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A Solid Polarized Target Development Facility at Jefferson Lab

J. Brock,[a,*] **C. Carlin,**[a] **T. Kageya,**[a] **C. Keith,**[a] **J. Maxwell**[a] **and D. Meekins**[a]

[a] *Thomas Jefferson National Accelerator Facility,*
12000 Jefferson Ave, Newport News VA, 23606, USA

E-mail: jbrock@jlab.org

Solid polarized targets are a crucial tool used in scattering experiments to investigate different properties of the nucleon. The equipment used to produce, measure, and maintain conditions for the polarized material is complex in comparison to nonpolarized targets. This, in combination with the frequency of polarized experiments run at Jefferson Lab, limits the opportunity to refine and build upon experience gained from previously constructed targets. To address this, the Jefferson Lab Target Group has begun the construction of a permanent facility dedicated to creating and testing material for Dynamic Nuclear Polarization. The design elements and manufacture of the test cryostat along with the incorporation of other DNP associated equipment will be presented.

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*Speaker

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1. Introduction

During the 6 GeV Jefferson Lab era there were twelve solid polarized targets used in experiments, and many more are scheduled in the near term for the 12 GeV beam. All of these experiments utilizing polarized targets to study properties of the nucleon have benefited from the technical expertise of the Jefferson Lab Target Group and the long tenure of its members. However, despite the complexity required to design, construct, and characterize new polarized target systems, a dedicated laboratory for polarized solid target R&D does not currently exist at Jefferson Lab. Instead, these activities typically occur in the short lead-up time before an experiment's commissioning or during the experiment itself. In these proceedings we describe the design and construction of a Target Development Laboratory for characterizing new and existing polarized solid target materials and techniques. We also discuss a potential new spectroscopic method for studying the radiation damage incurred by polarized targets during electron scattering experiments which is partly motivated by our observation of light emission from frozen ammonia samples as they were removed from the CEBAF electron beam in a recent experiment at JLab.

2. Solid Polarized Target Materials

The method used for polarizing solid materials at Jefferson Lab is Dynamic Nuclear Polarization (DNP). This involves transferring spin from unpaired electrons, with a high gyromagnetic ratio, to the nuclei with a much smaller ratio. The spin exchange occurs under conditions of high magnetic field and low temperature where the unpaired electron spins are highly polarized. In electron scattering experiments at JLab, these conditions are typically 5 T and 1 K. Off-center saturation of the electrons' spin resonance (ESR) frequency transfers their polarization to nearby nuclear spins, which are coupled to the electrons via the dipole-dipole interaction. The exact nature of the spin-transfer mechanism (solid effect, cross effect, or thermal mixing) depends largely on the width of the ESR line [1]. The nuclear polarization that builds around the unpaired electrons spreads throughout the target sample via spin diffusion.

The free electrons needed for the DNP process are generally provided by paramagnetic radicals within the target material. These can be added by chemically doping or by irradiating the material with ionizing radiation. While DNP has been successfully demonstrated in numerous materials [2], solid ammonia (NH_3 or ND_3) has been by far the most frequently utilized material for polarized solid targets at Jefferson Lab. Under the 1 K and 5 T conditions favorable for intense beams, it exhibits high polarizations (greater than 90% for protons and 50% for deuterons) relatively high ratios of polarized to unpolarized nucleons (3/17 and 6/20), and a strong resistance to the damage caused by the CEBAF electron beam. Ammonia is initially prepared for DNP by irradiation with an electron beam. This is typically performed to a fluence of about 10^{17} e^-/cm^2, with the sample submerged in a bath of liquid argon. Once irradiated, the material can be stored under liquid nitrogen until it is used in a scattering experiment.

Currently the only other solid polarized material scheduled to run in experiments at Jefferson Lab is ^{7}LiD, which is also prepared for DNP using ionizing radiation [3]. Development of material for DIII-D National Fusion Facility project [4] will also require ^{7}LiD.

