ISSN 0030-400X, Optics and Spectroscopy, 2020, Vol. 128, No. 10, pp. 1583–1587. © Pleiades Publishing, Ltd., 2020.
Russian Text © The Author(s), 2020, published in Optika i Spektroskopiya, 2020, Vol. 128, No. 10, pp. 1473–1477.

SPECTROSCOPY OF CONDENSED MATTER

On the Transparency of Alkali-Halide Crystal in the Terahertz Spectral Range

I. A. Kaplunov[a, **], G. I. Kropotov[b, *], V. E. Rogalin[c, *], and A. A. Shakhmin[b]**

[a] Tver State University, Tver, 170100 Russia
[b] Tydex LLC, St. Petersburg, 194292 Russia
[c] Institute of Electrophysics and Electric Power, Russian Academy of Sciences, St. Petersburg, 191186 Russia
*e-mail: grigorykropotov@tydex.ru
**e-mail: kaplunov.ia@tversu.ru
***e-mail: v-rogalin@mail.ru

Received April 7, 2020; revised May 19, 2020; accepted June 16, 2020

Abstract—The transmission of THz (to 3000 μm) radiation by alkali halide single crystals of sodium chloride (NaCl), potassium chloride (KCl), potassium bromide (KBr), and rubidium iodide (RbI), which are widely used in the IR spectral region, is studied. The spectral dependences of the absorption coefficients of these materials in the range of 0.9–3000 μm are determined. These materials are transparent in the range of 1000–3000 μm, which makes it possible to use them in terahertz (millimeter) devices.

DOI: 10.1134/S0030400X20100136

INTRODUCTION

Terahertz (THz) radiation takes an intermediate position between infrared and radio waves and is of considerable interest for a number of fundamental (chemistry, physics, astronomy) and applied fields. The THz range contains the emission spectra of astronomical objects and complex organic molecules, including molecules of explosives, toxins, narcotics, air pollutants, proteins, and DNA. Many materials, including textile, plastic, and biological tissues, are transparent for THz waves. Due to the low photon energy, THz radiation causes no injury typical for ionizing radiations [1–3], owing to which it is used in medical diagnostics and for non-destructive quality control and/or protection of materials.

Until recently, the technologies of generation, conversion, and recording of THz radiation have been developed weaker than those of radiation in other regions. Because of this, this spectral range even got the name "terahertz gap" [1]. This situation is related to the fact that THz frequencies are too high for efficient application of radio-frequency generators but too low for application of infrared sources.

However, the THz region has been extensively studied in recent years [4, 5]. Narrow but quite suitable for use THz transmission bands are found in the spectrum of the atmosphere [1–3]. Various sources of THz radiation, including lasers, have been developed and have already demonstrated high powers (up to 10^9 W per pulse) [6–11]. Rather sensitive THz detectors are also developed [1–3]. Nevertheless, progress in the use of the THz region is slow due to a significant lack of high-quality optical materials transparent in the THz region [12–14]. Unfortunately, the available THz-transparent materials have significant drawbacks. A comparatively new optical material—polycrystalline diamond [15–17]—stands out from them. However, the high cost and treatment problems in many cases still hinder its wide application.

In practice, it is often necessary to use an optical material transparent not only in the THz but also in other spectral regions. This is important, for example, for two-range thermal imagers of the IR + THz spectral region [18, 19]. In addition, optical pumping of THz lasers is usually performed by CO_2 lasers emitting in the range of 9–11 μm [8–10], because of which it is often necessary to use optical elements transparent in both ranges.

In the present work, we study the transmission and absorption of alkali halide single crystals (AHCs) sodium chloride (NaCl), potassium chloride (KCl), potassium bromide (KBr), and rubidium iodide (RbI) in the THz region. This work continues our studies [12, 20] of optical materials for the THz region.

EXPERIMENTAL

The crystals studied in this work have a low refractive index, which is extremely important for applications in the THz region, in which it is almost impossible to use traditional antireflection coatings for optical elements [21]. High Fresnel reflection losses strongly affect the operation of optical systems. However, there

