1. Physics Motivation

Nuclear beta decay was important in motivating the Standard Model, and it continues to be of importance in searches for physics beyond the Standard Model. The simplest system for studying beta decay is the neutron, where there is a long history of increasingly accurate measurements of the parameters that characterize the decay. In particular, there have been many experiments in the past two decades that have measured the neutron's beta asymmetry A. This is the angular correlation between the neutron spin and the direction of emission of the electron in polarized neutron beta decay. A measurement of the beta asymmetry involves a determination of the forward-backward asymmetry of the beta with respect to the direction of the neutron polarization. In this proposal we detail our plans for developing and constructing neutron guide tubes and storage cells critical to a new experiment [2] at the Los Alamos Neutron Science Center (LANSCE) that will make the most accurate measurement of the neutron's beta asymmetry A. A measurement of A, when combined with results from the neutron lifetime, provides a determination of the fundamental vector and axial vector weak coupling constants G_V and G_A. This information can be used as a sensitive probe of possible physics beyond the Standard Model. Particular extensions that this measurement are sensitive to include right-handed currents (due to the presence of heavy right-handed exchange bosons), tensor and scalar interactions, and deviations of the Cabibbo-Kobayashi-Maskawa (CKM) matrix from unitarity (due to the presence of an additional generation of particles with very heavy neutrinos, for example).

Beta decay is described within the Standard Model via the emission of heavy $W^{+,-}$ bosons from quarks. The exchange bosons couple to the quarks and leptons with vector and axial-vector couplings. The couplings are such that the interaction is purely left-handed (V−A). To describe nuclear beta decay one uses the following Hamiltonian:

$$\mathrm{H} = \sum_{\mu} G_V \left[\bar{p}\gamma_\mu n\right]\left[\bar{e}\gamma_\mu (1+\gamma_5)\nu\right] - G_A\left[\bar{p}\gamma_\mu\gamma_5 n\right]\left[\bar{e}\gamma_\mu(1+\gamma_5)\nu\right] + \text{h.c.}$$

Here, p, n, e, and ν are spinors, and G_V and G_A are the weak vector and axial-vector coupling constants. This Hamiltonian is fully consistent with the Standard Model, so it includes no scalar or tensor terms, and it assumes only left-handed currents. Failure of this Hamiltonian to consistently predict all measured nuclear beta decay observables could be interpreted as evidence of physics beyond the Standard Model. By measuring the neutron's lifetime (from the Ft value) and the neutron's beta asymmetry A, one has enough information to independently determine G_V and G_A:

$$A = -2\,(-G_A G_V + G_A^2) / (G_V^2 + 3G_A^2)$$

$$Ft_n = \text{constant} / (G_V^2 + 3G_A^2)$$

The most recent accurate measurements of the neutron lifetime are in good agreement with each other [1]. The same cannot be said for measurements of the neutron's beta asymmetry A. Since 1985, there have been four accurate (combined statistical and systematic uncertainty of about 1% each) measurements of A [1]. Unfortunately, the agreement between these four A measurements is poor: the reduced χ^2 per degree of freedom for these four measurements is 4.6. The situation is illustrated in Figure 1, which shows the A measurements as bands in G_V–G_A space. In particular, the most recent and accurate measurement (PERKEO II) is in significant disagreement with the average of the prior three efforts. All of these experiments were carried out with cold neutron beams from nuclear reactors. Another such experiment will not resolve this discrepancy. What is needed is a new experiment with comparable or better statistical errors, but very different systematic errors. The LANSCE neutron beta asymmetry experiment fits that bill. It will be carried out with ultra-cold neutrons from a spallation neutron source. This will reduce the systematic errors that plague the in-beam reactor experiments, and with the expected UCN source flux, it should be possible to significantly improve the statistical error as well.

Two of the significant sources of systematic error in reactor based experiments stem from uncertainty in the neutron beam polarization and contributions from the neutron beam-associated background. A measurement of the beta asymmetry using ultra-cold neutrons (UCN) addresses both of these issues. UCN are neutrons whose wavelengths are sufficiently long (typically greater than 500 angstroms, which corresponds to 8 m/s neutron velocity) that they can undergo total external reflection at all angles from the surface for a variety of materials. These properties can be exploited to achieve significant reductions in the systematic errors associated with polarization and background.

Neutron beam polarizations of 100% can be achieved with UCN by passing the UCN through a strong magnetic field gradient. In practice, passage through a field of 6 Tesla is sufficient to polarize 100% of the UCN. This is due to the fact that the kinetic energy of the UCN is so small that UCN of one spin state cannot overcome the potential barrier due to the interaction of the neutron spin with the magnetic field. This polarization process can be contrasted with the technique used for the in-beam measurements, which use supermirror polarizers. The neutrons interact with the material (layers of Co, Ti, and Gd in a strong magnetic field), which produces a high (98%) polarization. The disadvantages are that the beam polarization is not completely uniform over the entire beam, the beam divergence is increased by a factor of two, and an intense gamma background is created. In all of the experiments done to date, the uncertainty in the neutron beam polarization has been one of the limiting systematics.

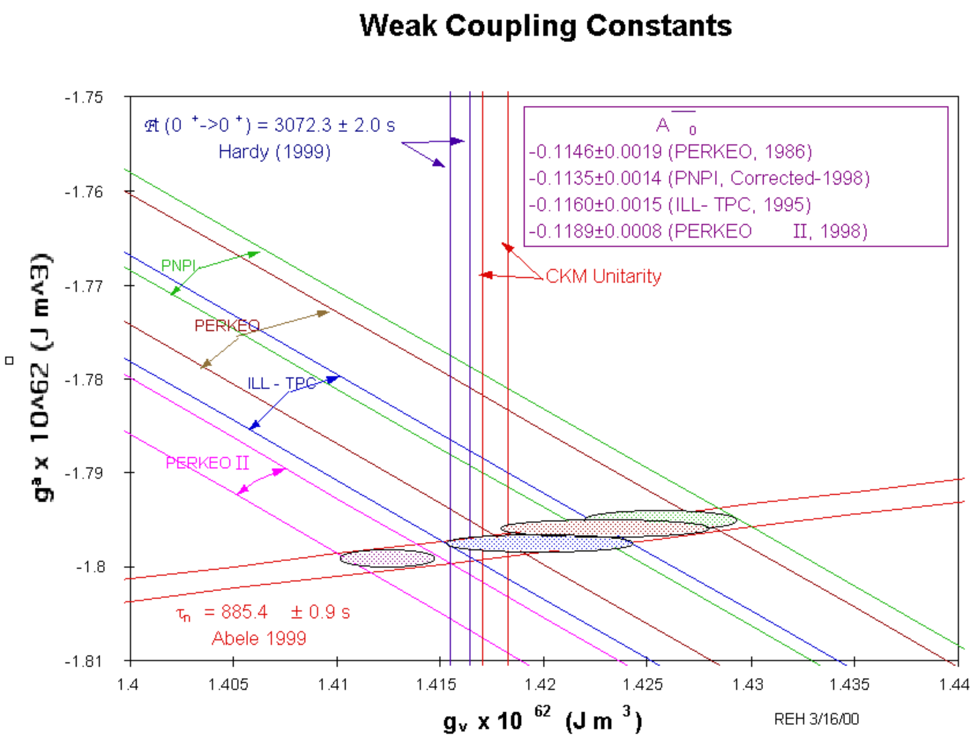

Figure 1. Results for the neutron decay beta asymmetry measurements as of 1999, and the values of G_V determined from $0^+ \rightarrow 0^+$ superallowed beta decay and from requiring unitarity of the CKM matrix.

