| x | 1 ‰ | 5 ‰ | 1 % | 5 % |
|----------------|---------|---------|---------------------|---------|
| $B_0 = 0.13 T$ | 5.7 104 | 8.1 104 | 4.6 10 ⁵ | 1.2 106 |
| $B_0 = 0.24 T$ | 3.3 104 | 1.1 105 | 3.4 105 | 8.1 106 |

Table 1-SI. Coefficient (in s⁻¹K⁻¹) of the linear fit of the variation of $\sqrt{\rho_1 \rho_2}$ with temperature in La_{1-x}Gd_xPO₄ for four gadolinium concentrations and two measuring fields.

| $T(\mathbf{K})$ | 12 | 15 | 20 | 30 |
|---------------------|---------------------|---------|---------|----------|
| $B_2 (\mathrm{mT})$ | ~ 0.1 | 0.6 | 3 | 18 |
| $\rho_2(s^{-1})$ | $\sim 6.5 \ 10^{6}$ | 3.8 107 | 1.9 108 | 1.15 109 |
| τ_2 (ns) | ~ 160 | 26 | 5.3 | 0.89 |

Table 2-SI. Temperature broadening of the ESR line of LaPO₄:Nd 1‰ determined by fitting the lines by the convolution of the 8 K line by a Lorentzian line of width B_2 . The transverse relaxation rate (ρ_2) and time (τ_2) are related to the broadening width B_2 by $\rho_2 = 1/\tau_2 = \gamma_{Nd} B_2$.

| <i>T</i> (K) | 4.6 | 5.5 | 8.0 | 10.0 | 12.0 |
|---|---------------------|----------|---------------------|---------|---------|
| $B_{\rm s}({\rm mT})$ | 6 10-4 | 1.2 10-3 | 0.010 | 0.047 | 0.084 |
| $\sqrt{\rho_1 \rho_2}$ (s ⁻¹) | 3.9 10 ⁴ | 7.3 104 | 6.4 10 ⁵ | 3.0 106 | 5.3 106 |
| $\sqrt{\tau_1 \tau_2}$ (ns) | 2.6 104 | 1.4 104 | 1.6 10 ³ | 330 | 190 |

Table 3-SI. Saturation of the ESR line of LaPO₄:Nd 1‰. The saturation field B_s is determined by fitting the variation of the intensity of the ESR signal at 0.23 T as a function of the microwave power using Eq. (1). The longitudinal and transverse relaxation rates (ρ_1 , ρ_2) and times (τ_1 , τ_2) are related to B_s by $\sqrt{\rho_1 \rho_2} = 1/\sqrt{\tau_1 \tau_2} = \gamma_{Nd} B_s$.

Figure Caption

Figure 1-SI. Variation of the ³¹P NMR signal intensity without MAS rotation in $La_{1-x}Nd_xPO_4$ for *x* varying from 0 to 10%. The broken line is the linear adjustment.

Figure 2-SI. Details of the ESR spectra of LaPO₄:Gd at 20 K and 30 dB for three Gd^{3+} concentrations: 1‰ (a), 1% (b) and 5% (c). The 1% and 5% spectra were fitted (broken red lines) by the convolution of the 1‰ spectrum with Lorentzian lines 2.5 mT and 16 mT linewidths, respectively.

Figure 3-SI. (a) Temperature variation of the main part of the ESR spectrum of $La_{0.999}Nd_{0.001}PO_4$ at 20 dB. (b) Fit of the 20 K spectrum by the convolution of the 8 K spectrum with a lorentzian line of 3 mT linewidth.



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