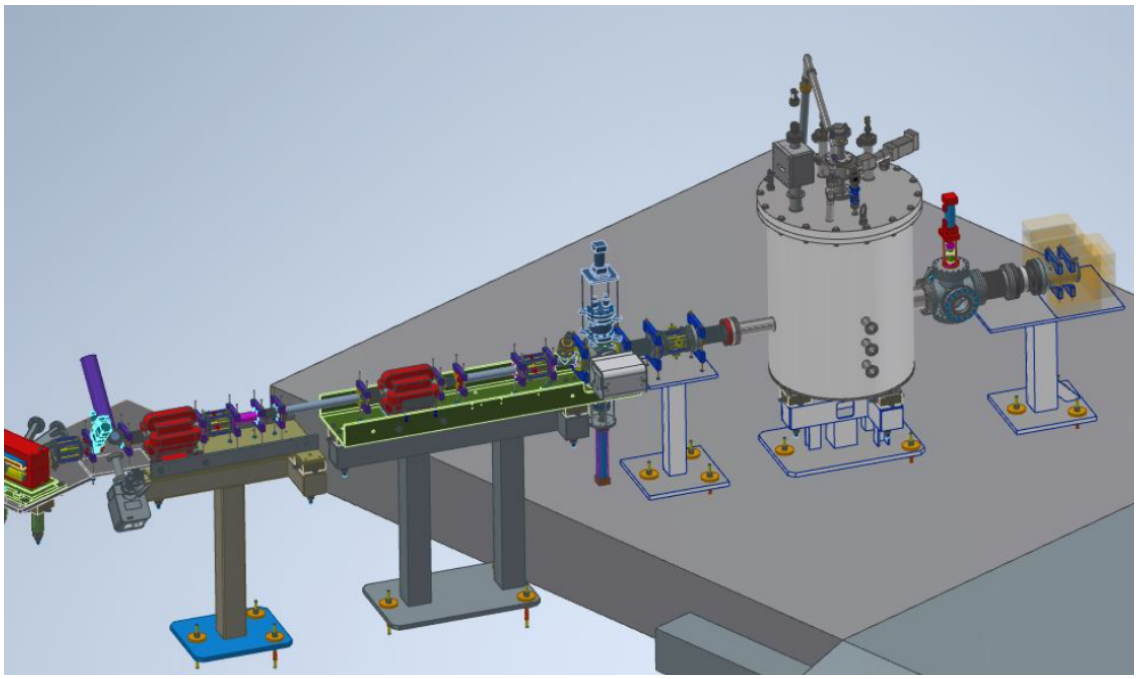

Figure 1: Irradiation Cryostat shown installed in the CEBAF injector area.

3. The Target Development Lab

Historically, irradiated ammonia used in the scattering experiments at Jefferson Lab have been supplied by the Solid Polarized Target Group at the University of Virginia with the initial irradiation of the target material taking place at the Medical-Industrial Radiation Facility (MIRF) at NIST. The Target Development Lab described here will enable the *on-site* production and characterization of target samples, and will allow detailed studies as a function of both irradiation dose and temperature that may improve targets' performance during scattering experiments. For example, a slight under-dosing prior to the experiment may result in a higher average polarization in the CEBAF beam and extend a sample's viable lifetime.

Two simultaneous projects are currently being undertaken to establish the Target Development Lab. The first is a collaborative effort between the Target Group, the Injector Group, Hall B, and the Cryo Group, and is a variable-temperature (2-300 K) cryostat for irradiating target materials in the CEBAF Injector (Figure 1). The temperature will be held fixed at a selectable value, the beam energy will be approximately 10 MeV (the injector's maximum), and the beam fluence on the sample will be measured using a Faraday cup. In parallel, the Target Group is constructing a vertical DNP 1 K test stand (Figure 2) where the irradiated samples will be tested for maximum polarization, characterized by electron spin resonance (ESR) and a potential new method, IR spectroscopy, described below.

