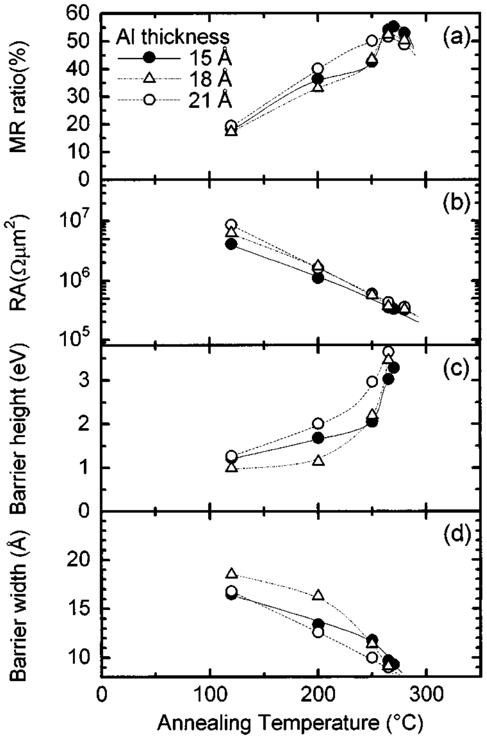

Fig. 4. Annealing-temperature dependence of (a) MR ratio, (b) RA product, (c) barrier height, and (d) barrier width for the MTJs with different Al thickness, oxidized with same conditions.

of the insulating barrier thickness. In other words, the changes of the fitted barrier width [Fig. 2(d)] give a real feature for the changes of the barrier layer structure. Assuming loosely bonded oxygen in as-prepared MTJs, which may be thermally activated and easily migrated into the barrier layer to form a more stable oxide, such as Al_2O_3, one can understand the change of the barrier thickness during thermal annealing.

The barrier widths obtained for all the MTJs in Fig. 2(d), except for the case of He + O_2 plasma, are thinner than the expected value from the Al layer thickness of 15 Å before oxidation. It follows that there may remain an unoxidized (or inadequately oxidized) Al layer at the interface between the barrier and the ferromagnetic electrodes of the MTJs. In order to investigate the influence of the unoxidized Al layer on the magnetotransport properties, we fabricated the MTJs, having thicker Al layer. Fig. 4 shows the T_a dependence of (a) MR ratio, (b) RA product, (c) barrier height, and (d) barrier width for the MTJs, having an Al-layer thickness of 18 Å and 21 Å subsequently oxidized by Kr + O_2 plasma under the same condition, shown in Table I. For comparison, the results for the MTJ with 15-Å-thick Al case, which has been shown in Fig. 2, are also plotted. One finds that the MR ratio and the RA values of the three MTJs are very close for all annealing temperatures and that the very similar tunneling barriers are formed for the three MTJs, although the Al thickness before oxidation is quite different. If we assume that the Al–O layer is formed perfectly flat and homogenously in the film plane without any "hot spot" in conduction, this result means that the MR ratio is not so sensitive to the probable remaining unoxidized Al layer. However, to confirm this conjecture, further investigation such as cross-sectional TEM observation is required.

IV. SUMMARY

Fabrication of MTJs having a high MR ratio of about 50% is successfully achieved. The RA shows a monotonous decrease with an increasing T_a, which is the essential property for the MTJ using RLSA plasma. The decrease of the RA is due to the decrease of the effective barrier width. The barrier width of the MTJs, especially after annealing, is thinner than the expected value from the Al thickness before oxidation. One, thus, expects to fabricate the MTJ having a thin barrier width easily by using an RLSA plasma compared to the other oxidation method.

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