

Spin polarization of salt nuclei by optical pumping of Cs atoms and laser heating of Cs salt

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We report hyperpolarization of alkali-metal salts by optical pumping of alkali-metal vapor. The spin angular momentum is transferred from the polarized alkali-metal atoms to the salt nuclei at the surface, and the spin polarization diffuses into the depth of salt. We find laser heating to be useful for facilitating spin diffusion, leading to the nuclear polarization of approximately 0.1 % at the magnetic field 0.56 T and the temperature 100 °C. The signal enhancement is so large that we are able to detect a small amount of solid salts without averaging and to measure the real-time development of nuclear spin polarization.

One of ways to increase the signal-to-noise ratio in conventional NMR experiments is to enhance the nuclear spin polarization compared to the value in the thermal equilibrium. Recently, we developed way to enhance the spin polarization in alkali-metal salts by optical pumping of alkali-metal vapor [1-4]. The polarized atoms in the gas phase transported the angular momentum to the surface of the salt. The incoming flow was characterized by the electron spin current and the nuclear spin current [1]. The enhancement of NMR signal in the salt was well correlated with the nuclear spin current predicted for the optically pumped atomic vapor. We therefore found that the nuclear polarization of the adsorbed atoms was transferred to the nuclei of the salt, and then the spin polarization diffused from the surface into the depths of the salt [2]. Since the NMR signal was enhanced for the stable cesium chloride as well as the dissociative cesium hydrides, the spin transfer at the surface was not due to the exchange of the adsorbed atoms and the salt ions but primarily to the nuclear dipole interaction among them [3]. When thin crystallites were in contact with the polarized vapor, the nuclear polarization was efficiently transferred at the relatively large surface area and also quickly built up since they were similar in size to the spin diffusion length. The spin polarization, however, quickly decayed in the dark because the accumulated angular momentum easily leaked via the surface of thin crystallites [4]. The spin diffusion, which is limited by the dipolar interaction between stationary nuclei at room temperature, is facilitated by the ion movement at the high temperature and thus the nuclei can be polarized in the entire volume of macroscopic particles. As well as optical pumping of atomic vapor, laser heating of crystals makes it possible to hyperpolarize the alkali-metal salt.

A permanent magnet was used for optical pumping and the subsequent NMR detection. The free induction decay signal was detected by applying a single rf pulse. The spectral area of the Fourier transform was the measure of the nuclear polarization in the salt. The Cs chloride and the Cs metal were contained in a glass cell filled with quartz-glass wool and N₂ gas. The Cs salt was formed on the glass fibers for having a large surface area in contact with the polarized Cs vapor. The glass cell

placed at 0.56 T was electrically heated at 100°C [2]. The Cs atoms confined in small spaces of the glass wool were optically pumped by a single-mode laser at the D₂ line. Since the pump light was multiply scattered by the glass wool, the directions of polarization and propagation of the light were randomized in the optical pumping cells. Nonetheless, the Cs atoms can be polarized very near the salt surface because a particular polarization component of light was selectively absorbed due to the electron Zeeman splitting [3]. The laser frequency was precisely tuned to one of the absorption lines to maximize the NMR enhancement. The Cs NMR signal of the salt was greatly enhanced when the laser beam was focused at the glass cell. The enhanced line was narrower than the thermal equilibrium signal at 100°C. The spin polarization was built up within 10 s, while the longitudinal relaxation time T_1^{bulk} was approximately 650 s for bulk crystal in the dark.

The above observation suggests the motional narrowing caused by the ion movement. The ions move not only in the molten salt but also in the crystal and can transport the angular momentum. For the crystal, the diffusion coefficient is obtained from the electric conductivity measured with applied voltage [5]. The cation and anion conductivities are predominantly due to the activation energy of single vacancies and are greatly increased by heating. Since the anion nuclei most affect the linewidth and since the anion conductivity is larger than the cation's, the narrowing of Cs NMR signal is mostly due to the anion movement. On the other hand, the cation movement contributes to the NMR enhancement because the nuclear polarization was transported by the cations into the entire crystal. The characteristic distance of the spin diffusion is $\sqrt{DT_1}$, where the time T_1 is due to the electric quadrupole relaxation, and the diffusion coefficient D , which depends on the density and the diffusion coefficient of cation vacancies, increases several orders of magnitude by heating.

The enhanced signal decayed in the dark with the time constant similar to T_1^{bulk} . The slow relaxation suggested that the paramagnetic sites were sparse in the salt although the Cs metal coexisted in the optical pumping cells. Once the salt was cooled in the dark, the ion conductivity was too small to transport the nuclear polarization to the surface of macroscopic particles. As a result, the time T_1 was switched by laser such that the nuclei were quickly polarized in the entire crystallites with the help of the ion movement, and that the enhanced polarization slowly decayed in the dark because the spin diffusion was determined by the nuclear dipole interaction in the cooled salts.

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