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Quantum coherent transport versus diode-like effect in semiconductor-free metal/insulator structure

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Quantum coherent transport in double barrier tunnel junctions has been exploited for building micrometric-size semiconductor-free diodes. At room temperature, we observe strongly asymmetric current–voltage characteristics with an asymmetry ratio increasing with the bias voltage, reaching a maximum of 20 at 1 V. Our experimental data can be perfectly explained using a theoretical model involving resonance-assisted tunneling. The coherent/resonant tunneling regime is achieved using metallic 3 nm diameter monodisperse Cu clusters, sandwiched between two Al_2O_3 barriers. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1426685]

Magnetic tunnel junctions (MTJ) are promising candidates as storage elements in nonvolatile magnetic random access memories (MRAM), where each magnetoresistive MTJ cell corresponds to one single bit of information.[1] The most promising MRAM technology adds an additional semiconductor (SC) switch in series with the memory cell for enhancing the read/write contrast and avoiding the current shortcuts in the memory array. Integrating complementary metal-oxide semiconductor transistors or *p*–*n* junctions with submicron MTJ produces a system whose packing density is limited not by the active memory cell size (MTJ) but by the semiconductor correlation length needed by the silicon technology. Indeed, the minimum cell size of in-plane transport SC devices is limited by quantum effects in the semiconductors occurring when the lateral size of the device reaches the order of magnitude of the associated scale lengths. To circumvent this limiting size factor, one of the most promising solutions consists of the use of a metal-insulator-metal switch integrated vertically within the tunnel memory such that the total cell size is now the size of the active memory.

In this letter, we present the experimental concept of a metal/oxide switch acting as a diode, integrated vertically in a magnetic tunnel junction. We show that resonant effects and hot electron transport in this double barrier metal/insulator/metal (MIM) system are used for achieving a large asymmetry in the current–voltage characteristic at room temperature (diode-like effect) in a semiconductor free device. In such a double barrier system [see Fig. 1(a)], the electrons are injected by tunneling from the bottom ferromagnetic electrode (FM_1) across the first tunnel barrier (I_1). When resonant effects take place in the middle metallic interlayer (M), sandwiched between the two insulators, an energy selection of electrons occurs and modulates the electron current which is subsequently injected by tunneling across the second insulating barrier (I_2) and detected by the top ferromagnetic electrode (FM_2). Moreover, the energy selection gives rise to an asymmetry of the current depending on the orientation of the current vector or, in other words, of the sign of the applied voltage. Since the transport occurs in the CPP geometry (current-perpendicular-to-plane), this physical effect is not laterally sensitive unlike the semiconductor switch, making this concept suitable for vertical integration as a blocking element in new generations of nanoscale spin electronic devices.

The first concept reported in literature to obtain asymmetric current–voltage ($I-V$) characteristics using metal/oxide junctions exploited the asymmetric potential profile

