Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supporting Information

Long-range Li ion diffusion in NASICON-type $Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$ (LAGP) studied by ^{7}Li pulsed-gradient spin-echo NMR.

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1. Estimation of the eddy current effects (Figs. S1, S2 and S3).

4. Fig. S4 (a) ^{31}P and (b) ^{27}Al spectra of LAGP at 30 ºC.

5. Fig. S5 Wide-range ^{7}Li NMR spectrum of the LAGP at 30 ºC.

6. Fig. S6 Echo attenuation spectra in real mode with Δ = 15 ms, g = 14.3 Tm^{-1} and δ varied from 0.1 to 4 ms (40 points).

7. Fig. S7 Echo attenuation spectra in real and magnitude modes with Δ = 15 ms, g = 10.0 Tm^{-1} and δ varied from 0.1 to 4 ms (40 points).

8. Fig. S8 The echo attenuation plots on Δ = 10 ms and g = 4.2 to 18.1 Tm^{-1} (six plots) at 38 ºC

Estimation of eddy current effects.

The eddy current effects were estimated by the pulse sequence in Fig. S1

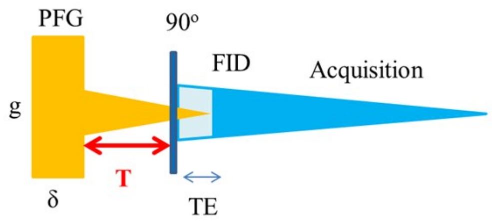

Fig. S1. The pulse sequence to estimate the eddy current effects. The interval time *T* between PFG and 90 ºpulse was set variable.

Using a test sample of the garnet LLZO-Ta ($Li_{6.6}La_3Zr_{1.6}Ta_{0.4}O_{12}$, Ref. 35), ^{7}Li spectra were measured at 78.45 MHz at 25 ºC. The ^{7}Li 90º pulse width was 11.0 μs. Examinations were made by several measuring conditions, and an example is shown in Fig. S2 with g = 10.0 Tm^{-1}, δ = 2 ms and the T was varied from 0.1 to 0.4 ms. When T was very short, the signal was small and its shapes were distorted. In the T longer than 0.24 ms, the signals were recovered to the pattern without the PFG.

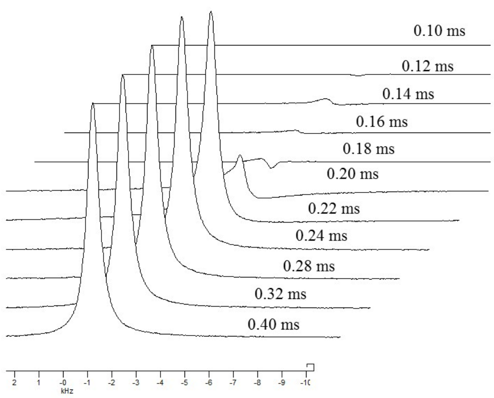

Fig. S2. ^{7}Li spectral patterns for TE = 0.1 ms, δ = 2 ms, g = 10.0 Tm^{-1}, and T = 0.10 to 0.40 ms.

For various g values from 2 to 14.3 Tm^{-12}, the peak heights depending on T are shown in Fig. S3.

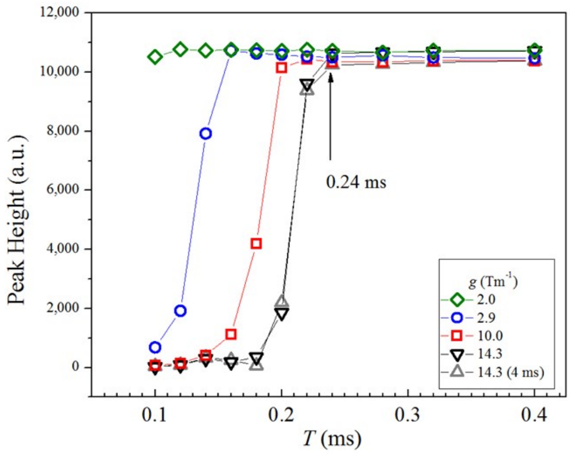

Fig. S3. The peak heights are plotted versus *T* for various *g* values.

When the *g*-value was small, the recovery time was short. For $g = 14.3\ \mathrm{Tm^{-1}}$, the longer recovery time was required. When the PFG width δ was changed between 2 and 4 ms, the recovery time was almost the same and insensitive to δ. We found that after strong *g* imposition of $14.3\ \mathrm{Tm^{-1}}$, the recovery time was found to be longer than 0.35 ms including the interval between 90 º pulse and acquisition (TE, 0.1 ms) by our PGSE NMR machine. This short recovery time (0.35 ms) is accomplished by the good rectangular shape of the PFG shown in Supplementary Material 2 in Ref. 36 and a wide-bore SCM without a room-shim unit. The conditions have been fulfilled throughout our diffusion measurements.

The pulse sequence in Fig. S1 is used to check the eddy current effects in MRI and TE is one of variable parameters to estimate shapes of the eddy current.

