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Measurement of Electron Beam Polarization from Unstrained Bulk GaAs via Two Photon Photoemission

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Abstract. This paper describes measurements of the beam polarization and quantum efficiency for photoemission using two-photon excitation from unstrained bulk GaAs illuminated with pulsed, high intensity 1560nm laser light. Quantum efficiency is linearly proportional to 1560nm peak laser intensity, which was varied in three independent ways, indicating that the emitted electrons are promoted from the valence to the conduction band via two-photon absorption. Beam polarization was measured using a microMott polarimeter, with a value of 16.8(4)% polarization at 1560nm, which is roughly half the measured value of 33.4(8)% using 778 nm light.

1. Introduction

Polarized electron sources are important components of particle accelerators like the Continuous Electron Beam Accelerator Facility (CEBAF) at Jefferson Lab, where the polarized electron beam is used to study nuclear structure, the dynamics of strong interactions, electro-weak nuclear physics including parity-violation, and physics beyond the Standard Model. The first GaAs-based polarized electron source used at an accelerator [1] provided beam polarization $\sim 35\%$, with polarization limited to less than 50% by the energy level degeneracy in the $^2P_{3/2}$ valence band state of unstrained bulk GaAs (Figure 1a) [2]. Significantly higher beam polarization was obtained in the 1990s by introducing an axial strain within the GaAs crystal structure to eliminate this degeneracy [3,4]. Today, beam polarization routinely exceeds 80% using strained-superlattice GaAs/GaAsP structures [5,6,7,8]. However, these high polarization photocathodes are expensive and delicate compared to unstrained bulk GaAs, making them more difficult to use.

This paper presents a summary of measurements of quantum efficiency and polarization of photo-emitted electrons excited through two-photon absorption from unstrained bulk GaAs, as analyzed by a compact retarding-field microMott polarimeter.

2. Theoretical background

Nakanishi first proposed using two-photon absorption with unstrained bulk GaAs to obtain high polarization [9]. He reasoned that quantum mechanical selection rules associated with the absorption of two photons of circularly-polarized light at half of the bandgap energy would allow excitation of only one spin state to the conduction band (Figure 1b). Although the efficiency of this non-linear optical process would be small, the observation of beam polarization $> 50\%$ from unstrained bulk

GaAs would be an exciting development, giving insight into the band structure and allowed quantum mechanical transitions in GaAs, and would be useful in a practical sense because research projects with polarized electrons often require only very low current beam. Furthermore, recent advances in the lasers used for telecommunications have made high power light sources at 1560nm inexpensive and abundant. Photoluminescence experiments support this theory with polarization measured at 58% and extrapolated to infer that the polarization at excitation was 95% [10].

A more sophisticated theory by Bhat et al. [11] suggests electrons make transitions from the valence and conduction bands via two-photon absorption as governed by quantum mechanical selection rules (as suggested by Nakanishi), but also via “forbidden” intraband transitions within conduction or valence bands, with angular momentum of the photons transferred to carrier orbital motion and the crystal structure, and not just the electron spin. They predict polarization via two-photon absorption will be comparable to conventional one-photon induced polarization, a value less than 50%. Experimental results supporting this theory used differential transmission pump/probe techniques to measure nascent polarization equal to 48% [11,12].