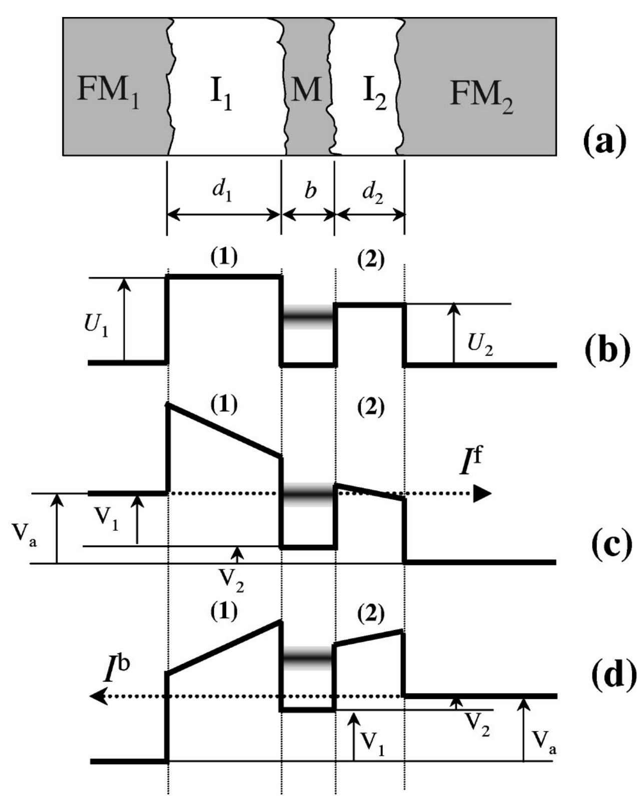

FIG. 1. (a) Sketch of a double barrier tunnel structure and its corresponding potential profile without applied voltage (b), under positive (c) and negative (d) applied voltage (V_a). A broad resonant level is depicted in the metallic interlayer. The parameters of the barriers are (1) height U_1, width d_1 (2) height U_2, width d_2, and b is the thickness of the metallic interlayer. The voltage drops in each barrier are denoted by V_1 and V_2.

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(trapezoidal barrier) of a single insulating barrier configuration. Such a profile arises from the difference in the work functions of the two metallic electrodes. Experimental results at room temperature[2] show that the current asymmetry in this case is small ($A=I^{f}/I^{b}\leqslant 1.3$, $I^{f(b)}$ indicates forward(backward) current), making this concept unsuitable for potential applications as blocking diode. A new concept is proposed here that exploits in addition to the asymmetry of the potential profile the coherent electronic transport in double barrier MIM systems. Indeed, in the case of a double barrier system, by varying the two insulating layers one can get different barrier parameters (Fig. 1). Beyond an ''intrinsic'' asymmetry of the $I-V$ characteristics, related to the asymmetric potential profile of the two barrier configurations, the asymmetry can be ''enhanced'' by two main features of electron tunneling in the MIM quantum well system: (a) coherent resonant tunneling and (b) hot electron transport.

Few fundamental aspects may be detrimental to resonant tunneling at room temperature in experimental systems implicating MIM quantum wells: the effect of the fluctuations related to interfacial roughness combined with a larger Fermi wave vector in metals with respect to SC quantum wells and also a high density of scattering centers in metals. However, even when the resonance contributions are completely destroyed by fluctuations, a large asymmetry can be achieved by exploiting the hot electron transport, as qualitatively illustrated in Fig. 1. Indeed, for forward (positive) applied voltage V_a, one can see that if the voltage drop V_1 in the first barrier gets superior to the second barrier height U_2, the electrons become hot for the second barrier. Therefore, they only undergo the first barrier in terms of tunneling [Fig. 1(c)]. The corresponding current is high. For a similar backward (negative) V_a, the electrons undergo both barriers [Fig. 1(d)]. Thus, a high asymmetry ratio A is theoretically expected. The asymmetry between V_1 and V_2 is related to the difference in barrier parameters ($U_1\neq U_2$, $d_1\neq d_2$) and to the scattering in the middle metallic layers.[3] Moreover, in case of resonant tunneling a much stronger current asymmetry ratio A is expected. This can be easily explained by the model illustrated in Fig. 1 with one resonant energy level located in the quantum well. Indeed, when applying a forward voltage, the current is strongly enhanced when the energy of the electron injected by tunneling across the first barrier reaches the resonance level. Therefore, a strong increase in the current is expected when the forward V_a is increased. This increase of the current follows exactly the distribution in energy of the resonance level in case of realistic systems, where the resonance level has a given width due to limited fluctuations and electron scattering in the interlayer. Thus, similar to the case of SC quantum well structure,[4] we expect a kink in the forward branch of the $I-V$ curve, corresponding to the situation where the energy of the injected electron crosses the ''core'' of the resonance level. The width of this kink is directly related to the width of the resonance level. On the other hand, for a backward voltage the energy of the injected electron moves away from the resonant level and no resonant tunneling occurs. Consequently, in this case the current is much lower than the one corresponding to resonant assisted current.

