

# Synthesis and THz properties of rare earth double molybdate $\text{KLu}(\text{MoO}_4)_2$

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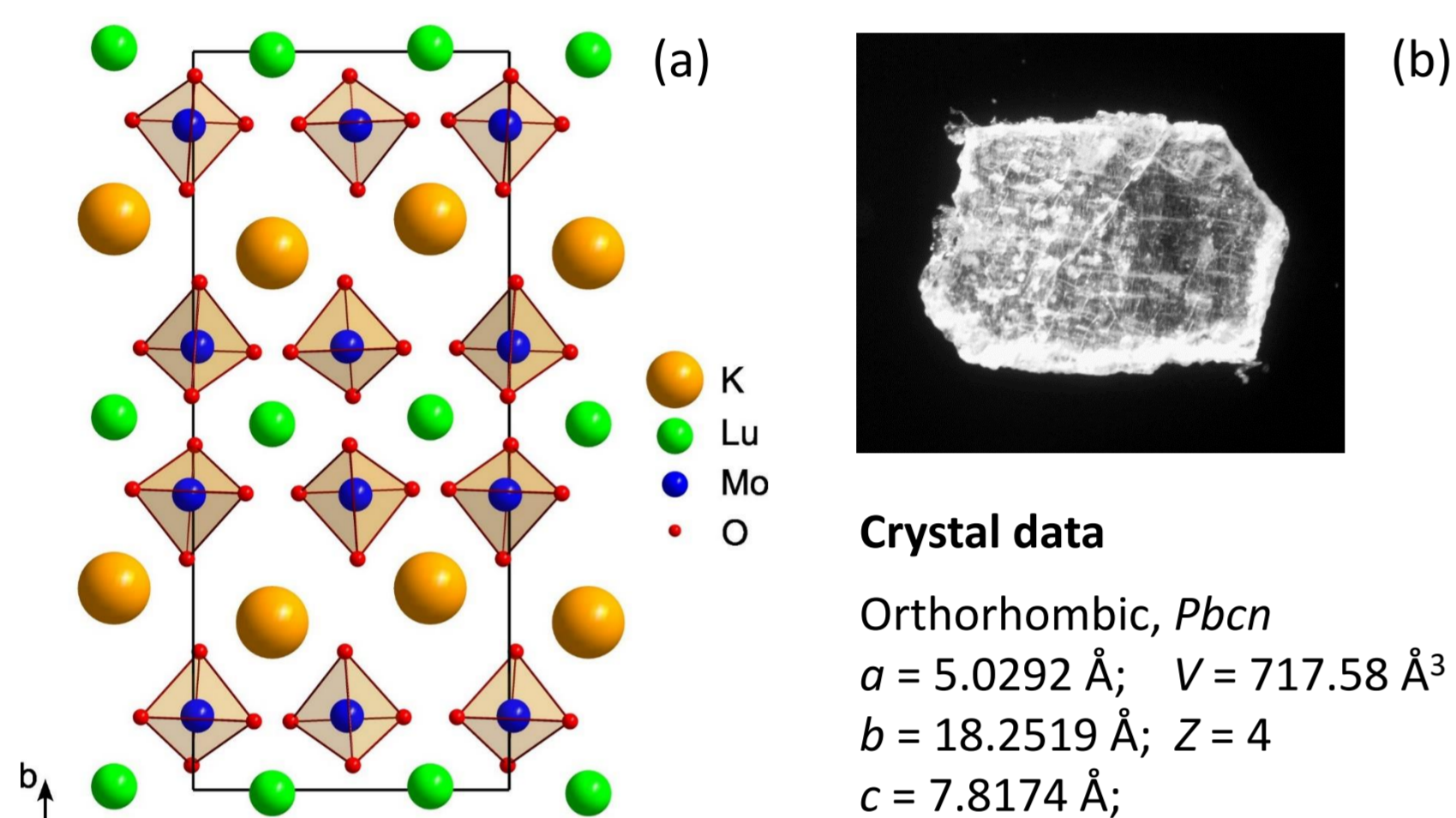
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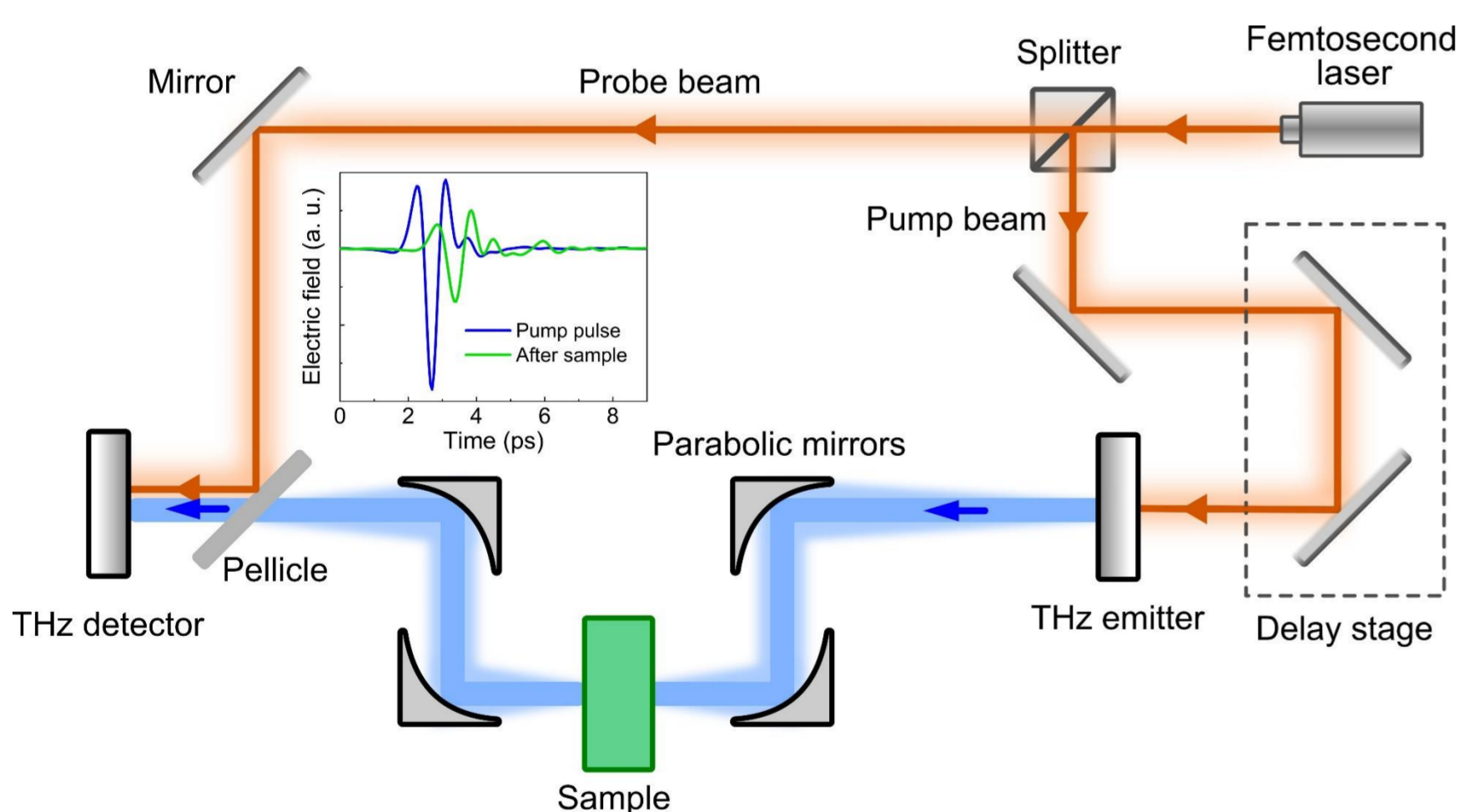
## Introduction