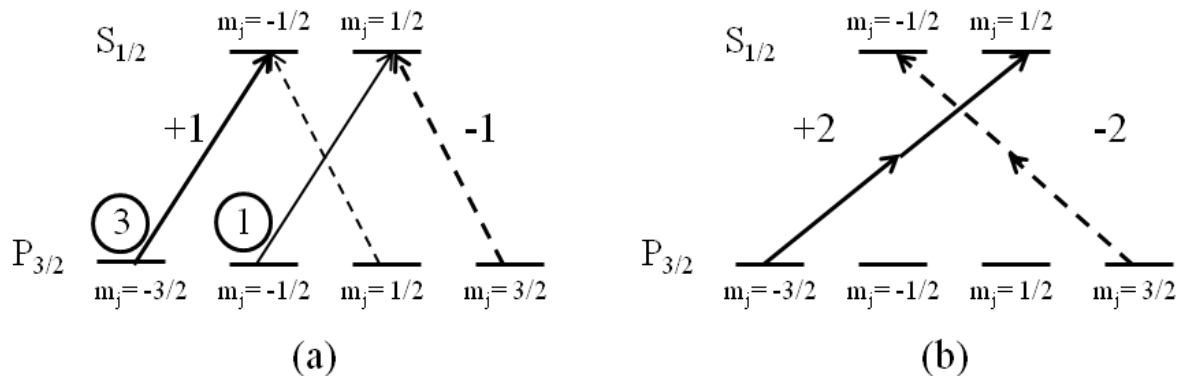


Figure 1. Optical transitions in GaAs with circularly polarized light from the $^2P_{3/2}$ valence band to the $^2S_{1/2}$ conduction band: a) one-photon excitation, and b) two-photon excitation, as suggested by Nakanishi. The circled values indicate transition probabilities. Maximum polarization from un-strained bulk GaAs would be 50% for one photon excitation and 100% for two photon excitation.

Two-photon absorption is a non-linear optical process [13] that only occurs within crystals that lack inversion symmetry, such as GaAs. The cross-section for this process is small, meaning the likelihood that two photons with energy less than the band gap can be absorbed simultaneously to excite an electron from the valence band to the conduction band is small compared to conventional one-photon absorption with light having more energy than the band gap.

For conventional, one-photon photoexcitation, the number of electrons emitted is proportional to the number of incident photons, or laser power, and is constant with laser intensity. Equation 1 [11, 14,15] relates the expected QE at 1560 nm to that at 778 nm:

$$\frac{QE_{1560}}{QE_{778}} = 4\pi e^{hc} \frac{2(2h\omega - E_{gap})}{h\omega} \frac{I}{m_c \omega^3} = 2 * 10^{-12} * I \left(\frac{W}{cm^2} \right), \quad (1)$$

where $\omega = \frac{2\pi c}{\lambda}$ is the frequency of the light, I is the light intensity, m_c is the electronic effective mass, and E_{gap} is the band gap energy.

There are two noteworthy features of this equation. First, QE_{1560nm} due to two-photon absorption is proportional to laser intensity, and secondly, QE_{1560nm} will be very small unless intensity is quite large. There are three relatively simple ways to provide high intensity for this experiment: a) illuminate the photocathode with high average power, b) use short-pulse light to generate high peak power, and c)

focus the incident laser beam to a small spot at the photocathode. Laser intensity was varied through each of these methods to verify the two-photon absorption and photoemission process.

3. Experiment

A load-locked vacuum apparatus was employed, consisting of three chambers (Figure 2a): a low-voltage source chamber for installing and activating photocathodes, a beam transport section, and a microMott retarding-field polarimeter. Un-strained bulk GaAs was mounted to a long stalk that could be lowered into the source chamber and activated to negative electron affinity (NEA) with Cs and NF_3 . The photocathode was biased at -268V and the emitted electron beam was delivered to the microMott polarimeter using a 90° electrostatic deflector [16] and electrostatic steering lenses [17]. The beam transport system and the microMott polarimeter are described thoroughly in another publication [18]. Optical systems for each wavelength, 778nm and 1560nm , could be quickly and reproducibly moved in and out of position beneath the vacuum chamber. They comprised numerous elements manufactured for the appropriate wavelength: optical attenuators, linear polarizers, quarter-wave plates, and insertable half-wave plates. When the 1560nm laser system was employed, long pass optical filters (two at 1350nm and one at 850nm) were inserted to ensure that no light below 850nm could enter the vacuum apparatus. This was very important, because fiber amplifiers (described below) produce a small amount of light in the visible and near-IR wavelengths via second harmonic generation. Even a small amount of undetected light in the visible or near-IR wavelength range could easily overwhelm the beam produced at 1560nm where two-photon photocurrent is comparatively small. Insertion of additional 1350nm long pass filters had no effect on the amount of photocurrent, indicating that only 1560nm light reached the photocathode. Finally, a mirror system mounted to dual translation stages directed the laser beam vertically into the source chamber and provided a means to “QE scan” the photocathode (see below).

