

Radiation-tolerant polarized solid target

K. Tateishi^{1,2,*}, Y. Saito¹, D. Takahashi³, K. Sekiguchi^{1,3,4}, K. Aradono⁵, K. Hirasawa⁵, Y. Maeda⁶, Y. Nagao⁵, H. Nishibata⁵, S. Otsuka^{2,7}, H. Sakai¹, H. Sugahara³, K. Suzuki³, T. Uesaka^{1,2,7}, T. Wakasa⁵, and A. Watanabe¹

¹Nishina Center for Accelerator-Based Science, *RIKEN*, Saitama 351-0198, Japan

²Pioneering Research Institute, *RIKEN*, Saitama 351-0198, Japan

³Department of Physics, *Institute of Science Tokyo*, Tokyo 152-8551, Japan

⁴Department of Physics, *Kyoto University*, Kyoto 606-8502, Japan

⁵Department of Physics, *Kyushu University*, Fukuoka 819-0395, Japan

⁶Department of Applied Physics, *University of Miyazaki*, Miyazaki 889-2192, Japan

⁷Department of Physics, *Saitama University*, Saitama 338-8570, Japan



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Polarized targets evolved into indispensable tools in particle and nuclear physics. However, the polarized solid target is degraded by high-intensity beam irradiation, known as radiation damage due to target heating and radical generation. We demonstrated a radiation-tolerant polarized solid target operating at room temperature. An annealing allows the spontaneous repair of the damage by reducing unwanted radicals. Using a single crystal of *p*-terphenyl doped with 0.01 mol% pentacene-*d*₁₄, dynamic nuclear polarization using photoexcited triplet electrons was applied to proton spins at room temperature and in 0.39 T. For the proof of concept, a deuteron beam with an energy of 135 MeV/u and intensities of 10⁷–10⁹ counts per second (cps) was irradiated. The proton polarization was determined to be 3.0% ± 0.2% (stat.) ± 0.1% (sys.) from a scattering asymmetry. The polarization was almost not attenuated up to 10⁹ cps, but the target crystal was yellowed. The visible-light absorption spectroscopy suggested irreversible radiation damage due to missing protons by the knockout reaction. The room-temperature polarized solid target allows impractical experiments with the conventional target system, leading to a next-generation spin-dependent accelerator science.

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Within decades, polarized solid targets as well as polarized beams evolved into indispensable tools to explore the structure and reaction in particle and nuclear physics. Recently, the origin of proton spin has been intensively investigated from the viewpoint of quantum chromodynamics using them [1]. The polarization also gives us important information about spin-dependent interactions, such as spin-orbit, tensor, and three-body forces in nuclei [2,3]. However, the small magnetic moment of nuclei leads to significant disturbances from thermal energy, resulting in only slight polarization. Thus, several methods for dynamic polarization of nuclei have been developed [4].

Dynamic nuclear polarization (DNP), a means of transferring spin polarization from electrons to nuclei by microwave irradiation, is applied to polarized solid targets [5,6]. The target material has to be cooled to cryogenic temperatures below 1 K and placed in a magnetic field of several teslas (>2.5 T) to polarize thermally equilibrated electrons [1]. Proton spins can polarize almost unity with DNP under these conditions

but are degraded by high-intensity beam irradiation, called radiation damage. The damage originates from two effects, target heating [7,8] and radical generation [1,9]. The former became more serious at cryogenic temperatures because of smaller heat capacity. To suppress the latter, a variety of materials were developed such as lanthanum magnesium nitrate (LMN), butanol, ammonia, and lithium hydride [10]. The collision with the high-energy beam particles induces chemical bond breaking of the target material, producing radicals that accelerate spin relaxation. Consequently, radiation damage is an important consideration for beam intensity and statistical accuracy in experiments using a polarized target.

