# GADOLINIUM RICH OXIDE/SILICON HETEROJUNCTION DIODES FOR SOLID STATE NEUTRON DETECTION

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## Introduction

There are several approaches to detecting and monitoring neutrons. One problem is that neutrons emitted by typical fissile materials have energies of about 1.5 MeV and higher, while most available neutron detectors are most sensitive to slow (epithermal) neutrons. For this reason, neutron detectors based on $^{3}$He or Cd need a thick moderator layer (at least several centimeters) in order to slow the neutrons down to about 30 meV or less and need a bulky high(er)-voltage power supply. Solid-state neutron detectors based on a gadolinium containing semiconductor may potentially address some of these problems. The extremely large thermal neutron absorption cross section of gadolinium (Gd) is an attractive property for creating a high efficiency neutron detector. Natural Gd has a thermal neutron capture cross section of 46,000 barns, while 15.65% abundant $^{157}$Gd has a cross section of 255,000 barns. Additionally, the Gd cross section remains significant out to neutron energies of about 200 meV. Indeed for a 15% Gd doped $HfO_2$ the neutron absorption for 100 meV neutrons is comparable to a boron carbide device for 30 meV neutrons; requiring a layer in the region of 30-40 microns for opacity. Hafnium oxide ($HfO_2$) with Gd incorporation [1-4] along with $Gd_2O_3$ [4] are candidates for one side of a heterojunction diode detector, and proof of principle can now be demonstrated.

## Experimental

*Materials*

The *p*-type 15% Gd-doped $HfO_2$ films were deposited on *n*-type single crystal Si(100) substrates using pulsed laser deposition (PLD) [1-2]. (Throughout, *x*% Gd means the nominal composition $Hf_{1-x}Gd_xO_{2-0.5x}$.) The *n*-type $Gd_2O_3$ films were deposited on *p*-type single crystal Si(100) substrates using pulsed laser deposition (PLD) [4] and using supercritical deposition [5]. X-ray diffraction (XRD) patterns indicated that the $HfO_2$ films with 15% Gd are in the cubic phase with strong texture growth [1,2]. For $Gd_2O_3$ films fabricated by PLD, there is strong texture growth and the films are in a monoclinic phase when deposited on Si(100) [4], but when deposited by supercritical deposition, the $Gd_2O_3$ films favor the lower energy cubic phase expected for a free standing film.

*Apparatus and Procedures*

The pulse height spectra were taken using a ~5.2 Curie plutonium-beryllium source which provided 2.2 x $10^4$ thermal or epithermal neutrons/$cm^2$-second, as calibrated by foil activation methods. The diodes were reverse biased with 3 V.

## Results and Discussion

Thermal neutron reactions with Gd nearly always results in an (n,γ) reaction, as in:

$$^{157}Gd(n,\gamma) \rightarrow {}^{158}Gd + \gamma + \text{X-rays} + \text{IC } e^- + \text{ACK } e^-$$

reactions which lead to the emission of low-energy gamma rays and internal (IC) and Auger (ACK) conversion electrons. Gd does have a high internal conversion coefficient of nearly 39% for emitting a conversion electron.

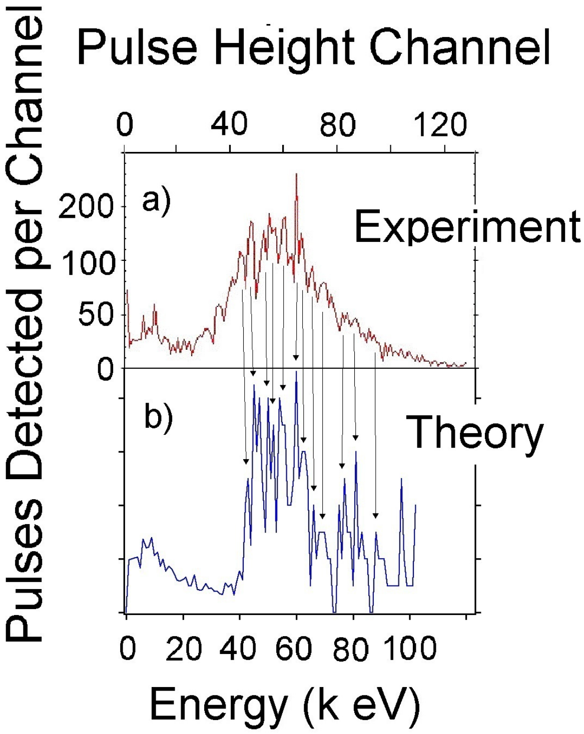


Fig. 1 The pulse height spectra of 15% Gd doped $HfO_2$ on n-type Si(100) samples with thermalized neutrons from a PuBe source with a flux of 600 neutrons $s^{-1}$ incident on the diode, compared to expectations.

We do observe rectification or diode like characteristics with both *p*-type 15% Gd-doped $HfO_2$ films deposited on *n*-type single crystal Si(100) substrates [1-2], and with *n*-type $Gd_2O_3$ films deposited on *p*-type single crystal Si(100) [4].

With the PuBe neutron source, the neutron detection efficiencies for 15% Gd doped $HfO_2$ on n-type Si(100) samples are $6x10^{-3}$, or about a factor 5 times smaller than the ideal expected. While the devices are seen to be effectively gamma blind, the effectiveness of the neutron detection, though pulse counting, was confirmed by comparison with an MCNP numerical experiment was conducted using MCNP5.0 [6]. To compare with theory, pulse height spectra were taken with electronic noise and signal suppression for pulses below 42 mV and with long time constants (100 microseconds), thus permitting many of the nuclear capture and decay resonances to be resolved in the pulse height spectra, as seen in the Figure.

In the model calculations, a planar source of $10^{11}$ neutrons was simulated calculated spectrum from 30 eV to 14 MeV. There are corrections at the low energy end of the model pulse height spectra to account for the large number of counts rejected by electronic noise and signal suppression below 42 mV pulse height. As seen in the Figure, with these corrections, the agreement is surprisingly good between the observed pulse height spectra and expectation for *p*-type 15% Gd-doped $HfO_2$ heterostructures formed on *n*-type Si(100) substrates. There are deviations between theory and experiment occurring for the larger pulse heights. The deviations indicate that there is likely incomplete charge collection in the experiment. This incomplete charge collection is to be expected, given that the 15% Gd-doped $HfO_2$ film is less than a micron thick.

## Conclusions

There is now a basic proof of principle to the idea that a Gd rich semiconductor containing heterostructure can detect neutrons. The optimization of both the materials and device structure is far from complete and the materials growth properties are not fully understood as yet, but successful solid state devices can be fabricated based on the concept.

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