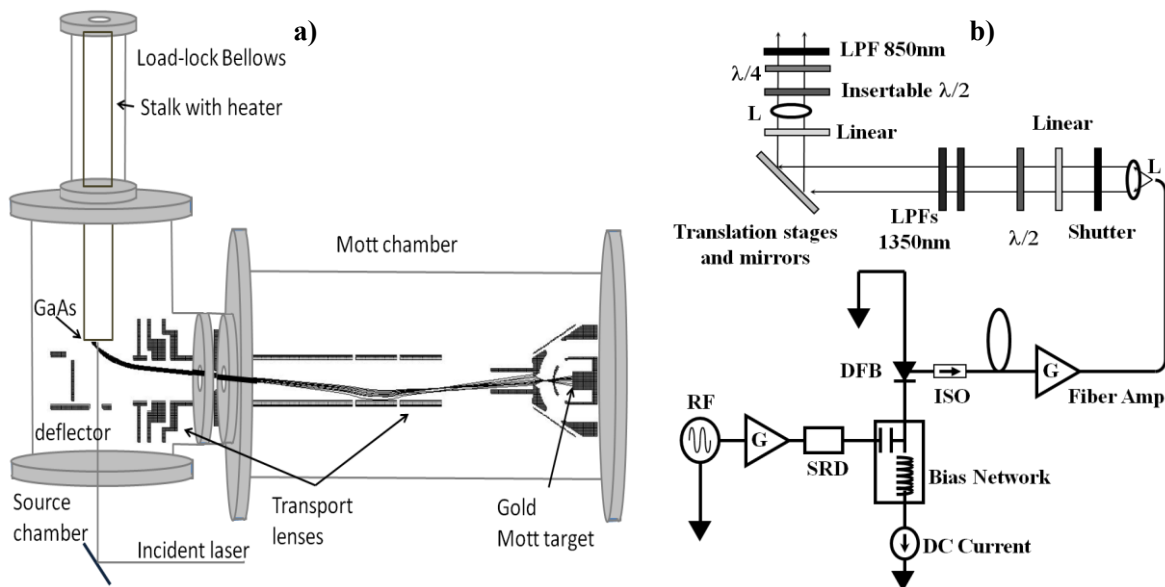


Figure 2. a) Schematic of the experimental apparatus, b) Schematic of 1560nm light source used to generate two-photon photo-emitted beam from un-strained bulk GaAs. DFB, distributed feedback Bragg reflector diode laser; ISO, fiber isolator; SRD, step recovery diode; L, lens; Linear, linear polarizer; $\lambda/2$ and $\lambda/4$, half-wave and quarter-wave plates; LPF, long pass filter.

The 778nm laser was a simple low-power diode laser that could be operated in DC or RF pulsed-mode via gain-switching at repetition rates from 250 to 2000 MHz. The 1560nm laser was composed of a gain-switched fiber-coupled diode “seed” laser and fiber-amplifier (Figure 2b), that could produce up to 5W average power with 35ps optical pulses over the same range of pulse repetition rates as the 778nm laser system [19]. By using short pulse length light, high peak power was obtained to enhance the two-photon absorption process.

