Evidence for a positron bound state on the surface of a topological insulator

K Shastry¹, A H Weiss¹*, B Barbiellini², B A Assaf², Z H Lim¹, P V Joglekar¹, D Heiman²
¹ Physics Department, University of Texas at Arlington, Arlington, TX 76019-0059 USA.
² Physics Department, Northeastern University, Boston, MA 02115 USA.
*weiss@uta.edu

Abstract. We describe experiments aimed at probing the sticking of positrons to the surfaces of topological insulators using the Positron Annihilation induced Auger Electron Spectrometer (PAES). A magnetically guided beam was used to deposit positrons at the surface of Bi₂Te₂Se sample at energy of ~2eV. Peaks observed in the energy spectra and intensities of electrons emitted as a result of positron annihilation showed peaks at energies corresponding to Auger peaks in Bi, Te and Se providing clear evidence of Auger emission associated with the annihilation of positrons in a surface bound state. Theoretical estimates of the binding energy of this state are compared with estimates obtained by measuring the incident beam energy threshold for secondary electron emission and the temperature dependence positronium (Ps) emission. The experiments provide strong evidence for the existence of a positron bound state at the surface of Bi₂Te₂Se and indicate the practicality of using positron annihilation to selectively probe the critically important top most layer of topological insulator system.

1. Introduction

Positron annihilation spectroscopies have been shown to have a high degree of surface selectivity in systems in which positrons can become trapped in an image potential surface state at the time of annihilation [1]. Positrons in such states annihilate primarily with atoms in the topmost atomic layers and annihilation related signals convey information regarding the elemental composition and electronic structure of these layers [2, 3]. Topological Insulators (TI) [4] are a novel class of materials that behave as conductors on the surface and insulators in the bulk. In these materials the electronic and spintronic states of interest are confined to the topmost layer of the surface [5], [6].

In this paper we present the results of experiments that provide strong evidence that positrons can be efficiently trapped in surface localized states on TI surfaces. These experiments show that it is likely that positron spectroscopic techniques such as Two Dimensional – Angular Correlated Annihilation Radiation (2D- ACAR), Doppler broadening [7] and Spin-polarized positron spectroscopies [8] can be successful used to selectively probe the momentum distribution and spin properties of the topologically constrained electron surface states on TI materials.

Shastry et al. [9] have reported preliminary results showing positron annihilation induced Auger peaks from a Bi₂Te₂Se sample which provided strong evidence for the localization of the positron in a surface state at the time of annihilation. Subsequently, theoretical calculations by R.Sainz and B. Barbiellini [10] based on the model in which the positron is assumed to be bound to the surface in the...
form of a physisorbed Ps atom [11] indicated that the positron would be bound to the surface of Bi$_2$Te$_2$Se. They calculated the binding energy of the Ps to be between -0.12 eV and -0.4 eV.

Here we provide details of the PAES measurements on Bi$_2$Te$_2$Se along with the results of two additional experiments, Auger mediated sticking and Ps thermal desorption to confirm the existence of the surface state and help provide limits on the binding energy.

2. Experimental Details

The Bi$_2$Te$_2$Se topological insulator sample used in our experiments was grown on a Si (111) substrate using Molecular Beam Epitaxy (MBE) [6]. The positron measurements were carried out using a magnetic bottle analyzer type Time-Of-Flight Positron Annihilation induced Auger Electron Spectrometer (TOF-PAES) at University of Texas at Arlington [12],[13],[9]. The positrons are generated via $\beta$ decay from a $^{22}$Na radioactive source and are moderated using a polycrystalline tungsten (W) moderator. A combination of electric and axial magnetic field is used to guide the slow positron beam to the sample. A magnetic field of 4mT was used to efficiently transport the low energy positrons to the sample and the resulting electrons from the sample. The beam energy of the incident positrons is given by

$$E_p = e(V_m - V_s) + \phi_m^+$$

where $\phi_m^+$ is the positron work-function of the moderator (W). The values $V_m$ and $V_s$ represent the biases on the moderator and sample respectively, $e$ is the magnitude of the electronic charge. The beam energy for PAES and Ps fraction measurement were 9 eV and 70 eV respectively. The beam energies for the AMS measurement ranged from 2.7 eV to 6.2 eV.