One of the goals of the new DNP test stand is a reduction in the consumption of helium. The 1 K refrigerator will operate on a closed helium loop (Fig. 3), while the 5 T polarizing field will be generated by a "dry" (cryogen-free) superconducting magnet with a 4 inch vertical warm bore. Suspending the magnet in an existing 3 m deep pit makes access to the top of the 1 K refrigerator for

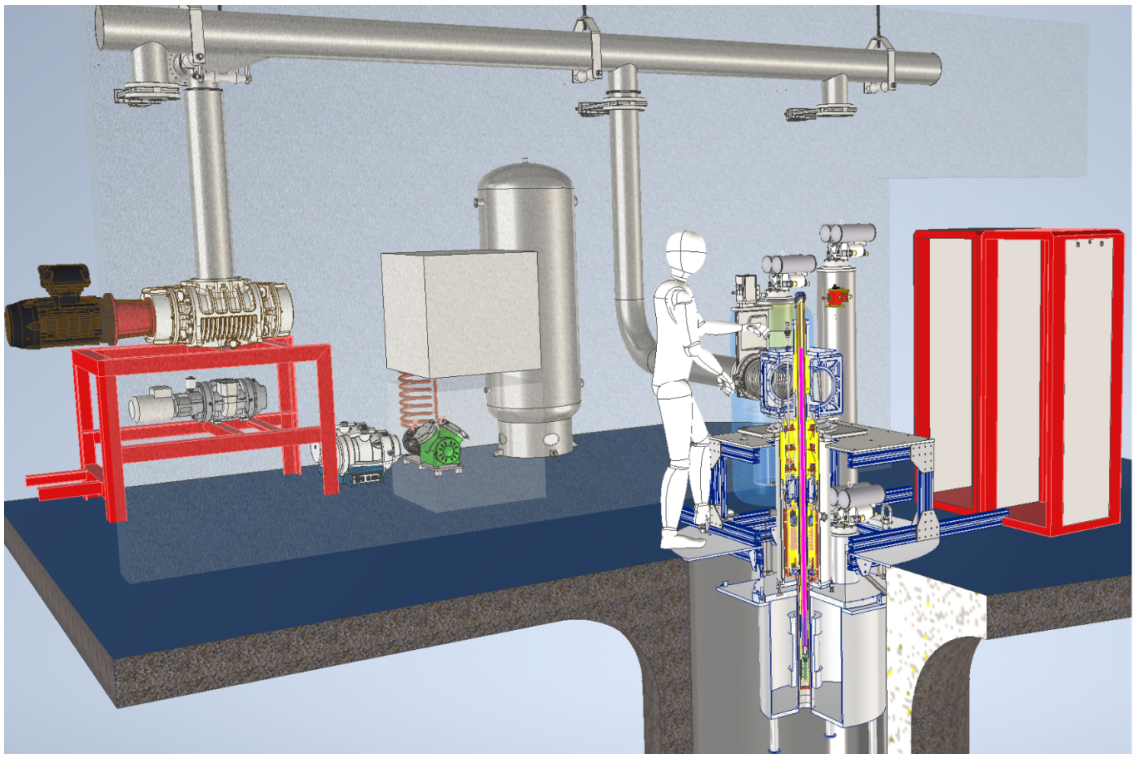

Figure 2: Shown is the Target Development Lab with the DNP 1 K refrigerator installed in the 5 T polarizing magnet suspended in a pit. In a separate pump room the large roots pumps are visible along with the backing scroll pumps, compressors and the helium ballast tank. A vacuum manifold has been installed as infrastructure to service additional evaporative refrigerators.

sample insertion more convenient. Precise alignment of the magnet within the pit is accomplished with a series of six adjustable aluminum tie rods in a triangular arrangement that provides all six degrees of freedoms.

Construction methods developed for the RGC horizontal refrigerator will be duplicated in the Development Lab refrigerator, with extra attention paid to maximizing efficiency. The vertical orientation of the refrigerator will reduce the heat load from convection in the helium vapor and permit a more compact design compared to the RGC refrigerator. Thermal shielding and material choices in the construction of the helium liquefier, the refrigerator, and the transfer line connecting them maximize the uses of the available enthalpy. A pulse tube[1] condensing 28 liquid liters of helium per day will supply the refrigerator, enabling continuous operation while allowing for one, possibly two, sample changes per day. The efficiency of the transfer line is maximized by keeping its length as short as possible and utilizing a thin-walled, stainless steel non-corrugated inner line, thermal standoffs, and a multiple layers of super-insulation at the optimal density of 27 layers/inch. This design maintains moderate flexibility while eliminating oscillations at low flows due to vapor lock. A phase separator inside the refrigerator removes the vapor from the liquid helium transfer to cool thermal shielding around the refrigerator and radiation baffles inside its pump tube. The liquid helium flows through a series of louvered and perforated-copper heat exchangers to a metering