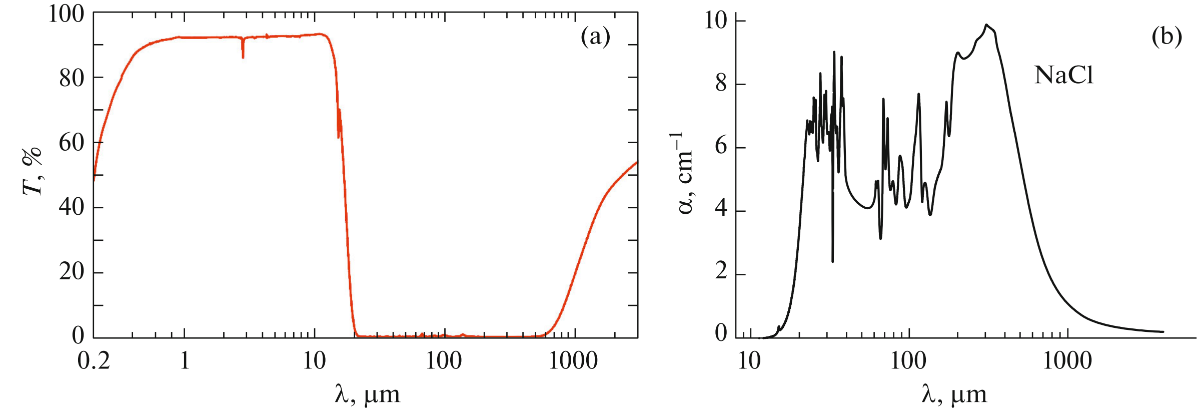

Fig. 1. (a) Transmission and (b) absorption coefficient spectra of a NaCl crystal (thickness 10.15 mm).

exists the possibility to partially reduce the reflection of optical elements for the THz region. It is known that silicon and quartz elements can be antireflection coated with polyethylene and parylene films [22].

Due to poor mechanical properties, the crystals studied in the present work are sensitive to the operation conditions, namely, they are fragile and easily cleaved along cleavage planes (⟨100⟩ planes). These crystals are soluble in water and can be used only under low-humidity conditions. In some cases, AHCs are coated with special moisture-protective coatings [23]. These materials were almost not used previously in the THz region. Owing to good optical quality, AHCs are widely used in IR devices, and we used for investigation standard crystals industrially grown by the Kyropoulos method from melt in air.

The spectral transmission was measured using a Photon RT spectrophotometer (Essent Optics) and Bruker Vertex 70 Fourier spectrometer in the spectral range of 0.185–670 μm. The absolute wavelength error for the Photon RT spectrophotometer in the range of 185–1700 nm was 1 nm, and the error in determination of the wave number with the Bruker Vertex 70 spectrometer was 0.3–0.5 cm^{-1}. The measurements in the range of 100–3000 μm were performed using a Tera K8 spectrometer (MenloSystems). It should be noted that this spectrometer allows quantitative measurements in the range of 100–1500 μm and only qualitative measurements in the range of 1500–3000 μm. The error of measurements of transmission coefficients was ~0.5% for all spectrometers.

The absorption (attenuation) coefficients α were calculated using a standard method taking into account multiple reflections by a known formula [24, 25]. The wavelength dependences of the refractive index in the considered spectral ranges were determined using data from [26–29].

The absolute calculation error Δα can be found using the following formula known from the error theory:

$$\Delta f = \sum_{i=1}^{b} \Delta\alpha_i \left| f'_{x_i}(a_1 \ldots a_k) \right|, \qquad (1)$$

where Δf is the absolute error of the function, a_i is the ith argument of the function, and Δa_i is the absolute error of this argument. As applied to the formula for absorption (attenuation) coefficients α, we obtain

$$\Delta\alpha = \frac{\Delta h}{h^2}\left|\ln\frac{(1-R)^2+\sqrt{(1-R)^4+4T^2R^2}}{2T}\right| + \frac{2\Delta R}{h}\left|\frac{(1-R)\sqrt{(1-R)^4+4T^2R^2}+(1-R)^3-2RT^2}{[(1-R)^2+\sqrt{(1-R)^4+4T^2R^2}][\sqrt{(1-R)^4+4T^2R^2}]}\right| + \frac{2\Delta T}{hT}\left|\frac{2R^2T+4T^2R^2+(1-R)^4}{[(1-R)^2+\sqrt{(1-R)^4+4T^2R^2}][\sqrt{(1-R^4+4T^2R^2)}]}\right|, \qquad (2)$$

where ΔR and ΔT are the absolute errors of reflection and transmission coefficients, respectively.