The PAES and AMS measurements were done in the reverse timing scheme where the signal from the Micro Channel Plate is used as the START signal and the delayed BaF$_2$ signal is the STOP signal to a Time to Amplitude Converter (TAC). The resulting spectrum is a histogram of the time of flight of electrons from the sample to the MCP. A calibration procedure detailed in (Ref .13) is used to reference the energy of the electron with respect to the vacuum band. For the Ps fraction, the gamma spectrum from the NaI detector is passed to a timing amplifier and then to a multi-channel analyzer. A heater button behind the sample is used to raise the temperature of the sample in-situ.

3. Results and Discussion

3.1. Positron Annihilation Induced Auger Electron Spectrum (PAES) Measurement:

The TOF PAES of the Bi$_2$Te$_2$Se sample are shown in Fig. 2. A corresponding energy spectra (shown in Fig. 3) was obtained using a transformation [9]. The energy spectrum shows the Auger peaks at 240 eV and 100 eV due to the Bismuth N-VV transition [14], at 30 eV due to the Tellurium N-VV transition, at 42 eV due to the Selenium M-VV transition [15] and a Carbon Auger S-VV peak at 260 eV.

The observation of annihilation induced Auger peaks is a strong indication for the existence of a positron surface state on the surface of Bi$_2$Te$_2$Se. It should be noted that the beam energy used to obtain the Auger signal is very low to cause Auger transitions due to impact ionizations. In addition, if the positrons were in a bulk state at the time of annihilation only a tiny fraction of the positrons would annihilate close enough to the surface for an electron to escape without losing energy. This distance is in the order of 10 Å for a 60 eV electrons. The Bi$_2$Te$_2$Se sample is 40 nm thick; if the Auger signal is from below 10 Å the Auger signal intensity would be 100 times weaker than that from the surface. It is therefore clear that for a significant Auger signal to be observed for a given material for an incident beam energy below the core ionization energy, the positrons must be trapped in an image correlation well at the surface at the time of annihilation. Hence the observation of a positron annihilation induced Auger signal for a 9eV positron beam provides strong evidence for the existence of a surface localized positron bound state. A further implication of the observation of the PAES signal from the TI surface is that positron spectroscopies such as 2D-ACAR and Doppler Broadened Annihilation Gamma Spectroscopy can be used to sample the characteristics of the top atomic layers of these materials.
3.2. Auger Mediated Sticking (AMS) Measurement:
We report here the results of a series of measurement aimed at estimating the surface state binding energy by determining the cut-off energy for positron induced secondary electron emission from the Bi$_2$Te$_2$Se sample. Previous experiments by Mukherjee et al. [16] have demonstrated that measurements of the secondary electron yield as a function of incident positron beam energy yield estimates of the surface state binding energy in agreement (within the errors) with previous estimates of the surface state binding energy obtained from Ps thermal desorption [20]. Following reference [16], the maximum energy that a secondary electron can escape from the sample for a process in which a positron makes a direct transition from a positive energy scattering state to a surface bound state may be found using:

\[ E_{K_{max}} = E_p + E_{bss} - \varphi^- \]  

(2)

where \( E_{K_{max}} \) is the maximum energy a secondary electron can escape from the surface, \( E_p \) is the kinetic energy with which the positrons hit the sample, \( E_{bss} \) is the surface state binding energy, \( \varphi^- \) is the work-function. Figure 4 is the secondary electron spectrum at different beam energies. It may be observed in figure 3 that the area under the secondary electron peak is going to zero as the beam energy decreases.

\[ E_{Th} = \varphi^- - E_{bss} \]  

(3)

where \( E_{Th} \) is the threshold kinetic energy above which it is energetically possible for the positrons to knock out secondary electrons. The data points plotted in Figure 4 show the values of the integrated intensities under the measured positron induced secondary electron energy distributions (shown in figure 3) as a function of incident positron kinetic energy. A straight line fit to the data shown in figure 4 was used to estimate the value, \( E_{Th} \), at which the integrated secondary electron yield would go to zero. Using this method we obtained a value of \( E_{Th} = 1.8 \) eV for the Bi$_2$Te$_2$Se sample measured. The surface state binding energy was then calculated from the resulting value of \( E_{Th} \) using equation (3). Reported values of the electron work-function values for Bi$_2$T$_2$Se ranged from 4.32 eV [17] to 5.23 eV [18] were taken from literature. The surface state binding energy was calculated to be between 2.5 eV to 3.4 eV.