To address this issue, we proposed the radiation-tolerant polarized solid target. The idea was spontaneous repair of the damage by “annealing.” The reduction of the unwanted radicals in ammonia was known by heating the target to 75–95 K [11]. The mechanism was carefully investigated and confirmed to occur around 190 K in organic materials [12], indicating that the radiation damage can be *effectively* canceled if the repairing rate exceeds the damage.

In this Letter, we present the polarized solid target with DNP using photoexcited triplet electrons (Triplet-DNP) operating at room temperature [13–15]. The photoexcited triplet electrons have large nonequilibrated polarization independent of temperature and magnetic field, e.g., >70% for pentacene-*d*₁₄, which is the standard polarizing agent of Triplet-DNP [16]. This method has been applied to the neu-

*Contact author: kenichiro.tateishi@riken.jp

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tron spin filter [17–19] and the polarized target for light-ion beams [2,20]. Proton spins were polarized up to 80% at 25 K and in 0.3 T using a single crystal of naphthalene doped with pentacene- d_{14} [17]. That of 30% was obtained even at 100 K [20]; however, naphthalene was rapidly evaporated at room temperature by laser irradiation. Therefore, we introduced *p*-terphenyl as a host molecule of the target [21]. The new system operates at room temperature and in 0.39 T. For the proof of concept, a 135 MeV/u deuteron beam was irradiated with the intensities of 10^7 – 10^9 counts per second (cps). The nuclear magnetic resonance (NMR) signal, which is proportional to polarization, decreased slightly during beam irradiation. This was attributed to laser degradation rather than radiation damage.

The procedure of Triplet-DNP in 0.39 T was as follows: Pentacene- d_{14} was photoexcited with a 556 nm pulsed laser. Then, a 10.2 GHz microwave was irradiated to transfer the polarization from electron spins to proton spins in *p*-terphenyl. The external field was swept during the microwave irradiation to improve the transfer efficiency. The sign of proton polarization can be varied by switching the direction of the field sweep. This scheme was called the integrated solid effect (ISE) [22]. By repeating the ISE sequence, the proton polarization can be build up until it is balanced with the spin relaxation. A single crystal of *p*-terphenyl doped with 0.01 mol% pentacene- d_{14} was grown with the Bridgman method [Fig. 1(a)]. The crystal with a size of $\phi 10\text{ mm} \times 2.5\text{ mm}$ was mounted in a Teflon holder and set the long axis of the pentacene- d_{14} parallel to the external magnetic field. The Triplet-DNP system is shown in Figs. 1(b) and 1(c). The electromagnet was designed for the detection of both longitudinally and transversely scattered particles. A diode-pumped solid-state laser (CNI, HPL-589-Q) was used for pentacene- d_{14} excitation. The wavelength, pulse width, pulse energy, and repetition rate were 556 nm, $\sim 600\text{ ns}$, 4.0 mJ, and 1.0 kHz, respectively. The resonance frequencies of triplet electron and proton in 0.39 T were 10.2 GHz and 16.7 MHz, respectively. A 20- μs microwave pulse amplified to 2.2 kW using a pulsed traveling wave tube amplifier (IFI, PT188-1KW) was applied, while the field was adiabatically swept with a voltage of $\pm 50\text{ V}$. The polarized proton signals were monitored using an OPENCORE nuclear magnetic resonance spectrometer [23]. The NMR signal was calibrated using the proton polarization independently determined from the measured scattering asymmetry. The technical difficulties were to prevent the target crystal and cavity heating due to high-power laser and microwave irradiation. The high temperature accelerates a spin relaxation, resulting in decreasing the proton polarization. For the crystal, air was blown with a rate of $\sim 200\text{ l/min}$. For the cavity, a Peltier cooler (TAISEI, UT-7070J-HS) was utilized via heat pipes.

