HOT-ELECTRON ATTENUATION IN THIN Al_2O_3 FILMS

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A number of recent reports have shown that much information concerning the barrier heights in tunnel barrier structures of the type Al-Al_2O_3-Al can be obtained from internal photoemission experiments.[1-5] The observed photocurrents in these structures are interpreted as being due to hot electrons, generated in the aluminum electrodes, which are energetic enough to overcome the 1.5- to 2-eV interfacial barriers and thus enter the Al_2O_3 conduction band—the net current is then the difference of the currents from the two electrodes. In order to understand the results under all conditions of applied voltage and incident photon energy it has been necessary to make two assumptions. First, there are unequal densities of hot electrons reaching the two Al-Al_2O_3 interfaces due to the optical-absorption process and the hot-electron transport in the electrodes. Second, the current due to electrons which are traversing the Al_2O_3 conduction band in the direction opposite to the net field in the Al_2O_3 (the sum of the applied field and the internal field that is always present[6]) is attenuated due to scattering or other energy- or momentum-loss processes. Electrons which are traveling with the field will reach the opposite electrode in spite of any such processes, while those opposed to it will have a high probability of being returned to the electrode in which they originated.

Most of the reported measurements have been taken with the light incident on the upper or counter electrode of the structure, which is the one with the higher interfacial barrier height. In two cases, however, experiments were also done with the light incident on the electrode which was deposited on the glass substrate, and it was discovered that the photocurrent changed sign.[1,5] It is the purpose of this Letter to show the spectral dependence of the photocurrent with this direction of illumination and to show that it can be used to give quantitative verification of the two assumptions made above.

Experimental samples were prepared as previously described,[3] with the exception that the electrodes were both semitransparent and of equal thickness. The Al_2O_3 thickness was approximately 40 Å. The measurements were taken with an optical system which had two beams of equal intensity which illuminated the two electrodes. Shutters could be placed in either beam so that photoemission could be measured with the electrodes separately illuminated or with both illuminated simultaneously.

Figure 1 shows the photoresponses obtained as a function of photon energy with zero applied bias voltage when the electrodes are separately illuminated. The R_{21} current is the one which has usually been observed. When the square root of R_{21} is plotted against photon energy, the result is linear with an extrapolated photothreshold of 1.96 eV, which is interpreted as the Al-Al_2O_3 barrier height for the top electrode of the structure (generally denoted by φ_2) at zero bias. The R_{12} current is opposite to R_{21} at low photon energies, rises to a maximum at 2.96 eV, is zero at 3.47 eV, and is in the same direction as R_{21} thereafter. R_{12} and R_{21} are approximately equal and opposite from the 1.96-eV threshold to 2.5 eV as shown by the curves in Fig. 2.

With the aid of the two assumptions stated above and the further assumptions of zero absolute temperature and approximate equality of the optical-absorption distance and the hot-electron attenuation length in the aluminum electrodes,[7] the currents R_{12} and R_{21} may be calculated theoretically as a function of the incident photon energy $h\nu$. The details will be published elsewhere[7]; for our present purposes it is only necessary to state the results. They are

$$R_{12}=\frac{4\pi\alpha LT}{h^3}\left\{\left(\frac{\lambda_0}{s}\right)^2\exp\left(-\frac{s\Delta\varphi}{2\lambda_0}\right)\left[\exp\left(-\frac{sx}{\lambda_0}\right)-1+\frac{s}{\lambda_0}x\right]-\frac{\delta}{2}x^2\right\} \tag{1}$$

and

$$R_{21}=-\frac{4\pi\alpha LT}{h^3}\left\{\frac{x^2}{2}-\delta\left(\frac{\lambda_0}{s}\right)^2\exp\left(-\frac{s\Delta\varphi}{2\lambda_0}\right)\left[\exp\left(-\frac{sx}{\lambda_0}\right)-1+\frac{s}{\lambda_0}x\right]\right\}, \tag{2}$$

