Chapter 2

Apparatus *

The ultrafast electron crystallography (UEC) apparatus in our lab is built for the study of thin crystals and surfaces, based on experience of three generations of gas phase ultrafast electron diffraction (UED). It includes a homemade ultrahigh vacuum (UHV) chamber system, which sits on a supporting frame assembly, and a commercial femtosecond (fs) laser system with optical interfaces (see figure 2.1 and figure 2.2). The fs laser system provides both the laser pulses for the generation of ultrashort electron pulses, and those initiating the reactions to be studied. The UHV chamber system is where the samples are mounted, stored and prepared, and where the experiments take place. It connects to the electron gun and imaging system, where the electron pulses are generated, focused and recorded after they are diffracted by the samples. The experiments are controlled and automated by personal computers with Labview programs developed in UEC laboratory.

2.1 UHV chamber system

Since electrons have large scattering cross sections, high vacuum is needed for electrons to travel through without being scattered by the gas molecules. A typical modern transmission electron microscopy (TEM) has a pressure inside the column of $P \approx 10^{-7}$ torr. However, ultrahigh vacuum (UHV), which refers to pressure P of 10^{-9} to 10^{-12} torr, is required to carry out the surface experiments. It reduces surface

*This apparatus was constructed in collaboration with Dr. Chong-Yu Ruan, Dr. Franco Vigliotti, and Dr. Vladimir A. Lobastov, see reference [24].

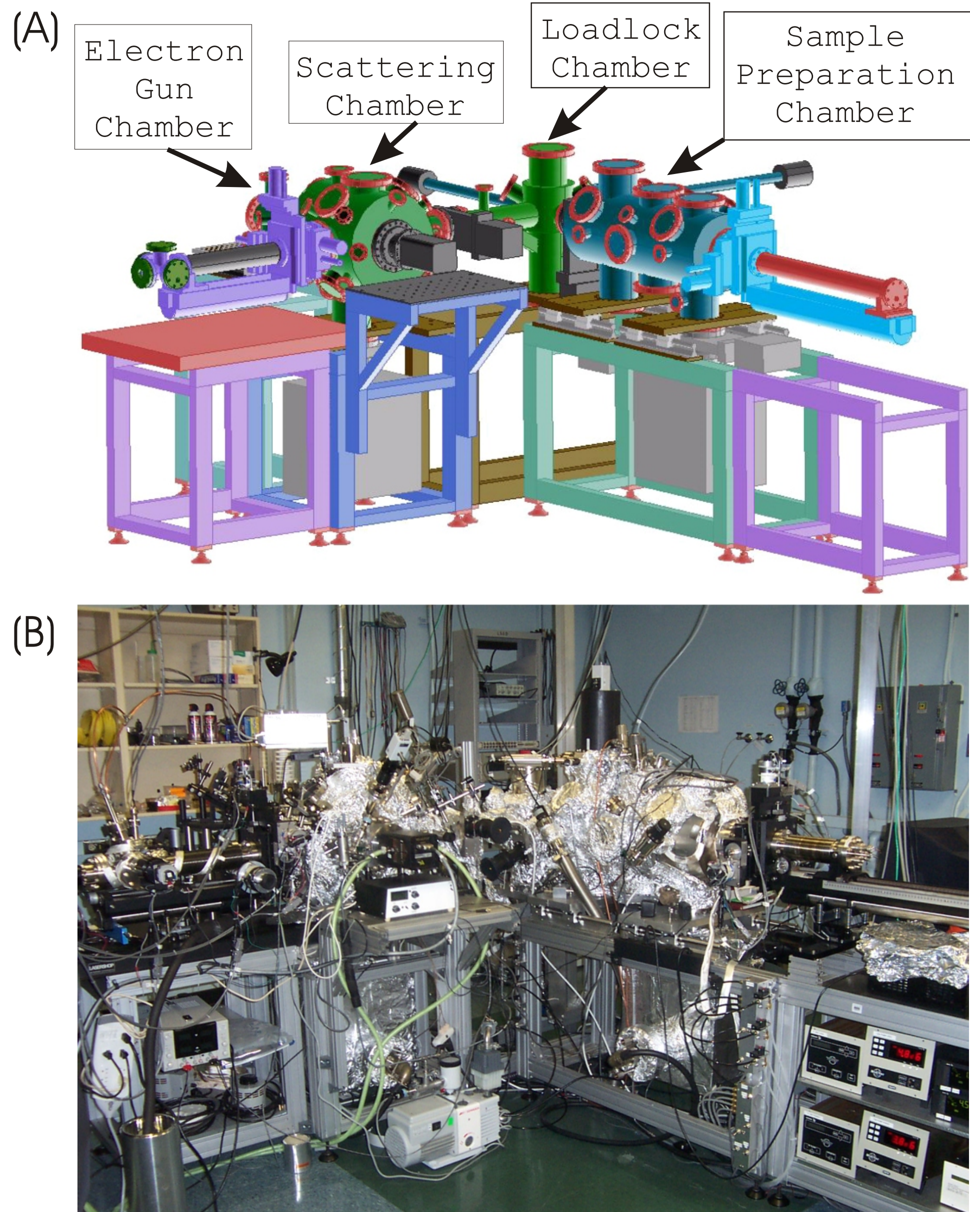

Figure 2.1: The chamber system with frame support: (A) schematic diagram; (B) photo of the same view in the lab.

contamination by reducing the number of molecules reaching the sample over a given time period.

The number of gas molecules hitting the surface per unit time and area is given by

$$\Phi = \frac{1}{4} n \bar{v} \quad ,$$

where $\bar{v}$ is the average velocity of the gas molecules determined by temperature, T, and the molecular weight, M,

$$\bar{v} = \sqrt{\frac{8RT}{\pi M}} \quad .$$

And n is the number of molecules per unit volume given by

$$n = \frac{P}{k_B T} \quad .$$

Giving the molar gas constant, R, and Boltzmann constant, k_B, the gas impingement flux, Φ, is calculated in the unit of $\mathrm{cm}^{-2}\mathrm{s}^{-1}$

$$\Phi = \frac{P}{\sqrt{mT}} \times 3.51 \times 10^{22} \quad ,$$

where P is in torr, m is in amu and T is in K. The number of adsorption sites, i.e., the number of atoms on the surface, is $\sim 10^{15}$ cm^{-2}. So at $P \approx 10^{-6}$ torr, it only takes a few seconds for a monolayer to form on the surface. Whereas at P below 10^{-9} torr, it takes hours or even days for a monolayer to form and effectively reduces the reaction of the sample surface with the gas molecules, thus allowing experiments for the sample surface to be performed.

The chamber system in the UEC lab consists of three integrated main UHV chambers (figure 2.2): the scattering chamber, the sample preparation chamber and the load lock chamber. Two manual gate valves (from MDC Vacuum Products, LLC, Hayward, CA) separate the load lock chamber from each of the scattering chamber and the sample preparation chamber. The scattering chamber is also connected to the electron gun chamber and the CCD (charge-coupled device) imaging system through differential pumping.