A deuteron beam with an energy of 135 MeV/u was irradiated at RIKEN RI Beam Factory (RIBF) to confirm the spontaneous repair of the radiation damage of the polarized target and to determine the proton polarization from scattering asymmetry. The deuteron beam was transported to the E3A course in the E3 experimental room. The polarimetry was made using deuteron-proton elastic scattering [24,25]. The scattered deuterons and recoil protons were detected under kinematical coincidence conditions using a detector system

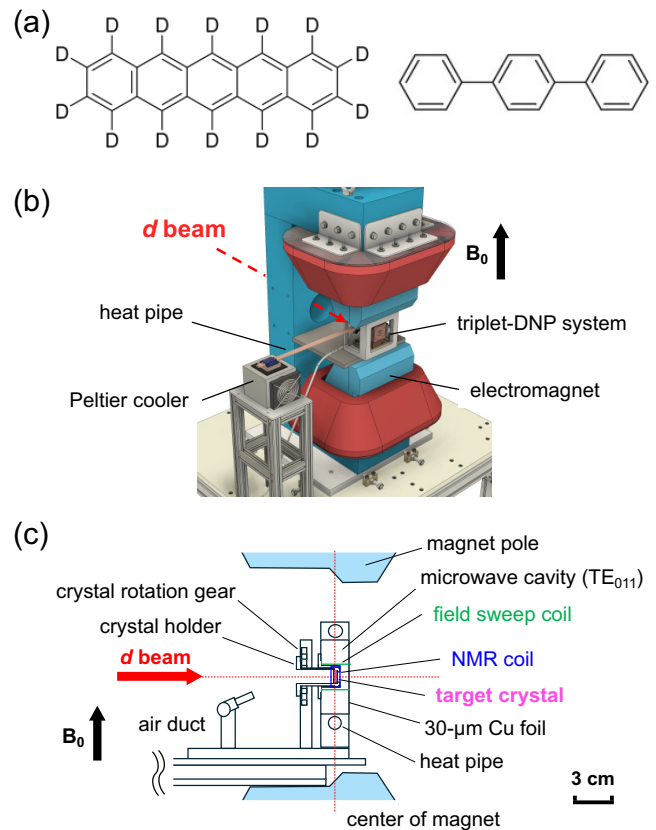


FIG. 1. (a) Molecular structures of pentacene- d_{14} ($\text{C}_{22}\text{D}_{14}$) and *p*-terphenyl ($\text{C}_{18}\text{H}_{14}$). (b) Polarized proton solid target based on Triplet-DNP operating at room temperature and in 0.39 T. (c) Side view of Triplet-DNP system installed inside the electromagnet.

consisting of plastic scintillators and multiwire drift chambers, arranged symmetrically in the left, right, up, and down directions relative to the beam direction [26]. The measured angles were 68.8° , 78.6° , 88.6° , 98.6° , 108.8° , 119.0° , 129.4° , 139.8° , and 150.3° in the center-of-mass system. The beam was stopped using a tungsten-based beam dump located 2.5 m downstream of the target. The beam counting rate was extracted by using the existing differential cross-section data for the deuteron-proton elastic scattering [27]. Together with this, the relative value of the beam counting rate was monitored by using the plastic scintillator placed upstream of the target.

The 135 MeV/u deuteron beam was irradiated for 25.5 h at 2.0×10^7 cps, 16.0 h at 7.0×10^7 cps, and 1.0 h at 1.0×10^9 cps, respectively. Figure 2 shows the NMR signal intensities as a function of time, which is proportional to the polarization. The signals were taken every 3 min. The target polarization was switched several times between positive, negative, and unpolarized to reduce systematic errors of the proton polarization estimation. The polarization buildup typically takes about 30 min, and the polarization can be reversed within 1 h. The negative polarization was slightly lower than the positive due to the improper crystal cutting. There is almost no signal attenuation for *p*-terphenyl target operating at room temperature, whereas a 10% signal intensity was attenuated in 8 h at 4×10^6 cps for the naphthalene target operating at 100 K and in 0.3 T [20]. The slight attenuation with increasing beam

