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A photoconductance method has been used to study directly the barrier asymmetry in TaO_x magnetic tunnel junctions. Due to optical electron-hole pair generation in the barrier itself and subsequent transport in the electric field, the sign and magnitude of the barrier asymmetry can be determined quite accurately. The reliability of the technique is demonstrated by the independence on the direction of illumination. The oxidation time where the asymmetry becomes zero is found to coincide with a maximum in the magnetoresistance ratio. This is argued to be due to the complete oxidation of the barrier material, resulting in a symmetric tunnel barrier. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1759778]

In the emerging field of magneto-electronics, one of the most relevant structures for the actual application in devices is the magnetic tunnel junction (MTJ).[1] The presence of large room-temperature magnetoresistance (MR) ratios ensure both scientific and industrial interest. The most crucial part in a MTJ, both from a fundamental as well as a processing point of view, is the tunnel barrier. The potential landscape of this barrier layer determines to a large extent the transport properties of the junction.[2]

The potential barrier is often described in terms of an effective barrier height ϕ, which directly affects the electron tunneling probability. Experimentally ϕ has either been determined *indirectly* by fitting the current–voltage characteristics to the Simmons equation,[3] or *directly* by ballistic electron emission microscopy[4,5] or photoconductance.[6] However, while instructive in their own way, these techniques only result in an effective, or a maximum ϕ, respectively. Moreover, the potential landscape can be strongly modified by an electric field, e.g., due to different (effective) work functions for the two electrodes, and/or a nonhomogeneous barrier layer. The barrier asymmetry $\Delta\phi$ therefore plays an additional important role in the spin dependent transport through the tunnel barrier.[2]

The modification of the Simmons equation by Brinkman, Dynes, and Rowell[7] to explicitly include $\Delta\phi$ is often used to get an indication of the barrier asymmetry. The resulting asymmetry parameter is determined by the voltage dependence of the junction conductance. However, the influence of the barrier shape itself can be masked by effects on the $I(V)$ curve, such as a nonhomogeneous barrier thickness, as well as density of state effects. To overcome some of these issues, Rottländer, Hehn, and Schuhl have applied an alternative method to determine $\Delta\phi$.[8] From the logarithmic derivative of the conductance curves and their temperature dependence the barrier heights at each interface can be determined, giving an indication of the asymmetry of the potential landscape. However, this method requires the application of bias voltages in excess of the barrier height, which may lead to electrical breakdown. Also, for a nonhomogeneous barrier, the tunnel current is concentrated in small areas, namely there where the transmission is highest. This results in barrier parameters which can differ significantly from the integral characteristics. The above issues necessitate the use of a different technique to determine the complete barrier asymmetry independently and reliably.

In this letter we present a photoconductance method to *directly* probe the barrier asymmetry in magnetic tunnel junctions and relate this to the oxidation state of the tunnel barrier. By illuminating the junction structure with photons, with energies above the band gap of the tunnel barrier, electrons in the insulator will be excited from the valence to the conduction band. Those electrons which have upon generation a k-vector component perpendicular to the barrier layer, and a kinetic energy that is smaller than that which is required to reach the top of the barrier, will all leave the barrier in the ''downhill'' direction. This induces a directional preference for the excited electrons and will thus result in a net photocurrent. The sign and magnitude of $\Delta\phi$ can then be easily derived by determining the applied bias voltage at which the photocurrent changes sign.

To demonstrate this technique, TaO_x based MTJs have been fabricated by sputter deposition through metallic shadow masks. The following layer stack is used: glass// 3.5 Ta/3.0 $Ni_{80}Fe_{20}$/10.0 $Ir_{20}Mn_{80}$/2.5 $Ni_{80}Fe_{20}$/1.5 $Co_{90}Fe_{10}$/ TaO_x barrier/4.0 $Co_{90}Fe_{10}$/10.0 $Ni_{80}Fe_{20}$/3.5 Ta, with all thicknesses in nanometers. The tunnel barrier is formed by first depositing a thin Ta layer over the complete substrate, followed by oxidation in an oxygen plasma. A more detailed description of the fabrication process can be found elsewhere.[9] TaO_x was used as the barrier material, because the low band gap makes interband excitations accessible with a simple UV illumination system. Photoconductance experiments were carried out by measuring the short-circuit photocurrent under illumination of the top electrode by photons of variable energy. In all our photoconductance mea-