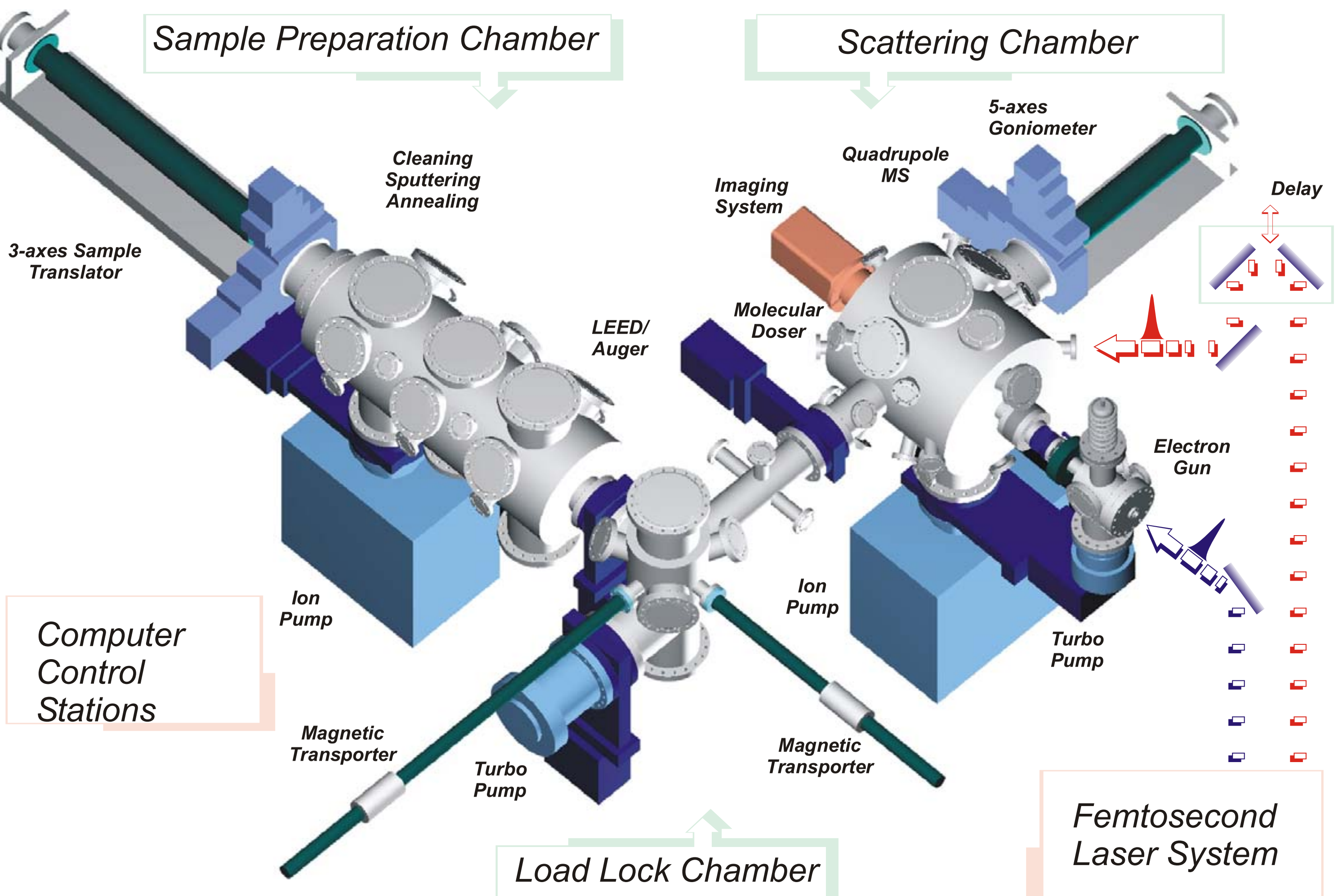

Figure 2.2: Schematic diagram of the experimental setup [24].

There are 3 pumping stages for the main chambers (figure 2.2). The system is rough pumped by a dry mechanical pump (QDP40 Drystar Pump from BOC Edwards, Inc., Wilmington, MA) through a turbomolecular pump (STP451 Seiko Seiki from BOC Edwards, Inc., Wilmington, MA), which is connected through an electropneumatic gate valve (from MDC Vacuum Products, LLC Hayward, CA) to the load lock chamber. This turbo pump provides intermediate pumping for the three main chambers, and can take the load lock chamber alone to low 10^{-10} torr. Two ion pumps connected to the main chambers through electropneumatic gate valves (from MDC Vacuum Products, LLC Hayward, CA) provide fine pumping for the sample preparation chamber and the scattering chamber, respectively. One ion pump (VacIon Plus 500 Diode Ion Pump with Titanium Sublimation from Varian, Inc., Palo Alto, CA) is underneath the scattering chamber. The other ion pump (VacIon Plus 500 Starcell Ion Pump with Titanium Sublimation from Varian, Inc., Palo Alto, CA) underneath the sample preparation chamber has better performance for large amounts of gas. The electron gun chamber and the CCD system are pumped separately and connect to the scattering chamber through differential pumping. The electron gun is pumped by a small turbo pump (Model EXT255H from BOC Edwards, Inc., Wilmington, MA), which is backed by a dry mechanical pump (Model XDS10 from BOC Edwards, Inc., Wilmington, MA). The image intensifier is pumped separately by a mechanical pump (RV5 Rotary Vane Pump from BOC Edwards, Inc., Wilmington, MA). The vacuum is measured by a multigauge system (from Varian, Inc., Palo Alto, CA), which use thermocouple (TC) type gauges for pressure from atmosphere (760 torr) to 10^{-3} torr, and ion gauges for pressure below 10^{-3} torr.

The load lock chamber is used to load the samples from the ambient pressure and store in UHV, and to retrieve the samples from the system. So the sample preparation chamber and the scattering chamber can keep under UHV at most time. It houses a sample cassette (from Transfer Engineering and Manufacturing, Inc., Fremont, CA, see figure 2.3), which has shelves to hold up to 5 sample holders and can be moved up and down by a Z-slide translator underneath. The load lock chamber is relatively small, so the pump down time is fast, usually only takes 4–5 hours to pump from

atmosphere pressure (7.6×10^2 torr) to 10^{-9} torr. With the multiholder cassette, it also allows for minimum exposure of the samples to air before the experiment, and changing samples while the experiment is going on. To mount the sample holders, the load lock chamber is opened by removing the 8" flange on the top and the sample holders are put on the shelves by hand.

The sample preparation chamber is where the sample surface can be prepared *in situ* under UHV and characterized (figures 2.2 and 2.3). It has three sections, and is augmented with LEED/Auger Spectrometer (from SPECS Scientific Instruments, Inc., Sarasota, FL) and sputter cleaning (ion source package and gas inlet system from SPECS Scientific Instruments, Inc., Sarasota, FL) in two sections, with the third one (in the center) open for additional surface techniques. The sample preparation chamber is home to a 3-axis sample translator (from Transfer Engineering and Manufacturing, Inc., Fremont, CA), which allows x, y and z translation of the sample with high precision (10 μm). The sample can be moved to different sections, and the position be adjusted for different surface experiments. The sample can also be heated up to 1000℃ through the feed through wires on the sample translator.

