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HTML ABSTRACT + LINKS

Fabrication of ferromagnetic single-electron tunneling devices by utilizing metallic nanowire as hard mask stencil

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The stacked magnetic tunnel junctions (MTJs) are microfabricated into ferromagnetic single-electron tunneling devices (F-SETs) by using electron-beam lithography. The F-SETs have a couple of small MTJs ($30\times 500\ \mathrm{nm}^2$–$0.1\times 100\ \mu\mathrm{m}^2$), which are connected via a metallic nanowire. The large tunnel magnetoresistance ratio as much as 40% (at RT) and small junction area dependence of the *RA* (resistance×area) are obtained. The electrostatic energy of F-SETs estimated from the minimum junction area corresponds to the temperature of 1 K, which is high enough to observe Coulomb blockade phenomena in a dilution refrigerator. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1850408]

Ferromagnetic single-electron tunneling devices (F-SETs) are the combination of very small magnetic tunnel junctions (MTJs) and a small island electrode. As is predicted theoretically, phenomena such as enhancement of the tunnel magnetoresistive (TMR) effect and TMR oscillation with bias voltage occur in these systems.[1] They could achieve highly functional devices, such as a "switchable" MTJ, which will be strong candidates for the cell of the future ultrahigh-density magnetic random access memory. Much attention, therefore, has been paid to the origin of the phenomena. While the oscillation of TMR is well described by semi-classical orthodox theories,[1] the enhancement of TMR is still controversial. The enhancement was believed to result from the higher order electron tunneling in the strong tunneling regime, wherein the junction resistance of the F-SET is nearly equal to the quantum resistance R_Q ($=h/e^2 \sim 25.8\ \mathrm{k}\Omega$). This description, however, cannot explain the amazing enhancement of up to ten times larger than the normal state, which is reported in small Ni/Co/Ni MTJ arrays.[2] In addition, recent experimental work by Matsuda *et al.* completely conflicts with this description in that the increment of the TMR ratio exhibits no apparent dependence on the junction resistance of their F-SETs.[3] Therefore, in order to say for certain whether the assertion is valid, more precise examination is required. From this point of view, the traditional fabrication procedure—so-called Dolan method[4,5]—seems unsuitable for making F-SETs, since it restricts the way to form the MTJs within evaporation and surface oxidation of the bottom layer, both of which are far from the trend in MTJs. Such restrictions result in a worse signal-to-noise ratio of the MR and prevent us from making quantitative discussions.

Previously, we fabricated MTJs on a nanometer scale by using top-down-type microfabrication techniques.[6,7] The size of the island was, however, too large to observe single-electron phenomena. In this work, we have made marked progress in the fabrication procedure for F-SETs by employing both top-down techniques and a hard mask stencil method. The developed method only reduces the multilayer of MTJs, so that we are able to obtain both small and highly optimized MTJs. Furthermore, the island electrode with nanometer width is self-aligned on the MTJs. These are significant advantages for making F-SETs that enable us to make a direct comparison with aforementioned theories in various tunneling regimes.

The multilayer, consisting of $Ni_{80}Fe_{20}$ (10 nm)/$Co_{75}Fe_{25}$ (10 nm)/Al-O (1 nm)/$Co_{75}Fe_{25}$ (5 nm)/Ta (10 nm), is prepared on Si (100) substrate with Al_2O_3 (100 nm) buffer layer. The deposition is performed by inductively coupled plasma (ICP) rf magnetron sputtering. The insulating layer is formed by sputtering of Al with nominal thickness of 1 nm and ICP plasma oxidation for 120 s. The bottom and the top are soft and hard magnetic layers, respectively. The magnetization of both layers is oriented in the same direction by the permanent magnet (~100 Oe) during the deposition. The entire process of deposition is performed without breaking vacuum.

At the first stage of microfabrication, the multilayer is separated into 81 pieces of electrode by using Ti (15 nm) hard mask stencil and electron cyclotron resonance Ar ion milling. The electrode consists of two large pads connected via a bridge with a width of 100 μm [Fig. 1(a)]. Next, the substrate is spin-coated by a 500 nm positive e-beam resist (Nippon Zeon, ZEP-520) and baked on a hot plate. The substrate is then exposed by the facility of a 20 kV field-emission-type e-beam writer (ELIONIX, ERA-8000FE). After development, long stripe patterns are formed across the middle of the bridges. The dimensions of the stripe are 0.5, 1, and 10 μm in width and 200 μm in length. The exposed area of the multilayer is etched away by using Ar ion milling, and filled with thick Al_2O_3(80 nm) by sputtering and lift-off [Fig. 1(b)].