Use of UCN also significantly reduces the backgrounds. Due to their reflection properties, the UCN can be piped out to a spectrometer located 10 m or more away from the primary neutron beam. Thus it is possible to provide 100% coverage with shielding and to have a background which is close to the intrinsic background of the spectrometer. In addition, since the LANSCE facility uses a short-pulse spallation source (2% duty cycle), one can suppress backgrounds significantly by using timing information so that data are excluded during the period just after the proton pulse strikes the spallation target. In contrast, in-beam experiments typically have detectors located relatively close to the neutron beam. Great care must be taken with issues such as neutron beam halo and dumping of the primary neutron beam. For these reasons, the beam-associated background is typically an important systematic concern in these experiments. For the UCN experiment the contribution should be very small.

With a reliable measurement of the neutron's beta asymmetry A, there are several ways to confront the Standard Model to search for new physics. Among the most discussed tests are the following:

Comparison to superallowed $0^+ \rightarrow 0^+$ nuclear beta decay: The value of G_V can be determined from $0^+ \rightarrow 0^+$ superallowed nuclear beta decay. To date, this decay rate has been measured in nine nuclei. Extraction of G_V involves applying corrections for finite nuclear size, Coulomb effects, and radiative corrections. The current best value from this type of analysis is shown by vertical bands in Figure 1. It is inconsistent with the values from the neutron and with the values obtained from unitarity of the CKM matrix (see below). One interpretation is that the quoted precision on G_V is underestimated due to the Coulomb effects. If the quoted precision is correct, then a possible interpretation is the existence of new physics like scalar or tensor interactions, which would mean that the Hamiltonian above does not provide a complete description.

Unitarity of Cabibbo-Kobayashi-Maskawa(CKM) mixing matrix: The CKM matrix describes the mixing among the quarks in the Standard Model. Assuming there are only three generations, the CKM matrix must be unitary. One can use the assumption of unitarity, along with measurements of various hadronic and meson decays to extract a value for the up-down quark mixing matrix element V_{ud}. An independent determinaton of V_{ud} comes from combining G_V and G_A as determined in neutron beta decay with the Fermi coupling constant G_F determined from the muon lifetime. As shown in Figure 1, for some values of the neutron beta asymmetry, one has significant deviation from unitarity. However, the scatter in the data on A prevents a definite conclusion at this time. A confirmed violation of the unitarity of the CKM matrix would call into question the assumption of only three generations. While three generations of light leptons have been precisely confirmed at LEP, this result does not rule out the possibility of heavy lepton generations. Thus, this test of the CKM matrix is complementary to information obtained at higher-energy machines.

Right-handed currents: The Standard Model assumes purely left-handed currents, so any observation of right-handed currents would be physics beyond the Standard Model. The conventional way to parameterize this possibility is to assume the existence of a right-handed gauge boson W_R in addition to the conventional left-handed gauge boson W_L. The relative unimportance of right-handed currents is explained by the presumed large mass of the right-handed boson. This formalism is parameterized by the mass ratio of the two bosons and the mixing angle between them. Definite predictions can be made for the neutron beta asymmetry in this model, so constraints on the mixing angle and mass ratio can be obtained by its measurement. If the LANSCE measurement achieves its goals, it will provide the tightest bounds on these parameters.

The LANSCE neutron beta asymmetry experiment will be the first such experiment performed with UCN. It will use a new solid deuterium UCN source which should provide the world's highest flux of UCN. The neutrons will be transported to a dedicated spectrometer where the decays of the neutrons in a holding cell will be observed. By exploiting the properties of the UCN it will be possible to minimize the most important systematic errors in the current generation of beta asymmetry experiments. The goal of the first generation of this experiment is to measure the neutron beta asymmetry to a relative precision of 0.2% (0.2% statistical, 0.04% systematic). The implications of a measurement of this accuracy on the determination of the weak coupling constants G_V and G_A is shown in Figure 2.

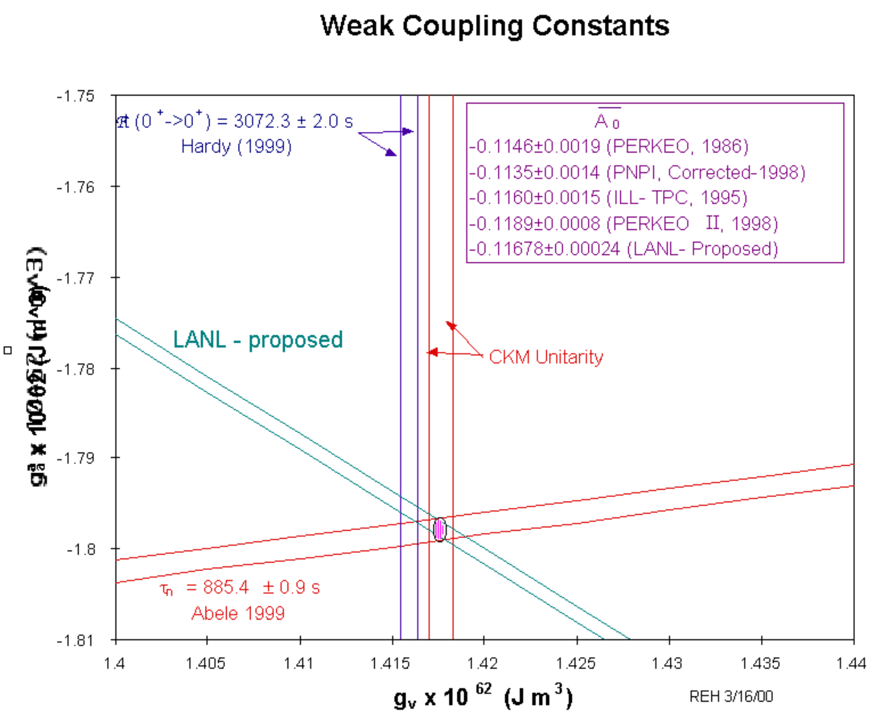

Figure 2. Projected sensitivity for G_V and G_A for the LANSCE beta asymmetry experiment and results for the most recent neutron lifetime experiments as of 1999 showing the sensitivity of this measurement. The central value for the new measurement has been arbitrarily set to coincide with the value of G_V determined by requiring unitarity of the CKM matrix.