Figure 1. (Color Online)The TOF Auger spectrum for Bi$_2$Te$_2$Se sample after sputtering. The Auger transitions due to C (K VV) at 257 eV, Bi (N VV) at 240 eV and (N-VV) at 100 eV, Te (N-VV) at 30 eV, Se (M-VV) at 40 eV are shown.

The emission of secondary electrons is energetically precluded when \( E_{K_{max}}=0 \). Inserting this value into equation (2) we find:

The data points plotted in Figure 4 show the values of the integrated intensities under the measured positron induced secondary electron energy distributions (shown in figure 3) as a function of incident positron kinetic energy. A straight line fit to the data shown in figure 4 was used to estimate the value, \( E_{Th} \), at which the integrated secondary electron yield would go to zero. Using this method we obtained a value of \( E_{Th} = 1.8 \) eV for the Bi$_2$Te$_2$Se sample measured. The surface state binding energy was then calculated from the resulting value of \( E_{Th} \) using equation (3). Reported values of the electron work-function values for Bi$_2$T$_2$Se ranged from 4.32 eV [17] to 5.23 eV [18] were taken from literature. The surface state binding energy was calculated to be between 2.5 eV to 3.4 eV.
respectively. The initial theoretical calculations based on first principles, for the surface state binding energy varied from 1.61 eV to 2.61 eV.

Figure 2. (Color Online) The energy spectrum for Bi$_2$Te$_2$Se sample after sputtering. The peaks due to the Auger transitions C (K VV) at 257 eV, Bi (N VV) at 240 eV (N-VV) at 100 eV, Te (N-VV) at 30 eV, Se (M-VV) at 40 eV are indicated by arrows. Inset: magnified region between 200 and 400 eV.

Figure 3. (Color Online) The energy distribution of secondary electrons obtained at a series of incident positron kinetic energies ranging from 2.7eV to 6.2 eV.

Figure 4. (Color Online) The data points represent the integrated intensity under the secondary electron energy distributions show in figure 3. The line is a fit to the data.

The AMS measurement on Bi$_2$Te$_2$Se sample constitute a second piece of evidence for the existence of a positron surface state. If the AMS signal came from the bulk and not from the surface, the maximum energy of the secondary electron would be given by

\[ E_{K,max} = E_p - \varphi^- + \varphi^+ \]  

(4)
where $E_P$ is the kinetic energy of the positrons incident on the surface, $E_{K_{\max}}$ is the maximum Kinetic energy of the outgoing electron measured outside the sample surface, $\varphi^-$ and $\varphi^+$ are the electron and positron work function respectively. For secondary electrons to be ejected out of the sample ($E_k > 0\text{eV}$), $E_P$ should be greater than the difference between the electron and positron work functions. Typically for materials the value of $\varphi^-$ is $\sim 5\text{eV}$ while $\varphi^+$ is $< 1 \text{eV}$. Therefore, from equation (4) the minimum incident beam energy for secondary electron emission should be $> 4 \text{eV}$, but in our experiment the beam energy was $2 \text{eV}$. The lower value of $E_{Th}$ observed in our experiment is consistent with the existence of a surface state whose binding energy is of order $2 \text{eV}$ below the bulk state.

3.3. Positronium (Ps) Fraction Measurement:

In the third experiment an estimate of the energy, $E_a$, required to remove the positron from a surface bound state was estimated from a measurement of the temperature dependence of the fraction of incident positron emitted as positronium, $f_{Ps}$, from the Bi$_2$Te$_2$Se surface using the method of Mills [19, 20]. The positron fraction, $f_{Ps}$, was determined from the annihilation gamma spectra measured using a NaI (TI) detector [19] using a calibration procedure detailed in reference 9. The Ps fraction obtained from the Bi$_2$Te$_2$Se at room temperature was $f_{Ps} = 0.11$ which is significantly lower than the value of $f_{Ps}$ obtained from metal surface (e.g. $f_{Ps} = 0.51$ for Cu (100)[20]). The data points shown indicate the measured values of $f_{Ps}$ for 5 different temperatures. The solid lines are functions obtained using the thermal activation model described in reference 20 for three values of the term $E_a$, representing the activation barrier for thermal emission. The value of the activation barrier, $E_a$, can be estimated from the surface state binding energy [20] using equation 5

$$E_a = E_{bss} + \varphi^- - 6.8 \text{eV}$$

$E_{bss}$ is the surface state binding energy, $\varphi^-$ is the electron work-function, 6.8 eV is the binding energy of ground state Ps. Based on published values of $\varphi^-$ and the corresponding values of $E_{bss}$ found from the AMS measurements discussed above, the values of $E_a$ determined from equation 5 range from 0 eV and 1.8 eV. It may be seen in figure 5. That the data are not consistent with models in which $E_a = 0 \text{eV}$ corresponding to spontaneous Ps emission, nor $E_a = 1.8\text{eV}$.