Thereafter, both e-beam lithography and e-beam evaporation followed by lift-off are performed to make a nanowire. The narrow line consisting of Ti (10 nm)/Pd (20 nm)/Ti (15 nm) is left across the Al_2O_3 stripe [Fig.

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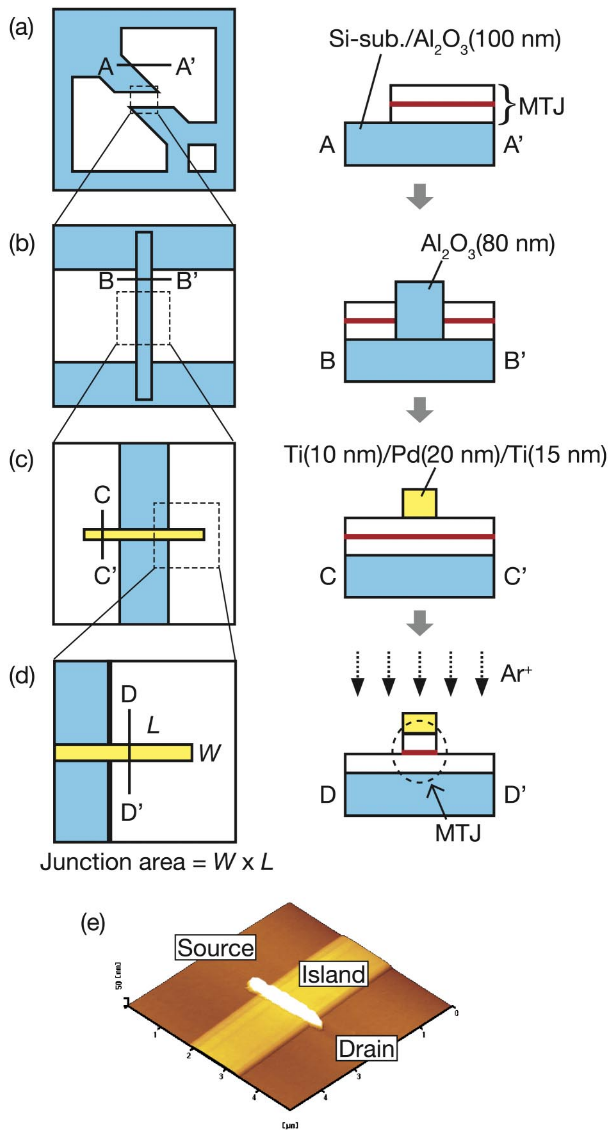

FIG. 1. (a)–(d) Schematic illustration of fabrication procedure for F-SET. (e) AFM image of F-SET with the junction area of 30×500 nm^2.

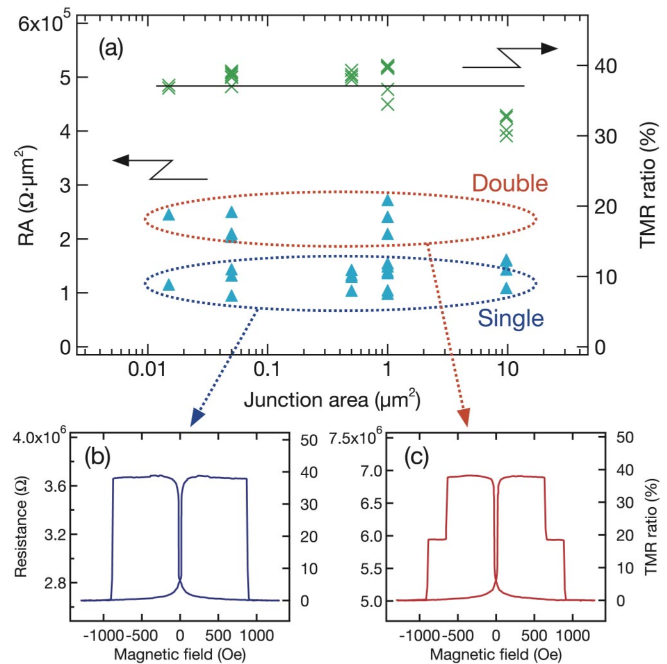

FIG. 2. (a) Dependence of RA and TMR ratio on the junction area. Typical R-H curve of the F-SET with (b) single and (c) double MTJs, both of which were measured at room temperature.