As mentioned above, the QE_{778nm} for clean and undamaged GaAs should be independent of incident laser power, intensity, peak power or laser spot size. For two-photon absorption, QE_{1560nm} should vary linearly with laser intensity. In this experiment, there were three “knobs” to vary the intensity at the photocathode: the average power of the light, the spot size of the laser beam on the photocathode, and the pulse repetition rate (while keeping pulsewidth and average power constant). Equations 2 and 3 illustrate how these parameters affect intensity at the photocathode:

$$I_{avg} = P_{avg}/A \quad , \quad (2)$$

$$I_{peak} = \frac{I_{avg}}{D} = \frac{P_{avg}}{A*D} = \frac{P_{avg}}{A*t*f} \quad , \quad (3)$$

where P_{avg} is the average power of the laser, D is duty factor equal to the product of the optical pulsewidth t and the laser pulse repetition rate f , and A is the spot size of the laser on the photocathode. High intensity was obtained by increasing the average power of the incident laser light using an optical attenuator, and by decreasing the size of the laser beam. Different laser spot sizes were obtained by simply using lenses of different focal length. A 30cm focal length lens produced a beam waist (i.e., the smallest spot) at the photocathode and longer focal length lenses placed in the same location below the vacuum window produced larger spots at the photocathode. Because changing lenses in the optical system can steer the beam, QE scans were done after each lens change (see below).

The rf-pulsed nature of the laser systems provided a means to easily vary the peak intensity of the light at the photocathode. The optical pulsewidth of the gain-switched laser system remained approximately constant across the range of pulse repetition rates: 35ps FWHM. The fiber amplifier provided the same output power for each repetition rate, so the peak intensity of the laser system could be varied by changing the pulse repetition rate, with the highest intensity obtained at lower pulse repetition rates.

The unstrained bulk GaAs wafer was activated, and reactivated as needed by heating to $\sim 550^\circ\text{C}$ to clean the surface and by applying Cs and NF_3 to make a NEA surface [20]. Each measurement comparing one and two-photon emission was taken during the same activation. Due to non-uniformity of the Cs deposition at the photocathode, the QE of the GaAs wafer was not constant over its surface. Where relevant, data are presented at the location of highest QE and at the location of the highest transmission to the polarimeter, which were not identical. The highest transmission - about 20% of the extracted beam delivered to the polarimeter - occurred from a specific location of the GaAs photocathode about 2mm in diameter. A QE scan of the photocathode was performed each time a laser system was installed, to identify the region of highest transmission and to ensure that comparative measurements between 778 and 1560nm laser systems were made using beam extracted from the same region of the photocathode.

4. Results

4.1 Two Photon QE

The unstrained bulk GaAs was first evaluated with 778nm light. As expected, QE_{778nm} remained nearly constant versus input power and pulse repetition rate (Figure 3a). In fact, there was a slight decrease in QE at higher laser power (made more apparent by plotting with zero suppression),

observed for all repetition rates; this was attributed to the surface charge limit effect [21]. The QE behavior at 1560nm however, was markedly different (Figure 3b), as higher intensity produced higher QE, increasing linearly with power and intensity, indicative of a two-photon absorption process. As described above, average power (and average intensity) was varied by adjusting an optical attenuator, whereas peak intensity was varied by selecting different pulse repetition rates. It should be noted that high average power ($\geq 2\text{W}$) heated the photocathode, which significantly degraded its lifetime [22]. Cold nitrogen gas was applied to the back surface of the photocathode stalk to help maintain the QE during measurements. The slight roll-over of QE plots at high average power is likely a result of photocathode heating.

The repetition rate of the laser was also changed while keeping the average power constant, which according to equation 3 changed the peak laser intensity. As can be inferred from Figure 3b, the QE was linear with the laser intensity, which was again indicative of a two-photon process.