[1] Cryomagnetics PT420

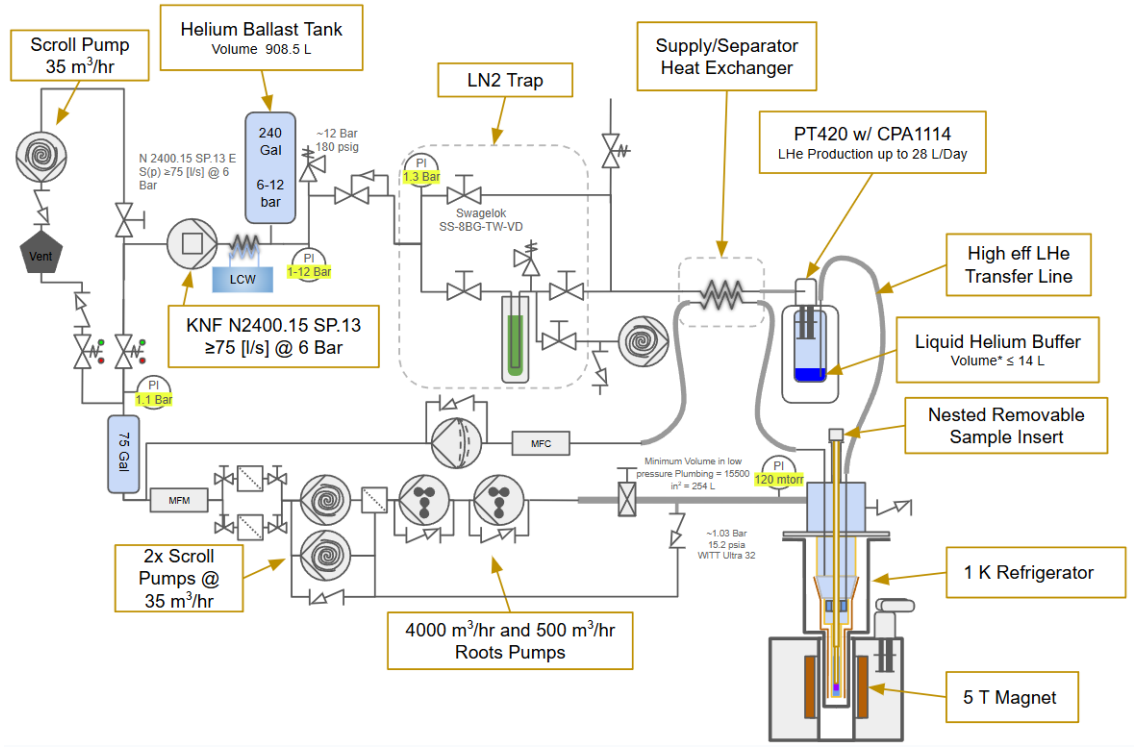

Figure 3: Irradiation Cryostat shown installed in the CEBAF injector area.

valve where Joule-Thomson expansion occurs, cooling the liquid to 1 K. Non-superconducting solder in the this heat exchanger is used to maintain good thermal conductivity regardless of the magnetic field. Attention to detail in low-flow, high-efficiency refrigerator construction is crucial, and building all components in-house allows for stricter quality control.