It is clear that one of the most important features of the new LANSCE experiment is UCN and their special properties. One can generate highly uniform 100% polarized beams of UCN. To take full advantage of this, one must be able to transport and store these neutrons in a holding cell with minimal polarization losses. Since the experiment appears to be limited mainly by statistics, it is also essential to transport the neutrons with minimal loss. The development and practical implementation of appropriate coatings to achieve these goals is essential to the success of the experiment. Our group at Virginia Tech has been given responsibility for UCN transport for the LANSCE experiment (see supplementary documentation letter). Since the transport of the polarized neutron beams places the most stringent demands on the guide tube coating properties, we have begun a program to develop appropriate coatings at Virginia Tech. This proposal describes our progress to date and outlines the resources needed to deliver a working transport system based on this technology to the LANSCE experiment.

2. Experimental Overview

The LANSCE neutron beta-decay asymmetry experiment is an approved experiment with 10 collaborating institutions (see collaboration responsibility list in the supplementary documentation). Virginia Tech has been given responsibility for developing and procuring suitable materials to transport and store the UCN. In this section, we give an overview of the experiment as a whole, and in subsequent sections we discuss Virginia Tech's responsibility in more detail.

The experimental systems divide into three functional groups (Fig. 3): the UCN source, the transport and polarization system, and the decay spectrometer. The source uses a unique solid deuterium moderator which produces a significant fraction of neutrons with speeds below the critical velocity of its Ni-58 coated containment wall. The neutrons are then held in a diffusive collection box to make sure that no neutrons above the critical velocity remain. At least one near-normal collision with the wall will assure that no neutrons above the critical velocity are left. The strength of the polarization magnet is such that neutrons below this critical velocity with the wrong polarization can not pass through. Once polarized, an adiabatic fast-passage (AFP) spin flipper is used to change the spin direction. Once inside the solenoidal field of the spectrometer, subsequent decay of polarized neutrons produce betas which spiral along the field-lines to one of two detectors, whose difference measures the A coefficient.

Source: The 800 MeV proton beam from the LANSCE accelerator impinges on a stopping-length tungsten spallation target. The MeV spallation neutrons are reflected back in a flux trap consisting of a LN-temperature Be reflector. Within the Be reflector is a LN-temperature polyethylene premoderator that surrounds a UCN guide tube that contains a few hundred cm^3 of SD2. The proton beam is on target for less than 1 second and is turned off after 40 microCoulombs of charge (4 macropulses) have been delivered. The UCN produced in the SD2 move up into the UCN storage bottle where they are filtered for 1 second to remove any neutrons above the critical velocity of the ^{58}Ni coated bottle. The bottle is pumped out through a 3.3 m

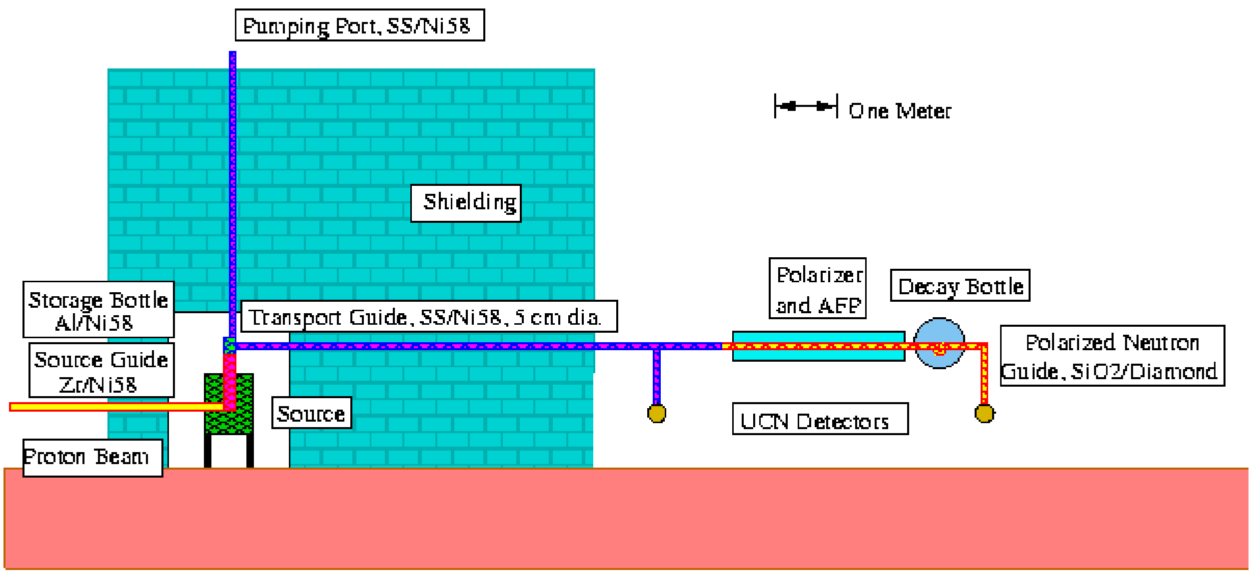

Figure 3: Overview of experimental layout

high vertical pumping port that consists of 4 sections of standard 8-cm diameter ^{58}Ni-coated Petersburg Nuclear Physics Institute (PNPI) stainless-steel guide. The height is chosen so that UCN with V < 8 m/s are gravitationally confined and cannot reach the top of the guide tube. The exit valve on the bottle is then opened and the UCN flow out to the experiment. This cycle is repeated every 10 seconds. This results in a time-averaged beam current on target of 4 µA. Eventually (after about 10 beam cycles) an equilibrium density of UCN is produced in the storage bottle. The beta asymmetry experiment sees essentially a constant flow of UCN.

Transport: The UCN storage bottle is joined to a long horizontal section that feeds the UCN to the beta asymmetry spectrometer. The first horizontal section is a 7-m long, 5 cm diameter ^{58}Ni-coated guide tube that penetrates the 4 m-thick shielding surrounding the target position. This guide transitions to a 2-m long, 5 cm diameter diamond-coated UCN guide in the region of the polarizer and AFP spin flipper. (This region must be electrically non-conducting to allow RF spin flipping of the UCN.) After passage through the AFP spin flipper, the horizontal guide transitions to a 50-cm long, 3-cm diameter diamond-coated UCN guide that is required for injection of the UCN into the beta decay trap through the

4-cm diameter warm bore penetration through the side of the superconducting solenoid. The horizontal guide also contains an UCN switch and a set of UCN valves that allow depolarization measurements. The horizontal UCN guides are at room temperature and they transition to LN temperature at the UCN storage bottle and the beta decay trap.