The scattering chamber is where the ultrafast diffraction experiments take place (figure 2.2). It connects to the electron gun chamber and the CCD assembly through differential pumping. It encases a 5-axis goniometer (from Transfer Engineering and Manufacturing, Inc., Fremont, CA) on which the sample is mounted, giving the sample high precision in x, y and z translation and in rotation of θ and ϕ (10 μm in translation and 0.01° in rotation), as shown in figure 2.4. The goniometer head (figure 2.4(B)) has two sections, one is the stage for the sample holder and the other has three needles for alignment purposes. For the goniometer, x-axis is along the horizontal direction to the load lock chamber, y-axis is pointing up along the vertical direction, and z-axis is horizontal along the direction from electron gun chamber to the CCD camera. The rotation in the sample plane around the normal of the sample surface is represented by ϕ. And the out-of-plane rotation around x-axis is represented by θ. Note that in the figure, the sample holder is in the horizontal position where the y-axis and the axis for ϕ rotation (the normal of the sample surface) is

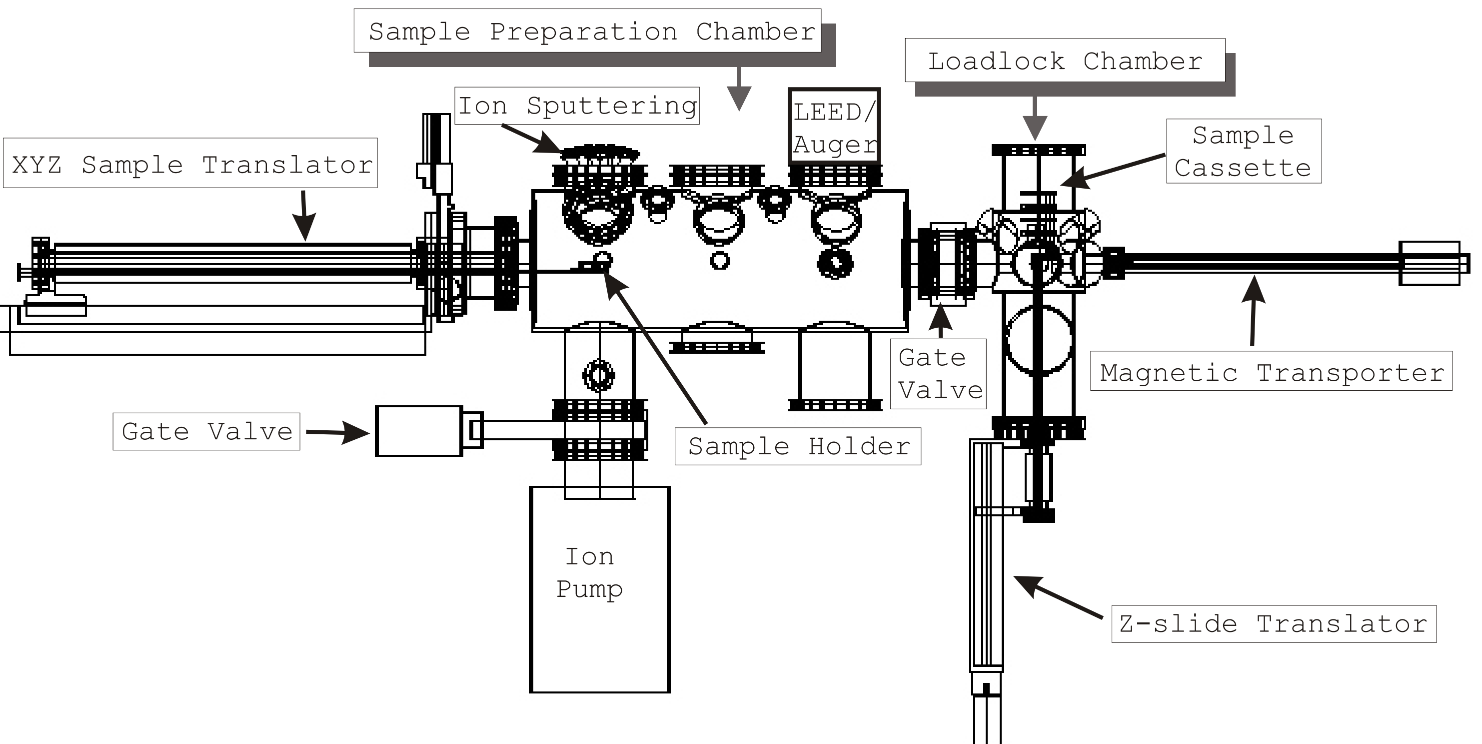

Figure 2.3: Schematic side view of the sample preparation chamber and the load-lock chamber.

the same. In experiments, depending on θ, these two axes usually are not the same. The sample holder can be heated to over 200℃ through the wires on the goniometer head, and cooled down to −180℃ through liquid nitrogen (N_2) running in the tubing underneath the stage for the sample holder (not shown in the figure). The temperature of the sample can be measured by the platinum resistance temperature detector (RTD) buried in the stage for the sample holder, or more precisely, using thermocouples directly connected to the sample.

2.2 Electron gun system

The ultrafast electron pulses used in the experiments of UEC are generated in the electron gun chamber (see figure 2.2 and figure 2.5) by the fs laser pulses ($\lambda = 266$ nm) through photoelectric effect. The photo cathode is made of a thin silver film (∼45 nm) deposited on a sapphire window (∼16 mm OD from Rolyn Optics Co. Covina, CA), and is confined in a close-fitting grove on the stainless steel cathode set by conductive silver paste (PELCO Colloidal Silver Paste from Ted Pella, Inc., Redding, CA), which also provides electric contact. The cathode set allows the fs laser illuminate the photocathode from the back through the sapphire window. The anode is made of stainless steel with a gold mesh (Gilder Fine Bar Grids from Ted Pella, Inc., Redding, CA) covered hole at the cathode side to provide a uniform electric field to extract photo-electrons. A μ-metal disk (from Magnetic Shield Corporation, Bensenville, IL) on the other side of the anode provides shielding from the electric magnetic field, with a hole of 150 μm diameter to let the electrons pass. The cathode set and the anode are supported and connected to the flange by macor rods, which also serve as electric insulators. The cathode set is connected to the high voltage (30 kV) through vacuum feedthrough, whereas the anode is grounded to the whole chamber system. By adjusting the length of the macor rods, the distance between the cathode and the anode can vary to give different extraction field strength. In our experiments, the field strength is 30kV/3mm = 10 kV/mm.