Recently, we have demonstrated that in the dielectric compound  $\text{KY}(\text{MoO}_4)_2$ , low-frequency optical phonons excited by broadband THz pulse exhibit long-lasting (lifetime over 100 ps) narrowband re-emission of THz electromagnetic radiation [1]. Similar effect has been found in other compounds of  $\text{KRe}(\text{MoO}_4)_2$  family including compounds with magnetic lanthanide ions ( $\text{Re}^{3+}$ ). Here, we investigate the THz properties of another non-magnetic compound of this family,  $\text{KLu}(\text{MoO}_4)_2$ , with heaviest lanthanide ion  $\text{Lu}^{3+}$ . The absence of magnetic contributions together with mass-related effects influences the lattice dynamical parameters and, consequently, the frequencies of the low-frequency phonons involved in the re-emission process.

## Sample and method

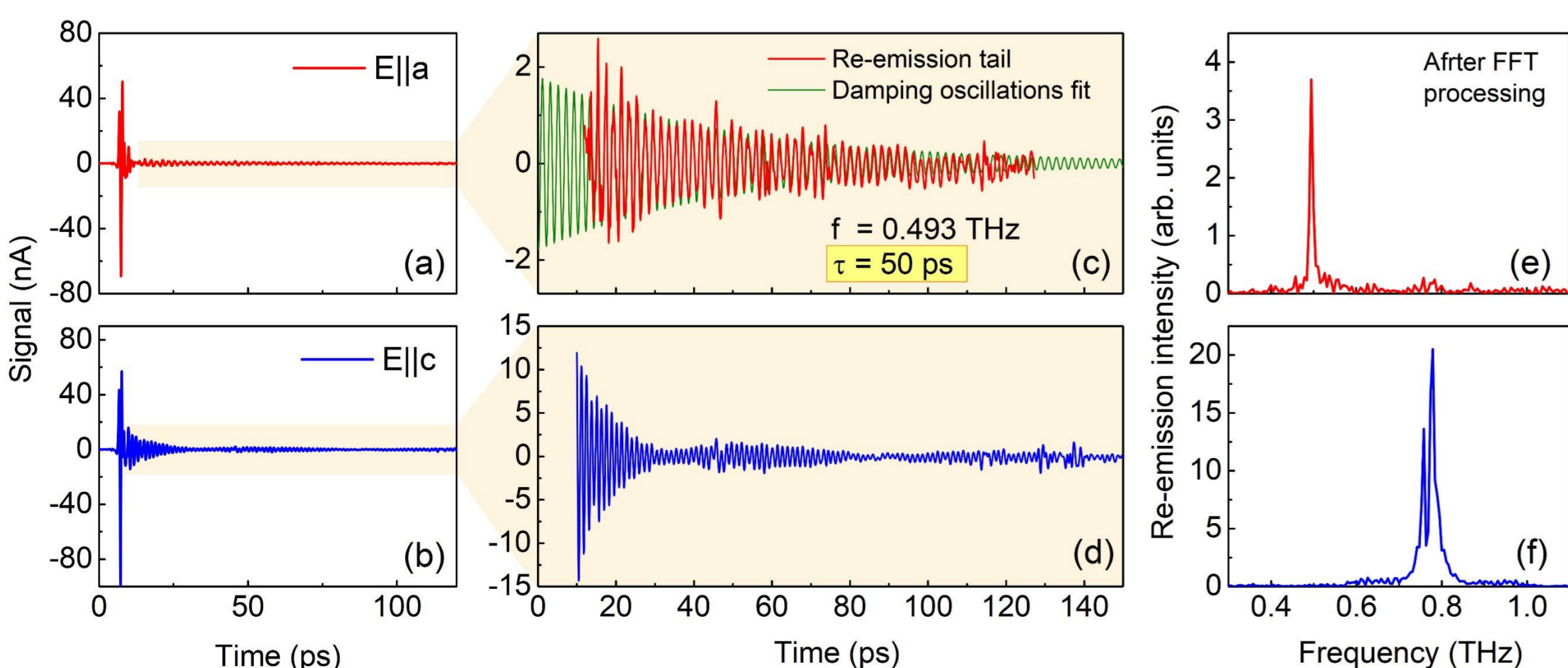


**Figure 1.** (a) Crystallographic structure of  $\text{KLu}(\text{MoO}_4)_2$  at  $T = 100 \text{ K}$  [data from S. Chong et al, *Acta Crystallogr., Sec.E* 76, 1871 (2020)]; (b) The photo of single crystal sample.



**Figure 2.** Schematic diagram of a representative THz-TDS system. The insert shows time diagram of the THz pulse with and without the sample.

## Re-emission spectra



**Figure 5.** (a, b) The waveforms of the THz pulse after passing through the  $60 \mu\text{m}$  thick sample for polarizations along  $a$  (red) and  $c$  (blue) axes (measurements are done at  $T = 5 \text{ K}$ ). (c, d) Zoomed waveforms for the  $a$  (red) and  $c$  (blue) polarizations 10 ps after the start of the pulse. Damping oscillations were fitted with equation  $E = A_0 \exp(-t/\tau) \cdot \cos(\omega t + \varphi)$ . (e, f) FFT of the waveforms is shown in (c) and (d), respectively.

