Development of spin-polarized transmission electron microscope

M. Kuwahara, Y. Takeda, K. Saitoh, T. Ujihara, H. Asano, T. Nakanishi, N. Tanaka

1 EcoTopia Science Institute, Nagoya University, Nagoya, Aichi 464-8603, Japan
2 Graduate school of engineering, Nagoya University, Nagoya, Aichi 464-8603, Japan
3 Graduate school of science, Nagoya University, Nagoya, Aichi 464-8602, Japan

E-mail: kuwahara@esi.nagoya-u.ac.jp

Abstract. In order to study spin related phenomena in nano-size materials, spin-polarized electron source (PES) has been employed for the incident beam in transmission electron microscope (TEM). The PES has been designed and constructed with optimizing for spin-polarized TEM. The illuminating system of TEM is also designed to focus the spin-polarized electron beam emitted from a semiconductor photocathode with a negative electron affinity (NEA) surface. The beam energy is set to below 40 keV which is lower energy type as a TEM, because the spin interaction with condensed matters is very small corresponding with a Coulomb interaction. The polarized electron gun has realized in an extra high vacuum (XHV) condition and high field gradient of 4 MV/m on a surface of photocathode. Furthermore, it demonstrated that 40-keV polarized electron beam was operated with a sub-milli second pulse mode by using the backside excitation type photocathode. This high performance PES will make it possible to observe dynamically a magnetic field images with high contrast and high-speed temporal imaging in TEM.

1. Introduction

The polarized electron beam plays important roles in high-energy physics and materials sciences related to electron spin. The polarized electron source (PES) technology has been developed intensively for accelerators, and recently it was also applied to materials science instrumentation to observe the surface magnetic phenomena [1-4]. Furthermore the electron spin plays very important roles in not only fundamental physics but also some key technologies such as spintronics and quantum-information technology [5-7].

We have started the research and development of PES for applying to a transmission electron microscope (TEM) [8]. The spin-polarized electrons can be generated by using an optical orientation of the proper III-V type semiconductors and a extraction mechanism into vacuum using a negative electron affinity (NEA) surface, as explained by the following three-step model. First, the circularly polarized laser light illuminates the semiconductor and spin-selectively excites the valence band electrons to the conduction band. Secondly, these polarized electrons in the conduction band diffuse and some of them drift to the surface region. Finally, these polarized electrons are extracted into
vacuum through the NEA surface under applying a negative electrostatic field. To realize the polarized electron beam experiments in the future International Linear Collider (ILC) project, the PES technology has been developed intensively from a middle of 80’s [9]. As results, several beam performances were drastically improved, which include electron spin polarization (ESP), quantum efficiency (QE), a sub-nanosecond multi bunch structured beam and low beam emittance. The high ESP of 92% and high QE of 0.5% were already realized by a GaAs-GaAsP strained superlattice (SL) photocathode with the active layer with of 100 nm and the excitation wavelength of 780 nm [10, 11]. It was also demonstrated that the normalized thermal beam emittance can be minimized to as low as 0.15 π mm mrad.

This high performance PES will make it possible to dynamically observe magnetic structure images with high-resolution spatial imaging and high-speed temporal imaging in TEM. Then we have started the development of spin-polarized TEM (SPTEM). In this paper, we would like to share an overview of the PES and the pulse performance. We will also report about the construction of the new PES for TEM.

2. Overview of spin-polarized TEM

The overview of the SPTEM system is shown in figure 1. The SPTEM consists of a PES, illumination-lens system including spin manipulator, objective-lens system, projector-lens system and image-detection system. The polarized electron beam emitted from the PES first passes through a spin-rotator and a condenser-lens system in order to concentrate the beam on the specimen. Then the beam passes through the specimen. Some of the electrons pass all the way through, and others are scattered by some interactions that are the Coulomb interaction, spin-orbit interaction and spin-spin interaction. The modified beam then passes through the objective lens, the projector-lens and onto a fluorescent screen where the final image is observed. Hence the electron beam passes entirely through the specimen, the diffraction pattern gives the observed comprehensive view of the interior of the specimen including magnetic distribution. In the whole parts of vacuum system in the SPTEM excepting for the detection chamber, these vacuum conditions are maintained in ultra-high vacuum (UHV) condition to avoid a destruction of an NEA surface due to the residual gas. Especially, the illumination-lens system was newly developed in this project, which consists of a spin-rotator for transverse rotation (SRB), and a condenser lens (CL) system including in-plane spin-rotation lens magnet (SRL). A 90-degrees bending system was employed for the SRB to apply both an electric field and a magnetic field for rotation in any azimuth angle. The system also can realize to correct the over-rotated angle due to the anomalous magnetic moment of electron and the Thomas precession in the

