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Measurement of electron beam polarization from unstrained GaAs via two-photon photoemission

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ABSTRACT

Two-photon absorption of 1560 nm light was used to generate polarized electron beams from unstrained GaAs photocathodes of varying thickness: 625 μm, 0.32 μm, and 0.18 μm. For each photocathode, the degree of spin polarization of the photoemitted beam was less than 50%, contradicting earlier predictions based on simple quantum mechanical selection rules for spherically-symmetric systems but consistent with the more sophisticated model of Bhat et al. (Phys. Rev. B 71 (2005) 035209). Polarization via two-photon absorption was the highest from the thinnest photocathode sample and comparable to that obtained via one-photon absorption (using 778 nm light), with values $40.3 \pm 1.0\%$ and $42.6 \pm 1.0\%$, respectively.

1. Introduction

Polarized electron sources are important components of particle accelerators, like the Continuous Electron Beam Accelerator Facility (CEBAF) at Jefferson Lab, where the spin of the 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 [1]. The first GaAs-based polarized electron source used at an accelerator [2] provided beam polarization ~35%, with a theoretical maximum polarization limited to 50% [3,4] due to the heavy-hole, light-hole energy level degeneracy of the $^2p_{3/2}$ valence band state (Fig. 1a). Significantly higher beam polarization was obtained in the 1990s by introducing an axial strain within the GaAs crystal structure [5–7] which eliminates this degeneracy (Fig. 1b). Today, beam polarization at accelerators routinely exceeds 80% using strained-superlattice GaAs/GaAsP structures [8,9]; however, these high-polarization photocathodes are thin compared to typical unstrained bulk GaAs and with respect to the photon absorption depth. As a result, strained-superlattice photocathodes exhibit significantly lower quantum efficiency (QE) than that of bulk GaAs samples [7,10].

Matsuyama et al. proposed using two-photon absorption, which is a non-linear optical process [11] that occurs only within crystals that lack inversion symmetry, as a mechanism to obtain high polarization from unstrained GaAs [12]. They reasoned that quantum mechanical selection rules associated with the simultaneous absorption of two photons of circularly-polarized light at half the band-gap energy would provide a means to populate the conduction band with electrons of just one spin state, yielding, in principle, completely polarized electrons in the conduction band immediately following excitation (Fig. 1c). According to this prediction, the electron polarization should be of opposite sign to that produced using single photon excitation. Subsequently, Matsuyama et al. [13] performed an experiment that relied on electron–hole photoluminescence measurements (but not photoemission) with electron–hole recombination fluorescence polarization measured to be 58%. This value was used to infer an electron polarization of 95% at the time of excitation to the conduction band [13]. While this result was consistent with an electron polarization of unity, the sign of the fluorescence polarization was inconsistent with their prediction.

In contrast to Ref. [13], a full examination of the quantum selection rules indicates that the transition depicted in Fig. 1c is not allowed, as two photons of like circular polarization must excite electron transitions with a change in azimuthal quantum number $\Delta\ell = 2$, which precludes a $^2p_{3/2}$ to $^2s_{1/2}$ transition at the Γ point. Bhat et al. [14] provided a detailed analysis of two-photon absorption in semiconductors, and predicted that polarization via two-photon absorption should be less than 50%. Photoluminescence

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experiments using differential transmission pump/probe techniques indicated a nascent polarization equal to 48%, in support of their predictions [14,15].

This paper presents the first direct measurement of electron beam polarization resulting from two-photon excitation of GaAs. We determined the electron polarization for three GaAs sample thicknesses using a compact retarding-field micro-Mott polarimeter. Two-photon absorption with 1560 nm light was verified by noting that quantum efficiency varied linearly with laser intensity, which was adjusted by various means. For each photocathode the degree of spin polarization of the photoemitted beam was less than 50%, with the same sign as that from our one photon measurements, contradicting both the prediction and photoluminescence measurements of Matsuyama et al. [12,13]. Polarization via two-photon absorption was the highest from the thin photocathode samples and comparable to that obtained via one-photon 778 nm absorption (~43%).

2. Theoretical considerations

Although spin–parity selection rules would bar a two-photon transition for a free atom, Bhat et al. [14] show that dipole-forbidden transitions are allowed in GaAs near the Γ point. To summarize the findings of Bhat the structure of a GaAs crystal violates the spherical symmetry of single-atom fields, which leads to the non-conservation of angular momentum for $\vec{k} \neq 0$. The perturbation of this spherical symmetry is very small for electrons near the center of the Brillouin zone, which is why the angular momentum is still an approximate quantum number for electron transitions induced by one-photon absorption in a direct band-gap transition. For two-photon absorption, the picture is more complicated, as spherical symmetry forbids such transitions at the center of the Γ point. However, because of the crystal lattice perturbation, there can be weak two-photon absorption in this region. In this interaction, there is a lack of angular momentum conservation between the incident photon and excited electron, which precludes electron polarization for two-photon absorption from being above 50%. Indeed, Bhat et al. [14] predict a polarization just above the band-gap threshold of ~+49% at 1560 nm, while the work in Ref. [12] predicts complete polarization of the opposite sign. Our experiment was performed to differentiate between the two predictions.