## THz transmittance

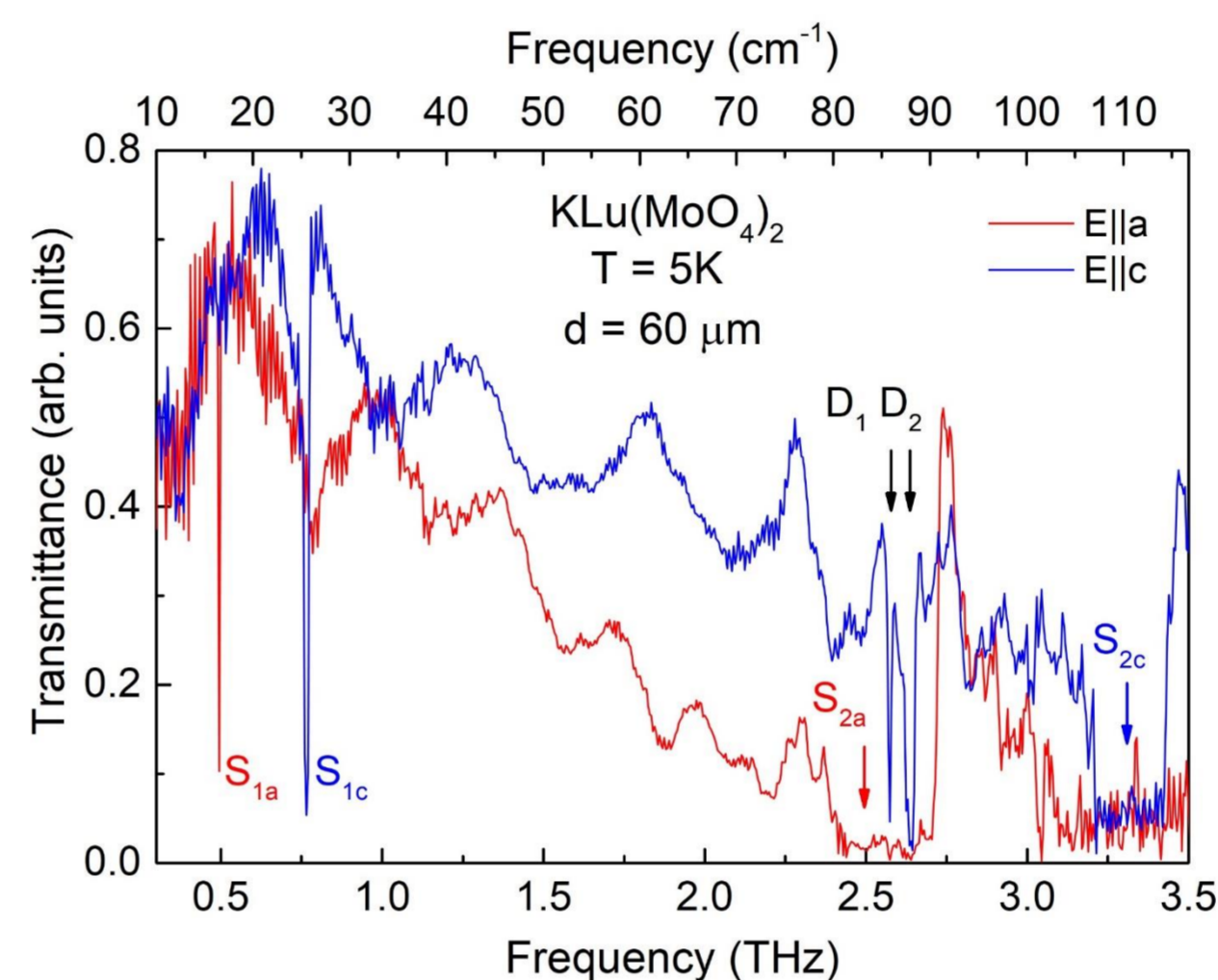