![Figure 1. Schematic overview of SPTEM](image-url)
relativistic effect. The electron beam optics in the PES and the illumination lens system is shown in figure 2.

3. Polarized electron source

3.1. Polarized electron gun

The PES is required to produce high polarization and high brightness beam simultaneously. The 40-keV PES system was newly designed and built to install the transmission type photocathode instead of ordinal LaB$_6$ electron gun. The PES consisted of three components in vacuum chambers; 40-kV DC-gun, a differentially pumping chamber equipping a collector lens and a photocathode preparation chamber. The photocathode preparation chamber connected to the 40-keV gun is used for activation of photocathode surface to create NEA surface. In order to maintain the NEA surface for long time, two kinds of advanced techniques; the ultrahigh vacuum and the dark current (between the high field gradient gun-electrodes) reduction are absolutely required. The PES apparatus is designed to satisfy the vacuum condition of UHV. Especially the vacuum around the photocathode is kept in XHV ($\sim$several$\times$10$^{-10}$ Pa) condition to protect the NEA surface from (1) adsorption of the residual gas and (2) positive ion back-bombardment caused by beam collisions with gas molecules. Each chamber are evacuated by ion pumps and non-evaporable getter (NEG) pumps. The components of 40-kV gun are shown in Figure 3. The cathode and anode are made of molybdenum and titanium respectively, since it showed the best performance in our test experiment, where the dark current was suppressed below 1nA under applied field gradient of 134 MV/m on the molybdenum cathode surface [12]. As the final metal surface polishing process, both procedures of electrochemical-etching and fine buffering were simultaneously employed to obtain a mirror-flat surface [13].

An activation process of NEA surface is carried out in a preparation chamber. A photocathode sample is attached to the molybdenum cathode-pack stage, and heated to remove the surface arsenic layer to obtain clean surface by a PBN (pyrolytic boron nitride) heater. Cesium is evaporated from a commercially available dispenser which is placed in front of the photocathode by using a linear manipulator during the cesiation process. Oxygen is introduced into the preparation system from a pure oxygen bottle through a variable leak valve. Circular light is led to the gun by an optical fiber and finally focused on to the photocathode surface from the backside through the focusing lens. A high field-gradient for acceleration is necessary to suppress the space-charge-dispersion effect to keep the high beam brightness. This gun could supply the high gradient of 4 MV/m to the photocathode surface, since a gap separation of electrodes and a supplied voltage are 8.0 mm and 40-kV, respectively. In this condition the dark current in the vacuum still does not exceed a level of 0.1nA. The PES, after the 40-
keV gun constructed and tested, was connected to a beam diagnosis system. The figure 4 shows the temporal current profile of polarized electron beam emitted from the 40-keV PES and a phosphorous screen image of the spatial profile using a transmission-type photocathode. It was demonstrated that the PES could emit polarized electron beam using a transmission-type photocathode with the new laser focusing system.

Figure 3. The photographs of the electron gun including an optical system and collector lens.