An estimated range for the combined flow of the separator and refrigerator will be 10 to 14 SLPM(~42 mg/sec). Large roots blowers, a 4000 m^3/hr backed by a 500 m^3/hr, will be used to maintain the pressure and flow rate necessary to achieve and sustain operation at 1 K. This includes a 0.3 W load from microwaves when polarizing. Two 35 m^3/hr scroll pumps in parallel will back the roots pumps and the flow will be monitored by a mass flowmeter. The separator flow and thermal shield cooling will be controlled by a mass flow controller and separator pump. A filtration system including a micron particulate filter followed by a series of molecular sieve filters and activated carbon filter will condition the helium exhaust gas prior to entering the compressor. A diaphragm compressor with an interstage cooler has been chosen to compress the exhaust gas to 6 atm, up to 12 atm max, before entering the ballast tank. The interstage cooler is necessary due to the increased amount of heat generated compressing helium vs nitrogen, which is typically the gas quoted in pump/compressor specifications. Further conditioning of the recirculated helium gas is done using liquid nitrogen traps. Two traps connected in parallel allow the alternating isolation for cleaning, baking and evacuating the traps. A precision two-stage, high-purity gas regulator with a flow restricting orifice maintains the pressure in the pulse tube liquefier at 1300 mbar and appropriately sized ASME safety relief and check valves prevent overpressure of the closed helium circuit. When changing target samples, a purge must be initiated inside the refrigerator to avoid

contamination. A flip-flop valve arrangement allows the refrigerator exhaust gas to be vented during this operation avoiding contamination of the pulse tube liquefier.

The refrigerator has a 50 mm diameter open axis to the 1 K bath to accommodate a series of nested components comprising the DNP insert, including the sample holder, NMR coils, waveguide for dynamic polarization and ESR studies, and optical instrumentation for IR spectroscopy. The insert will also include superconducting shim coils that may be used for ESR and to examine the impact of field gradients on the maximum achievable polarization. Construction techniques used for the Run Group C target will again be utilized on the DNP insert, such as thin-walled G10 tubes, PFA laminated NMR coils, and 3D printing methods like Direct Metal Laser Sintering (DMLS).

4. Thermoluminescence of Ammonia

The most recent polarized target constructed at Jefferson Lab was the longitudinally polarized target that was used for the Run Group C experiments in Hall B [5]. This was the first solid polarized target to run at JLab in its 12 GeV era. The target's design benefited significantly from a collaborative effort, drawing upon the collective experience of the lab's polarized target program and incorporating valuable knowledge and guidance from the broader scientific community.

The polarized target materials used in the RGC run, frozen ammonia and deuterated ammonia, NH_3 and ND_3, were contained in preloaded, removable cartridges and installed via a load lock into a mobile liquid helium bath inside a horizontal 1 K refrigerator [6]. Carbon, polyethylene (C_2H_4), and deuterated polyethylene (C_2D_4) samples were also utilized to quantify the scattering of electrons from nuclei other than hydrogen and deuterons. During RGC target samples were inserted and retrieved from the mobile helium bath on 75 occasions. This technique proved both highly reliable and efficient, reducing the overhead for target changes from approximately 14% of the scheduled run to under 3.5%. Other new processes were developed for material handling and sample loading, as well as a new NMR Q-meter for measuring the target polarization [7].

The load lock used for changing the target material included a fused quartz viewport, enabling observation and recording of the thermoluminescence (TL) of target samples as they were removed from the cryostat following irradiation by the 11 GeV CEBAF beam at 1 K. Blue light emissions near 480 nm were observed from both NH_3 and ND_3. Examples are shown in Figure 4, which includes green emission around 520 nm from the bath's downstream window. Observation of TL in these samples was unexpected, and our improvised attempts to measure the emission spectra in the UV-Vis-NIR region using a spectrophotometer were unsuccessful.

We believe the blue emission may be associated with excited hydrogen atoms because it was also observed in the C_2H_4, and C_2D_4 samples and in the PEEK[2] cartridges containing them, but it was not observed in PTFE material of the hydrogen bath or PCTFE cartridges containing the ammonia samples. The H-β line of the hydrogen Balmer series has a wavelength of 486 nm. The green light is probably from nitrogen or oxygen frozen from the air. The TL from all sources persisted for several seconds and varied in intensity as the materials warmed from 50 to 100 K. During the next operation of the RGC target, we hope to better instrument the load lock and accurately measure both the wavelength and temperature dependence of these emissions.