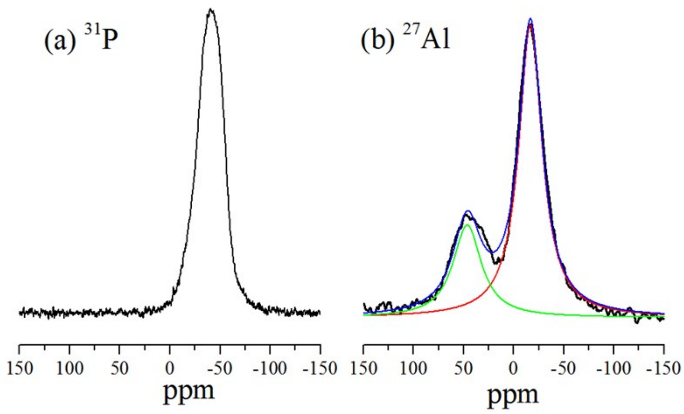

Fig. S4. (a) ^{31}P and (b) ^{27}Al spectra of LAGP at 30 ºC. The ^{31}P and ^{27}Al shifts were referred to H_2PO_4 and $AlCl_3$ in aqueous solutions, respectively.

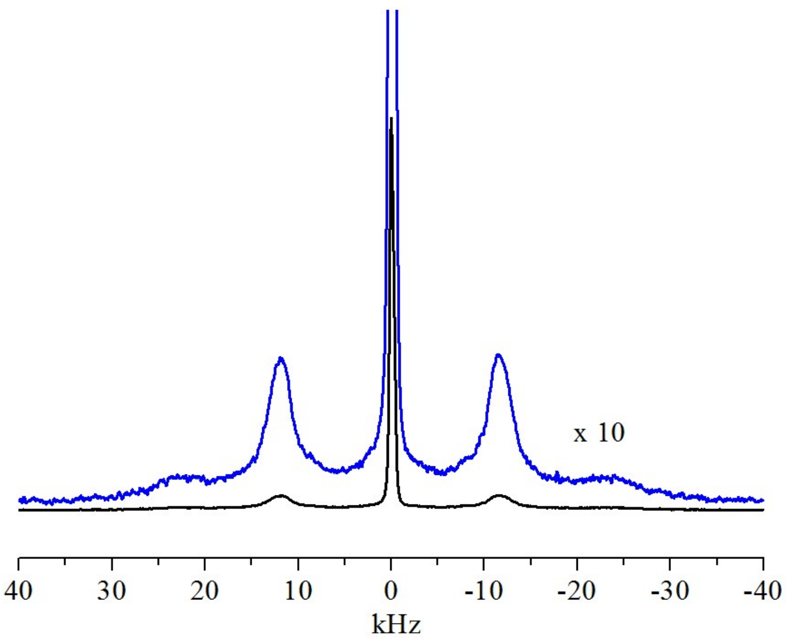

Fig. S5. ^{7}Li NMR spectrum in the wide range for the LAGP at 30 ºC.

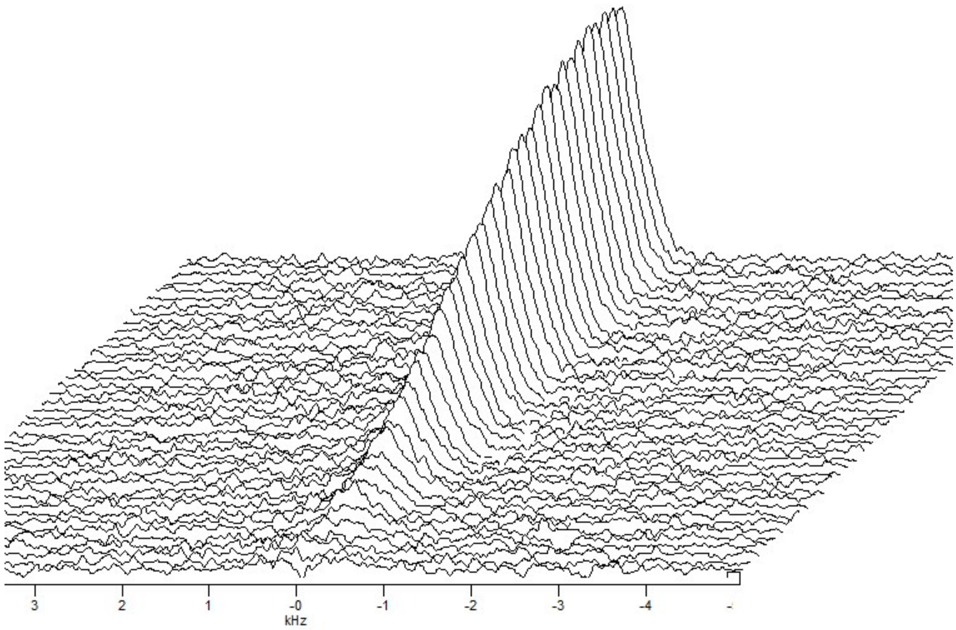

Fig. S6. Echo attenuation spectra of the LAGP at 38 ºC in real mode measured with Δ = 15 ms, ***g* = 14.3 Tm^{-1}**, δ = 0.1 to 4 ms (40 points), and τ = 4.5 ms.