![Figure 5](Image)

Figure 5. (Color Online)The data points correspond to the measured values of the Ps fraction, $f_{Ps}$, for 5 different temperatures. The curves represents values obtained using the model given in Ref. 20 for 3 different activation energies.

The curve corresponding to an activation energy of $E_a = 0.4\text{eV}$ is consistent with our $f_{Ps}$ vs temperature data. This value of $E_a$ is also consistent with our AMS measurements assuming $\varphi^- = 4.7 \text{eV}$. Based on the Ps fraction dependence on temperature of Bi$_2$Te$_2$Se, the value of $E_a$ that is most consistent with our data is $0.4 \text{eV}$. 

---

5
4. Conclusion

We have presented the results of three different measurements which provide strong evidence for a positron bound state on the surface of the topological insulator. The observation of positron annihilation induced Auger electron peaks from the Bi$_2$Te$_2$Se surface provide strong evidence that the positrons are localized at the surface at the time of annihilation. Measurements of the dependence of the positron induced secondary electron yield as a function of incident beam energy and the temperature dependence of the fraction of incident positrons forming Ps provided complementary evidence for the existence of the surface state. Latter experiments allowed us to estimate values for the activation energy for Ps thermal emission, $E_a = 0.4$ eV, and a positron surface state binding energy $E_{bss} = 2.7$ eV. These values obtained are within the range of preliminary theoretical calculations [10]. Though the effects observed in this paper are not topological in nature, the results indicate that low energy positrons can be trapped with high efficiency into a surface localized bound state. Consequently, the topological effects could be probed with positrons employing other positron techniques such as polarized positron beam, 2D- ACAR and Doppler broadening spectroscopies, which are of most interest for device applications.

We acknowledge Dr. Moodera for his help in growing the Bi$_2$Te$_2$Se samples. We also acknowledge Dr. Sainz and Dr. Barbiellini for their useful discussions. We also acknowledge funding from Welch Grant 1100 and NSF DMR 0907679.

References

[1] Weiss, A H and Coleman, P G Surface Science with Positrons, in Positron Beams and Their Applications, World Scientific Publishing, pp 129-189 (2000).
[2] Weiss, A H et al., Phys.Rev.Lett. 61, 2245 (1988).
[3] Asoka-Kumar, P et al., Phys.Rev.Lett. 77, 2097 (1996).
[4] Kane, C L Mele, E J Phys. Rev. Lett 95, 1456802 (2005).
[5] Moore, J E Nature 464, 194 (2010).
[6] Assaf, B A Ph.D thesis, Northeastern University, (2014).
[7] Coleman, P G Positron beams and applications, World Scientific Publishing, pp 129-189 (2000).
[8] Gidley, D W, Koymen A R et.al Phys.Rev.Lett. 49,24(1982).
[9] Shastry, K et al. Bulletin of the American Physical Society Volume 58, Number 1 M13.00004 (2013) and Shastry, K Ph.D dissertation, December (2014).
[10] Saniz, R et al., Journal of Physics: Conference Series 505 (2014) 012002.
[11] Platzman, P M et al., Phys. Rev. B 33 5900 (1986).
[12] Xie, S Ph.D. thesis, UT Arlington, (2002).
[13] Mukherjee, S F et.al, Rev.Sci.Instrum (under preperation).
[14] Roy Sumalay et al., Phys.Rev.B 90, 155456 (2014).
[15] Zhu, J G et al.,Journal of Applied Physics ,Volume:97 Issue:10.
[16] Mukherjee, S et al., Phys.Rev. Lett. 104, 247403 (2010).
[17] Hao, G Journal of Applied Physics 113, 024306 (2013).
[18] Niessner, D et al.,Journal of Electron Spectroscopy and Related Phenomena 195 (2014).
[19] Mills Jr, A P et al., Solid State Comm, Vol 31, (623-626).
[20] Mills Jr, A P et al., Phys.Rev.Lett, Vol 41, Number 26.