[2]Polyether ether ketone, $(C_{19}H_{12}O_3)_n$

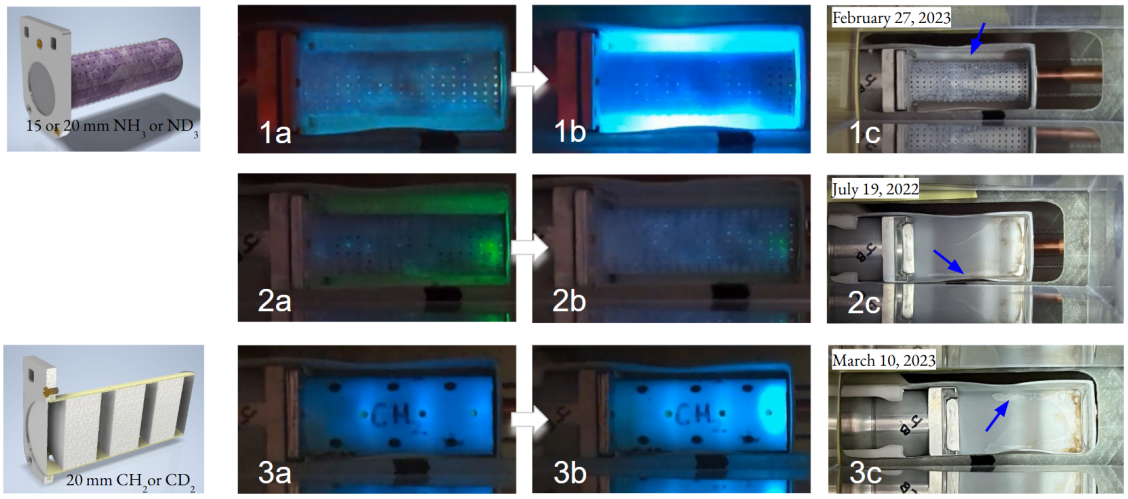

Figure 4: The two images on the far left are CAD renderings of the target cells containing frozen beads of ammonia or disks of polyethylene. The remaining nine images were taken on the dates indicated in the right column of images and the rows are sequential in time from left to right. 1a) Thermoluminescence (TL) of irradiated ND_3 target material seen immediately after retraction of the mobile bath to the cryostat's load lock. Initially dim, the blue glow increases in intensity. 1b) Intense flash of blue light from ND_3. 1c) The target cell just prior to removal. Frozen material can be seen at the bottom of the bath (blue arrow) and on top of the PCTFE cell. 2a) The target cell after retraction with a dim green TL emission from the downstream end of the bath. 2b) As the faint green glow fades, the blue hue returns from the TL of ammonia. 2c) Frozen residue in the bottom of the 1 K bath evaporates before the bath temperature reaches 100 K. 3a, 3b) TL from the polyethylene target and its PEEK container. 3c) Frozen residue remaining in the 1 K bath after removal of the polyethylene target cell.

5. Infrared Spectroscopy of Irradiated Ammonia

As previously described, ammonia is prepared with paramagnetic radicals using ionizing radiation at temperatures around 80 K, and the amino radicals NH_2 and ND_2 are believed responsible for DNP. However, continued irradiation by the CEBAF beam at 1 K slowly destroys the polarization in three ways. Firstly, this radiation can produce new radicals, not stable at 80 K, that do not contribute to the DNP process, but do promote spin-lattice relaxation of the nuclear spins. Secondly, it can create interstitial defects that introduce stress in the crystal lattice, which again promotes relaxation. Finally, it increases the concentration of the amino radicals beyond the optimum value. This "radiation damage" can be repaired multiple times (albeit with diminishing results) by annealing the sample for several minutes at temperatures 80–100 K. These trends are well known and have been the topic of many publications [2, 8–12], but after decades of operational experience at JLab, we feel there is a potential for significant improvement in the understanding of radiation damage, how to repair it, and possibly, reduce it.

While electron spin resonance is the traditional technique for identifying paramagnetic impurities in solids [13], an alternative method is infrared spectroscopy. The efficacy of IR spectroscopy in solid ammonia has been demonstrated by the astrochemistry community, who have observed the radiolytic products of various nitrogen hydrides found in interstellar clouds, Kuiper belt objects, Saturn's rings, and other near planetary objects and have studied their formation under similar

conditions in the laboratory (low pressure, cryogenic temperatures, and ionizing radiation) [14]. IR spectroscopy in these samples has identified not only paramagnetic radicals such as NH_2 and NH_4^+, but also nonmagnetic products such as H_2 and N_2 that are transparent to ESR (Table 1).