EXPERIMENTAL RESULTS AND DISCUSSION

The results of the present work show that the halides of alkali and alkaline-earth metals can be also used in the millimeter spectral range. Figures 1–5 show the measured transmission spectra and the calculated attenuation coefficient spectra of NaCl, KCl, KBr, and RbI single crystals in the range from near-IR to 3000 μm. The transmission of these crystals in the UV, visible, and IR regions are well known. In this work, we present the measured spectra of our samples

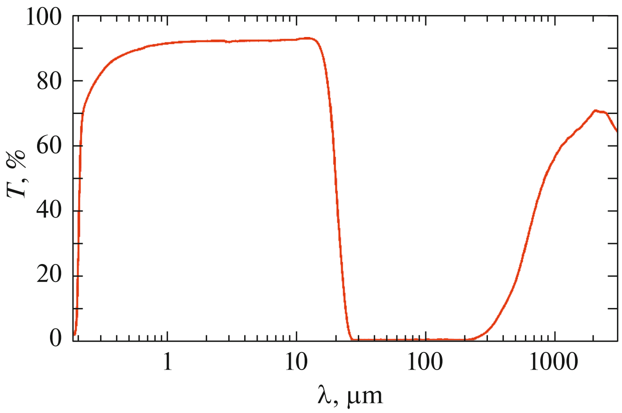

Fig. 2. (a) Transmission spectrum of a NaCl crystal (thickness 2 mm).

to provide the complete pattern of transparency of this group of materials in a wide spectral range.

From Figs. 1–5, one can see the IR transmission band and then the intense absorption caused by phonon processes [27]. It is seen that the phonon absorption edge shifts as a rule to longer wavelengths. These crystals are transparent in the range of 1000–3000 μm and can be used.

Among these crystals, the best transparency in the short-wavelength region of the THz range is demonstrated by RbI (Fig. 5). On the whole, it should be noted that RbI crystal plates are transparent in the range from 0.24 to 64 μm [28] but are rarely used in optics because their hygroscopicity is higher than that of KBr and CsI. These crystals are mainly used in detectors of high-energy particles [30]. The intense absorption band in the range of 7–8 μm observed in Fig. 5 is caused by barium (Ba) impurity specially introduced to improve the scintillation properties of the crystal in other regions [28, 30, 31].

Unfortunately, similar to other crystalline materials [12], transmission losses in AHCs in the millimeter range are considerably higher than in the IR region. This is clearly seen from the transmission spectra of NaCl crystals. Figure 1 shows the transmission spectrum of a sample with a thickness of 10.15 mm, while Fig. 2 presents the spectrum of a NaCl crystal 2 mm

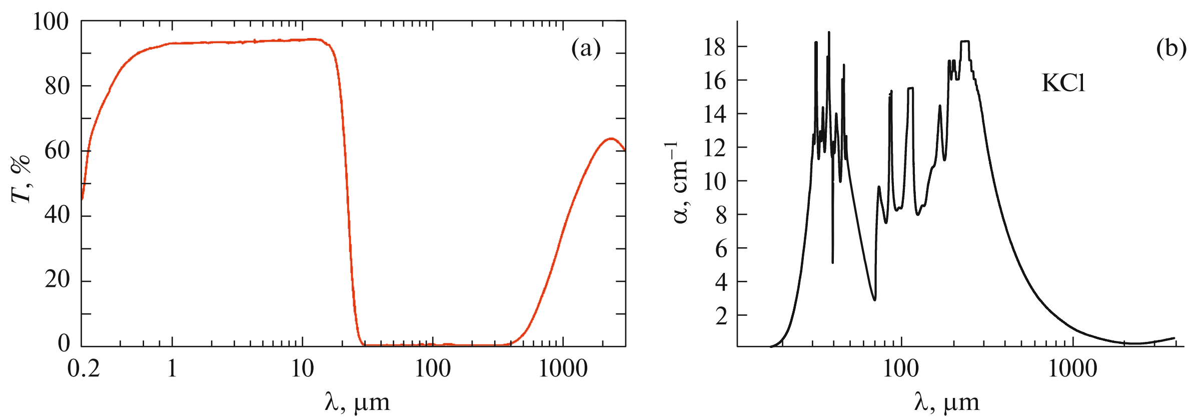

Fig. 3. (a) Transmission and (b) absorption coefficient spectra of a KCl crystal (thickness 6.1 mm).