Figure 4. The left figure shows temporal current profile of the polarized electron beam extracted from the transmission-type photocathode under operation beam energy of 40keV. The right figure shows a phosphorous screen image of spatial beam profile

3.2. Photocathode

Requirements for the photocathode are (1) high ESP and (2) high QE. The latter is assured by a clean GaAs surface layer with high NEA. Both were already satisfied by a GaAs-GaAsP strained SL layer photocathode. However the previous photocathode is not compatible with the laser backward illumination, because GaAs was used as a substrate. Therefore, the micro-structure of the reflection type photocathode is modified to the transmission type. It was fabricated by MOVPE in Nagoya-U. The active layer is composed of 12 pairs of GaAs-GaAsP strained superlattice (total thickness; 96 nm) with Zn-dopant concentration of $1.5 \times 10^{18}$ cm$^{-3}$. The active-layer is grown on a 1-µm GaAsP buffer-layer which is deposited on a 500-nm GaAs inter-layer. These micro-film-structures are grown on a GaP wafer which works as the transparent substrate. The active-layer is coated with a highly p-doped GaAs surface-layer with a Zn concentration of $6 \times 10^{19}$ cm$^{-3}$. The GaAs inter-layer plays an important role to achieve the highest ESP of ~90% by controlling the strain property of the GaAsP buffer-layer [14]. In the other 20-kV DC-gun, the photocathode could produce a polarized electron beam of the reduced brightness of $1 \times 10^7$ A m$^{-2}$sr$^{-1}$V$^{-1}$. It is noted that the brightness was dramatically improved, that is 1000 times larger than that of conventional PES [15]. The pulse performance of the
PES in creation of a sub-nanosecond bunch-beam was also realized by using a high p-doped surface that can relax a surface charge limit phenomena [11].

3.3. Laser illumination system

Figure 5 shows a schematic of the laser optical system. To make a compact optical system, a cw light emitted from a semiconductor laser diode, which has linear polarization and wavelength of 785 nm, is transferred to the PES through a polarization-maintaining fiber. The linearly polarized light injected from the fiber is converted to circularly polarized light by a quarter-wave plate that can switch the light helicity from +1 to -1. The circularly polarized light is then guided and focused on an active SL layer of the photocathode by an aspheric lens. The position of the lens is adjusted by monitoring the reflected laser spot size by projecting a magnified focus image on a charge coupled device (CCD) camera. Using the transmission type photocathode, the laser spot size could be minimized to a radius of 0.65 µm in half width at half maximum, which is the diffraction-limit of 785 nm wavelength due to the NA of the focusing lens.

4. Spin manipulation

4.1. Spin rotation in longitudinal axis

A spin-rotator is necessary to rotate a spin direction of polarized electron beam. The design of the SPTEM system also requires a 90 degrees deflection of electron beam because of the angle between a beam axis of the electron gun and that of lens barrels in the image-projection optics. Then the instrument has to realize both spin-rotation and beam deflection. An arbitrary angle rotation of the polarization is achieved by the combination of crossed electric and magnetic fields whose proportion defines the polarization angle. The angle between the beam direction and the spin direction can be controlled to any polar angles. The spin can be rotated in transverse plane azimuthally by a field of SRL magnet in the series of CL magnets. The spin rotation apparatus was designed carefully to maintain both of UHV and high brightness conditions. An electric field is created by a sectored spherical condenser with the curvature radius of 100 mm. A transverse magnetic field is generated by a magnetic pole pair with the gap separation of 30 mm. In order to avoid a degradation of UHV, magnet coils to provide the magnetic field is located outside of the vacuum chamber, and the magnetic field is lead through the magnetic poles into the vacuum.

4.2. CL magnets and spin rotation in-plane

A CL system essentially exists for collimating and concentrating the beam to a specimen. In this new system, the CL system also plays a role of spin-rotator for in-plane rotation. The system consists of four lens magnets, which are SCL1, SRL, SCL2 and SCL3. In beam optics, the electron beam extracting from the 90-degrees bending-type spin-rotator is focused in the center of SRL by the SCL1 solenoid magnet, since the beam trajectory does not have to be disarranged in the SCL2 regardless of
any magnitude of the magnetic field in the SRL. The SCL2 and SCL3 play the role of an ordinary CL in TEM. Therefore the newly designed CL system can realize an in-plane spin-rotation with concentrating the beam to a specimen.

5. Summary

The 40keV PES for SPTEM has been constructed and performed with a transmission-type photocathode. The 90-degrees bending spin-rotator and the illuminating-lens system including a CL system within in-plane spin-rotation have been made. The photocathode type PES has a high potential to observe a dynamics mechanics in a nano-structural matters with spin information [13, 17].