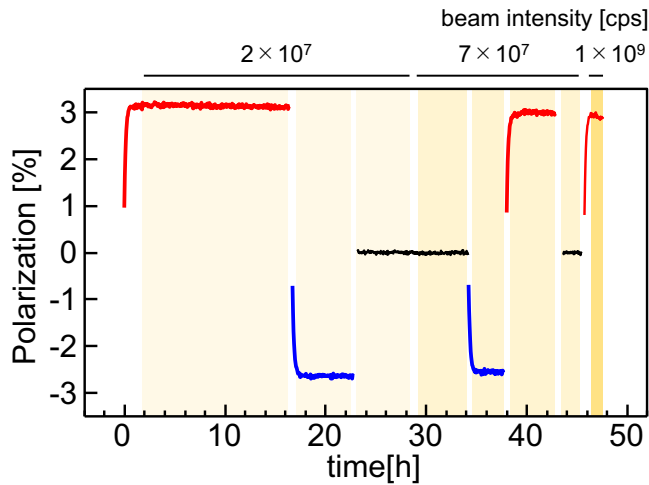


FIG. 2. Proton polarization buildup curve of the single crystal of *p*-terphenyl doped with 0.01 mol% pentacene-*d*₁₄ at room temperature and in 0.39 T. The polarization was switched several times between positive, negative, and unpolarized to reduce systematic errors of the proton polarization estimation. The 135 MeV/u deuteron beam was irradiated in the colored areas for 25.5 h at 2.0×10^7 cps, 16.0 h at 7.0×10^7 cps, and 1.0 h at 1.0×10^9 cps, respectively.

intensity was concluded to be laser pulse energy degradation due to the radiational environment. The laser pulse energy decreased from 4.0 to 3.4 mJ after the experiment. Details will be discussed in later paragraphs. Thus, a radiation-tolerant polarized solid target has been confirmed.

The target polarization can be determined from the deuteron-proton scattering asymmetry, independent of the NMR measurements. The target polarization was determined to be $3.0\% \pm 0.2\%$ (stat.) $\pm 0.1\%$ (sys.) averaged over the absolute values of both positive and negative polarizations in the first (2.0×10^7 cps) and second (7.0×10^7 cps) irradiations. The second and third values represent statistic and systematic errors, respectively. The polarization was $3.3\% \pm 0.4\% \pm 0.1\%$ in the first weak irradiation and $2.4\% \pm 0.3\% \pm 0.1\%$ in the second middle irradiation, respectively. Since the detectors could be damaged, the scattered particles were not measured during the third (1.0×10^9 cps) strong irradiation. The fact that the polarization from each measurement agrees within the error indicates that the repairing rate of the unwanted radicals is sufficiently fast, and no local depolarization has occurred. However, the achieved polarization was not high enough. An important direction for future work is to introduce a target crystal with a longer relaxation time, since the polarization is mainly limited by spin-lattice relaxation, which is caused by molecular vibrations of the host molecule [21,28].

For a better understanding, the depolarization at high-intensity beam was investigated. The polarization of the third irradiation area was $\sim 5\%$ lower than the first. As mentioned above, the laser pulse energy decreased from 4.0 to 3.4 mJ due to the radiational environment. Consequently, the polarizing power of Triplet-DNP would be reduced. In addition, the target crystal partially yellowed [Fig. 3(a)]. To clarify that, Triplet-DNP was carried out with the laser pulse energy of 4.0 mJ after the experiment. Figure 3(b) shows the proton