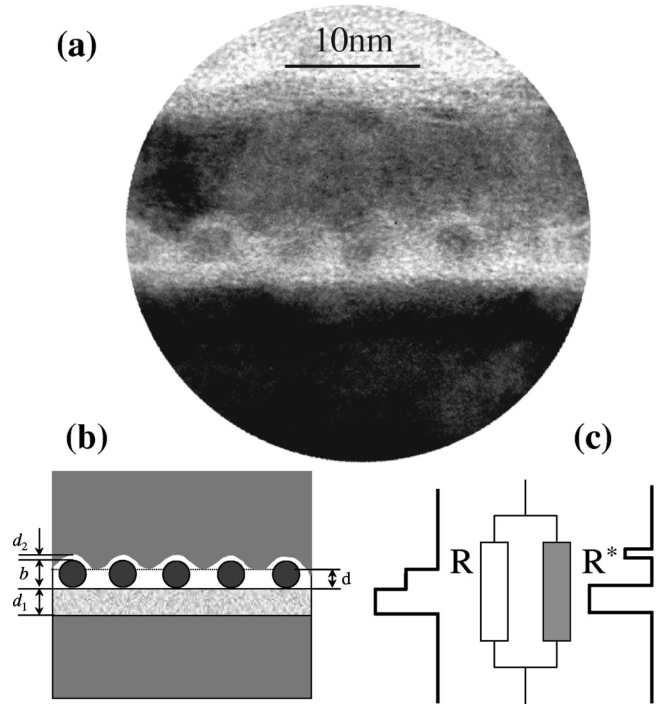

FIG. 2. (a) Cross section transmission electron microscopy image; (b) the corresponding sketch model for a double barrier system with discontinuous metallic interlayer. d_1, d_2 represent the thickness of the first and the second insulating barrier, respectively, b is the average cluster diameter, d indicates the thickness of the intergrain insulator; (c) two-conduction channels model vs the corresponding potential profile.

There are two important requirements for exploiting the concepts exposed above: (1) the two barriers have to be chosen properly in order to have the required asymmetry in the voltage drops V_1 and V_2 that would allow hot electrons to tunnel across the second barrier; (2) the level of fluctuations has to be very small, in order to allow the resonant assisted tunneling.[3]

These two criteria can be tested in a realistic experimental system if the thickness of the metallic interlayer sandwiched between the two insulators becomes less than the characteristic lengths of the coherent transport. Increasing the thickness of the interlayer would increase the probability of the elastic (inelastic) scattering leading to loss of the coherence in the transport so that the current asymmetry vanishes.[3] Our samples have been elaborated by sputtering and patterned in micronic junctions ($10\times 10\,\mu m^2$) by ultraviolet lithography.[5] They are composed of two ferromagnetic electrodes of CoFe(3 nm) separated by AlO_x(1.5 nm)/Cu(1 nm)/AlO_x(1 nm). As will be discussed later, the Cu metallic spacer layer is discontinuous and is formed by embedded spherical clusters with a very narrow distribution of size and intergrain spacing, as illustrated by the cross section transmission electron microscopy picture [Fig. 2(a)]. The cluster size was estimated to be about 3 nm with an average spacing of around 2 nm. Estimated barrier parameters are respectively $d_1=1.5$ nm, $U_1=2.3$ eV, and $d_2=1$ nm, $U_2=0.8$ eV. For this structure, one can expect Coulomb blockade effects at low temperature.[6]

Figure 3 shows strongly asymmetric $I-V$ characteristics, measured on a $10\times 10\,\mu m^2$ junction. The bias-voltage variation of the current asymmetry ratio [Fig. 3(a)] shows that it

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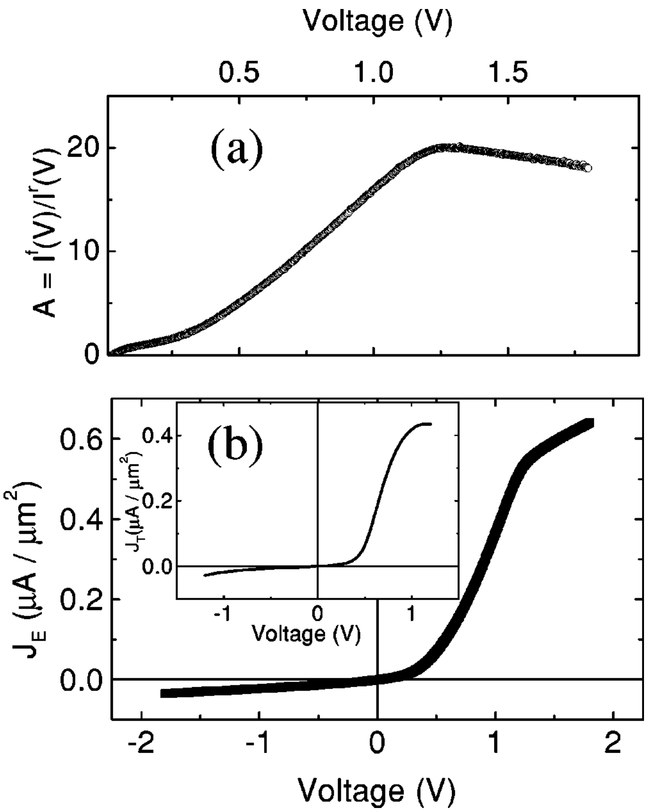