1(c)]. Finally, Ar ion milling defines the junction area, wherein the nanowire acts as an etching mask of the MTJs [Fig. 1(d)]. The milling is stopped at the surface of the bottom layer, which is monitored by the end-point detector. In the completed structure, small MTJs are connected via a narrow "island," which is the remainder of the hard mask stencil. The dimensions of MTJs, W and L, are varied as W $=30$, 50, 100 nm and $L=500$ nm-100 μm. The narrow island electrode is parallel to the easy axis of the MTJs. Both single and double MTJs are fabricated in the same substrate to evaluate the properties of each junction, where "single" means the combination of small and quite large MTJs. The resistivity measurements at room temperature are performed by standard ac lock-in techniques with the modulation of 10 mV_{rms}. The magnetic field up to 2 kOe is applied to the easy axis.

Figure 1(e) shows atomic force microscopy (AFM) image of the F-SET with the junction area of 30×500 nm^2. Narrow wire is confirmed to exist between source and drain. The exact width of the island cannot be evaluated due to the artifact of our AFM cantilever, but we can evaluate them "electrically" as discussed later. The shift from the proper position of the island seems less than 50 nm, corresponding to the nominal value of our e-beam facility. Therefore, further reduction of L to sub-100 nm is still achievable.

Figure 2(a) shows the dependence of both RA (resistance$\times$area) and TMR ratio on the junction area. The large TMR ratio as much as 40% is obtained in the most of MTJs with high reproducibility. This is one of the highest values among F-SETs previously reported. At this stage, we can assert that the process shrinks MTJs without affecting their properties at all. The slight decrease at 10 μm^2 is due mainly to the configuration of magnetization in both layers. The junction area dependence of RA is quite small, which suggests that the dimensions of the MTJs are exactly same as the designed one, even in the nanoscale devices. In addition, the resistance of the double structure is twice as large as the single one at each junction area, which confirms that the island electrodes are self-aligned with high accuracy.

Figures 2(b) and 2(c) show typical R-H curve of the F-SETs with the single MTJs and the double MTJs, respectively. The resistance increase at the magnetic field about 50 Oe, shows a slight decrease in the anti-parallel alignment, and then rapidly drops down to the initial value at about 800 Oe. The only discrepancy of their curve is the switching of the hard magnetic layer. While switching of the single MTJ occurs at once, that of the double MTJ occurs two times. This indicates that hard magnetic layer of both MTJs has a single-domain structure, wherein parallel or anti-parallel alignment to the longitude direction of the island is permitted. This multiple switching was reported in a $Co/AlO_X/Co$ small MTJ array by Urech *et al.*[8] They also pointed out experimentally that a single-domain state is achievable when the width of the junction is below ~70 nm.

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It seems to be consistent with our result, wherein MTJs with 100 nm width hardly show such multiple changes. Our F-SETs also show the increment of the switching field of the hard magnetic layer with decreasing the width of MTJs. This increment can be explained by the analysis of Stoner and Wohlfarth,[9] in which the coercive field of ferromagnetic wires is inversely proportional to their width.

The electrostatic energy of F-SETs with the minimum junction area $(30 \times 500\ \mathrm{nm})$ is estimated as 150 μeV, which corresponds to the temperature of 1 K. Therefore, we will be able to observe the aforementioned phenomena at $\sim$100 mK, which is easily achieved by a dilution refrigerator.

In conclusion, we developed a fabrication procedure for a F-SET by utilizing a nanowire as a hard mask stencil of MTJs. A large TMR ratio as much as 40% and small junction area dependence of the *RA* are obtained even in the nanometer-scale MTJ array. This highly efficient F-SET is the key device for us to handle phenomena, such as enhancement of the TMR effect, qualitatively. This method is also useful for other phenomena, such as an injection of spin current into nanojunction.

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