Spectrometer Design: The UCN are injected into the spectrometer through a 3-cm ID diamond-coated guide that penetrates through a specially-designed aperture in the side of the superconducting solenoid. (Fig. 4) The spectrometer consists of a 3-m-long UCN guide tube which defines a decay volume for the UCN. A 1.0 T magnetic field is generated along the axis of the UCN guide tube by a superconducting solenoid. At the ends of the decay tube, the magnetic field is expanded in the region before the detector. The strong magnetic field is used to determine the neutron spin direction and to guide the electrons from neutron decay out of the apparatus into a position-sensitive electron detector. This detector will be used to identify the location of the neutron decay within the decay volume. The betas will be stopped and their energy measured in a plastic scintillator. The spectrometer is shielded against both neutron and gamma backgrounds.

The UCN exiting the decay tube are monitored in an array of UCN detectors (surface barrier detectors with a thin ^{6}LiF layer plated onto them), transported down a diamond-coated UCN guide to a ^{3}He detector, or captured on ^{6}LiH surfaces in the region between the decay tube and the detectors. Thus, the UCN are effectively pumped away at the ends of the decay tube, thus strongly reducing the number of neutron decays in the field expansion region.

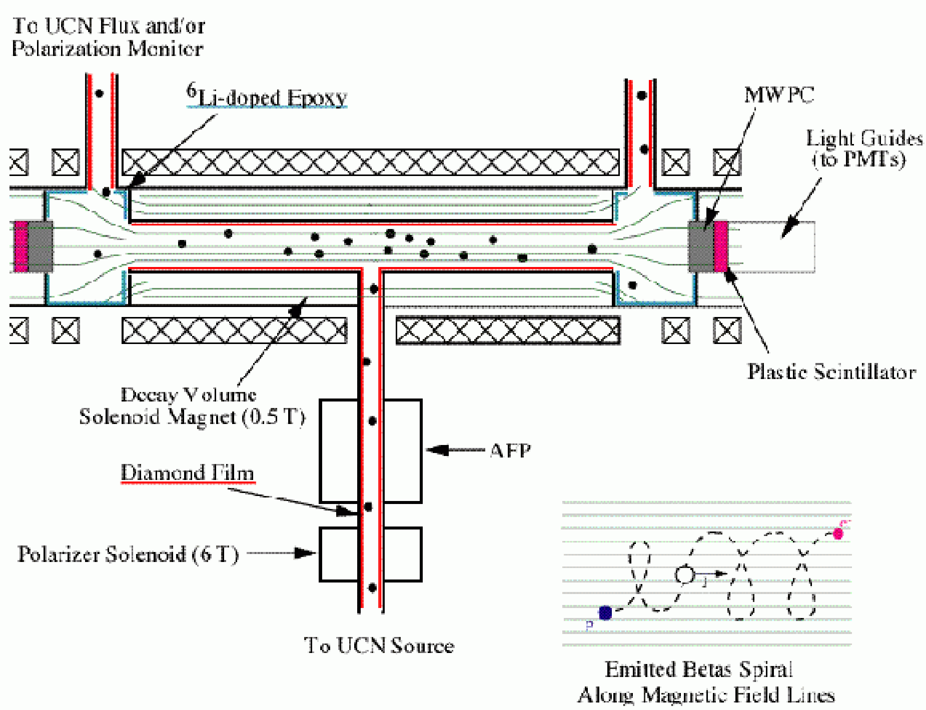

Figure 4. Top view of the A correlation spectrometer

Status: Here we briefly summarize the status of several of the major experiment components. Initial fluxes from the SD2 source were less than anticipated. This problem was overcome with the important discovery that it was necessary to remove the para-D2 from the mixture. With the para-D2 contamination reduced to 2%, the source has generated a very high UCN flux (~100 UCN/cm^3). The AFP spin flipper unit has been designed and is being constructed. The superconducting solenoid magnet, including the warm-bore feedthrough has been designed and is awaiting construction. The electron detector is currently planned to be a gas proportional counter backed by scintillators. Prototypes of these have been built and calibrated with monoenergetic electron beams. A time projection chamber is also being explored as a detector option. For the transport of unpolarized neutrons, 58Ni –coated stainless steel guide tubes will be sufficient. Finding suitable materials to satisfy the requirements of polarized neutron transport has had some preliminary investigation, and completing that job is the goal of this proposal.

3. Ultra-cold Neutron (UCN) Transport

As described in Section 1, an optimized UCN transport system is essential to achieve the statistical and systematic goals of the experiment. Extensive Monte-Carlo simulations have been carried out to optimize the transport system for the highest decay rate in the spectrometer bottle with as little depolarization as possible using the best-case scenario for guide-tubes (described later). Monte-Carlo calculations have been benchmarked with runs of our prototype source and by component testing at ILL where possible. The inputs to the Monte Carlo are the critical velocity (the velocity below which neutrons are externally reflected for all angles), a loss factor [3] to account for absorption at the surface, and the specularity (the probability of specular reflection). The characteristics of the primary ^{58}Ni guide-tube for unpolarized neutron transport are well known: critical velocity of 8 m/s, a loss factor of 0.0005, and specularity of 97.5%. The polarized guide section is modeled using our measured depolarization probability for carbon of $5x10^{-6}$, a conservative loss factor of 0.001, critical velocity of 7 m/s (corresponding to PLD diamond, see below) and specularity of 97.5%. The final decay rate in the spectrometer is over 100 Hz with the optimal configuration – enough to complete the experiment in the proposed time frame.

The ^{58}Ni guide tubes for unpolarized UCN transport are currently planned to be obtained from the Serebrov group at the Petersburg Nuclear Physics Institute. For the polarized UCN guide tubes, it remains to be shown that guide-tubes with these properties can be realized in fact. If we fail to achieve the parameters used in the Monte-Carlo simulations, the UCN rate could be dramatically reduced since during transport through the experiment the polarized UCN bounce off the walls several thousand times. The polarized neutron transport material properties that must be satisfied to achieve all of our goals are summarized below.

Critical Velocity: Coatings with the highest critical velocities must be used in all parts of the experiment to increase the final decay rate of the UCN in the spectrometer. The flux of UCN through the experiment increases proportional to the cube of the critical velocity of the guide. This is because spallation neutrons are down-scattered to a Maxwell speed distribution peaked around 20 to 30 m/s. The UCN speed distribution is the v^2 tail of this Maxwellian distribution, which integrates to v^3. Critical velocities of four to eight m/s are common for UCN guide and bottle materials. The difference in critical velocity between these extremes would lead to an eight-fold gain in the flux to the spectrometer. The increased flux could also be used to lessen the time spent in the spectrometer by the UCN, which would lessen the amount of depolarization in the spectrometer. This could be accomplished by not using a diffuse surface for the spectrometer bottle, thereby lowering the number of bounces in the bottle and the chance of depolarization. In general a coating with a high critical velocity makes the experiment design more flexible and likely to succeed.

