Monochromatic THz emission by optically excited lattice vibrations in K*Re*(MoO_4)$_2$

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Last decades significant achievements in application of the photo-Dember effect boost the Time-Domain Terahertz Spectroscopy (THz-TDS). While this broad-band THz light generation utilizes the ultrafast formation of a dipole due to the difference in mobilities of holes and electrons in semiconductors, excitation of lattice vibrations in solid dielectrics also induces an electric dipole with light emitting capability, which may provide opportunities for generation of monochromatic THz radiation. Here, we demonstrate the emission of monochromatic terahertz electromagnetic radiation by coherent lattice vibrations via ultrafast excitation of optical phonons in dielectric materials K*Re*(MoO_4)$_2$ (where *Re* stands for a rare-earth or Y^{3+} ion). Layered structure of K*Re*(MoO_4)$_2$ compounds leads to infrared-active shear lattice vibrations with the energies below 3 meV [1], which corresponds to the frequencies below 700 GHz where solid state monochromatic radiation sources are rare.

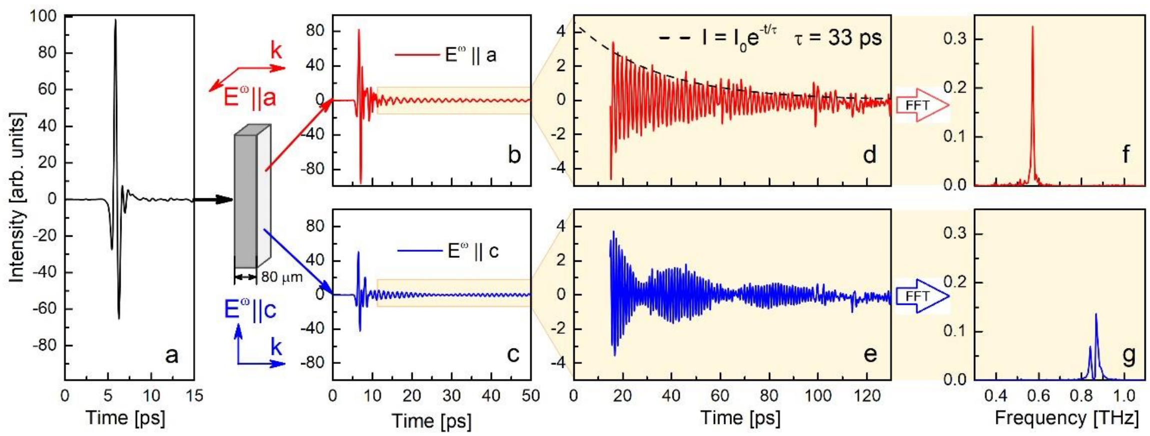

Figure 1. (a) The electric field waveform of the THz pulse before the sample. (b, c) Waveforms of the THz pulse passed the sample for polarisations along *a* (red) and *c* (blue) axes ($E^\omega \parallel$ a and $E^\omega \parallel$ c, respectively). (d, e) Zoomed waveforms for the *a* (red) and *c* polarisations 10 ps after the start of the pulse. (f, g). Squared FFT of the waveforms shown in d and e, respectively.

Our findings are summarized in the Figure 1. Coherent infrared-active optical phonons in KY(MoO_4)$_2$ are excited by a broadband THz pulse (Fig.1a) and re-emit monochromatic THz radiation with the dipole decay time of 33 ps (Fig. 1d,e). Such slow decay is an exception for the oscillators in solids with frequencies below 1 THz. By studying dependencies of the THz-TDS spectra on the sample temperature and thickness, we show that the beating observed in Fig.1c is due to a strong absorption for the radiation polarized along the c axis ($E^\omega \parallel c$).

Long lifetime of the coherent emission allows for the detection of more than 50 periods of radiation with frequency of 568 GHz for $E^\omega \parallel a$. Thus, the remarkably long decay time of the optical phonons in K*Re*(MoO_4)$_2$ together with the chemical stability of these compounds suggest a variety of possible applications of such class of dielectrics in THz technology.

References

[1] S. Poperezhai, P. Gogoi, N. Zubenko, K. Kutko, V. I. Kutko, A. S. Kovalev and D. Kamenskyi. “Terahertz lattice dynamics of the potassium rare-earth binary molybdates” J. Phys.: Condens. Matter **29**, 095402 (2017).