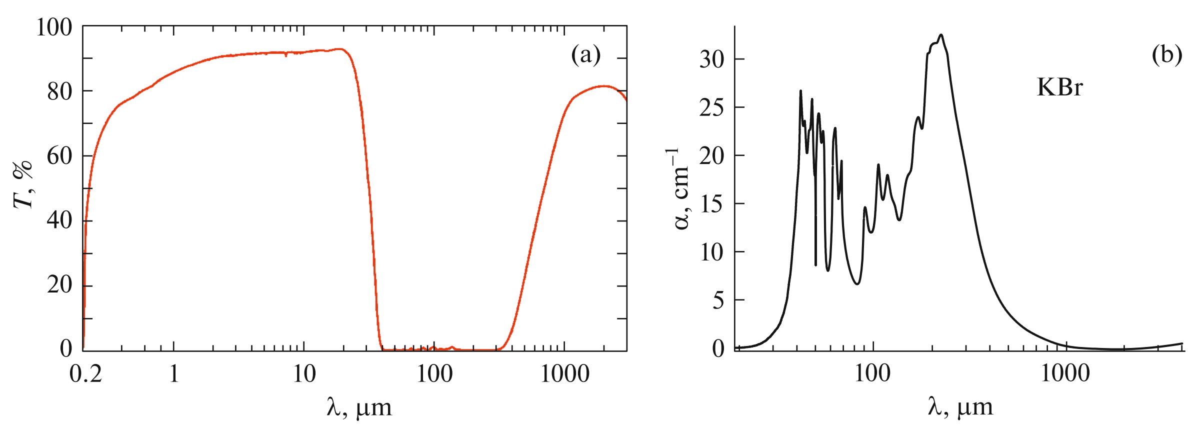

Fig. 4. (a) Transmission and (b) absorption coefficient spectra of a KBr crystal (thickness 3.2 mm).

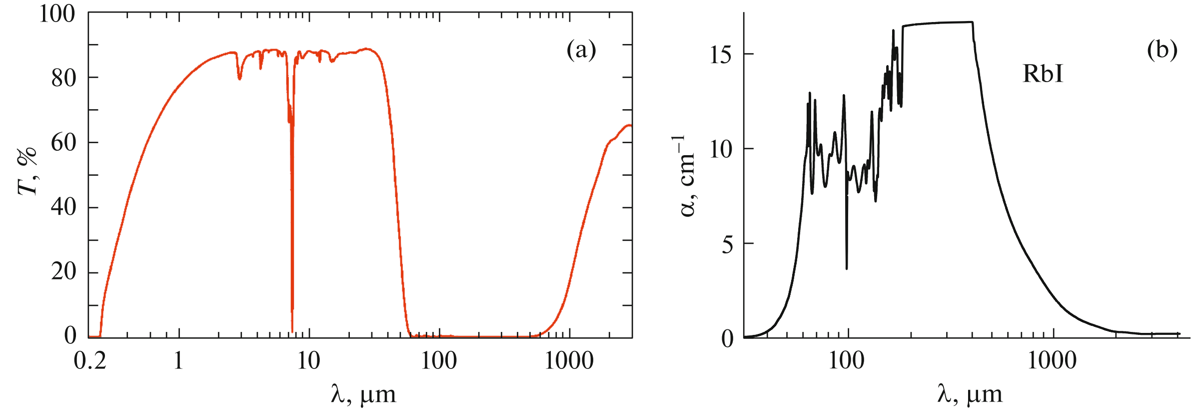

Fig. 5. (a) Transmission and (b) absorption coefficient spectra of a RbI crystal (thickness 6.78 mm).

thick. One can see that the increase in the thicknesses of the samples almost does not affect the absolute absorption in the IR region but noticeably increases absorption in the THz range even in the transmission peak. In particular, in the region of 2000 μm, the thin sample transmits about 70% of radiation, while the thick sample transmits only about 50%. Similar results were obtained for KCl and KBr crystals.

CONCLUSIONS

The transmission coefficient spectra are measured and the absorption coefficient spectra are calculated for alkali halide single crystals NaCl, KCl, KBr, and RbI in the THz range of 30–3000 μm. The IR spectra of these materials are known, and our spectra obtained in the THz region are presented to characterize the complete behavior of transparency of these materials in a wide spectral range. It is found that these crystals have transmission bands in the millimeter range, which makes it possible to use them in devices of this range.

FUNDING

This study was performed using resources of the Tydex LLC and the Center for Collective Use of the Tver State University within State contract no. 3.5786.2017/8.9.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