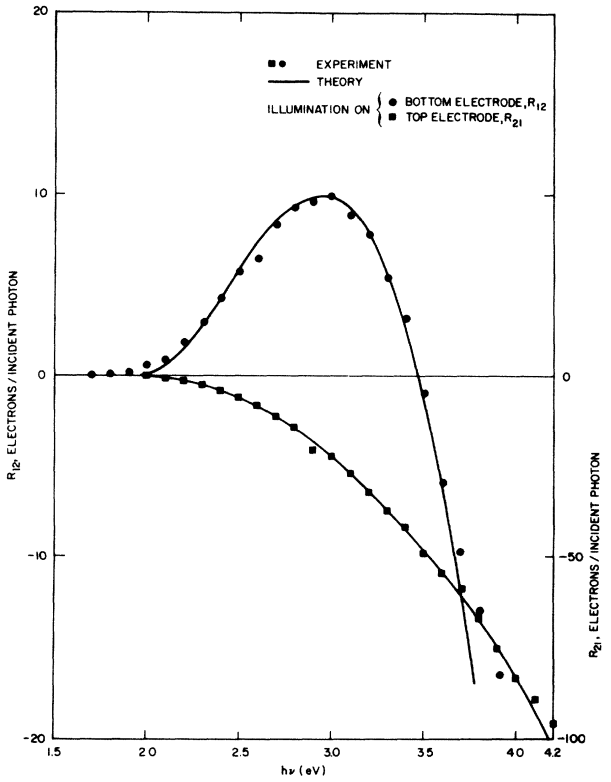

FIG. 1. Relative photoresponse (arbitrary units) versus photon energy for an Al-Al_2O_3-Al structure with both electrodes transparent.

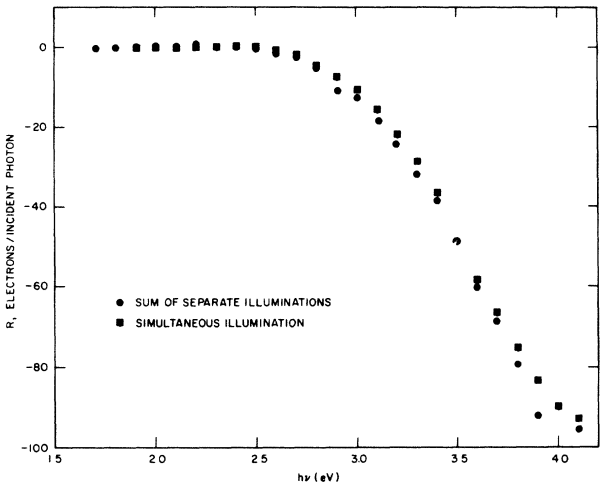

FIG. 2. Relative photoresponse (arbitrary units) versus photon energy with light incident on both electrodes of the sample of Fig. 1. The sum curve is the sum of the two curves of Fig. 1.

where α, L, and T are, respectively, the optical absorption constant, thickness, and optical transmission of the electrodes (which were assumed the same). h is Planck's constant, x is $h\nu-\varphi_2$, s is the Al_2O_3 thickness, and $\Delta\varphi$ is the internal field in the Al_2O_3. δ is the ratio of the number of electrons reaching the two Al-Al_2O_3 interfaces and is given by

$$\delta = (1-T^2)/2\alpha L. \tag{3}$$

The attenuation in the Al_2O_3 for an electron of energy E has been described by an attenuation length, λ, given by

$$\lambda = \lambda_0/(E-E_c), \tag{4}$$

where E_c is the energy of the Al_2O_3 conduction band edge.[8]

In order to fit the data of Fig. 1 to (1) and (2), the parameters s/λ_0 and δ were determined by numerical solution of a transcendental equation involving the photon energies at which R_{12} is maximum and zero.[9] The results were $s/\lambda_0 = 0.53\ (\text{eV})^{-1}$ and $\delta = 0.70$. The curves labeled "theory" in Fig. 1 were then calculated from (1) and (2). The tail at low energy on the R_{12} curve is due to photon-assisted tunneling which was neglected in the theory.

The simple model assumed gives an excellent fit to experimental data with reasonable values of the parameters and thus supports the assumptions concerning optical absorption and hot-electron transport and loss on which it is based. Although we have used the simple mean-free-path variation with energy given in Eq. (4), other dependences such as $\lambda \sim (E-E_c)^{-2}$ also give reasonable results. Experiments on samples of varying Al_2O_3 thickness should provide the information required to determine the correct form of this law.