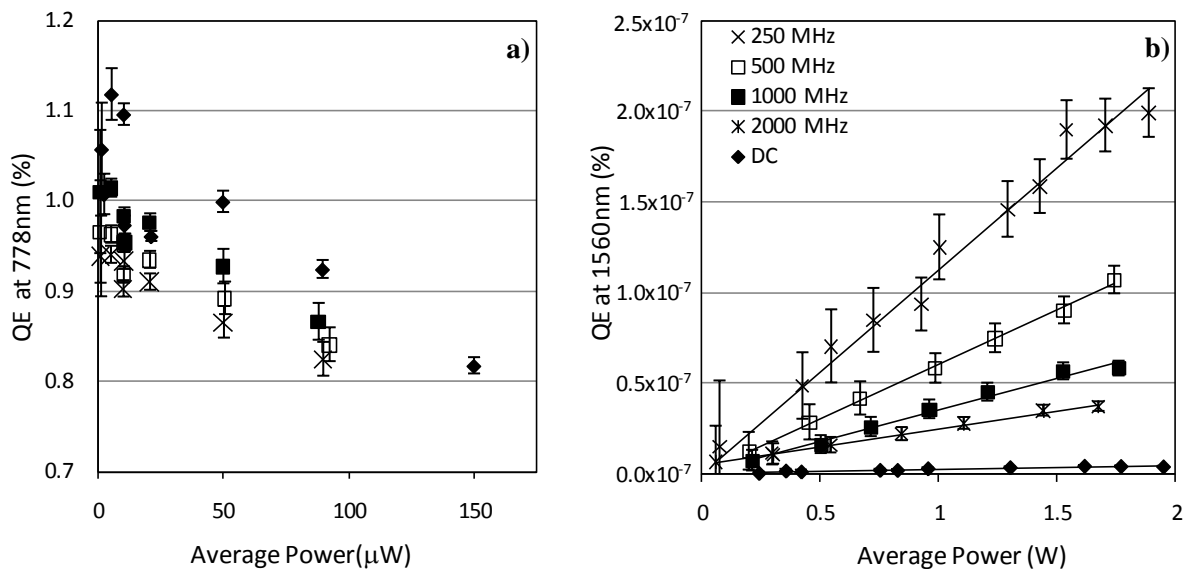


Figure 3. a) QE vs. input power at 778nm, adjusted by optical attenuator for a given repetition rate, and b) QE at 1560nm with the same repetition rates used in Figure 3a.

Further verification of two-photon photoemission was obtained by changing the size of the laser spot at the photocathode for both laser wavelengths, with results shown in Figure 4. The pulse repetition rate was 250MHz. As noted above, the photocathode activation was not uniform across the surface, so that the location of maximum QE was not coincident with the location of maximum beam transmission to the microMott polarimeter. The QE scans were performed to ensure that comparative measurements with both wavelengths were always made at the same photocathode location. Measurements in Figure 4 show results from both locations. At 778nm, the QE was nearly constant versus peak intensity, with maximum QE rising slightly at the smallest spot sizes due to the non-uniformity of the QE of the photocathode. The behavior at 1560nm however, was quite different from that at 780nm, namely, QE increases linearly with laser peak intensity (Figure 4b). Also shown is the theoretical predicted QE using a base QE at 778nm of 1.32%. The QE is proportional to the light intensity and is within an order-of-magnitude agreement with the predicted QE values, again indicating that two-photon absorption is the cause of the photocurrent.

