Efficient excitation of photo-excited triplet electrons of pentacene for dynamic nuclear polarization

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Dynamic Nuclear Polarization (DNP) has been successfully applied to a polarized target. DNP is a methods of transferring spin polarization from electrons to nuclei with microwave irradiation. However, as long as electron spins in thermal equilibrium are used as a polarizing agent, cryogenic temperatures of approximately 4.2 K will be required for hyperpolarization in the order of 10% even under strong magnetic fields of several Tesla.[1)] One approach for overcoming the limitation is to use non-thermalized electron spins as a polarizing agent instead.

DNP using the photo-excited triplet-electron spin of pentacene[2)] requires neither a cryogenic system nor a superconducting magnet, but a laser for creating hyperpolarized electron spins. There are four important requirements in the light source: wavelength, pulse width, repetition rate, and output power. The wavelength affects the excitation efficiency from the ground state to the excited singlet state. The pulse width influences the transition efficiency from the excited singlet state to the triplet state. The repetition rate is related the initial buildup rate of ^{1}H spin polarization. The output power is determined by the number of excited electrons, i.e., the sample volume.

To this end, a new laser system is developed, which is constructed using two neodymium-doped yttrium aluminum garnet (YAG) lasers with wavelengths of 1064 nm and 1319 nm and a LiB_3O_5 crystal for sum-frequency generation (Fig. 1). [3)] The wavelength, pulse width, repetition rate, and output power are 589 nm, 126 ns, 3 kHz, and 1 W, respectively. We developed oscillators independently using laser diode pumped YAG crystals. The acousto-optic Q-switch (A/O Q-sw) was chosen to generate light pulses because of the high repetition frequency. The pulse width can be varied by adjusting the input current of the laser diode, and in this experiment we used final output pulse widths of 84 and 126 ns. In order to generate 589-nm light, the infrared lights were overlapped at the LiB_3O_5 crystal. Finally, residual infrared lights were separated by visible-coated mirrors.

Using a single crystal of *p*-terphenyl doped with deuterated pentacene[4)], we carried out the DNP experiments in 0.35 T and at 300 K with laser pulses of 84 ns and 126 ns. The maximum polarization of a 84 ns laser pulse is 16% with a repetition rate of 400 Hz. The maximum polarization of the 126 ns laser pulse is 22% with a repetition rate of 600 Hz. The optimal repetition rate was different because the excited electrons play not only the role of a polarized agent but also that of a relaxation source. When the repetition rate was increased, the initial buildup rate increases, but the paramagmetism acceralates spin-lattice relaxation of ^{1}H spins. The final achievable polarization is determined by the balance of these two effects. Furthermore, to obtain higher ^{1}H spin polarization, our results indicated that a longer laser pulse width to increase the initial buildup rate or a higher external magnetic field to decrease the spin-lattice relaxation of ^{1}H spins is necessary.

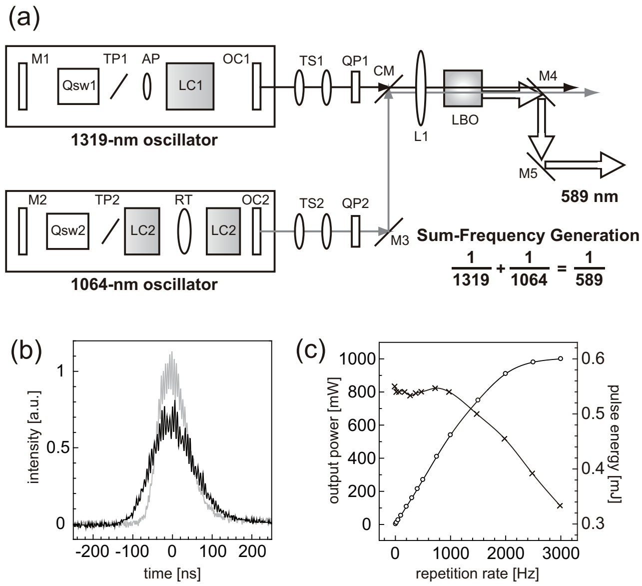

Fig. 1. (a) Schematic view of 589-nm laser constructed with two Nd-doped YAG lasers with wavelengths of 1064 nm and 1319 nm and a LiB_3O_5 crystal for sum-frequency generation. M1,M2: Oscillator mirror; Qsw1,Qsw2: A/O Qsw; TP1,TP2: Thin Plate; LCP,LC2: Laser Chamber; RT: Rotator; OC1,OC2: Output coupler; AP: Aperture; TS1,TS2: Telescope; QP1,QP2: Quarter-wave plate; CM: Combiner mirror; L1:Lens; M3,M4,M5: Mirror; LBO: LiB_3O_5 crystal. (b) Pulse shape. (c) Output power (circles) and pulse energy (crosses) as a function of repetition rate.

References

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