The electron gun chamber is kept in high vacuum with pressure at 10^{-7} torr to

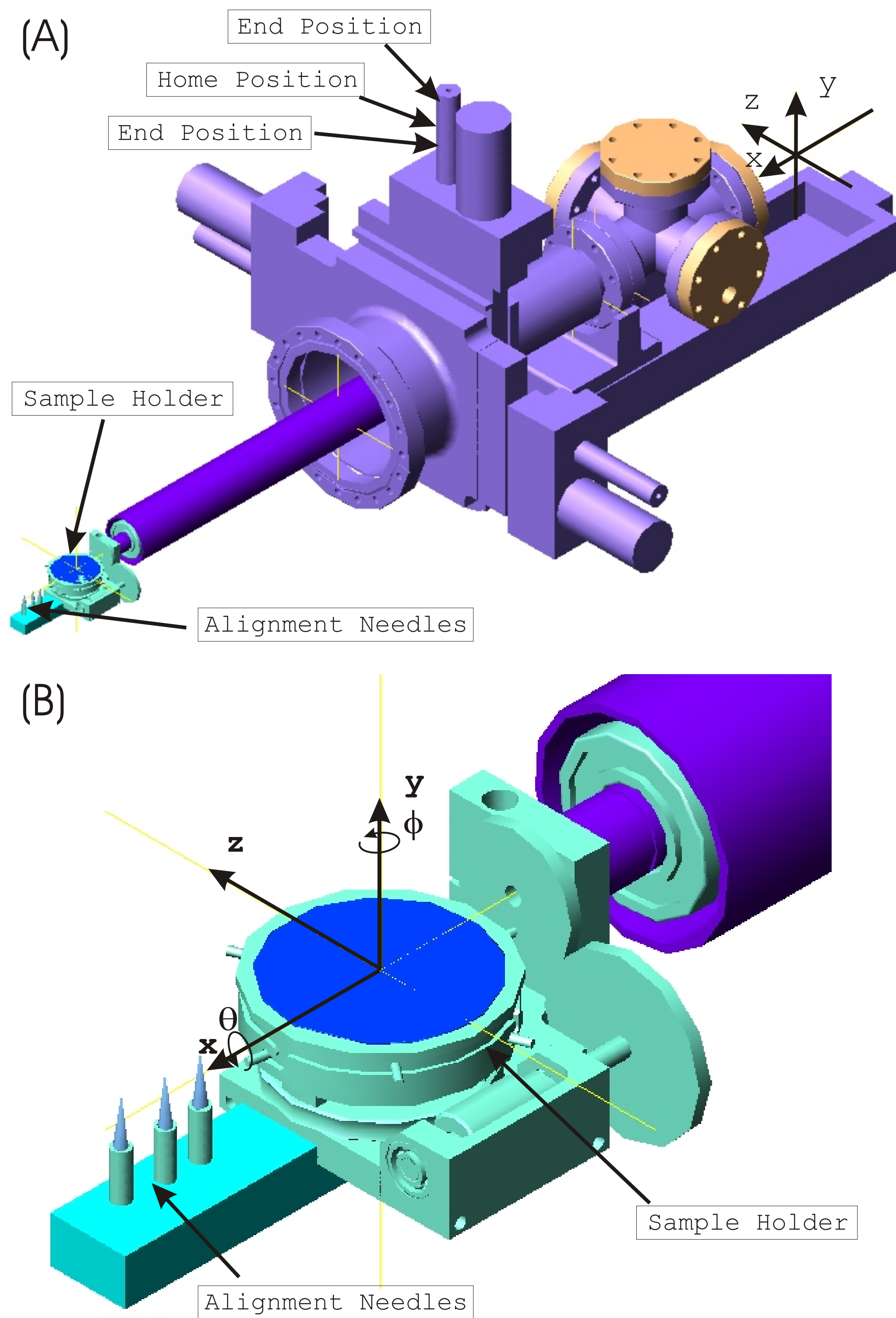

Figure 2.4: Schematic view of the goniometer: (A) the whole body; (B) an enlarged view of the goniometer head. The liquid nitrogen tubing is omitted for clarity.

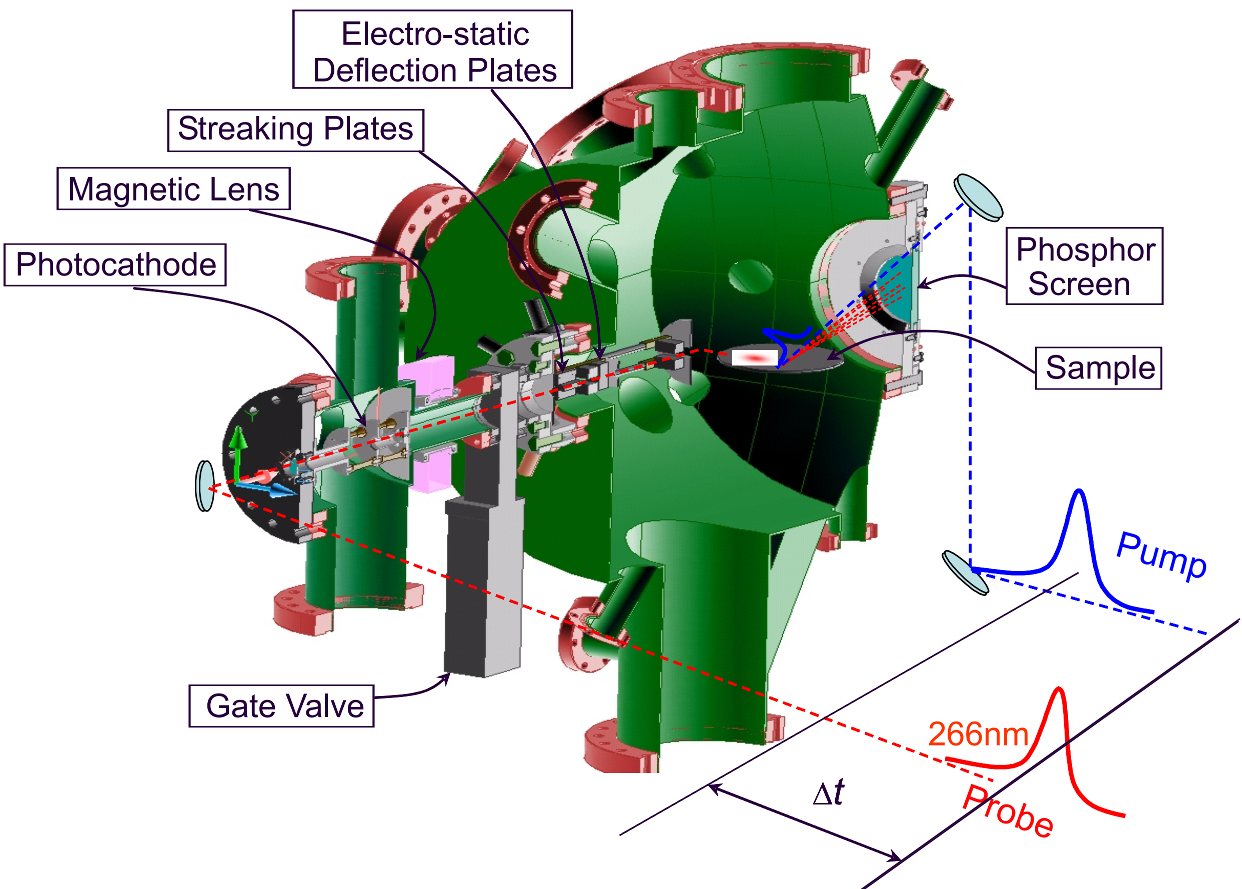

Figure 2.5: A cut through the electron gun chamber and the scattering chamber, showing the paths of the electron pulse and the excitation laser pulse onto the sample. Note that the sample is on the goniometer, which is omitted for clarity. Also the CCD imaging system is omitted; only the phosphor screen is shown.

let the electrons pass without scattered by gas molecules. The high vacuum also provides the insulation between the high voltage (30 kV) cathode and the anode, which is grounded to the rest of the chamber. The electron gun chamber is connected with the scattering chamber through an electropneumatic gate valve (from MDC Vacuum Products, LLC Hayward, CA). When the gate valve is open, a hole with diameter of 5 mm allows the passage of the electrons into the scattering chamber. In the mean time, it keeps the scattering chamber at UHV through differential pumping.

The electrons are focused using a homemade magnetic lens located right after the electron gun chamber. The magnetic lens is a homemade solenoid inside a magnetic coil enclosure (from Magnetic Shield Corporation, Bensenville, IL), and sits outside the vacuum tube where the electron pulses pass (see figure 2.5). It has a weak focusing power on the electrons, with the focusing length determined by the electric current. There are three sets of fine-threaded screws, with two screws in a set at each side of the solenoid, evenly placed around the vacuum tube on the enclosure. By turning the screws, the position and the angle of the magnetic lens can be finely tuned.

There are four sets of electro-static plates inside the scattering chamber. The first set is the streaking plates, used in the streaking experiment to characterize the electron pulses (see section 3.2.1). There are three sets of deflection plates after the streaking plates that can steer the electron beams by electric static force, one in the horizontal direction and two in the vertical direction.

In UEC experiments, the electron pulse is directed onto the sample and scattered to form the diffraction pattern in the far field on the phosphor screen. Figure 2.5 shows the case in reflection mode. In transmission or at low angles in reflection, only some of the electrons get diffracted, and many electrons still go through in the so-called main beam. A beam stopper, which is a grounded metal tube to catch the electron main beam, is put in front of the phosphor screen (not shown in figure 2.5). The beam stopper can be moved around in the plane of the screen through a UHV manipulator system (from MDC Vacuum Products, LLC Hayward, CA).