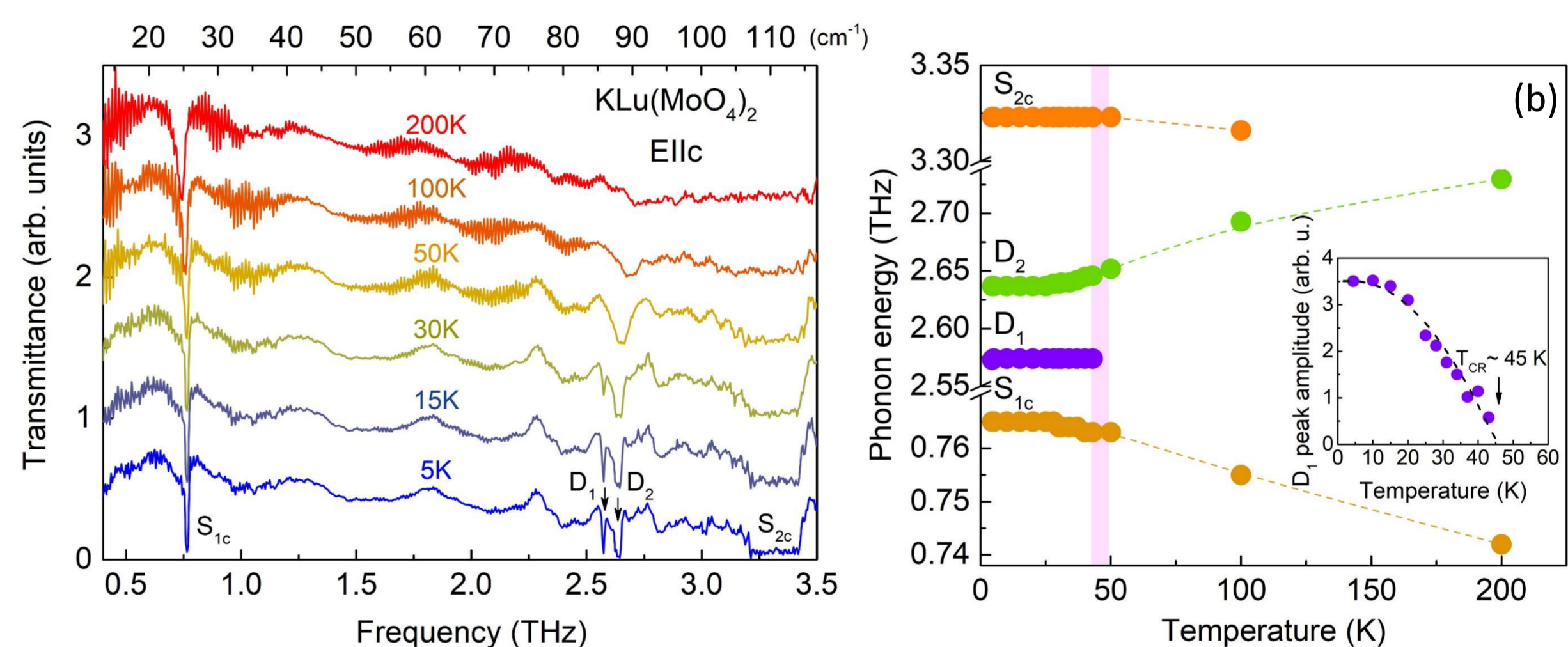