Hydrogen free: One mechanism for up-scattering of neutrons to higher speeds, and subsequent loss, is the presence of hydrogen in the walls. This effect can be reduced by substituting deuterium for the hydrogen; the higher deuterium mass results in less energy transfer to the scattered neutrons.[4]

Smoothness: In addition to a high critical velocity the guides need to have near specular reflection for UCN to reduce the average number of bounces. UCN have a wavelength one tenth that of visible light, so the surfaces need to have a better than optical finish. This demands that the guide tube have roughness on the order of nanometers over micrometer distances.

Non-depolarizing: To preserve the neutron's polarization, gradients in the magnetic field must be minimized. To accomplish this, materials with low magnetic susceptibility must be used for the construction of the polarized guide sections in addition to having well designed magnetic fields. In addition, the polarized guide passes through a 10 Gauss, 30 MHz RF field in the AFP, which imposes an additional restriction that the coating and guide be non-conducting or very thin to avoid RF field attenuation.

Neutron cross-section: Another material property to consider is the neutron absorption cross section of the guides. This is especially important since the cross section increases for lower energy neutrons. This needs to be balanced against the other properties of the material to obtain a high flux through the spectrometer.

Mechanical Stability: The guides will undergo large changes in temperature during the running of the experiment; this needs to be considered when designing the guides. During calibration, activated Xe gas will be introduced into the spectrometer; as part of this calibration the gas will be cryopumped onto the walls to look at the effect of betas scattered off the walls back into the fiducial volume. In order to lower wall losses due to up-scattering of UCN by water the guide will need to be baked out at high temperatures under vacuum. To lower the UCN wall losses due to phonon interaction the guide will be lowered to liquid nitrogen temperatures during data runs. These changes in temperature will put extra demands on the coating-guide interface.

4. Material Selection of Polarized UCN Guides

For the **polarized** guide section, none of the standard UCN guides satisfy all of our criteria. Therefore, we have begun a program of testing some relatively new commercial coating options and developing and testing our own coating and substrate.

Coatings: A survey of material properties of many guide materials is contained in Table 1. Many of the materials are ferromagnetic, conducting, or have too low of a critical velocity to be of use with our current UCN flux. Copper and beryllium are both good as far as depolarization goes, but they are conducting.

Carbon appears to be the most promising material. It has low magnetic susceptibility, high critical velocities, is non-conducting, and has a low neutron absorption cross section. There are currently three types of carbon which look promising for use in the experiment. One option is sputtered graphite or chemical vapor deposition amorphous diamond (CVD diamond) which have been tested with neutrons and used for UCN; our collaboration has been testing these to see if they satisfy our other criteria. The other options are pulsed laser deposition amorphous diamond (PLD diamond), which has had limited neutron testing, and carbon nitrogen compounds. These coatings are currently being developed and characterized by our group at Virginia Tech.

The Russian members of our collaboration have made and tested sputtered graphite. As described below, depolarization tests were done on this carbon and it performed very well. Neutron reflectometry found this coating to have two critical velocities, 4.5 and 5.7 m/s [5]. The two critical velocities also show up when looking at UCN storage times in graphite traps. Both critical velocities are below the theoretical 6.11 m/s for crystalline graphite.

The CVD diamond coating currently in use by the UCN community is made by Surmet [6]. It is made in a reaction chamber below 400 C from a mixture of hydrogen and hydrocarbon gas. The hydrogen in the coating causes excessive loss of UCN by up-scattering due to the equivalent masses of the UCN and hydrogen. To minimize the up-scattering, the hydrogen is replaced with deuterium (DCVD), which has a scattering length similar to carbon and twice the mass of hydrogen. The final product has about 20 percent deuterium in it [4]. The Surmet coating is as smooth as the substrate it is deposited on up to one-micron thickness. Neutron reflectometry studies have measured the Surmet coating to have a critical velocity of 6.49 m/s.

PLD diamond is made by laser ablation of graphite onto a substrate in a vacuum. The resulting coating is an amorphous tetrahedral (sp_3) bonded film with diamond-like properties. This produces a coating free of hydrogen that is denser than DCVD diamond. This process also produces coatings with smoothness similar to the substrate used. There is currently very little data for PLD carbon with regard to neutron interactions. A neutron reflectometry study by Findeisen, et al [7] found atomic densities, which give a projected critical velocity of 6.95 m/s. Reported densities for PLD films [8] would also give critical velocities around 7 m/s. We are in the process of making and testing PLD diamond; this will be covered in depth in a later section.

Carbon nitride compounds (CN_x) are also made by laser ablation of graphite but in a nitrogen atmosphere at around .1 torr. These are interesting since nitrogen has a bound coherent scattering length 1.5 times larger than carbon. This could give even higher critical velocities if the coating densities are held high. The theoretical compound carbon nitride (C_3N_4) could have critical velocities around 8 m/s. We are currently making CN_x films for testing with neutron reflectometry. These films also have roughness the same as the substrate they are applied to.

Recent measurements [9] by members of our collaboration (led by Serebrov) have made the first quantitative assessment of depolarization of UCN in material traps. This study introduced UCN to a trapping volume coated with beryllium, and then introduced various foils to measure changes in the depolarization rate.

The apparatus used for making these measurements is a 100-liter Be-coated UCN bottle filled to a density of about 6 UCN/cm^3 from the turbine source at the ILL. This bottle was connected to a similar UCN bottle by a UCN guide that passed through a 4.5T superconducting solenoid. Valves at the entrance and exits could be opened and closed so that only UCN in one polarization state can enter the second bottle. That bottle is then shut for a period of time (up to several hundred seconds) and then allowed to exit through the solenoid. Any UCN that underwent a spin flip are then trapped in the second bottle. The spin-flipped UCN are detected by opening a valve to a UCN detector on the second bottle. It was possible to place thin sheets of other materials in the UCN bottle, thus providing a means to study depolarization from different materials.

The results are shown in Table 1. The depolarization rates for C, brass, Cu, and Teflon are all reasonably consistent with about 10^{-6} spin flip probability per collision, which meets the needs of our experiment. The sensitivity of the measurement was limited by the use of Be (which turned out to have a rather high probability of spin flip) as the liner material for the UCN storage bottle. We are now carrying out further depolarization measurements at the ILL using a Cu-coated UCN bottle. At present, all available information indicates that carbon (in the form of a diamond-like coating) will make a good wall material for polarized UCN.

There is also data on the depolarization rate of UCN on Surmet-produced DCVD diamond surfaces from the ILL EDM experiment.[4] The EDM experiment uses a diamond-coated cell for holding the UCN while their spin precesses. Measurements with two size cells (50 and 100 liters) have been made and a lower limit on the depolarization rate of 1000 s been set. All of the evidence indicates that the depolarization time in the EDM cell is limited by magnetic field nonuniformities in the cell (mainly due to the UCN valve located at the entrance to the cell) and that the intrinsic relaxation rate is much longer than 1000 s. Thus, experimental data provide a conservative limit of a depolarization of $< 5 \times 10^{-3}$ in a bottle with a holding time of 5 seconds. Thus, all data and calculations indicate that the depolarization relaxation time is quite likely to be sufficiently long that depolarization in the UCN bottle in the beta asymmetry experiment will be negligible.