3. Experimental setup

Our apparatus consisted of a low-voltage polarized electron source chamber for installing and activating photocathodes, a beam transport section, and a micro-Mott retarding-field polarimeter (Fig. 2). Unstrained bulk GaAs was mounted on a long stalk that could be lowered into the source chamber. The GaAs was then activated, and reactivated as needed, by heating to ~550 °C to clean the surface and then by applying Cs and NF_3 to create a surface with negative electron affinity [16]. Three different wafers of unstrained GaAs were used. One sample, known as the "thick" sample (625 μm), was epi-ready unstrained bulk GaAs, with a (1 0 0) surface, p-doped with a Zn density of $\sim 5 \times 10^{18}/\text{cm}^{-3}$. The "thin" samples (0.18 and 0.32 μm) were grown via MOCVD with p-doping of Zn (density $\sim 4 \times 10^{18}/\text{cm}^{-3}$) on thick GaAs substrates, with an intervening barrier layer of p-$Al_{0.3}Ga_{0.7}As$ that was ~0.9 μm thick. The band gap of this barrier layer is much larger than that of GaAs, which ensured that no photoemission originated in the barrier layer from either 780 nm or 1560 nm light and also that any electrons excited in the substrate material did not reach the photocathode surface. For all GaAs samples, the photocathode was biased at −268 V using batteries and the emitted electron beam was delivered to the micro-Mott polarimeter using a 90° electrostatic deflector and electrostatic steering lenses [17,18]. The beam transport system and the micro-Mott polarimeter are described more thoroughly in another publication [19].

Two laser wavelengths were used: i.e. 778 and 1560 nm for one- and two-photon absorption, respectively. Optical systems for each wavelength could be quickly and reproducibly moved in and out of position beneath the vacuum chamber. When the 1560 nm laser system was in place (Fig. 3), long-pass optical filters (two at 1350 nm and one at 850 nm) were inserted into the laser path to

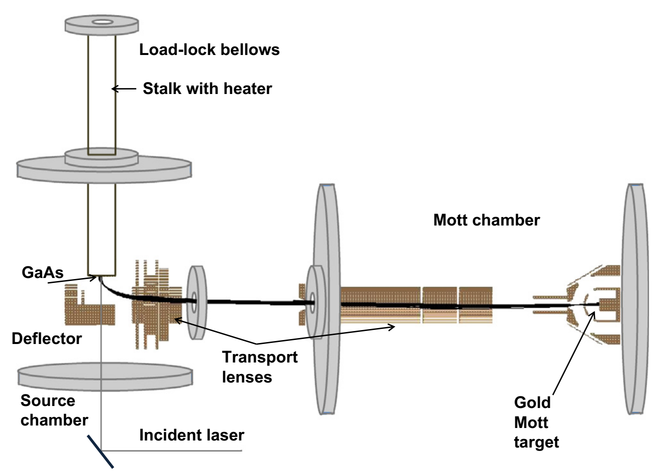

Fig. 2. Schematic of the experimental apparatus, with source chamber, transport, and Mott chamber sections. The black lines directed through the lenses represent electron trajectories simulated by SIMION.

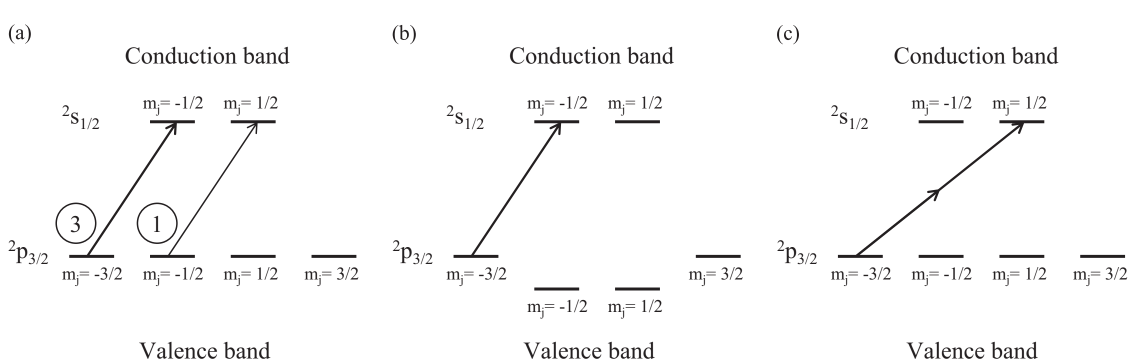

Fig. 1. Various means to populate the conduction band of GaAs with circularly-polarized light: a) one-photon excitation of unstrained GaAs, b) one-photon excitation of strained GaAs, and c) two-photon excitation of unstrained GaAs with photons having energy equal to half that of the bandgap. The circled values in (a) indicate relative transition probabilities for unstrained GaAs. The maximum theoretical polarization is 50% from unstrained bulk GaAs via one-photon absorption, 100% from strained GaAs via one-photon excitation, and −100% from unstrained GaAs via two-photon excitation, at least in the simple selection-rule picture of Ref. [12].