REFERENCES

1. Xi-Cheng Zhang and Jingzhou Xu, *Introduction to THz Wave Photonics* (Springer, New York, 2010). https://doi.org/10.1007/978-1-4419-0978-7
2. E. Brundermann, H. W. Hübers, and M. F. Kimmit, *Terahertz Techniques* (Springer, Heidelberg, 2012).
3. Y. S. Lee, *Principles of Terahertz Science and Technology* (Springer, New York, 2009).
4. N. N. Rosanov, M. V. Arkhipov, R. M. Arkhipov, A. V. Pakhomov, and I. V. Babushkin, Opt. Spectrosc. **123**, 100 (2017). https://doi.org/10.7868/S0030403417070236
5. N. N. Rosanov, Opt. Spectrosc. **107**, 768 (2009).
6. K. L. Vodopyanov, Opt. Express **14**, 2263 (2006).
7. A. A. Vedenov, G. D. Myl'nikov, and D. N. Sobolenko, Sov. Phys. Usp. **25**, 833 (1982).
8. S. Ya. Tochitsky, Chieh Sung, S. E. Trubnick, Chan Joshi, and K. L. Vodopyanov, J. Opt. Soc. Am. B **24**, 2509 (2007).
9. S. Ya. Tochitsky, J. E. Ralph, C. Sung, and C. Joshi, J. Appl. Phys. **98**, 26101 (2005).
10. D. Haberberger, S. Tochitsky, and C. Joshi, Opt. Express **18**, 17865 (2010).
11. N. A. Vinokurov and O. A. Shevchenko, Phys. Usp. **61**, 435 (2018). https://doi.org/10.3367/UFNr.2018.02.038311
12. V. E. Rogalin, I. A. Kaplunov, and G. I. Kropotov, Opt. Spectrosc. **125**, 1053 (2018). https://doi.org/10.1134/S0030400X18120172
13. V. V. Parshin, Int. J. Infrared Millimeter Waves **15**, 339 (1994).
14. THz Materials. http://www.tydexoptics.com/ru/products/thz_optics/thz_materials/.
15. V. G. Ral'chenko and A. P. Bol'shakov, *Carbon Photonics*, Ed. by V. I. Konov (Nauka, Moscow, 2017) [in Russian].
16. V. E. Rogalin, M. I. Krymskii, and K. M. Krymskii, J. Commun. Technol. Electron. **63**, 1326 (2018). https://doi.org/10.1134/S1064226918110098
17. V. E. Rogalin and S. M. Aranchii, Integral, No. 5 (67), 7 (2012).
18. V. V. Korotaev, G. S. Mel'nikov, S. V. Mikheev, V. M. Samkov, and Yu. I. Soldatov, *Principles of Thermal Imaging* (ITMO, St. Petersburg, 2012) [in Russian].
19. D. G. Manolakis, R. B. Lockwood, and Th. Cooley, *Hyperspectral Imaging Remote Sensing (Physics, Sensors, and Algorithms)* (Cambridge Univ. Press, Cambridge, 2016).

20. I. A. Kaplunov, A. I. Kolesnikov, G. I. Kropotov, and V. E. Rogalin, Opt. Spectrosc. **126**, 191 (2019). https://doi.org/10.1134/S0030400X19030093

21. J. D. Wheeler, B. Koopman, P. Gallardo, P. R. Maloney, S. Brugger, G. Cortes-Medellin, R. Datta, C. Darren Dowell, J. Glenn, S. Golwala, C. McKenney, J. J. McMahon, Ch. D. Munson, M. Niemack, S. Parshley, and G. Stacey, in *Millimeter, Submillimeter, and Far-Infrared Detectors and Instrumentation for Astronomy VII,* Ed. by W. S. Holland and J. Zmuidzinas, Proc. SPIE **9153**, 91532Z (2014).

22. http://www.tydexoptics.com/ru/products/thz_optics/thz_coatings/.

23. http://www.tydexoptics.com/ru/products/coatings/ik-pokrytiya/#ik.

24. M. V. Kurik, J. Appl. Spectrosc. **4**, 199 (1966).

25. I. A. Kaplunov, A. I. Kolesnikov, I. V. Talyzin, L. V. Sedova, and S. L. Shaiovich, J. Opt. Technol. **72**, 564 (2005).

26. H. H. Li, J. Phys. Chem. Ref. Data **5**, 329 (1976). https://doi.org/10.1063/1.555536

27. S. S. Mitra, *Optical Properties of Highly Transparent Solids* (Plenum, New York, London, 1975).

28. I. Ohlídal and D. Franta, in *Handbook of Optical Constants of Solids,* Ed. by E. D. Palik (Elsevier, Amsterdam, 1997), Vol. 3, p. 857. https://doi.org/10.1016/B978-012544415-6.50136-9

29. Refractive Index. INFO. https://refractiveindex.info/about.

30. F. W. Meyer, H. F. Krause, and C. R. Vane, Nucl. Instrum. Methods Phys. Res., Sect. B **205**, 700 (2003). https://doi.org/10.1016/S0168-583X(02)01957-2

31. K. Shunkeyev, A. Lushchik, L. Myasnikova, Sh. Sagimbaeva, Zh. Ubaev, and Z. Aimaganbetova, Low Temp. Phys. **45**, 1127 (2019). https://doi.org/10.1063/1.5125992

Translated by M. Basieva