Supporting Information Available

Table A. Refined Rietveld fit parameters and isotropic thermal parameters for $SrRAlO_4$ (R = Sm, Nd, La)

	$SrSmAlO_4$	$SrNdAlO_4$	$SrLaAlO_4$
Number of Reflections	145/2	152/2	153/2
Refined Variables	25	28	25
Volume ($Å^3$)	170.8	173.16	178.36
Al B_{iso} ($Å^2$)	0.589(66)	0.818(68)	0.752(59)
(Sr,R) B_{iso} ($Å^2$)	0.437(15)	0.492(15)	0.570(13)
O(1) B_{iso} ($Å^2$)	0.376(110)	0.468(107)	0.559(92)
O(2) B_{iso} ($Å^2$)	1.390(126)	0.739(112)	0.137(90)

Validity of Calculated *Q*×*f* Values Using the Classical Oscillator Model. The Fourier-transform infrared reflectivity measurements (*R*-FTIR) have proven to be largely independent of sample quality, and it can therefore be assumed that for the materials under investigation the phonon parameters defined by R-FTIR are of intrinsic origin.[38] At the room temperature, the $SrLaAlO_4$ single crystal presents *Q*×*f* values of 628,000 GHz (parallel to the *c* axis) and 181,000 GHz (perpendicular to the *c* axis).[11] So the calculated *Q*×*f* value of the $SrLaAlO_4$ ceramic are expected larger than 237,000 GHz (1/[(1/3/628000)+(2/3/181000)] ≈ 237000). Our current calculated *Q*×*f* value of $SrLaAlO_4$, 150,000 GHz, is not high enough. Such a great deviation should mainly attribute to the unjustified extrapolation down from the optical frequency range to microwave frequency range using the classical damped harmonic oscillator model.[38,39] Another reason may be that the anisotropy of *R* is averaged out in ceramic samples which might lead to effective broadening of some modes not corresponding to real mode damping. The more justified method is using the microscopic theory of intrinsic losses in dielectrics [40] to obtain a dielectric loss formula which is applicable within the frequency range of

$\omega \ll \Omega_{TO}$ (Ω_{TO} is the phonon frequency of the lowest-frequency transverse optical mode).[38-39] But such a method is too complicated for the materials under consideration, which are not centrosymmetric cubic (pseudo-cubic) crystals. Fortunately, the microscopic theory yields loss values $\mu\left(\varepsilon_0/\varepsilon_\infty\right)^x$ times higher than the classical oscillator model, where μ is the anharmonic factor and is of the order around 0.01, ε_0 the static permittivity, ε_∞ the permittivity at optical frequency, x lies within the range $1/2<x<3$.[38] The materials under investigation have ε_0 around 18 and ε_∞ around 4, so that $\mu\left(\varepsilon_0/\varepsilon_\infty\right)^x$ keeps nearly constant. For $SrRAlO_4$ (R=Sm, Nd, La), the trend predicted by the classical oscillator model are expected to be similar to that predicted by the microscopic theory given that the effects of the anisotropy on the dispersion parameters are almost the same for $SrRAlO_4$ (R=Sm, Nd, La).

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