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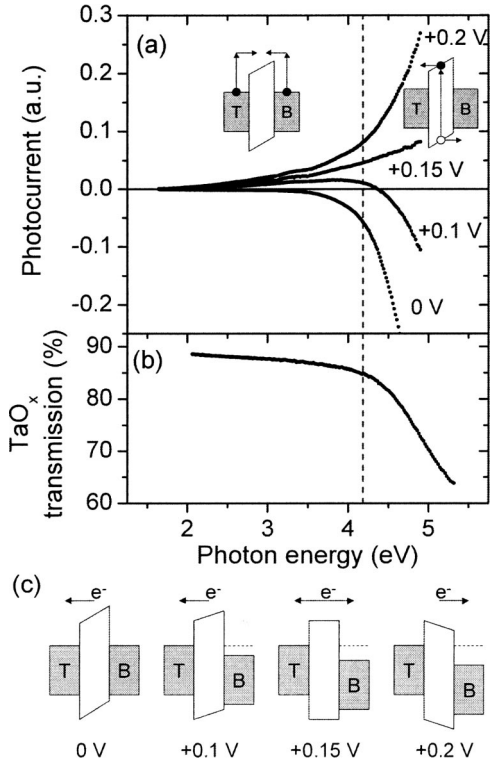

FIG. 1. Example (a) of photocurrent vs energy curves for different applied bias voltages for a TaO_x MTJ (1.2 nm Ta and 70 s oxidation), as well as (b) the optical transmission of a single TaO_x layer on a quartz substrate. The insets show energy diagrams, illustrating the dominant contribution to the photocurrent above and below the 4.2 eV band gap (for 0 V bias voltage). In (c) energy diagrams, demonstrating the change of the electric field inside the barrier (relative to the top T and bottom B electrode) with applied bias voltage and the resulting hot-electron current direction, are shown.

surements, a positive photocurrent corresponds to a flow of electrons from top to bottom electrode.

In Fig. 1(a) typical photoconductance curves are shown for a TaO_x MTJ under the application of different bias voltages. Two different regimes can be distinguished. For low-energy photons electrons will be excited in both electrodes. Some of these photoexcited electrons will be able to cross the barrier, and contribute to a photocurrent. In this letter, this process will not be addressed further. For photon energies above the band gap of the insulator, also electron-hole pairs will be generated in the insulator. The transport of these excited electrons and holes is not hampered by scattering processes or interface reflections, in contrast to excitation from the electrodes. Due to this larger collection efficiency, the contribution from interband transitions will dominate at higher energies. To directly demonstrate the onset of excitations in the insulator, an optical transmission spectrum of a single TaO_x layer on a quartz substrate is shown in Fig. 1(b). From this a band gap of roughly 4.2 eV is determined, which is in good correspondence with values found by others.[10]

From the high-energy part (>4.2 eV) of the photoconductance curves in Fig. 1(a) it can be seen that, for this junction, the photocurrent due to interband excitations changes sign around an applied bias voltage of ~150 mV, with an uncertainty of ~15 mV due to the background photocurrent from electrode excitations. As illustrated in Fig. 1(c) this implies that the average electric field in the insulator becomes zero, and that the above voltage therefore directly

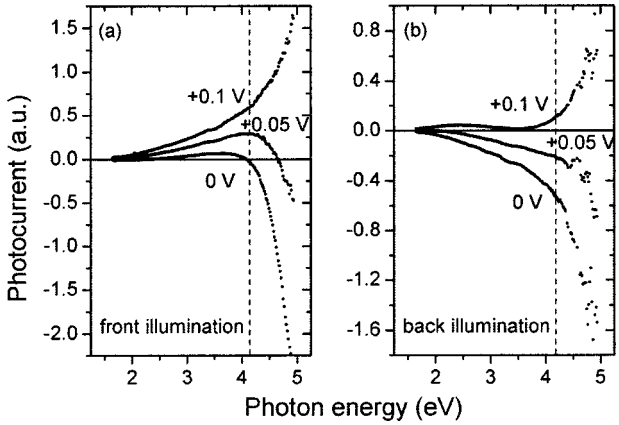

FIG. 2. Photoconductance curves of a TaO_x MTJ (1.2 nm Ta and 50 s oxidation) for (a) illumination of the top electrode and (b) illumination of the bottom electrode. Although the low-energy (<4.2 eV) photocurrent does depend on the illumination direction, the voltage dependence of the *additional* photocurrent due to interband excitations at high energies (>4.2 eV) shows the same behavior.

corresponds to the barrier asymmetry $\Delta\phi$. In this way $\Delta\phi$ can be determined quite accurately in complete junction devices.