2.3 Imaging system

The electrons and the diffraction patterns are recorded by a homemade intensified CCD imaging system, as shown in figure 2.2 and figure 2.5. Figure 2.6 is a cut view of the CCD imaging system. It is contained in a homemade holder/adapter. And it consists of four basic components: 1) a phosphor screen (P47 deposited onto fiber-optic faceplate) to convert electrons into photons; 2) a fiber-optic taper (1.5x, from Incom, Inc., Charlton, MA) which optically reduces and transmits the image; 3) an image intensifier (from Hamamatsu Corporation, Hamamatsu City, Japan) to amplify the image signals; and 4) a CCD camera system (Princeton Instruments Medium X-ray Digital CCD Camera System from Roper Scientific, Inc., Trenton, NJ) for digital readout of the image. It is connected to the scattering chamber through a homemade adapter, which houses the imaging system and provides deferential pumping between the low vacuum (10^{-2} torr) required for the image intensifier and the UHV chamber.

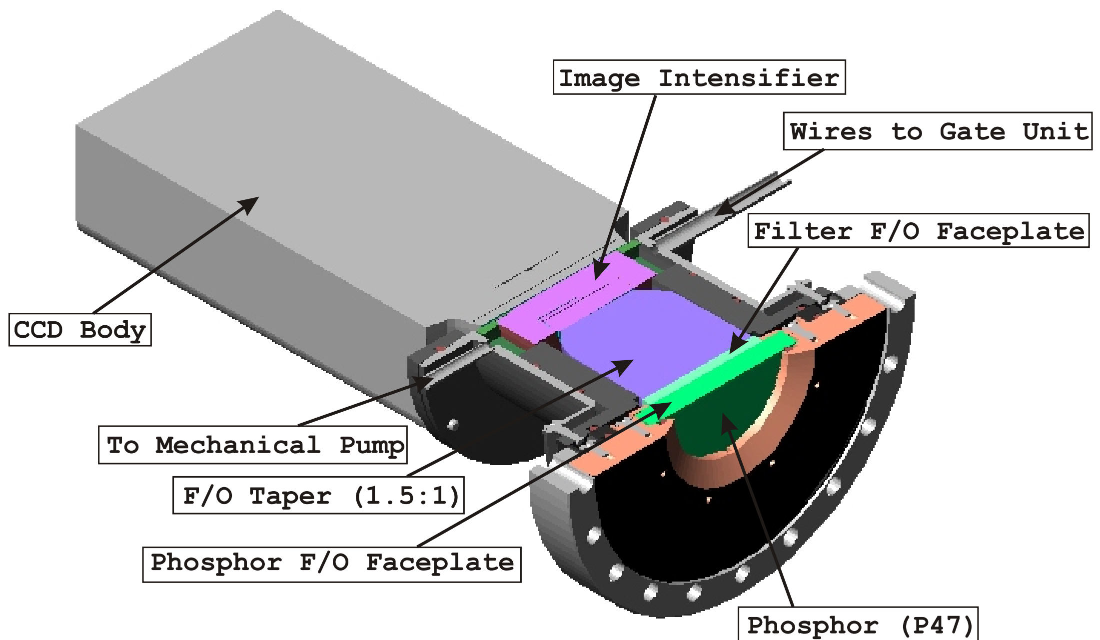

Figure 2.6: A cut through the CCD imaging system.

To minimize the noise generated by the image intensifier, the high voltage is

gated and synchronized to the 1 kHz laser pulse repetition. The image intensifier is controlled by a homemade controller, which provides gated high voltage signals subject to the voltage and the gate width settings.

The CCD camera is a 16-bit digital camera, and the intensity range is 0–65535. It is controlled through a CCD camera controller (from Roper Scientific, Inc., Trenton, NJ) by the software WinView (from Roper Scientific, Inc., Trenton, NJ), which can be programmed using Microsoft Visual Basic or LabView (National Instruments Corporation, Austin, TX). It has 1340 pixels in horizontal direction and 1300 pixels in vertical direction. The pixel size of the CCD camera is measured using a 1951 USAF glass slide resolution target (2" × 2", Edmund Optics, Inc., Barrington, NJ) to be 44.94 ± 0.25 μm in both horizontal and vertical directions.

2.4 Sample translation and manipulation

The samples, with sizes up to 2" in diameter, are placed on the sample holder using either metal (molydenum) clips or carbon double-sided tape (from SPI Supplies and Structure Probe, Inc., West Chester, PA). The sample holder can be moved under UHV between the load lock chamber and the scattering chamber, and between the load lock chamber and the preparation chamber. The sample holder (from Transfer Engineering and Manufacturing, Inc., Fremont, CA; see figure 2.4(B)) is made of molydenum or stainless steel, and has six pins distributed evenly at the side. Among the six pins, three pins are at the same lower level while the other three are at higher level.

The sample holder is put on the shelf of the sample cassette in the load lock chamber from the ambient pressure. Under vacuum, the sample holder can be picked up by the magnetic transporter (from Transfer Engineering and Manufacturing, Inc., Fremont, CA; see figure 2.2 and figure 2.3) and moved to the sample preparation chamber. To transfer the sample holder onto the XYZ sample translator in the preparation chamber, the sample holder is placed just above the sample stage of the translator. And then raise the stage until the sample holder sits on top of the stage.

The sample holder can also be transferred between the load lock chamber and the scattering chamber under vacuum, as illustrated in figure 2.2 and figure 2.7. The sample holder is picked up by the magnetic transporter in the load lock chamber and transferred between the sample cassette and the goniometor in the scattering chamber.

As shown in figure 2.4(B), the goniometer head has a sample holder cup, on which the sample holder can sit with its three lower pins fixed through spring-loading in the side groves of the cup. The head of the magnetic transporter is a flipped sample holder cup, with the side groves in the opposite direction (rotation) from the goniometer head cup, which holds the sample holder's three upper pins. To embed the sample holder in the sample holder cup, the sample holder is first put in place (pins inside the groves) by adjusting the position of the goniometer. Afterwards, rotation of the magnetic transporter in one direction will both release the sample holder with the upper pins from the transporter cup and fix it on the goniometer cup through the lower pins. Rotation in the other direction will have the opposite effect, i.e., release the sample holder from the goniometer head and pick it up by the magnetic transporter.

The sample translator in the preparation chamber and the goniometer in the scattering chamber are both driven by stepper motors and automated using Lab-View programs through electronic control boards. In experiments, the motions are controlled with personal computers through LabView programs.

The ranges in which the translators can travel are restricted by the space of the chambers and the flange sizes. To prevent the translators hit the walls of the chambers, two end positions are set for each translation axis (illustrated for the y-axis stage in figure 2.4), on the axles of the worm gears outside the chamber. The end positions are determined physically when the end plate hit the sensor and triggers the electronic switch. To provide a reference point, a home position is also set for each translation axis within the two end positions. The home position is set through an photoelectric switch where the diode laser is on and sends the beam across two slits. When the edge of the end plate goes through the slits and the beam is blocked, the home signal is given. This provides a very precise home position.