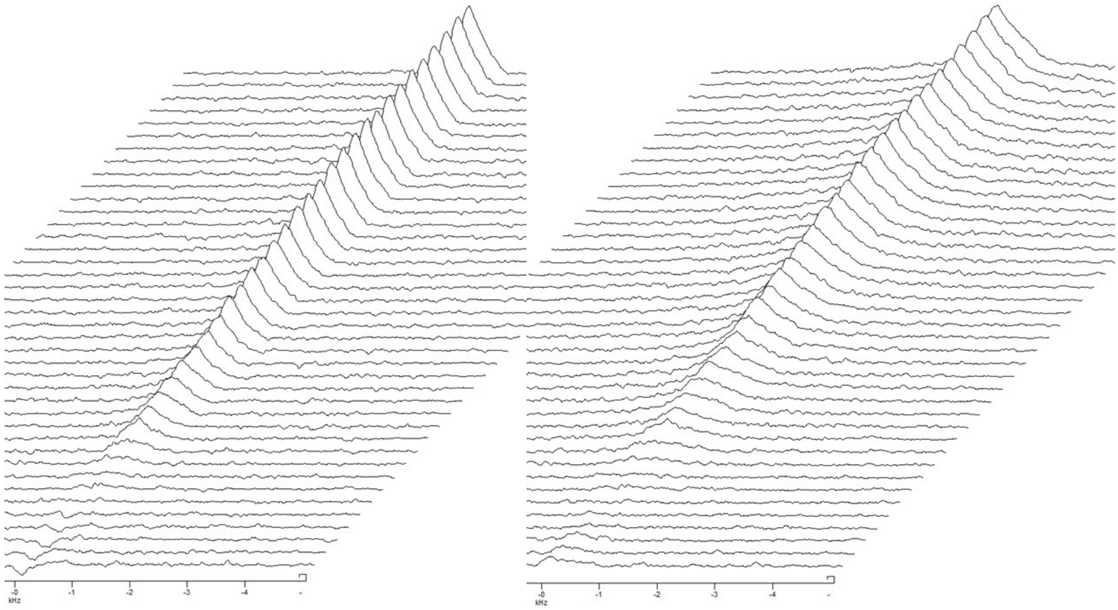

Fig. S7. Echo attenuation spectra of the LAGP at 38 ºC measured with Δ = 15 ms, ***g* = 10.0 Tm^{-1}**, δ = 0.1 to 4 ms (40 points), and τ = 4.5 ms. **Left**: real mode and **right**: magnitude mode. The echo signals changed their phases near the diffraction and the peak heights of the negative signals disappear in logarithmic plots. The signals with the magnitude mode were used for the echo attenuation plots in Figs. 4, 8, and S8.

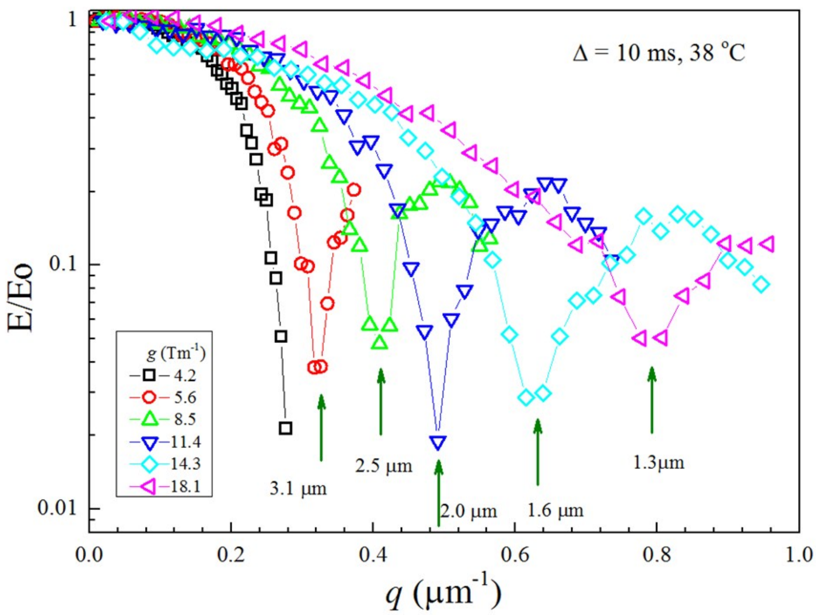

Fig. S8. The echo attenuation plots on Δ = 10 ms at 38 ºC measured with a fixed *g*-value and varying δ = 0.1 to 4.0 ms (40 points) for *g* = 18.1 (left-triangle), 14.3 (diamond), 11.4 (down-triangle), 8.5 (up-triangle), 5.6 (circle) and 4.2 Tm^{-1} (square). The measurements were also performed for *g* = 12.9, 10.0, and 7.1 Tm^{-1}.