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[6]See, for example, T. E. Hartman, J. Appl. Phys. 35, 3283 (1964), and references contained therein.

[7]A. Braunstein, M. Braunstein, and G. S. Picus, to be published.

[8]The energy dependence of λ is not based on any theoretical justification; it is made only because it permits the energy integrals in the theory to be carried out explicitly. However, it is believed that a more realistic theory will not greatly change the size of the attenuation lengths, but only the details of their energy dependence. It is hoped that the correct energy dependence may be determined by more extensive experiments in which the Al_2O_3 thickness is varied.

[9]$\Delta\varphi$ was assumed to be 0.4 eV in accord with past experience with Al-Al_2O_3-Al structures, φ_2 is known from the threshold found for R_{21}, and the initial constant was determined by adjusting the heights of the theoretical curves after the shapes had been determined by the other parameters.

FERRIMAGNETIC STRUCTURE OF MAGNETOELECTRIC $Ga_{2-x}Fe_xO_3$

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In this Letter we report microscopic magnetic studies of $Ga_{2-x}Fe_xO_3$ via high-field Mössbauer measurements and macroscopic magnetic-moment measurements. Studies on both single-crystal and polycrystalline samples show that this material is ferrimagnetic, with magnetic moments directed close to or along the c-axis, instead of a canted antiferromagnetic structure as previously assumed[1] or inferred.[2] A similar ferrimagnetic structure has been determined for the isomorphic compound $Al_{2-x}Fe_xO_3$ which is also piezoelectric. A consistent analysis was possible only when high magnetic fields were applied in order to align the net magnetization, thereby eliminating one of the major difficulties with earlier analyses, viz., the large anisotropy of this material which could easily be misinterpreted as an indication of a canted antiferromagnetic structure. Two distinguishable hyperfine fields are observed corresponding to inequivalent sites with approximately 5 μ_B per Fe^{3+} ion, thus eliminating possible questions concerning low-moment states. The qualitative features of the measurements vary uniformly for $0.8 \leq x \leq 1.2$, indicating no unusual stoichiometry-dependent characteristics. These measurements also eliminate some earlier suggested models for the magnetoelectric effect, but remain consistent with the symmetry requirements for the observation of a magnetoelectric effect in this material. Our results firmly establish the model used by Bertaut et al.[3] to interpret their unpublished zero-field neutron diffraction measurements with powders.

The piezoelectric material $Ga_{2-x}Fe_xO_3$ ($x \approx 1$), first prepared by Remeika,[4] has aroused great interest because in addition to its spontaneous magnetic moment observed by Remeika and studied in detail by Nowlin and Jones,[5] very large magnetoelectric effects were reported by Rado.[1] The detailed crystal structure has been determined by Abrahams, Reddy, and Bernstein[2] who find four cation sites, two octahedral and two tetrahedral, with Ga^{3+} occupation of one tetrahedral site and Ga^{3+} and Fe^{3+} distribution over the other three sites. From the available data a magnetic structure consisting of a canted spin system (with large canting angles) was inferred.[2] While Rado used a theoretical model which assumed the material to be a canted antiferromagnetic structure, i.e., a weak ferromagnet, in order to explain the nondiagonal character of the magnetoelectric susceptibility, he pointed out[1] the importance of knowing the actual magnetic structure before a complete understanding of the magnetoelectric properties of $Ga_{2-x}Fe_xO_3$ could be achieved.

We have examined the magnetic structure of $Ga_{2-x}Fe_xO_3$ for $x = 0.8$, 1.0, and 1.2 with intense external longitudinal magnetic fields using Mössbauer techniques and a vibrating sample magnetometer modified for axial fields in superconducting magnets.[6] Both Fe^{57}-enriched polycrystalline samples and a mosiac of x-ray oriented single crystals were investigated with a constant-acceleration Mössbauer spectrometer at temperatures from 320 to 4.2°K.

In some experiments the absorbers consisted of crushed crystals grown from a melt with a 20% enrichment of Fe^{57} and embedded in Lu-