Acknowledgments

This research is supported by Grants-in-Aid for Scientific Research (S) (No. 51996964) and Grants-in-Aid for Young Scientists (B) (No. 22740157) from the MEXT of Japan, a Research fund of KEK for cooperative development of polarized electron sources and Technology Development Program for Advanced Measurement and Analysis of Japanese Science and Technology Agency (Group Leader; T. Nakanishi).

References

[1] Prescott CY, Atwood WB, Cottrell RLA, Destaebler H, Garwin EL, Gonidec A, Miller RH, Rochester LS, Sata T, Sherden DJ, Sinclair CK, Stein S, Taylor RE, Clendenin JE, Hughes VW, Sasao N, Schuler KP, Borghini MG, Lubelsmeyer K, Jentschke W 1978 Phys. Lett. B 77 347
[2] SLD Collaboration 1995 Phys. Rev. Lett. 74 2880
[3] Zdyb R and Bauer E 2002 Phys.Rev. Lett. 88 166403
[4] Kohashi T and Koike K 2001 Jpn. J. Appl. Phys. 40 1264
[5] N. Gisin, G. Ribordy, W. Tittel, and H. Zbinden 2002 Rev. Mod. Phys. 74, 145
[6] Loss D and DiVincenzo DP, 1998 Phys. Rev. A 57 120
[7] Kuwahara M, Kutsuwa T, Ono K, Kosaka H 2001 Appl. Phys. Lett. 96 163107
[8] Tanaka N, Nakanishi T, Takeda Y, Asano H, Saitoh K, Ujihara T and Kuwahara M 2010 Proceedings of 17th International Microscopy Congress (Rio de Janeiro) I20.2
[9] ILC collaboration 2007, ILC Global Design Effort and World Wide Study
[10] Nishitani T, Nakanishi T, Yamamoto M, Okumi S, Furuta F, Miyamoto M, Kuwahara M, Yamamoto N, Niiwa K, Watanabe O, Takeda Y, Kobayakawa H, Takashima Y, Horinaka H, Matsuyma T, Togawa K, Saka T, Tawada M, Omori T, Kurihara Y, Yoshioka M, Kato K and Baba T 2005 J. Appl. Phys. 97 094907
[11] Togawa K, Nakanishi T, Baba T, Furuta F, Horinaka H, Kurihara Y, Matsumoto M, Matsuyma T, Nishitani T, Okumi S, Omori T, Suzuki C, Takeuchi K, Wada K, Yamamoto M and Yoshioka M 2000 Nucl. Instrum. Methods Phys. Res. A 455 118
[12] Furuta F, Nakanishi T, Okumi S, Gotou T, Yamamoto M, Miyamoto M, Kuwahara M, Yamamoto M, Niiwa K, Yasui K, Matsumoto M, Yoshioka M and Togawa K 2005 Nucl. Instr. and Meth. A 538 33
[13] Kato S, Aono M, Sato K and Baba Y 1990 J. Vac. Sci. Technol A 8 2860 (1990)
[14] Jin X, Yamamoto N, Nakagawa Y, Mano A, Kato T, Taniokum U, Ujihara T, Takeda Y, Okumi S, Yamamoto M, Nakanishi T, Saka T, Horinaka H, Kato T, Yasue T and Koshikawa T 2008 Appl. Phys. Exp. 1 045002.
[15] Yamamoto N, Nakanishi T, Mano A, Nakagawa Y, Okumi S, Yamamoto M, Konomi T, Jin X, Ujihara T, Takeda Y, Ohshima T, Saka T, Kato T, Horinaka H, Yasue T, Koshikawa T, Kuwahara M 2008 J. Appl. Phys. 103 064905
[16] King WE, Campbell GH, Frank A, Reed B, Schmerge JF, Siwick BJ, Stuart BC, Weber PM 2005 J. Appl. Phys. 97 111101
[17] Flannigan DJ, Park ST, Zewail AH 2010 Nano Lett. 10 4767