Substrates: Quartz tubing is the currently favored candidate for a substrate to deposit the coating on. Commercial quartz tubing was tested for transport during the May 2000 run of the prototype SD2 source at LANL. To test various parts of the transport system a thin aluminum window was placed in the guide system. This separated the vacuum system of the source from the rest of the guide system, which allowed different guide configurations to be tested without changing the source parameters. To test the quartz tube, a one meter section was used for horizontal transport of the UCN to the detector section. This section was then replaced with a polished stainless steel tube of the same length. The transport properties of the two tubes were found to be the same within the accuracy of the data. Since the quartz has a lower critical velocity than stainless steel this would indicate that the quartz tube is a very good start to a guide tube. The critical velocity for quartz is 4.26 m/s and stainless steel is 6 m/s implying that the flux through the stainless guide should be three times that of the quartz guide if the loss per bounce were the same. This indicates that quartz has a low loss per bounce. It appears that commercially available quartz tubing would be a good guide tube if a high critical velocity coating were applied to it.

Table 1: Guide Material Properties

Material	Critical Velocity m/s[10]	Magnetic Susceptibility (10^{-6}) cgs[11]	Probability of spin flip per collision (10^{-6}) [9]	absorption cross section for 2200 m/s neutrons [12]	Current UCN usage
C	Dia. 7.65, Gra. 6.11	Dia. -5.9, Gra. -6.0		.0035 b	
Graphite (Serebrov)	4.5, 5.7		1.9		bottles
CVD C (H_2 exp)	5.13, 4.68				
CVD C (D_2 NIM)	6.49				EDM bottle
PLD C (exp)	6.95,7				
CN_x	7.5				
Si	2.3	-3.12		.171 b	substrate
SiO_2	4.26	-29.6	14		substrate
Al	3.24	16.5		.231 b	guide backing
Zr	3.85	120		.185 b	guide backing
Cu	5.66	-5.6	1.2	3.78 b	guides
^{58}Cu	6.76	-5.6		2.17 b	
Be	6.89	-9.0	7.7	.0076 b	guides, bottles
BeO	6.99	-11.9	46		
Stainless Steel	6.0	ferro			guides, bottles
^{58}Ni	8.14	ferro		4.6 b	guide coating

Initial depolarization studies of quartz tubing indicate a depolarization rate an order of magnitude higher than carbon coatings. The depolarization could be caused by the native impurities in the tubing or from surface contamination. For example, ferromagnetic iron is typically present at the 0.2 ppm by weight level in commercial quartz.[13] The surface could also be contaminated with metal from the manufacturing process; this contamination should be reduced with surface cleaning in aquaregia. Further depolarization and surface studies are underway to determine if a surface cleaning and carbon coating can reduce the depolarization rate to that of carbon alone.

Current Preference: A guide made of quartz tubing with a carbon coating would meet all of the experiment requirements. Carbon has the lowest magnetic susceptibility and depolarization rates, and it is non-conducting. Deuterated CVD diamond is currently in use in the EDM experiments at ILL; tests to get a depolarization probability are planned by our collaboration. There is evidence that CVD diamond does not behave well under thermal cycling. Raman spectroscopy has shown the dislocation of CVD diamond coatings from substrates which were subjected to liquid nitrogen temperatures[14], although the particular coating manufactured by Surmet has not been subjected to this test. Our collaboration, with assistance from our group, will continue to test the Surmet DCVD diamond coatings.

Since the DCVD potentially may not meet all of our criteria, it is important to pursue other promising options. The highest critical velocity of the carbon coatings is PLD diamond, and carbon nitrogen compounds also look promising. PLD diamond coatings appear to survive thermal cycling better than CVD diamond; they have been shown to survived repeated dips into liquid nitrogen.[15] These coatings have not been tested yet for depolarization, but they should be similar to carbon. As described in the next section, we have begun a program at Virginia Tech to produce and characterize PLD diamond and carbon nitrogen compound coatings.

5. Pulsed Laser Deposition Diamond and Carbon Nitride Coatings

PLD diamond has been under development since the late 1980's [16]. It can be generally described as the deposition of carbon onto a substrate from laser ablation of graphite. The type of PLD diamond produced depends on the laser wavelength and power. In general, power densities of 10^8 – 10^{12} watts per cm^3 are needed. For smooth surfaces with high densities, ultraviolet (UV) wavelengths are better. This is due to the fact that the high absorption of UV light by graphite causes less power to be needed, thus lowering the amount of boiling on the surface and causing less particulate to be deposited.

In this section, we describe a UV laser, PLD deposition apparatus that we have set up at Virginia Tech. Results of tests on coatings we have produced are described along with future plans.

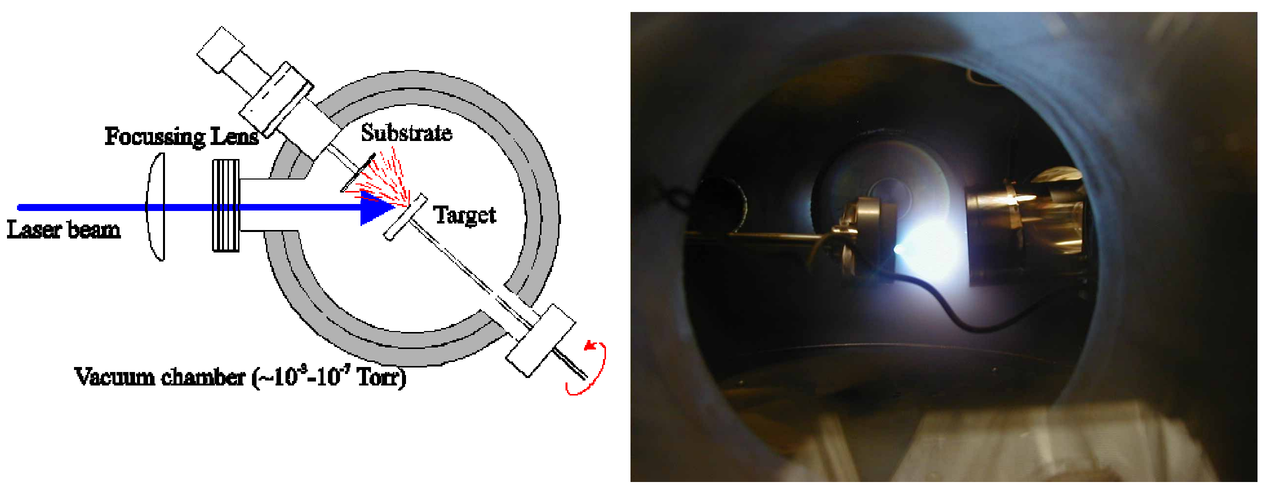

Figure 5: Schematic of pulsed laser deposition apparatus

Fabrication: The laser ablation system (Figure 5) being used for making test samples consists of an excimer laser, beam optics and a vacuum chamber. The excimer laser is operating at 248 nm wavelength and has a maximum energy of 1.2 joules per 25 ns pulse. The laser beam is focused into the vacuum chamber with a plano-convex quartz lens onto a pyrolitic graphite target. The resultant ablation plume is deposited onto a Si wafer or glass substrate. The laser is operated in the range from 100 – 600 mJ at a 20 Hz pulse rate. The lower end produces films with a graphitic nature while the higher energies result in diamond-like films. The energies above 400 mJ create coatings with a lot of graphite particulate; this can be decreased by defocusing the beam to lower the energy density.