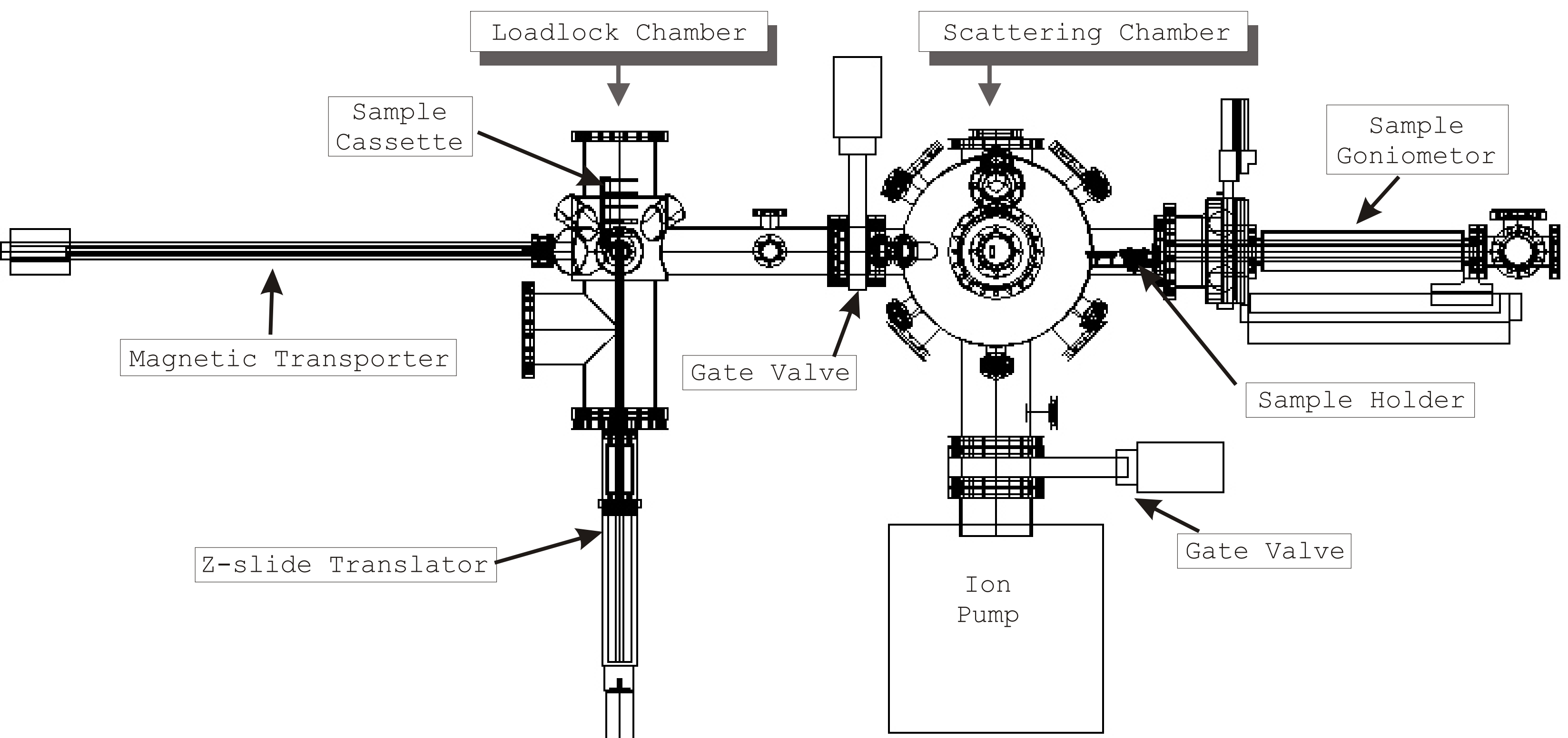

Figure 2.7: Schematic side view of the load lock chamber and the scattering chamber.

The rotations of the goneometer in the scattering chamber are produced through worm gears and worm wheels (figure 2.4(B)). Rotation of θ is realized by rotating the tube around the axle inside the tube, and the sample holder is driven to rotate around the axle as well. When the tube is fixed and the wheel #1 is rotated, wheel #2 is driven to rotate around its own axle, and through worm gears and another worm wheel under the sample holder stage, to rotate the sample holder stage with the sample holder around the surface normal and change the ϕ position of the samples. The precision is 0.01° for both the ϕ and θ rotation. The θ rotation is physically limited by restrictions on the wheel (not shown in figure 2.4(B)) to 0° to 180°. But without the counter balance block (not shown in figure 2.4(B)), the θ change should be confined within ±10° around 90°. Outside this region, the precision will be significantly worse. The ϕ rotation is, in principle, without limit. But, because of the presence of the tubes underneath the stage (not shown in figure 2.4(B)), the ϕ rotation is practically ristricted to the range of ∼180°. The θ and ϕ rotations are without reference point in space. $\theta = 90°$ is set usually to the position where the sample holder is horizontal.

2.5 Gas-handling system

To deliver gas adsorbates to the surface in a controlled and quantitative way, a gas-handling system with a microcapillary array beam doser [30] is designed and constructed as shown in figure 2.8. The system contains stainless steel tubes (OD 0.5”) connected by Ultra-Torr fitting (from Swagelok Company, Solon, OH) outside the chamber. A homemade stainless steel ballast serves as a gas reservoir of volume ∼1/4 L. The system can be rough pumped by a mechanical pump and fine pumped by a turbo pump. The inline valves are Nupro 8BG (from Swagelok Company, Solon, OH). A Nupro 4BRG (from Swagelok Company, Solon, OH) controls the flow from the gas bottles and provides some tunability. The gas-handling system connects to the beam doser through a gas feedthrough (from Kurt J. Lesker Company, Pittsburgh, PA) with Cajon VCR fitting inside the chamber.