FIG. 3. (a) Variation of the asymmetry ratio with the external voltage; (b) experimental $I(V)$ characteristics of double barrier system. Inset: theoretical curve, expected for coherent resonant tunneling.

increases monotonously with V_a and reaches a maximum value (about 20 around 1 V) whose origin is a direct proof for the presence of the resonant state in the metallic quantum well. This maximum value corresponds to the maximum value for the forward current when the energy of the electrons injected across the barrier matches the resonance level [as illustrated in the model exposed in Fig. 1(c)]. The maximum corresponds to the kink measured around 1 V on the positive branch of the $I-V$ curve. Extremely important for potential applications of the MIM double barrier diode is not only a high value for the asymmetry but also its variation with the external voltage. In our samples, a large asymmetry ratio is preserved for a wide voltage range of $1\ \mathrm{V} \pm 0.5\ \mathrm{V}$ around the applied voltage corresponding to the maximum value of the current asymmetry [Fig. 3(a)].

This system can be modeled as illustrated in Fig. 2(b) where the conduction involves two tunneling channels: (1) a low resistive highly asymmetric coherent channel R^* associated to metallic clusters paths [Fig. 2(c)]; (2) a high resistive channel R associated to the thick insulator regions located between the clusters. In this model the resistance of the junction is governed by the low resistive one. Their very small lateral size (<3 nm) insures very low fluctuations for the local transport in favor of resonant tunneling. Each cluster-associated channel can be modeled by a potential profile analogous to the one illustrated by Figs. 1(a)–1(d). The hot electron transport criteria is achieved for forward voltage when the voltage drop V_1 allows the injection of hot electrons across the second barrier with a small height.

A crucial step during the sample elaboration was the optimization of the electron tunneling across the first barrier. The extreme fluctuation level has been reduced up to rms ~ 1 Å for the top surface roughness and <1 Å for the barrier thickness.[7] Such a low level of fluctuations is a key parameter for achieving resonant-assisted tunneling and insures a very homogeneous current distribution upon the whole junction area.

Remarkably, as illustrated by the inset of Fig. 3(b), the experimental $I-V$ characteristic matches perfectly to the theoretical expectation, calculated in the framework of a quantum mechanical model developed in Ref. 3. Theoretically, the shape of the $I-V$ characteristics and its asymmetry is explained by the existence of quantum well states in the middle metallic layer which give rise to resonances in the current. Under applied voltage the positions of the resonant levels are shifted and for asymmetric structures this shift is different for the forward and backward bias voltage [Figs. 1(c) and 1(d)]. As a result, the $I-V$ curve is highly asymmetric, demonstrating a diode-like behavior [inset of Fig. 3(b)]. In general, the shape of the $I-V$ curve (in particular, the position and the shape of the kink) is related to the width of resonant levels which is defined by the amplitude of electron scattering in the middle metallic layer broadening the resonant peaks.

For potential application of the MIM double barrier diodes few perspectives are considered: (1) Reducing the forward resistance of the diode by using alternative insulating materials and by varying their elaboration techniques; (2) enhancing the asymmetry ratio and the position in voltage of the asymmetry maximum value (kink). This can be done by changing the interlayer material (i.e., Co, CoFe) or by varying the growth parameters (sputtering rate, pressure) and/or the thickness of the sputtered metallic layer in order to adjust the size of the clusters; (3) using a ferromagnetic interlayer represents an interesting challenge, since crossing this layer the electrons remain polarized. Therefore, one can envisage a magnetically controlled diode[3] which represents itself a storage and blocking element suitable for data storage potential applications.

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