Table I. The frequency of optical phonons in rare-earth molybdate compounds  $\text{KRe}(\text{MoO}_4)_2$ .

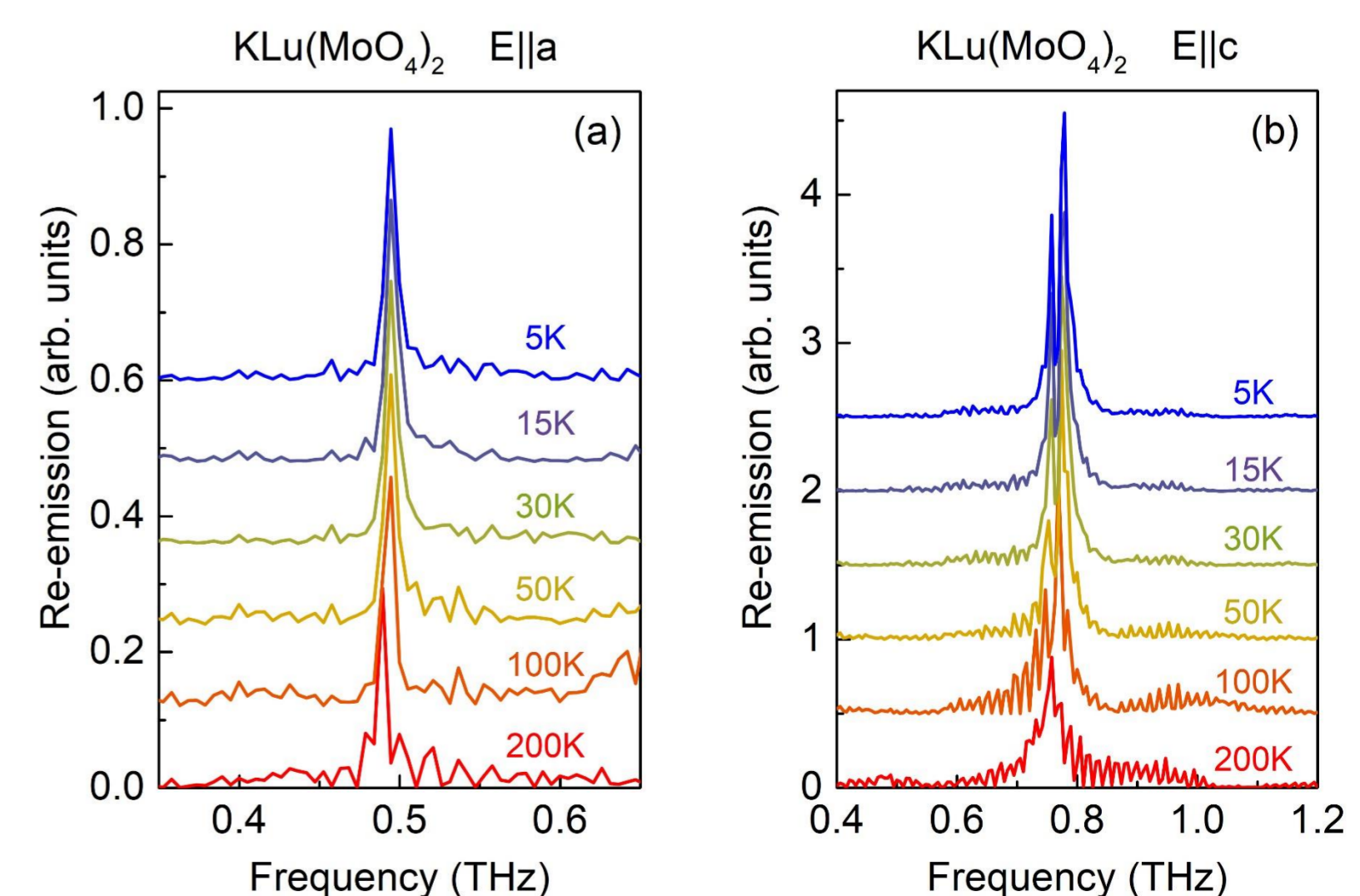
Compound	E  a, THz		E  c, THz	
	$S_{1a}$	$S_{2a}$	$S_{1c}$	$S_{2c}$
$\text{KY}(\text{MoO}_4)_2$	0.568	2.578	0.860	3.358
$\text{KDy}(\text{MoO}_4)_2$	0.525	2.548	0.794	3.238
$\text{KEr}(\text{MoO}_4)_2$	0.510	2.548	0.779	3.208
$\text{KTm}(\text{MoO}_4)_2$	0.501	2.458	0.764	3.268
<b><math>\text{KLu}(\text{MoO}_4)_2</math></b>	<b>0.493</b>	<b>2.476</b>	<b>0.755</b>	<b>3.304</b>

Data for  $\text{Re} = \text{Y}^{3+}, \text{Dy}^{3+}, \text{Er}^{3+}, \text{Tm}^{3+}$  was taken from: *Journal of Physics: Condensed Matter*, 2017, 29, 095402.

**Figure 3.** Transmittance spectra of  $\text{KLu}(\text{MoO}_4)_2$  at 5K for polarizations E||a and E||c (red and blue curves).  $S_1$  and  $S_2$  modes corresponds to shear lattice vibrations and the doublet ( $D_1, D_2$ ) – rotational vibrations of the  $(\text{MoO}_4)^{2-}$  tetrahedra.



**Figure 4.** (a) Temperature evolution of the transmittance spectra of  $\text{KLu}(\text{MoO}_4)_2$  for E||c. (b) Temperature behavior of the phonon modes energy. Inset shows the temperature dependence of the  $D_1$  mode peak amplitude.



**Figure 6.** Temperature evolution of the re-emission spectra (FFT after a 10 ps delay from the pump pulse front) for the  $60 \mu\text{m}$  thick  $\text{KLu}(\text{MoO}_4)_2$  sample at E||a (a) and E||c (b) polarizations.