IR spectroscopy has also been used to study the formation of NH_2 radicals via chemical induction processes at cryogenic temperatures [15]. Both NH_2 and NH radicals are observed in thin films of NH_3:N:N_2 at 3 K. These thin films are deposited at 3 K and thermally cycled to 10 K, inducing chemical reactions. Their production is attributed to the mobility of ground-state atomic nitrogen at 10 K, which can recombine briefly into metastable states of molecular nitrogen N_2^*. The metastable states decompose into more highly reactive excited atoms which can then produce the radicals. That is,

$$\begin{aligned} N + N &\rightarrow N_2^* \\ N_2^* &\rightarrow N^* + N^* \\ N^* + NH_3 &\rightarrow NH + NH_2 \end{aligned} \tag{1}$$

Studying reactions such as these could benefit our understanding of the annealing process in irradiated ammonia, and we intend to pursue IR spectroscopy as an investigative tool, supplemental to ESR, in the DNP insert.

With this information it is imaginable that the concentrations of these radiolytic compounds could be monitored using the IR signatures to investigate the relationship between the maximum polarization of irradiated frozen ammonia and the presence of these free radical species and other crystal defects. It could also lead to improved annealing techniques or better, a targeted selective annealing process using IR or UV radiation at a sufficiently high lattice temperature to initiate mobility and/or a reaction with another nearby species. The observed phase transitions and hydrogen abstraction observed in the IR spectrum during monitored warming and heavy ion bombardment hint at ranges to begin these investigations [14, 16].

6. Summary

Jefferson Lab has a history of successful polarized target development, most recently the RGC target in Hall B. The complexity of polarized targets necessitates a dedicated R&D facility, and the construction of such a facility has now begun. Two key projects are underway, a cryostat for controlled target material irradiation, and a 1 K test refrigerator and 5 T magnet for material characterization. With the completion of these projects we will be able to investigate the effects of radiation on solid ammonia and other materials, particularly the formation of paramagnetic radicals and their impact on polarization. Utilizing ESR and IR spectroscopy to study the formation of radiolytic products in irradiated targets, we will be able to explore potential methods to eliminate or reduce unwanted paramagnetic centers through targeted annealing or other techniques.

7. Acknowledgements

The authors would like to express their sincere appreciation to the members, both past and present, of the Jefferson Lab Target Group and to others in the polarized target community. CAD

Absorption (cm^{-1})	Species	Feature Assignments
NH3 amorphous sample at 14K		
4994	NH_3	$\nu_3 + \nu_4$
4478	NH_3	$\nu_3 + \nu_2$
4361	NH_3	$\nu_1 + \nu_2$
3478	NH_3	$\nu_1 + \nu_L$
3459	NH_3	ν_3 deg. N–H stretch
3211	NH_3	ν_1 sym. N–H stretch
1880	NH_3	$\nu_4 + \nu_L$
1625	NH_3	ν_4 deg. N–H deform.
1074	NH_3	ν_2 sym. N–H deform. / "umbrella"
Radiolytic products		
4131	H_2	ν_1 H–H stretch.
~3040	*cis*-N_2H_2	ν_5 N–H stretch.
2890	NH_4^+	$2\nu_4$
~2780	*iso*-N_2H_2	ν_5 N–H stretch.
2326	N_2	ν_1 N–N stretch.
2084–2060	$NH_4^+N_3^-$	N–N–N asym. stretch.
2022	N_3^-	N–N–N asym. stretch.
~1500	NH_4^+	ν_4 N–H bend.
	NH_2	ν_2 N–H bend.
~1280	*trans*-N_2H_2	ν_5, ν_6 N–H bend.
~895	N_2H_4	ν_6 NH_2 rock

Table 1: The infrared absorption features of solid ammonia and species formed after irradiation. Reproduced from V. Bordalo et al. [14].

illustration in Figure 2 is courtesy of Casey Flanagan of the JLab Target Group. This material is based on work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics under contract DE-AC05-06OR23177.

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