Figure 2.8(A) shows the design for the microcapillary array beam doser. A stain-

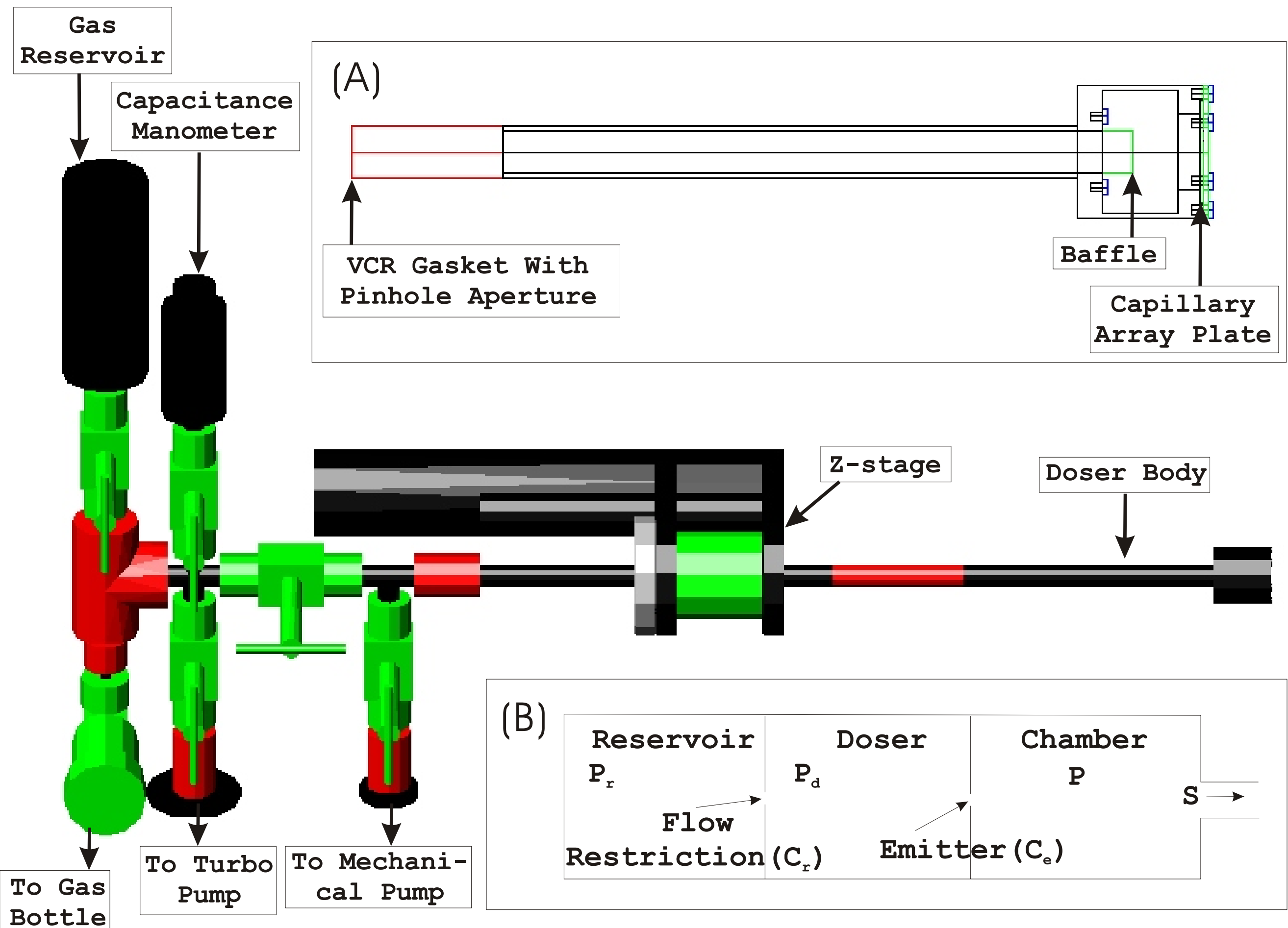

Figure 2.8: Schematic view of the gas-handling system. (A) Sketch of the microcapillary array beam doser. (B) Illustration of the gas flow, here P_r, P_d and P represent the pressure in the reservoir, doser and chamber, respectively; C_r and C_e are the conductance of the flow restriction and the emitter; S is the pumping speed for the vacuum chamber.

less steel pinhole aperture with 2 μm diameter (Drilled Swagelok VCR gasket from Lenox Laser, Glen Arm, MD) inside the VCR fitting serves as the flow restriction and provides convenient dosing fluxes. The flux of gas travels from the pinhole into the doser body, where it strikes a baffle made of copper shim stock, and the gas molecules' motion is randomized. The baffle is held into the place by the side bending toward the inside wall of the doser body and attached there by two screws. Collimation of the gas occurs in the high density capillary array (glass capillary array plate from Burle Technologies, Inc., Lancaster, PA) with perpendicular capillary size of 10 μm diameter and 800 μm long arranged in a hexagonal close packing arrangement. The capillary plate has a relatively high conductance for gases and is coated with NiCr to avoid charge accumulation. The plate is held in place by a stainless steel cap that, with a clearance of 0.040", confines the edge of the array in a close-fitting groove on the end of the doser body.

To estimate the gas flow quantitatively, the capillary array doser system can be simplified into three parts, reservoir, doser and chamber, as shown in figure 2.8(B). Assume the system in the molecular flow regime, the conductance of the flow restriction is

$$C_r = \frac{\frac{D_r^2}{4}\sqrt{\frac{\pi RT}{2m}}}{1 + \frac{3L_r}{4D_r}} \quad ,$$

where $D_r = 2$ μm is the diameter of the pinhole, L_r is the length of the pinhole and $L_r \simeq 76$ μm. For D_r and L_r in μm, temperature, T, in K, molecular mass, m, in amu, $C_r = (2.857 \times D_r^2\sqrt{\frac{T}{M}} \times 10^{-7})/(1 + \frac{3L_r}{4D_r})$ in the unit of l/s. And it gives $C_r = 4.438 \times 10^{-8}$ l/s for Nitrogen gas (N_2) at room temperature ($T = 300$ K). The same equation can be used also for calculating the conductance of the emitter, i.e., the capillary array plate, with D_r replaced by D_e, the diameter of the whole capillary area, $D_e = 18$ mm; and L_r by L_e, the pore length, i.e., the thickness of the plate $L_e = 800$ μm. And it gives $C_e = 293$ l/s for Nitrogen gas (N_2) at room temperature ($T = 300$ K).

The gas emitting flux, i.e., the number of gas molecules emitted from the doser in

every second, is given by

$$N_{tot} = \frac{C_e P_d}{k_B T} \quad . \tag{2.1}$$

In the experiment, a fraction of it FN_{tot} is intercepted by the sample, where the factor F is determined by the geometry arrangement of the doser emitter and the sample [31]. Let S_t denotes the sticking coefficient, the number of molecules absorbed on the sample surface is $S_t F N_{tot}$, and $(1 - S_t F)N_{tot}$ molecules get into the chamber. At equilibrium, the rate of the molecules entering the chamber is the same as that of the molecules leaving, i.e.,

$$(1 - S_t F)N_{tot} = \frac{SP}{k_B T} = (1 - S_t F)\frac{C_e P_d}{k_B T} \quad , \tag{2.2}$$

where S is the pumping speed and $S = 500$ l/s in the scattering chamber.

An enhancement factor E is defined as the ratio of the number of total gas molecules hitting the sample surface to the number of those from the background pressure, which is

$$N_{bg} = AP\sqrt{\frac{N_A}{2\pi m k_B T}} \quad ,$$

where A is the area of the sample surface. The definition of the enhancement factor gives $E = 1 + \frac{FN_{tot}}{N_{bg}}$. From equation (2.2), we can get

$$P = P_d \frac{(1 - S_t F)C_e}{S} \quad .$$

So E is given by

$$E = 1 + \frac{F}{1 - S_t F} \times \frac{S}{A} \times \sqrt{\frac{2\pi m}{RT}} \quad .$$

For S in l/s, A in cm^2, m in amu and T in K,

$$E = 1 + 0.275 \times \frac{F}{1 - S_t F} \times \frac{S}{A} \times \sqrt{\frac{m}{T}} \quad .$$

One geometry used in our experiments is the sample has a radius of approximately 5 mm and the distance between the capillary array plate to the sample is about 9 mm,

which yields $F = 0.6$ [31]. Assuming a sticking coefficient $S_t = 0.5$ at low temperature $T = 100$ K, we obtain an enhancement factor of ~ 65 for water vapor.

As shown by equation (2.1), the number of gas molecules emitted from the gas doser, and hence the number of those absorbed by the sample surface are determined by the pressure in the doser P_d. And P_d is given by the pressure in the reservoir P_r,

$$P_d = \frac{C_r P_r}{C_r + \frac{C_e S}{C_e + S}} \quad .$$

By tuning P_r, the dosing flux can be precisely controlled. In our setup, P_r is monitored accurately by a capacitance manometer (Baratron from MKS Instruments, Inc., Wilmington, MA). For the aforementioned case of water vapor at low temperatures, a flux of 5.1×10^{10} molecules per second is obtained for $P_r = 1$ torr, and the corresponding back pressure in the chamber is 1×10^{-10} torr.

2.6 Femtosecond laser system

A commercial amplified femtosecond laser system is used in UEC to provide two laser beams, one to generate ultrashort electron pulses and the other to excite the sample and initiate the dynamics.