To exclude an influence of the absorption profile on the determination of $\Delta\phi$, photoconductance curves were measured for different directions of illumination. From Fig. 2 we can see that by reversing the incident light direction from front to back illumination, the voltage dependence of the *additional* high-energy (>4.2 eV) photocurrent retains the same behavior. Since the optical penetration depth in TaO_x above 4.2 eV is much larger than the thickness of the barrier,[10] the excitation occurs homogeneously through the barrier. The electrons and holes, which are formed upon the excitation, have in a first approximation an isotropic momentum distribution. Combined with the independence on the direction of incident light, this directly shows that the asymmetry of the potential landscape determines the sign of the photocurrent.

In previous work asymmetric tunnel conductance curves were found for suboptimally oxidized MTJs with the same top and bottom electrode materials, suggesting a nonsymmetric barrier profile.[11–13] To directly investigate the relation between the oxidation state and the asymmetry of the tunnel barrier, a series of MTJs were deposited with 1.0 and 1.2 nm of Ta as the barrier material, and an oxidation time between 8–90 s. In Fig. 3(a) the MR ratio for each of the junctions is shown. For both thicknesses a clear maximum in the MR is present. For AlO_x this behavior is well known, and is attributed to the complete oxidation of the barrier layer, without oxidizing the underlying magnetic electrode.[12,13] For the above TaO_x junctions also the barrier asymmetry was determined by our photoconductance method, and is plotted in Fig. 3(b). For the oxidation time where the barrier is symmetric, the corresponding junction shows a maximum in the MR. This supports the simple picture, where a completely oxidized barrier, sandwiched between two identical electrodes, has a symmetric potential profile. This in contrast to results from a Brinkman fit, for which optimally oxidized junctions were found to have effective barrier asymmetries significantly different from zero: −50 and −100 mV for a

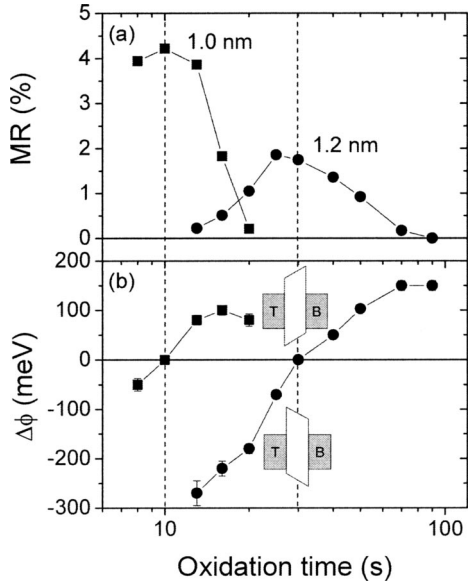

FIG. 3. The MR ratio (a) as well as the barrier asymmetry (b) as determined from photoconductance for TaO_x based MTJs. The vertical dashed lines correspond to the oxidation times where the barrier is symmetric and the MR ratio has a maximum. In (a) the thickness of the deposited Ta barrier layer is shown. The insets in (b) represent energy diagrams, defining the sign of the barrier asymmetry relative to the top T and bottom B electrode.

1.0 and 1.2 nm oxidized Ta layer, respectively. As stated before, these barrier parameters are determined for a possible heterogeneous tunnel current distribution and can be influenced by density of state effects. This in contrast to photoconductance which directly measures the integral characteristics.

Apparently under or overoxidation of the barrier material causes an asymmetry in the barrier potential with differing signs. Such an asymmetry is caused either by a difference in work function for the two electrodes (Ta has a lower work function than Co or Fe[14]), an inhomogeneous oxygen distribution in the barrier (resulting in a varying band gap), or a combination of the two. From photoconductance experiments on junctions where the complete bottom electrode is replaced by a 35 nm Ta layer, indeed a large negative barrier asymmetry of ~ -500 mV is found.

Summarizing, due to photoexcitation and transport in the barrier layer of MTJs, the integral barrier asymmetry has been determined and correlated to the oxidation state. This method provides an important tool that gives added information, which cannot readily be determined otherwise. One of these issues is, for example, the influence of an anneal treatment. From previous work it has been suggested that the observed increase in MR is related to an improvement of the barrier layer, and/or the interfaces.[15] Photoconductance can give additional information as to whether this coincides with a change in the electronic structure of the barrier, such that the asymmetry is modified.

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