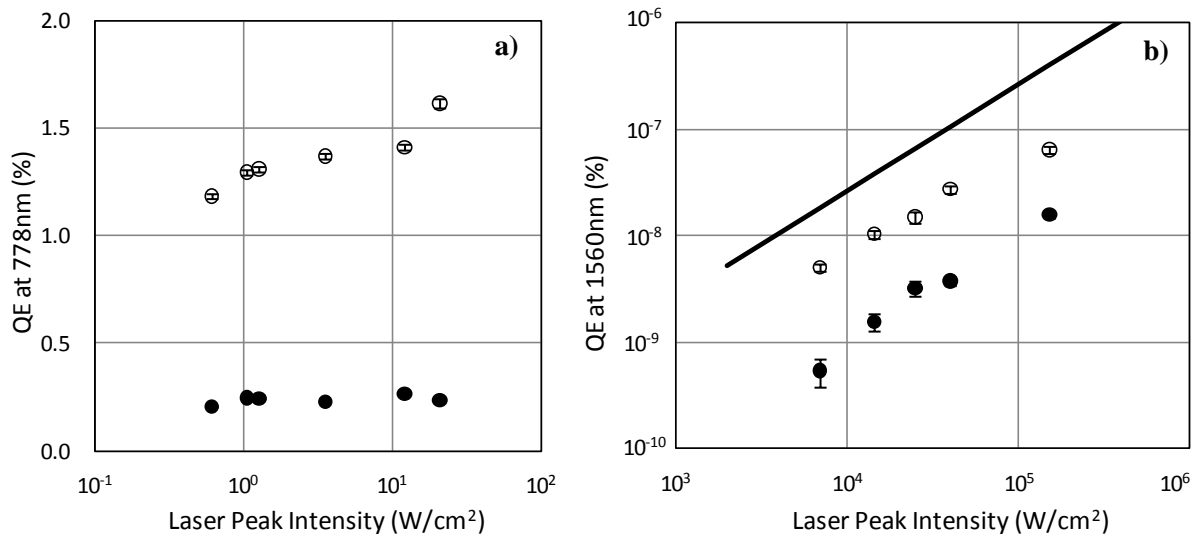


Figure 4. QE vs. peak intensity of the laser beam caused by changing the laser spot size at the cathode. The QE across the photocathode was not uniform: open circles represent maximum QE values, closed circles represent QE values at photocathode location for maximum transmission to the polarimeter. a) QE at 778nm. b) QE at 1560nm. The solid line is the theoretical QE (Eqn.1) for a two photon process at 1560nm, assuming a 1.3% QE at 778nm. Data were obtained using a laser repetition rate of 250MHz.

4.2 Two Photon Polarization

The polarization of electron beams generated with both one- and two-photon excitation was measured using the microMott polarimeter with its target biased at 20kV. For each polarization measurement, retarding field scans were performed to determine instrumental backgrounds. Repeated measurements, shown in Figure 5, indicate that the polarization for both wavelengths was stable with regard to extracted photocurrent. Photocurrents higher than 1nA were not tested using 1560nm light, as above this value the lifetime of the cathode was reduced to unusable levels due to laser heating of the GaAs. Measurements at 778nm were not made below 1nA in order to keep statistical errors low. Also included in Figure 5 is polarization data from a strained-superlattice GaAs photocathode taken using the same polarimeter, which indicates there is no instrumental dependence of the polarization on extracted photocurrent across the range of interest.

The effect of laser polarization on beam polarization is shown in Figure 6. The angle of the quarter-wave plate was rotated to vary the degree of circular polarization of the incident light. As expected, the electron polarization for both one- and two-photon excitation varies sinusoidally with the degree of circular polarization of the light. This behavior of the electron polarization with the degree of light polarization indicates that there is no issue with the optical arrangement that systematically affects the electron polarizations, which were measured to have maximum values of 16.8(4) % for 1560nm light and 33.4(8) % for 778nm light.