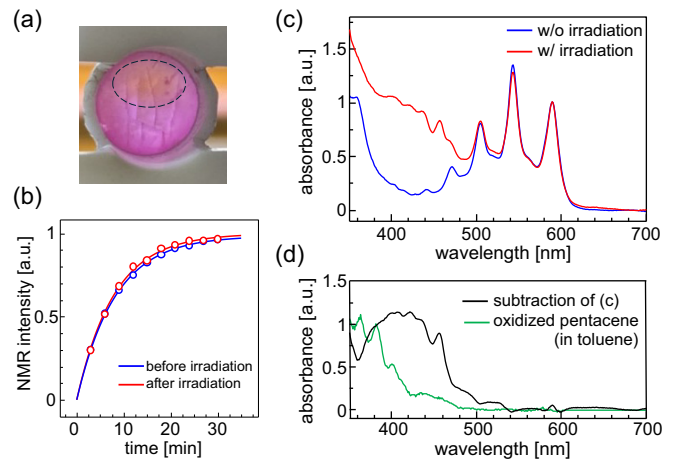


FIG. 3. (a) Photo of the target crystal after deuteron beam irradiation. (b) Proton polarization buildup curve before (blue) and after (red) deuteron beam irradiation. (c) VIS spectra of the target crystal with (red) and without (blue) deuteron beam irradiation. (d) VIS spectra of the subtraction of panel (c) (black) and oxidized pentacene in toluene (green).

polarization buildup curves before and after the beam irradiation. The initial buildup rate, the buildup constant, and the finally attainable polarization were reproduced. Therefore, we concluded that the depolarization at a high-intensity beam resulted in laser degradation due to the radiational environment.

We continued the analysis of the yellowing of the target crystal. There are two possibilities: oxidation of pentacene-*d*₁₄ by target heating and radical generation in *p*-terphenyl with knockout reaction. Visible-light absorption spectroscopy (VIS) was conducted to address that. Figure 3(c) shows the VIS spectra of the target crystal with and without the beam irradiation. The sharp peaks above 440 nm originate from the absorption of pentacene-*d*₁₄. *p*-Terphenyl dominantly absorbs ultraviolet light. The absorption around 350–500 nm increased with the beam irradiation, corresponding to the yellowing of the crystal. Compared with the spectrum of their subtraction and an oxidized pentacene in toluene, they were much different [Fig. 3(d)]. On the other hand, the subtraction was similar to the spectrum of chemically generated radicals in aromatic molecules [29]. Consequently, the yellowing is most likely to be radicals in *p*-terphenyl. One possibility for the radicals is due to the missing protons by the knockout reaction, resulting in irreversible damage. They induce the depolarization as paramagnetic relaxation sources, but their contribution was not observed in this experiment owing to the small amount. This will become more serious with increasing beam accumulation.

Finally, this target can be operated not only at high temperatures but also in low magnetic fields, indicating that it is suitable for use in nuclear physics experiments, i.e., low-energy charged particles. For example, three-body nucleon force research combined with a polarized deuteron beam is one of the promising applications. The force is essential to clarify nuclear phenomena such as discrete states of nuclei [30] and the equation of state of the neutron star [31]. Moreover, as large accelerators are constructed around the

world, the radiation damage of conventional polarized targets will soon become a bottleneck for statistical accuracy even in secondary beams. On the other hand, the target size limits the laser intensity; therefore, applications requiring large targets are more difficult.

In summary, we demonstrated the radiation-tolerant polarized solid target operating at room temperature. An “annealing” allows the spontaneous repair of the damage by reducing the unwanted radicals. Triplet-DNP is suitable for the concept because the photoexcited triplet electrons have large nonequilibrated polarization independent of temperature and magnetic field. We developed the Triplet-DNP based polarized target operating in 0.39 T and at room temperature using a single crystal of *p*-terphenyl doped with 0.01 mol% pentacene-*d*₁₄. A deuteron beam with an energy of 135 MeV/u and intensities of 10⁷–10⁹ cps was irradiated at RIKEN RIBF. The proton polarization was determined to be 3.0% ± 0.2% (stat.) ± 0.1% (sys.) from a scattering asymmetry and confirmed the spontaneous repair of radiation damage. After the experiment, VIS spectra suggested irreversible radiation

damage due to missing protons, indicating more serious with increasing beam accumulation. The room-temperature polarized solid target benefits not only the radiation damage but also the simplification of the experimental setup. This allows impractical experiments with the conventional target system, leading to a next-generation spin-dependent accelerator science.

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Data availability. The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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