The vacuum chamber is operated in a wide range of pressures with and without nitrogen atmosphere. The base pressure is around 10^{-6} torr , limited by rotary feedthroughs for the target and substrate. Nitrogen atmospheres of up to 10^{-1} torr have been used to create carbon nitride coatings.

Testing: A variety of PLD carbon and carbon nitride films have been produced under various conditions. Testing has begun to characterize all of the important material properties. They have been tested with atomic force microscopy (AFM), optical ellipsometery, and scanning electron microscopy (SEM) at Virginia Tech. Further studies with neutron reflectometry are starting at Los Alamos' SPEAR facility. We are also starting to design a depolarization test for our tubing in collaboration with Albert Young at NCSU.

Roughness – AFM: The PLD diamond films are inherently smooth due to the high kinetic energy of the carbon atoms in the ablation plume. The diamond bonding is optimized for 90 eV carbon atoms[17]; at this energy the atoms burrow into the substrate instead of wandering around the surface and forming islands which tend to grow and cause roughness. The main roughness is the occasional piece of graphite blown off of the target; aside from this the roughness is on the order of a nanometer.

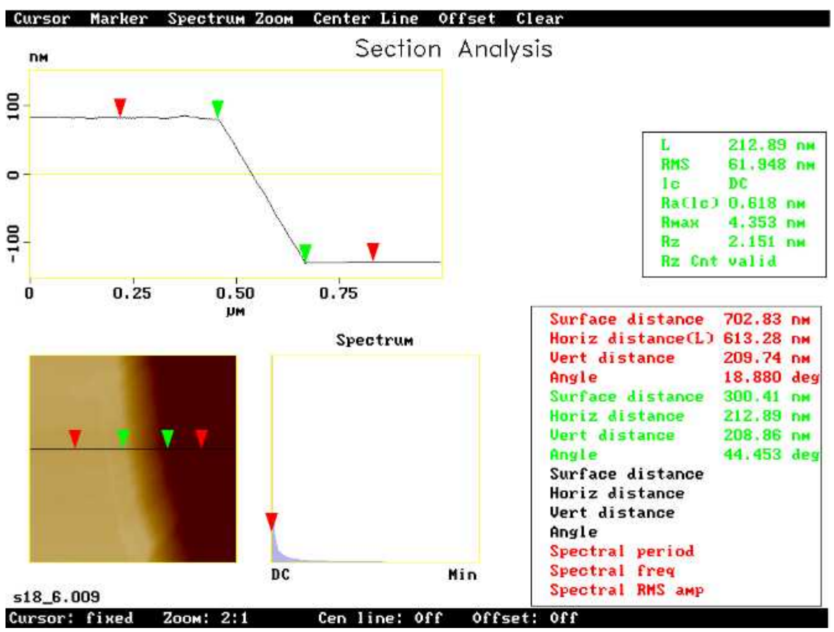

Figure 6: Atomic force microscopy cross section analysis of 200 nm PLD film. The right half has been peeled off. The upper graph is from a cross-section depicted in the lower left picture. Notice the smoothness of the substrate on the right compared to the film of the left.

For studying the roughness of the coatings the deposition is carried out on polished Si wafer. Coatings up to 600 nm thick have been studied. Float glass has also been tried with similar results. The roughness of the films is measured with an atomic force microscope. Figure 6 shows a cross section of a sample 200 nm thick. The image is 1 μm by 1 μm with 512 by 512 resolution. The maximum height in the image is 200 nm. The average roughness is less than 1 nm. Since the minimum wavelength of UCN is about 50 nm this will be more than adequate.

When compared to e-beam sputtering which is a standard technique to apply UCN coatings, these are very smooth. The roughness of a ^{58}Ni film evaporated onto Si wafer was ten times greater than the PLD diamond of the same thickness.

Graphite versus Diamond - Optical Ellipsometery: Spectrographic ellipsometry has been used to characterize the films and find their thickness. Since diamond and graphite have very different optical characteristics, ellipsometry is a very useful tool for optimizing the diamond-like properties of the film. Figure 7 shows results on one of our samples that demonstrates the presence of diamond.

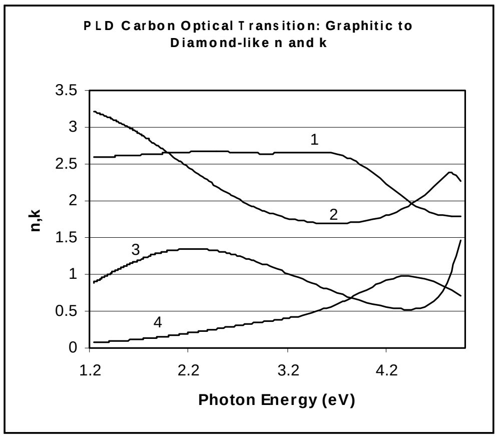

Figure 7: The initial ellipsometry data is converted to index of refraction and extinction coefficient data by using a Lorentz oscillator model. The lines 2 and 3 show the n and k for a low energy density PLD deposition; these n and k are predominately graphitic. This is indicated by the increase in n with decreasing photon energy and the k being peaked in the visible. Lines 1 and 4 show n and k for an energy density above the threshold for making PLD diamond. The n around 2.5 in the visible region and low k indicate a diamond-like film.

Composition – SEM: A SEM was used to perform energy dispersive spectrometry (EDS) composition studies of the films. The carbon nitride film was found to have a ten percent nitrogen content. If the atomic density is similar to the PLD carbon films, the critical velocity will be higher.

Critical Velocity – Neutron Reflectometry (SPEAR): A full spectrum of the films we have made are currently waiting to be tested with neutron reflectometry. The tests are being conducted at the LUJAN center on the SPEAR reflectometer. From the reflectometry data the atomic density of the films can be determined; using this information our SEM x-ray data can be calibrated to give the densities of our films [18]. From the composition and density measurements the critical velocities can be extracted.

Depolarization – UCN: There are two depolarization studies which can be carried out on our films. The first is using the depolarization experiment setup by Serebrov at ILL. This can be done with advanced scheduling during 2001. In addition we are developing a test using our prototype source and one of our superconducting magnets in conjunction with the AFP. The basic idea is to polarize the UCN and flip them with the AFP as they fill a test volume. The AFP is then turned off and only depolarized neutrons will exit the system back through the polarizing magnet where they will be detected with a ^{3}He detector.