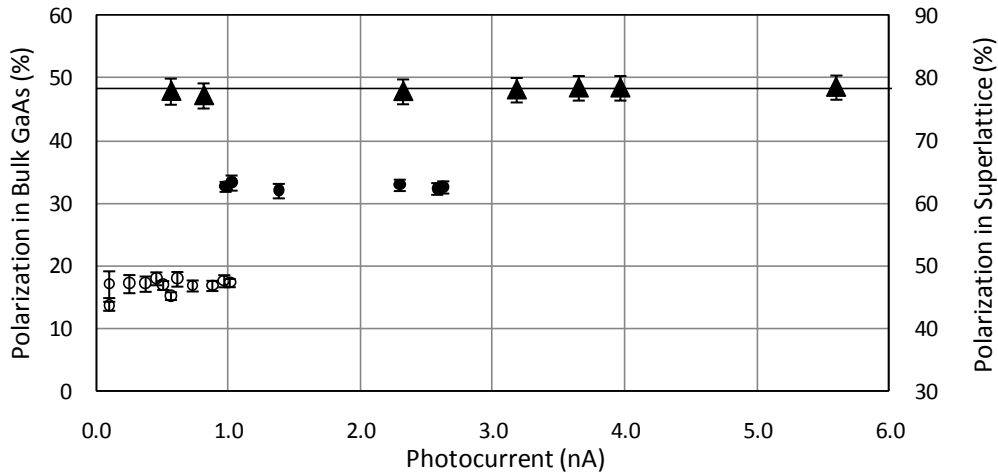


Figure 5. Polarization as a function of extracted photocurrent from unstrained bulk GaAs: 1560nm (open circles) and 778nm (closed circles); polarization measurements from strained-superlattice GaAs at 772nm (solid triangles), with the straight line representing the average value.

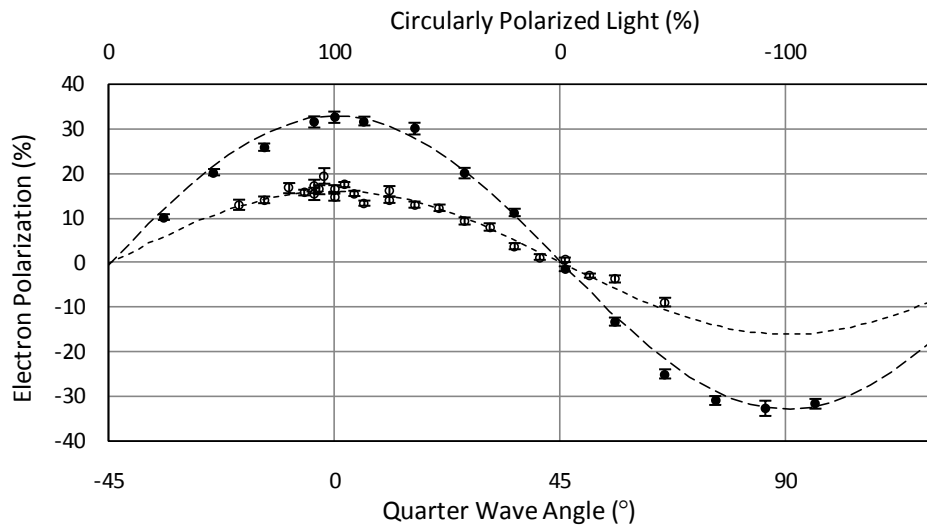


Figure 6. Polarization of the photoelectron beam at 778nm (closed circles) and at 1560nm (open circles) as a function of the orientation of the quarter-wave plate. Sinusoidal fits to the data are indicated by the dashed lines.

5. Conclusion

The polarization of electrons emitted by two-photon excitation of GaAs using 1560 nm laser light has been measured to be 16.8(4)%, roughly half the polarization using 778nm light, which is 33.4(8)%. The photoemission was confirmed to be due to two-photon absorption, as indicated by the linear dependence of the QE_{1560nm} on the incident laser intensity, varied using three different methods.

To explain this unexpected low polarization result, we speculate the electrons excited via two-photon absorption originate from relatively deep within the material, because the absorption coefficient is so small. Using a GaAs wafer polished on both sides, the absorption length for GaAs at 1560nm was measured to be $\sim 220\mu m$, while the absorption length at 778nm was $\sim 1\mu m$. Measurements using time resolved polarimetry [23] show electrons depolarize in the bulk prior to

emission from the surface. A larger proportion of electrons created via the two-photon process are created deeper inside the bulk material because of the attenuation lengths of the light used. The two-photon electrons have more time and distance to depolarize, thus decreasing the overall polarization of the photocurrent. Further studies are planned to confirm this hypothesis by using thin samples of bulk GaAs. Thinner GaAs samples should provide higher polarization, and it will be interesting to see if the two-photon polarization is always roughly half that of one-photon polarization.

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