The femtosecond laser pulses with wavelength centered at 800 nm and repetition rate of 80 MHz, are first generated by a mode-locked Ti:sapphire oscillator Spectra-Physics Tsunami (from Spectra-Physics Lasers, a Division of Newport Corporation, Mountain View, CA), which is pumped by a continuous-wave (cw) diode-pumped laser Spectra-Physics Millennia (from Spectra-Physics Lasers, a Division of Newport Corporation, Mountain View, CA). These laser pulses are then amplified in a 1 kHz Ti:sapphire amplifier Spectra-Physics Spitfire (from Spectra-Physics Lasers, a Division of Newport Corporation, Mountain View, CA), which is pumped by a diode-pumped, Q-switched Nd:YLF laser Spectra-Physics Evolution-30 (from Spectra-Physics Lasers, a Division of Newport Corporation, Mountain View, CA), to yield an output pulse energy of ∼2 mJ with pulse length ∼120 fs. The output fs laser

pulses are monitored by a single shot autocorrelator (Model SSA from Positive Light, Los Gatos, CA).

To generate the ultrashort electron pulses, the fs laser pulses with wavelength of 266 nm are used for photo-electric process in the electron gun chamber (see figure 2.5). As shown in the optics layout in figure 2.9, depending on the excitation wavelength wanted, part or all of the output from the Spitfire is frequency tripled by a tripler (Model TP-1A from U-Oplaz Technologies, Inc., Chatsworth, CA), with an efficiency $\sim$10% for 3 mJ input. Neutral density filters are used to deliver different powers to the photocathode, therefore to control the electron number and pulse length of the ultrashort electron pulses (see section 3.2.1).

The excitation laser pulses can be 800nm, 266nm or any wavelength from the Optical Parametric Amplifier (OPA-800CF-1 from Spectra-Physics Lasers, a Division of Newport Corporation, Mountain View, CA), which can provide laser wavelength from 1.1 to 3.0 μm with pulse-width $\sim$130 fs. The delay line is chosen to be in the path of the excitation through the translation stage (ATS150 series stage with 250 mm of travel from Aerotech, Inc., Pittsburgh, PA), because it is more precise to determine the relative timing of the optical pulses by the distance it travels, compared to determining the relative timing of the electron pulses. As shown in figure 2.9, it is designed so the excitation lasers with different wavelengths can use the same delay stage and go through the same optical path afterwards into the scattering chamber. When doing experiments under different wavelengths, the appropriate mirrors and lenses need to be added or changed to suit specific wavelength, while their positions only need to be adjusted slightly with the help of pinholes on the way. The optical path is experimentally determined to match the time deference between laser pulses of different wavelengths, in order to keep the time zero between the excitation laser and the electron pulses similar regardless of the wavelength of the excitation.

For example, to use 800nm excitation, 80% of the beam is reflected through BS2 (BS1 is removed from the beam path, see figure 2.9) from the output of the Spitfire before it enters the tripler, and sent into the chamber through the delay line. The rest 20% of the beam is frequency tripled and follow the beam path to the photocathode.

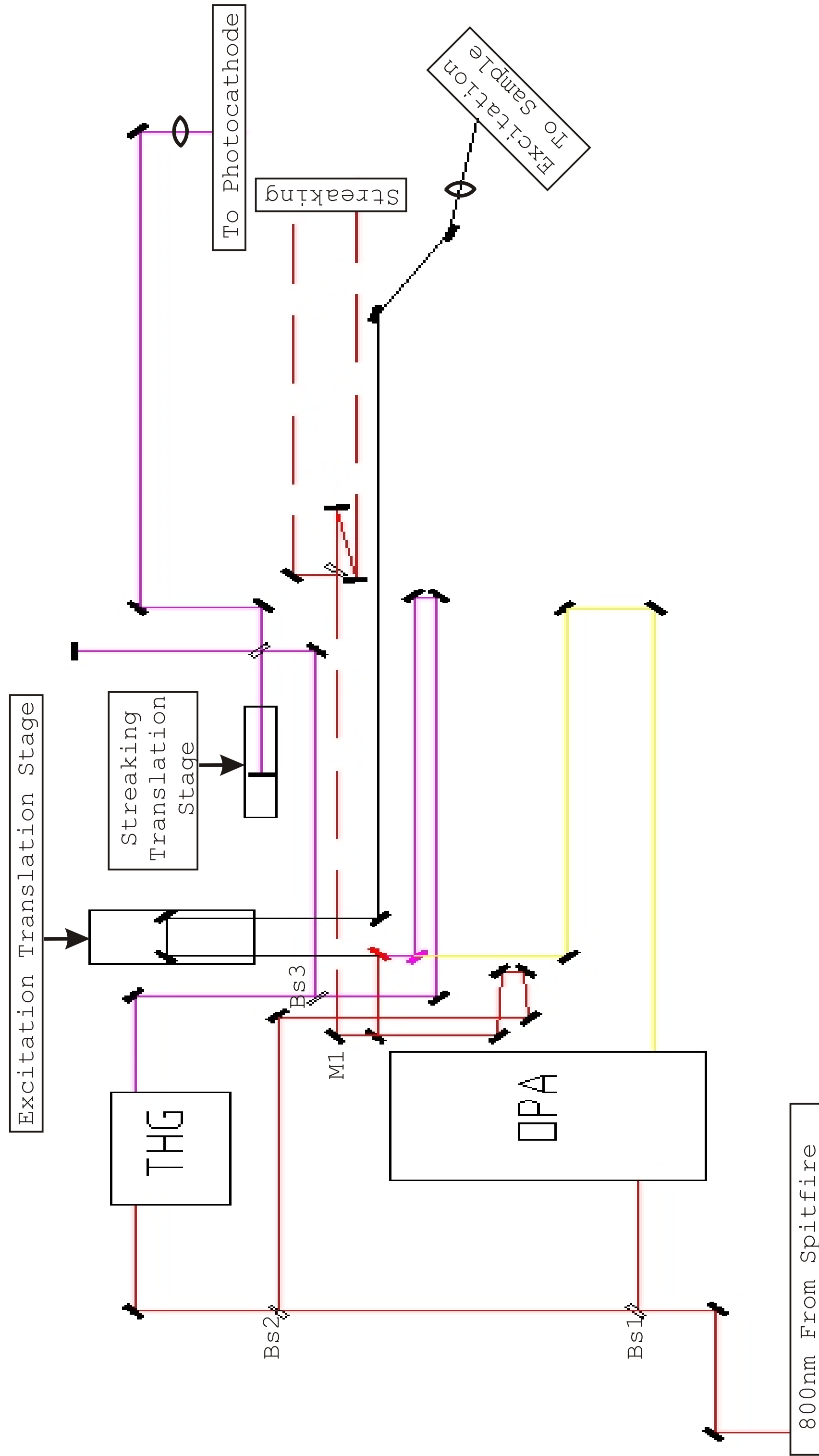

Figure 2.9: The optics layout.

For 266nm excitation, both BS1 and BS2 are removed and all the output of the Spitfire is frequency tripled. Then 50% of the beam is split through BS3 from the output of the tripler as excitation.

To minimize the damage caused by the fs excitation laser on the sample, an electronic shutter (IES electronic shutter with 1.6–25.4 mm clear aperture from Melles Griot, Inc., Rochester, NY) is used at the output of the Spitfire (not shown in figure 2.9). The shutter is synchronized with the CCD camera and controlled to open only when acquiring images.

The optics layout (figure 2.9) also contains the optical path for the streaking experiment, which characterizes the ultrashort electron pulses (see section 3.2.1). Instead of traveling into the scattering chamber as excitation, the 800nm laser is redirected by M1 to provide two pulses simultaneously to activate the two photoconductive switches in the streaking circuit (see section 3.2.1 and figure 3.4). The 266nm laser goes through a Michelson interferometer and gives two pulses in sequence with adjustable delay, τ, between them, to generate from the photocathode an identical pair of ultrashort electron pulses with delay τ. Additional to the streaking experiment, this configuration can also be used to do autocorrelation of the electron pulses [32].