Durability testing: As noted above, a potential problem with DCVD coatings is their possible inability to be thermally cycled. The coatings need to be tested in temperatures from liquid nitrogen to bake out temperatures. They also need to withstand glow discharge in a D_2 atmosphere. The PLD diamond coatings have been tested at temperatures up to 600 degrees C by Freidmann et al[19]. The effects of the heating are the release of compressive stress and an increase in the coating-substrate bond strength. Both of these are positive effects, which increase the durability of the coating for our uses. Collins et al [15] have dipped the films into liquid nitrogen and found no effect on the bonding to the substrate. We will test the coatings with multiple cycles of hot to cold and then check to see if any quantities have changed. The use of a glow discharge in deuterium gas is widely used in the UCN field to enhance the neutronics of guides and bottles. The effects of this process on the surface roughness will need to be studied for the diamond films with the AFM.

Development Program: Initial studies of our PLD diamond coatings are very positive. The remaining steps in the development program are to optimize the neutronics of the coating and develop a system for coating the inside of quartz tubing. During each step the durability of the coating needs to tested to arrive at a useful end product.

Neutronics: The neutronics of the coating need to be optimized for transport and depolarization. Neutron reflectometry studies will aid in the optimization of the coating's critical velocity and reflection quality. Once an optimal coating is determined, tubes can be tested with UCN for transport and depolarization properties. The depolarization will also be tested using the existing Serebrov system at ILL. Both the prototype source and the test beam at ILL are available for transport studies of the guides as we develop them.

Coating tubes: A system needs to be built for the coating of the inside of tubing using PLD diamond. The basic system is shown in Figure 8. The graphite target is held at a fixed position while the tubing is translated and rotated past it. We are currently redesigning the existing chamber to coat 15 cm sections of guide. This study will guide us in building a chamber to coat one meter sections of guide.

Schedule: The project schedule for the guide tube development is as follows. The testing of coatings and substrates for critical velocity and roughness will continue into early 2001. Concurrently, a new depolarization test setup will be under development with Albert Young at NCSU. During 2001, the system for coating the inside of tubing will be built and tube testing will start. The first generation of tubes will be tested during the summer of 2001 when LANSCE restarts. Guides for the A correlation experiment will be finished by early 2002 and installation will start. The current goal is to have the experiment started in 2002.

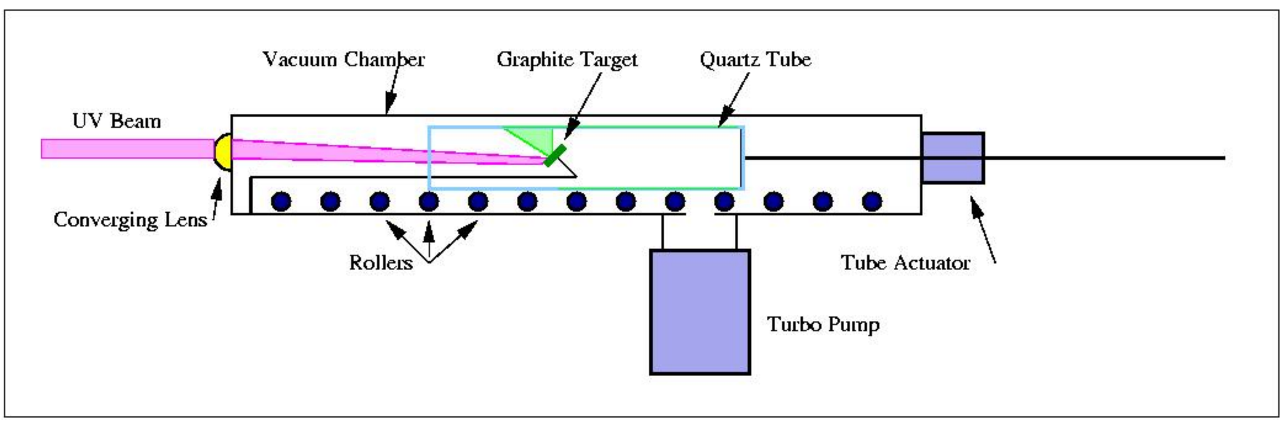

Figure 8: Schematic of proposed PLD coating apparatus

6. Other applications

The diamond coatings described here potentially may be useful in other applications. In particular, they might be interesting for spin-exchange optical pumping of ^{3}He and ^{129}Xe [20] because they are non-porous and hydrogen free. The dominant ^{129}Xe relaxation mechanism on silicone coatings currently in use is for the xenon to get trapped in pores in the coating and relax through interactions with essentially unpolarized hydrogen on the walls. Additionally, the toughness, and spin and chemical inertness of the diamond coatings might improve cell durability for higher temperature operation, where the chemical reactivity of the alkali metal vapors is problematic. Finally, the PLD coatings are transparent, which could prove vital in a variety of applications.

7. Results from Prior NSF Support

Vogelaar has received prior NSF support as co-PI or PI since 1993. Most of this work is directed towards measuring the Be-7 solar neutrino flux (Borexino), although he has also been involved in T-reversal studies and has been a participating member of the current experiment being described since it was first introduced while he was at Princeton. Mark Pitt has been supported by an NSF CAREER grant since May 1998. Significant accomplishments from this work include the first measurements of the asymmetry in backward angle parity-violating electron scattering from both the proton and deuteron. These measurements yield the first information of the strange quark contribution to the magnetic moment of the proton. Willi Graupner, while not currently funded by the NSF, has played a critical role in the characterization studies described in this proposal, bringing to bear skills not normally found in particle experimentalists. Such characterization studies are critical to the task at hand, and will allow investigation into other applications for these unique coatings.

8. Conclusion

The results of our progress to date were presented to the world UCN community at the LANL UCN Workshop September 6-8. The response to our research developments was very encouraging. There is a great need for a coating with the properties of PLD diamond. It will benefit new areas of research for UCN such as the A correlation experiment at Los Alamos, and also be useful as an upgrade for experiments like EDM of the neutron. It will also be much safer than beryllium, the standard non-magnetic coating with high critical velocity. PLD diamond will have the highest critical velocity of any non-exotic material. It was agreed that this coating will receive wide use in the next generation of UCN sources and experiments.

The funds requested in this proposal will allow us to carry out our responsibility for developing and delivering UCN guide tubes that fulfill the needs of the Los Alamos neutron beta asymmetry experiment. Of the numerous coatings being studied, PLD diamond films hold the greatest promise for an exciting new age of high density UCN experiments.

Note: Please see supplemental material as well:

1. Integration letter from co-spokesman of LANL UCN-A experiment
2. Appendix 16, Institutional Responsibilities from NSF/DOE proposal
3. LANL experimental approval from LANSCE deputy division director